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Polybrominated diphenyl ethers (PBDEs) in sediments and mussel tissues from Hong Kong marine waters

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Abstract

Sediments and green-lipped mussels, *Perna viridis*, were used to investigate concentrations of polybrominated diphenyl ethers (PBDEs) in Hong Kong's marine environment. PBDEs have been used extensively over the past two decades as flame retardants in polymer additives for a variety of plastics, computers, furniture, building materials, and fabrics. Many measurements of PBDEs in various environmental matrices have been reported from Belgium, Holland, Japan, Europe and North America, but few measurements are available for the southeast Asian region and Hong Kong. PBDE congeners ($n = 15$) were measured in 13 sediments and nine mussel samples, taken from Hong Kong marine waters. The $\sum_{15}$PBDEs in sediments ranged between 1.7 and 53.6 ng g^{-1} dry wt, with the highest concentrations located around the most heavily populated areas of Victoria Harbour and Sai Kung, while the lowest concentrations of $\sum_{15}$PBDEs were found at more remote locations of Sha Tau Kok, Wong Chuk Bay, Castle Peak Bay, and Gold Coast. $\sum_{15}$PBDEs ranged from 27.0 to 83.7 ng g^{-1} dry wt of mussel tissues. Although not identical, most of the congeners in sediments were found in mussel tissues, with BDE-47, BDE-99, BDE-153 and BDE-183 being the most prominent in both matrices. On the basis of a literature survey, the concentrations of PBDEs reported in Hong Kong sediments and mussel tissues are amongst the highest in the world.

Keywords: Polybrominated diphenyl ethers; PBDEs; Sediments; Mussels; *Perna viridis*; Hong Kong

1. Introduction

The marine environment of Hong Kong, situated on the southern coast of China, has experienced severe pollution stress through various anthropogenic discharges. Increased industrialization and urbanization in the Pearl River Delta (PRD) is considered to be one of the major sources of pollution in the Hong Kong marine environment, particularly in the western regions during the wet summer season (Richardson et al., 2000). Within Hong Kong, population growth and rapid urbanization have also resulted in increased releases of contaminants into Victoria and Tolo Harbours (Blackmore, 1998). Historically, discharges of largely untreated domestic and industrial wastewater into Hong Kong's coastal waters, along with the disposal of contaminated mud from harbour dredging and filling, have resulted in high levels of persistent organic pollutants in the local marine ecosystem (Connell et al., 1998a,b).

Brominated flame retardants (BFRs) are synthetic compounds which have been widely used to decrease the likelihood and intensity of fire in a variety of consumer products, such as automobile accessories, computers, foam furniture, building materials, electrical

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and electronic equipment, and textiles (de Wit, 2002). Included within the BFRs are the polybrominated diphenyl ethers (PBDEs), which have chemical properties and toxicological profiles similar to other persistent synthetic organic compounds. PBDEs have been associated with endocrine disruption, reproductive/developmental toxicity including neurotoxicity and cancer, and share similar chemical and toxicological profiles with the polychlorinated biphenyls (Schecter et al., 2004). In view of their bioaccumulation potential, environmental persistence, and potential human and wildlife toxicities (Martin et al., 2004), PBDEs are included amongst six environmentally hazardous substances (the others being lead, mercury, cadmium, hexavalent chromium, and polybrominated biphenyls, PBBs) in the European Union issued Directive 2002/95/EC regarding the restricted use of certain hazardous substances in electrical and electronic equipment (RoHS), which will come into force on July 1, 2006.

Regular surveillance programmes, such as those conducted by the Hong Kong Environmental Protection Department, have monitored the concentrations of heavy metals and persistent organic pollutants (PCBs, polycyclic aromatic hydrocarbons, and organochlorine pesticides) in sediments at 45 stations in open coastal waters and 15 stations in typhoon shelters of Hong Kong, but these studies have not included PBDEs. A number of recent studies in Hong Kong marine waters have examined persistent organic contaminant levels in biota samples (Monirith et al., 2003; Richardson et al., 2001; De Luca-Abbott et al., 2005), as well as perfluorinated compounds in sediments and biota (So et al., unpublished data).

In a recent review, Martin et al. (2004) examined the literature and published environmental data on PBDEs in Asia, and identified an urgent need to determine their distribution in coastal environments, particularly in regions with heavy industrialization, such as the southern coast of China. Within the southeast Asian region, Bayen et al. (2003) reported elevated PBDE concentrations in mussels collected from stations near an industrialized area and shipping lane in Singapore. PBDEs have also been reported from Singapore's coastal marine sediments (Wurl and Obbard, 2005). Significantly, a recent study on the sediment concentrations of PBDEs in the Pearl River Delta has revealed the presence of relatively high concentrations of these compounds in areas of major shipping/offload facilities, particularly those in enclosed harbours (Zheng et al., 2004).

The lower brominated PBDEs (e.g., tetra-BDEs and penta-BDEs) are thought to predominate and accumulate in biota, whilst the higher congeners (e.g., deca-BDEs) appear to be more prevalent in aquatic environments and sediments (Martin et al., 2004). Monitoring of PBDE levels in sediments and biota from northern latitudes generally shows an increasing temporal trend, with an apparent rapid acceleration in concentrations in the past 10 years. Indeed, PBDE levels in many environmental compartments have increased dramatically between the 1970s and 1990s (de Wit, 2002; Martin et al., 2004). The aim of this paper is to document concentrations of PBDE congeners in surface sediments and tissues of green-lipped mussels (*Perna viridis*) collected from coastal stations in Hong Kong.

2. Materials and methods

Samples (approximately 500 g) of surface sediments (up to approximately 10 cm depth) were removed close to the shoreline from each of 13 locations shown in Fig. 1 during the rainy season (September and October, 2004). Sediment samples were taken with a pre-cleaned (with tap water and distilled water) Eckman dredge, subtidally from approximately 2 m to 8 m water depth. The samples were transferred into pre-cleaned inert polypropylene containers (Decon 90® detergent, followed by three tap water rinses, three acetone rinses, and three dichloromethane rinses), and stored at −20 °C for subsequent extraction and chemical analyses. Samples of green-lipped mussels (*P. viridis,* shell lengths between 60 and 120 mm, approximately 30 individuals) were collected by hand at low tide from each of nine locations (Fig. 1). Mussels and sediments were collected at five co-located sites: Stations 4, 7, 13, 15 and 18. After collection, all samples were kept on ice until returned to the laboratory, separated by polypropylene containers within a $50 \times 50 \times 35$ cm polystyrene foam storage container. Mussels were shucked in the laboratory immediately upon return. Extracted tissues were transferred into pre-cleaned, capped, 50 ml BD Falco® centrifugal tubes (cleaned using the same procedures as for sediment containers), and the tubes were stored at −20 °C until extraction and chemical analyses.

Analytical procedures generally followed those of Zheng et al. (2004). The 15 BDE congeners investigated in this study were as follows: BDE-3, BDE-15, BDE-28, BDE-47, BDE-60, BDE-85, BDE-99, BDE-100, BDE-138, BDE-153, BDE-154, BDE-183, BDE-197, BDE-207, and BDE-209. BDE congeners are numbered according to the International Union of Pure and Applied Chemistry (IUPAC) system originally designed for PCBs (Ballschmiter and Zell, 1980). All ^{13}C-labeled BDE standards were purchased from Cambridge Isotope Laboratories, Inc., Andover, MA 02810–5413, USA. The selection of the BDE congeners analysed in our study was based upon the following: (1) reported presence in environmental media, including sediment, water, air, or biota; (2) availability of authentic reference standards for quantitation; and (3) publication of methods which had been previously utilized in PBDE evaluations (i.e., Hale et al., 2001; Strandberg et al.,

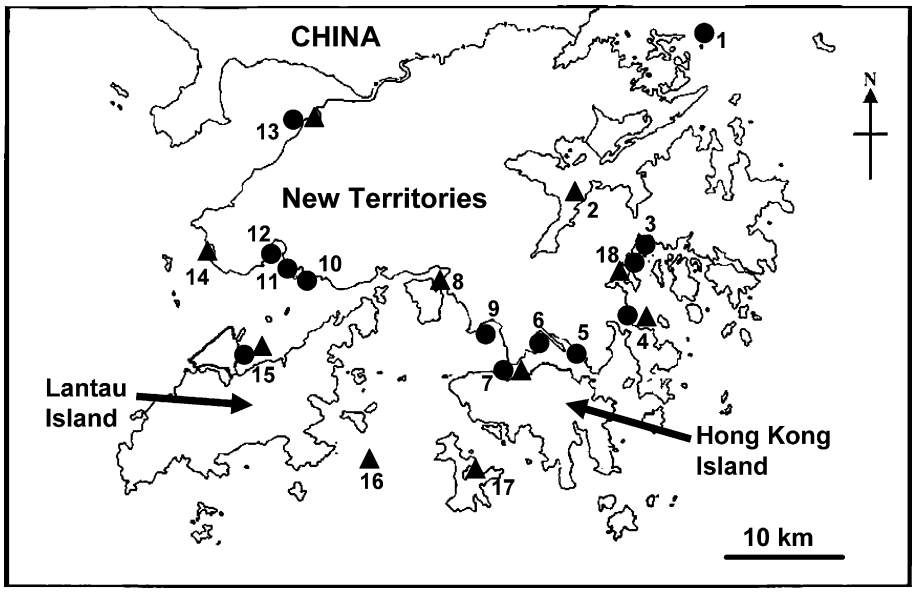

Fig. 1. Sediment and mussel sampling in coastal waters of Hong Kong. Sediment locations (●): 1 (Sha Tau Kok), 3 (Wong Chuk Bay), 4 (Hong Kong University of Science and Technology, HKUST), 5 (Kwun Tong), 6 (To Kwa Wan), 7 (Tsim Sha Tsui), 9 (Tai Kok Tsui), 10 (Gold Coast), 11 (Castle Peak Bay), 12 (Tuen Mun), 13 (Lau Fau Shan), 15 (Dung Chung Bay), 18 (Sai Kung). Mussel locations (▲): 2 (Chinese University), 4 (Hong Kong University of Science and Technology, HKUST), 7 (Tsim Sha Tsui), 13 (Lau Fau Shan), 14 (Lung Kwu Tan), 15 (Dung Chung Bay), 16 (Cheung Chau), 17 (Lamma Island), 18 (Sai Kung).

2001; Hyötyläinen and Hartonen, 2002; Dodder et al., 2002; Choi et al., 2003), and with modifications based upon two current reports (Ueno et al., 2004; Oros et al., 2005). The total PBDE ($\sum_{15}$PBDE) concentrations in the samples were calculated as the sum of these targeted analytes. Concentrations below the method detection limits (MDLs) were assumed to be zero for the summation of $\sum_{15}$PBDE in each sample.

Sediment or mussel samples were thawed at room temperature (approximately 20 °C), mixed well by stirring, placed in a wide mouth polyethylene tube, and freeze-dried for 7 days, during which time they reached constant weight. Dried sediments and mussels were then ground into powder with a stainless steel blender (National Super Blender®). The dried sediment sample (approximately 10 g) or dried mussel powder (approximately 2.5 g, and both weighed to 0.01 g accuracy) were transferred into solvent-rinsed Advantec® extraction thimble filters. Samples were Soxhlet extracted for 12 h in 150 ml of hexane:acetone:dichloromethane (1:1:1) solution to which 40 μl of a solution of 40 ng ml^{-1} of ^{13}C-labeled BDE congeners (BDE-3, BDE-15, BDE-28, BDE-47, BDE-99, BDE-153, BDE-183, BDE-197, BDE-207, and BDE-209) was added as an internal standard. The mixture was heated to 65 °C to gently boil the dried sediment or mussel tissues in the solvent solution. High purity solvents were used for analyses. Mussel samples were treated the same as the sediment samples.

Sediment. After extraction and cooling, the extraction solutions of sediment samples were reduced in volume to 1 ml by rotary evaporation prior to column chromatography. A chromatography column, consisting of 6 g of pre-activated silica gel and 1 g of activated copper powder to alleviate gas chromatographic interference from sulphur compounds, was washed with 15 ml acetone, followed by 15 ml of dichloromethane and 20 ml of hexane. The extraction aliquot (1 ml) was then added on the column, and the PBDEs were eluted with a 30 ml mixture of dichloromethane and hexane (1:1), followed by 15 ml of dichloromethane. The combined eluate (45 ml) was reduced in volume with rotary evaporation to 300 μl for quantitation by gas chromatography/mass spectrometry.

Mussel tissue. The extraction solutions of mussel samples were reduced in volume to 1 ml by rotary evaporation prior to column chromatography. The cleanup was performed through a multilayer glass column (1 cm i.d × 20 cm) packed from the bottom with 5 g aluminum oxide activated at 160 °C for 24 h and deactivated with 10% water; 1 g silica activated at 160 °C for 24 h; 5 g silica activated and impregnated with concentrated sulphuric acid (30 g silica + 10 ml 98% sulphuric acid); and 1 cm anhydrous sodium sulphate (Christensen et al., 2002). Prior to use, the column was eluted with 20 ml hexane, which was discarded. The sample was applied to the column with three 1 ml aliquots of *n*-hexane and eluted with a 50 ml mixture of dichloromethane and hexane (1:1), followed by 15 ml of dichloromethane. The combined eluate (65 ml) was reduced in volume by rotary evaporation to 300 μl for quantitation by gas chromatography/mass spectrometry.

All samples were analyzed for the 15 PBDE congeners using a Varian Saturn 2200 Ion Trap GC/MS/MS

with a 30 m DB-1 fused silica capillary column (0.25 mm diameter and 0.25 μm film thickness; 100% dimethyl-polysiloxane), with helium at a rate of 1 ml min^{-1}as the carrier gas. The 2 μl injections of the combined sample and internal calibration standard solutions or external calibration standards were made in the splitless mode. The injector temperature was 280 °C. The GC oven temperature was programmed as follows: 90 °C for 2 min, increasing 20 °C per minute to 230 °C, followed by 2 °C per minute to 247 °C, then 20 °C per minute to a final temperature of 280 °C, which was held for 25 min. Total run time was 46.15 min. The mass spectrometer was operated in the electron impact/selected ion monitoring (EI/SIM) mode. The following quantifying ions (*m*/*z*) were used to monitor BDE compounds in the EI/SIM mode: mono-Br 248–260, di-Br 328–340, tri-Br 406–418, tetra-Br 486–498, penta-Br 564–576, hexa-Br 644–656, hepta-Br 721–733. The BDE congeners were quantified using the isotope dilution method, with additions of ^{13}C-labeled BDEs. Concentrations of BDE were measured by using ^{13}C-labeled BDE internal standards. In those cases where no internal standards were added (BDE-60, BDE-85, BDE-100, BDE-138, BDE-154), we measured the BDE concentration with the "nearest neighbor" C^{13} internal standard. All BDEs were identified based upon the relative retention time of external standards and the computer library of mass spectra fragmentation patterns on the MS detector in EI/SIM mode.

A Shimadzu QP 2010 GC/MS with a short (11.5 m) DB-1 column (0.25 mm diameter and 0.25 μm film thickness; 100% dimethyl-polysiloxane) was used for measurement of BDE-197, BDE-207, BDE-209 concentrations, with helium as a carrier gas, in the EI/SIM mode. The following quantitation ions (*m*/*z*) were used to monitor BDE compounds in the EI/SIM mode: octa-, nona- and deca-Br 809–969. Concentrations of BDE-197, BDE-207, and BDE-209 were measured by using ^{13}C-labeled BDEs standards. BDE-197, BDE-207, and BDE-209 were identified based upon the retention times of external standards and the computer library of mass spectra fragmentation patterns on the MS detector. Oven temperature programming for analysis was: 120 °C for 2 min, increasing 8 °C min^{-1} to 300 °C, which was held for 15.5 min. Total run time was 40 min.

All samples were run in duplicate. Two quality control criteria were utilized to ensure the correct identification of the target compounds in sediment and tissues samples: (a) signal to noise ratio was greater than three; and (b) GC retention times matched (±0.05 min) those of standard compounds. Concentrations of BDEs in solvent blanks ($n = 2$) were low (<0.08 ng g^{-1}), and the detection limits were calculated as 3 times the procedural blank (=0.2 ng g^{-1}). All reported values were corrected using a mean blank value of 0.075 ng g^{-1}. All sediment and tissue individual BDEs and $\sum_{15}$PBDE concentrations are reported in ng g^{-1} dry wt.

Spike recoveries were performed on white sand, quartz HP Company, USA, with large grain size and low total organic carbon ($n = 2$): 108% for BDE-28, 89% for BDE-47, 81% for BDE-99, 85% for BDE-153 and 76% for BDE-183. Absolute recoveries of PBDEs in other investigations, ranging between 70% and 120% (Dodder et al., 2002), and between 50% and 120% (Oros et al., 2005), have been judged acceptable. The concentrations of analytes were not corrected for recoveries. Currently, there are no internationally recognized certified or standard reference materials for PBDE analyses in mussels or sediments matrices.

3. Results and discussion

3.1. Sediments

All of the stations had $\sum_{15}$PBDE concentrations above the detection limits. $\sum_{15}$PBDE concentrations ranged from 1.68 to 53.6 ng g^{-1} dry wt (Table 1). The highest concentrations of $\sum_{15}$PBDE in sediments was found at Sta 5, Kwun Tong (53.6 ng g^{-1} dry wt), which was an order of magnitude higher than the least contaminated station at Sta 10, Gold Coast (1.68 ng g^{-1} dry wt). Two of the stations had $\sum_{15}$PBDE concentrations that were similar to Sta 5, Kwun Tong (Sta 6 To Kwa Wan, 41.6 ng g^{-1} dry wt; and Sta 18, Sai Kung, 42.4 ng g^{-1} dry wt). Three stations had sediments with relatively low concentrations and were similar to those found at Sta 10, Gold Coast: Sta 1, Sha Tau Kok (4.81 ng g^{-1} dry wt); Sta 3, Wong Chuk Bay (4.39 ng g^{-1} dry wt); and Sta 11, Castle Peak Bay (4.14 ng g^{-1} dry wt). The remaining stations had intermediate concentrations, ranging from Sta 13, Lau Fau Shan (9.06 ng g^{-1} dry wt) to Sta 15, Dung Chung Bay (21.1 ng g^{-1} dry wt).

To evaluate the relative degree of PBDE contamination in Hong Kong marine sediments, we compared our data against other studies where PBDE concentrations have been reported. Although we recognize that the direct comparability is somewhat compromised by the fact that different studies evaluated different BDE congeners, we believe it is important to evaluate the qualitative patterns of PBDE contamination to get a sense of regional or area similarity. Surface sediments from San Francisco Estuary (USA) had ∑PBDE concentrations (sum of BDE-47, BDE-99, BDE-183, BDE-204, BDE-205) that ranged from non-detectable to 211.8 ng g^{-1} dry wt (Oros et al., 2005). Concentrations in sediments from Portugal ranged from 20 ng g^{-1} in riverine situations to 0.5 ng g^{-1}dry wt in marine sediments (Lacorte et al., 2003). Concentrations of PBDEs in Japanese river sediments have been reported as ranging from 21 to

Table 1
Concentrations of individual BDEs (ng g^{-1} dry wt) in surface sediments from Hong Kong coastal waters

BDE congener	Site, sample number per site, and mean value per site								
	1 (Sha Tau Kok)			3 (Wong Chuk Bay)			4 (HKUST)		
	1	2	Mean	1	2	Mean	1	2	Mean
BDE-3	0.08	0.06	0.07	0.64	0.34	0.49	0.52	0.18	0.35
BDE-15	0.72	0.68	0.70	0.63	0.54	0.58	1.60	0.66	1.13
BDE-28	0.96	0.56	0.76	0.00	0.00	0.00	2.80	0.82	1.81
BDE-47	2.20	1.56	1.88	1.30	0.97	1.14	2.80	2.30	2.55
BDE-60	0.06	0.04	0.05	0.03	0.01	0.02	0.16	0.28	0.22
BDE-85	0.21	0.12	0.16	0.00	0.00	0.00	0.47	0.51	0.49
BDE-99	0.36	0.23	0.30	3.30	0.98	2.14	4.56	0.96	2.76
BDE-100	0.00	0.01	0.01	0.00	0.00	0.00	0.14	0.10	0.12
BDE-138	0.11	0.17	0.14	0.00	0.00	0.00	0.35	0.47	0.41
BDE-153	0.24	0.22	0.23	0.00	0.00	0.00	0.33	1.40	0.86
BDE-154	0.17	0.32	0.25	0.00	0.00	0.00	0.23	0.34	0.28
BDE-183	0.24	0.11	0.18	0.00	0.00	0.00	1.28	3.20	2.24
BDE-197	0.05	0.07	0.06	0.02	0.00	0.01	2.88	2.62	2.75
BDE-207	0.01	0.01	0.01	0.01	0.00	0.01	0.67	1.15	0.91
BDE-209	0.02	0.03	0.02	0.00	0.00	0.00	0.00	0.00	0.00
$\sum_{15}$PBDE	5.43	4.19	4.81	5.93	2.84	4.39	18.8	15.0	16.9
	5 (Kwun Tong)			6 (To Kwa Wan)			7 (Tsim Sha Tsui)		
	1	2	Mean	1	2	Mean	1	2	Mean
BDE-3	0.40	0.28	0.34	10.9	4.68	7.79	1.70	2.43	2.06
BDE-15	0.24	0.15	0.20	0.72	0.48	0.60	1.40	1.69	1.54
BDE-28	1.70	1.23	1.46	1.40	9.60	5.50	0.72	0.87	0.80
BDE-47	0.96	0.78	0.87	0.00	0.00	0.00	0.56	0.47	0.52
BDE-60	0.17	0.21	0.19	0.03	0.00	0.02	0.14	0.18	0.16
BDE-85	0.65	0.34	0.50	0.00	0.00	0.00	0.17	0.22	0.20
BDE-99	9.36	7.56	8.46	2.90	4.68	3.79	1.40	1.54	1.47
BDE-100	0.04	0.00	0.02	0.00	0.00	0.00	0.02	0.03	0.02
BDE-138	1.06	1.32	1.19	0.00	0.00	0.00	0.16	0.09	0.12
BDE-153	6.16	4.56	5.36	1.40	1.50	1.45	0.96	0.87	0.92
BDE-154	2.34	2.56	2.45	0.43	0.57	0.50	0.35	0.21	0.28
BDE-183	18.9	9.75	14.3	6.20	7.45	6.82	2.20	1.65	1.92
BDE-197	11.3	12.1	11.7	10.5	11.4	11.0	0.00	0.00	0.00
BDE-207	5.18	4.96	5.07	4.07	2.25	3.16	0.00	0.00	0.00
BDE-209	1.34	1.72	1.53	0.67	1.27	0.97	0.00	0.00	0.00
$\sum_{15}$PBDE	59.8	47.5	53.6	39.2	43.9	41.6	9.78	10.2	10.0
	9 (Tai Kok Tsui)			10 (Gold Coast)			11 (Castle Peak Bay)		
	1	2	Mean	1	2	Mean	1	2	Mean
BDE-3	0.00	0.14	0.07	0.03	0.02	0.02	0.00	0.00	0.00
BDE-15	0.29	0.24	0.26	0.38	0.22	0.30	0.05	0.04	0.04
BDE-28	0.00	0.36	0.18	0.03	0.02	0.02	0.00	0.00	0.00
BDE-47	0.55	0.19	0.36	0.03	0.03	0.03	0.00	0.00	0.00
BDE-60	0.07	0.09	0.08	0.06	0.03	0.04	0.00	0.00	0.00
BDE-85	0.35	0.23	0.29	0.04	0.08	0.06	0.00	0.00	0.00
BDE-99	1.66	4.98	3.32	1.80	0.55	1.18	0.00	0.00	0.00
BDE-100	0.09	0.07	0.08	0.00	0.00	0.00	0.00	0.00	0.00
BDE-138	0.28	0.29	0.28	0.00	0.00	0.00	0.00	0.00	0.00
BDE-153	2.93	2.32	2.62	0.00	0.00	0.00	1.60	1.05	1.32
BDE-154	0.23	0.19	0.21	0.00	0.00	0.00	0.43	0.49	0.46
BDE-183	4.37	3.60	3.98	0.00	0.00	0.00	1.49	1.21	1.35
BDE-197	6.98	7.20	7.09	0.01	0.01	0.01	0.02	0.00	0.01
BDE-207	0.79	1.09	0.94	0.00	0.00	0.00	0.01	0.01	0.01
BDE-209	0.45	0.99	0.57	0.02	0.02	0.02	1.09	0.81	0.95
$\sum_{15}$PBDE	19.0	22.0	21.3	2.40	0.98	1.68	4.69	3.61	4.14
	12 (Tuen Mun)			13 (Lau Fau Shan)			15 (Dung Chung Bay)		
	1	2	Mean	1	2	Mean	1	2	Mean
BDE-3	0.32	0.12	0.22	0.17	0.11	0.14	0.47	0.97	0.72
BDE-15	0.76	0.87	0.82	0.67	0.56	0.62	0.08	0.28	0.18

(*continued on next page*)

Table 1 (*continued*)

BDE congener	Site, sample number per site, and mean value per site								
	12 (Tuen Mun)			13 (Lau Fau Shan)			15 (Dung Chung Bay)		
	1	2	Mean	1	2	Mean	1	2	Mean
BDE-28	0.06	0.12	0.09	0.58	0.62	0.60	2.13	2.67	2.40
BDE-47	0.36	0.45	0.40	1.80	1.17	1.48	0.89	0.55	0.72
BDE-60	0.00	0.00	0.00	0.14	0.21	0.18	0.14	0.20	0.17
BDE-85	0.05	0.03	0.04	0.32	0.19	0.26	0.25	0.15	0.35
BDE-99	1.40	0.76	1.08	1.85	1.98	1.92	1.25	1.63	1.44
BDE-100	0.00	0.00	0.00	0.12	0.34	0.23	0.03	0.07	0.05
BDE-138	0.09	0.07	0.08	0.17	0.53	0.35	0.51	0.61	0.56
BDE-153	0.96	0.45	0.70	3.84	1.34	2.59	4.23	5.37	4.80
BDE-154	0.08	0.03	0.06	0.32	0.24	0.28	0.23	0.37	0.30
BDE-183	0.02	0.09	0.06	0.39	0.29	0.34	3.01	3.79	3.40
BDE-197	1.17	0.97	1.07	0.00	0.00	0.00	0.61	0.93	0.77
BDE-207	0.96	0.74	0.85	0.03	0.07	0.05	4.12	5.26	4.69
BDE-209	0.14	0.22	0.18	0.02	0.02	0.02	0.21	0.95	0.58
$\sum_{15}$PBDE	6.37	4.92	5.64	10.4	7.67	9.06	18.2	23.8	21.1

	18 (Sai Kung)			BLANK			Recovery
	1	2	Mean	1	2	Mean	
BDE-3	0.46	0.35	0.40	0.00	0.00	0.00	
BDE-15	1.20	0.78	0.99	0.01	0.01	0.01	
BDE-28	1.20	0.96	1.08	0.00	0.00	0.00	107.64
BDE-47	4.60	2.76	3.68	0.01	0.02	0.02	89.14
BDE-60	0.19	0.53	0.36	0.00	0.00	0.00	
BDE-85	0.37	0.29	0.33	0.00	0.00	0.00	91.21
BDE-99	5.20	4.66	4.93	0.00	0.00	0.00	80.66
BDE-100	0.15	0.17	0.16	0.02	0.01	0.02	77.53
BDE-138	0.45	0.35	0.40	0.00	0.00	0.00	
BDE-153	4.20	2.71	3.46	0.00	0.00	0.00	85.17
BDE-154	1.54	1.43	1.48	0.00	0.00	0.00	
BDE-183	2.80	1.23	2.02	0.00	0.00	0.00	76.02
BDE-197	10.88	7.54	9.21	0.01	0.01	0.01	
BDE-207	13.05	9.43	11.24	0.00	0.00	0.00	
BDE-209	2.92	2.50	2.71	0.00	0.00	0.00	
$\sum_{15}$PBDE	53.2	39.7	42.4	0.05	0.05	0.05	

59 ng g^{-1}dry wt, although ranges between <25 and 11,600 ng g^{-1} dry wt as BDE-209 were also reported (Watanabe et al., 1987, in de Wit, 2002). A more recent report of sediment samples taken from industrialised areas in Japan, with a range of 0.013–2.349 ng g^{-1} appear to be inconsistent with earlier reports (Choi et al., 2003). PBDEs were determined in the upper layer of a sediment core collected in the Baltic Sea at 0.52 ng g^{-1} dry wt (Nylund et al., 1992). Marine surface sediments collected from 13 sites in Singapore had BDE-47 concentrations (the only congener detected) that ranged from 3.4 to 13.8 ng g^{-1} dry wt (Wurl and Obbard, 2005).

In Hong Kong sediments, all 15 BDE congeners were measured in at least one of the 13 sediment samples, and there were several congeners that occurred in the highest abundance at specific sites (Table 1). In the more contaminated sediments, BDE-183 at Sta 5, Kwun Tong (14.3 ng g^{-1} dry wt) was the most abundant congener, followed by BDE-197 (11.7 ng g^{-1} dry wt), BDE-99 (8.46 ng g^{-1} dry wt) and BDE-153 (5.36 ng g^{-1} dry wt). At Sta 4, HKUST, BDE-47 (2.55 ng g^{-1} dry wt), BDE-99 (2.76 ng g^{-1} dry wt), BDE-183 (2.24 ng g^{-1} dry wt), and BDE-197 (2.75 ng g^{-1} dry wt) were in nearly equal

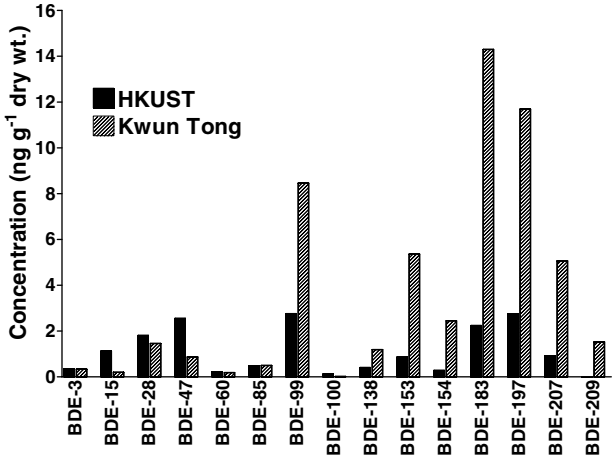

Fig. 2. Comparison of individual BDE congeners in sediments at HKUST (Sta 4) and Kwun Tong (Sta 5). Concentrations in ng g^{-1} dry wt.

abundances (Fig. 2). At Sta 18, Sai Kung, BDE-207 (11.24 ng g^{-1} dry wt) was the most abundant congener, followed by BDE-197 (9.21 ng g^{-1} dry wt) and BDE-99 (4.93 ng g^{-1} dry wt). At another station with higher concentrations of $\sum_{15}$PBDE (Sta 6, To Kwa Wan), the most abundant congener was BDE-197 (11.0 ng g^{-1} dry wt), followed by BDE-3 (7.79 ng g^{-1} dry wt), and BDE-183 (6.82 ng g^{-1} dry wt). At the least $\sum_{15}$PBDE-influenced sites (Sta 10, Gold Coast and Sta 3, Wong Chuk Bay), BDE-99 was the predominant congener, with these samples having a much less complex array of congeners than the more contaminated samples.

As with studies of $\sum_{15}$PBDE in sediments at other localities, BDE-47, BDE-99, BDE-153, BDE-154 and BDE-183 were almost always encountered in most of the samples, but were not necessarily the dominant congeners in the profiles. In the areas of highest $\sum_{15}$PBDE contamination (e.g., Kwun Tong and To Kwa Wan), the moderately and heavily substituted BDE congeners (BDE-153 and higher) predominated over the lesser brominated congeners. These patterns of congener abundance, with respect to the sampling locations or areas, are complex, reflecting several potential regulating factors which could include the source compositions, sedimentary dispersion/accumulation patterns, environmental degradation, or metabolism by sedimentary communities. Wurl and Obbard (2005) suggested that the main source of PBDEs in Singapore Harbour was likely to be industrial activities, shipyards and intensive shipping traffic.

Victoria Harbour. Two sites in Kowloon Bay (Sta 5, Kwun Tong and Sta 6, To Kwa Wan) had the highest sediment concentrations of $\sum_{15}$PBDEs in our survey. These sites have been previously identified as contaminated with a range of POPs, including PCBs, HCHs, petroleum hydrocarbons (PHCs), and PAHs (Richardson and Zheng, 1999; Zheng and Richardson, 1999). In certain instances (e.g., PHCs and PAHs), highest Hong Kong concentrations have been reported from these two sites.

The sites are industrial locations surrounded by densely populated urban areas, and a lack of sea water exchange, as well as a lack of resuspension energy for contaminants which have been deposited on the bottom, may contribute to their levels of contamination. Casual observations at the time of sediment collection were the presence of strong hydrogen sulphide smell, a distinctly black colour of the seawater, and a heavily oiled and colourful sheen on the sea surface. Kwun Tong sediments also had the highest toxicities of eight sediments tested in marine bioassays from a transect along Victoria Harbour (Wong et al., 1995).

The remaining two Victoria Harbour sites (Sta 7 and Sta 9) were consistent with POPs and PAHs distribution patterns described in previous studies (Zheng and Richardson, 1999; Richardson and Zheng, 1999). In spite of enormous shipping activity and local municipal waste discharges, Tsim Sha Tsui (Sta 7) is arguable one of the least contaminated sites in Victoria Harbour; the $\sum_{15}$PBDEs sediment concentrations at Tsim Sha Tsui are lower than surrounding Harbour sites (10 ng g^{-1}dry wt). The reasons for this seemingly lower accumulation in sediments is not known, but may be due to sedimentary characteristics (sandy), sources of parent compounds, and strong water circulation patterns. Tai Kok Tsui (Sta 9) shows an intermediate pattern of $\sum_{15}$PBDEs contamination (21.3 ng g^{-1}dry wt). Zheng and Richardson (1999) state that concentrations of trace organic contaminants tend to be lower at the western edge of the harbour, rise within the harbour itself (although Tsim Sha Tsui appears somewhat anomalous for several chemicals), and decline in eastern Hong Kong waters.

Eastern Hong Kong Zone. The high concentration of $\sum_{15}$PBDEs in sediment from Sta 18, Sai Kung was not consistent with previously reported patterns of POPs and PAHs distributions. Sta 18 had relatively high $\sum_{15}$PBDEs in sediments (42.4 ng g^{-1} dry wt), while three other sites (Sta 1, 3, 4) had low $\sum_{15}$PBDEs congener concentrations (4.81, 4.39, and 16.9 ng g^{-1}dry wt, respectively). Whilst the Eastern Hong Kong Zone is typically characterised as a "cleaner" Hong Kong area, with open exposure to sea currents from the south east, the reasons for the apparently elevated concentrations of $\sum_{15}$PBDEs, PCBs, and total DDTs at Sai Kung remain obscure. There may be local sources of these compounds (maritime activities, municipal waste releases, or non-point source runoff) or possibly unique depositional patterns which may result in the unexpected distributions of POPs at this site.

Pearl River Delta—Western Hong Kong Zone. Sediments from five locations, contiguous with the Pearl River Delta (Sta 10, 11, 12, 13, 15) contained $\sum_{15}$PBDEs concentrations between 1.68 and 21.1 ng g^{-1} dry wt. Dung Chung Bay (Sta 15) had the highest $\sum_{15}$PBDEs concentration of the Western Zone sites (21.1 ng g^{-1}dry wt), compared with Sta 10, 11, 12 and 13, which ranged between 1.68 and 9.06 ng g^{-1} dry wt. One apparent difference between Sta 15 and the others is its close proximity to the new airport facilities at Chek Lap Kok, but other activities at this site, such as recent and increasing urban development (sewage discharge) and sediment dumping (Hung et al., 2004), as well as busy maritime activities, including vessel refuelling and ship repair and construction activities, may have had some influence as well.

Southern Hong Kong Island Zone. Sediments were not collected in this zone, and, as described below, $\sum_{15}$PBDEs for this zone were evaluated using mussel tissue concentrations.

3.2. Mussels

Most of the mussel samples had $\sum_{15}$PBDEs concentrations above the detection limits for many of the

congeners (Table 2). $\sum_{15}$PBDEs concentrations in Hong Kong mussels from nine sites ranged from 27.0 to 83.7 ng g^{-1} dry wt. Mussels accumulated slightly higher concentrations of $\sum_{15}$PBDEs than sediments; however there was a smaller range between the lowest and highest stations when compared with the range of sediment con-

Table 2
Concentrations of individual BDEs (ng g^{-1} dry wt) in m mussels from Hong Kong coastal waters

BDE congener	Site, sample number per site, and mean value per site								
	2 (Chinese University)			4 (HKUST)			7 (Tsim Sha Tsui)		
	1	2	Mean	1	2	Mean	1	2	Mean
BDE-3	0.42	0.15	0.28	1.28	1.13	1.20	0.96	3.07	2.33
BDE-15	2.30	1.48	1.89	2.60	2.34	2.47	1.17	0.38	0.78
BDE-28	2.88	2.30	2.59	4.40	5.26	4.83	0.58	0.65	0.62
BDE-47	2.18	2.48	2.33	8.00	9.12	8.56	2.88	4.08	3.48
BDE-60	0.57	0.84	0.70	0.71	0.67	0.69	0.47	0.54	0.50
BDE-85	0.96	0.58	0.77	1.44	1.12	1.28	1.04	1.09	1.06
BDE-99	1.34	1.48	1.41	25.9	16.7	21.3	6.72	9.20	7.96
BDE-100	0.43	0.53	0.48	0.78	1.76	1.27	0.57	0.45	0.51
BDE-138	0.87	0.59	0.73	1.03	1.31	1.17	0.88	0.46	0.67
BDE-153	0.00	0.00	0.00	9.76	8.55	9.16	11.2	10.5	10.9
BDE-154	0.00	0.00	0.00	1.32	1.09	1.20	0.34	0.39	0.36
BDE-183	1.92	1.24	1.58	13.0	9.21	11.1	5.44	5.76	5.60
BDE-197	9.43	5.47	7.45	1.27	1.73	1.50	2.99	3.19	3.09
BDE-207	5.44	4.76	5.10	1.87	1.59	1.73	5.24	6.14	5.69
BDE-209	1.51	1.93	1.72	0.07	0.15	0.11	3.07	2.97	3.02
$\sum_{15}$PBDE	30.2	23.8	27.0	73.4	61.7	67.6	43.6	48.9	46.6
	13 (Lau Fau Shan)			14 (Lung Kwu Tan)			15 (Dung Chung Bay)		
	1	2	Mean	1	2	Mean	1	2	Mean
BDE-3	3.20	1.21	2.20	0.00	0.00	0.00	0.80	0.55	0.68
BDE-15	0.26	1.34	0.80	1.54	1.21	1.38	0.29	0.21	0.25
BDE-28	5.04	14.2	9.62	0.71	0.54	0.62	18.7	20.2	19.5
BDE-47	0.62	2.98	1.80	3.20	2.67	2.94	7.20	2.67	4.94
BDE-60	0.15	0.38	0.26	0.27	0.28	0.28	0.19	0.23	0.21
BDE-85	0.52	1.76	1.14	0.34	0.56	0.45	0.71	0.55	0.63
BDE-99	6.88	13.5	10.2	2.37	1.76	2.06	3.36	13.3	8.33
BDE-100	0.76	0.42	0.59	0.00	0.00	0.00	0.45	1.56	1.00
BDE-138	0.64	1.68	1.16	0.24	0.32	0.28	0.68	7.21	3.94
BDE-153	2.72	1.40	2.06	14.5	12.1	13.3	5.92	0.00	2.96
BDE-154	1.03	0.68	0.86	1.76	2.14	1.95	0.34	0.24	0.29
BDE-183	2.44	3.76	3.10	12.1	11.8	12.0	25.4	15.0	20.2
BDE-197	0.00	0.00	0.00	16.4	21.2	18.8	4.76	6.38	5.57
BDE-207	0.00	0.00	0.00	20.5	26.7	23.6	8.07	7.31	7.69
BDE-209	0.00	0.00	0.00	5.87	6.13	6.00	3.76	4.12	3.94
$\sum_{15}$PBDE	24.3	43.3	33.8	79.8	87.4	83.7	80.6	79.5	80.1
	16 (Cheung Chau)			17 (Lamma Island)			18 (Sai Kung)		
	1	2	Mean	1	2	Mean	1	2	Mean
BDE-3	1.44	0.65	1.04	2.04	2.34	2.19	2.11	2.38	2.24
BDE-15	0.49	0.18	0.34	0.12	0.23	0.18	4.16	3.94	4.05
BDE-28	0.29	0.34	0.32	29.8	20.8	25.3	6.72	4.59	5.66
BDE-47	5.44	5.31	5.38	1.54	1.29	1.42	5.16	2.59	3.88
BDE-60	0.79	0.67	0.73	0.46	0.73	0.60	0.36	0.65	0.50
BDE-85	2.24	3.01	2.62	0.68	0.46	0.57	0.69	2.58	1.64
BDE-99	5.88	3.45	4.66	15.6	10.7	13.2	5.20	5.34	5.27
BDE-100	1.11	0.73	0.92	0.93	0.67	0.80	0.54	0.49	0.52
BDE-138	1.97	1.43	1.70	0.55	0.45	0.50	0.33	0.58	0.46
BDE-153	23.0	11.9	17.4	2.40	2.14	2.27	4.16	3.41	3.78
BDE-154	1.64	1.47	1.56	0.76	0.87	0.82	0.00	0.00	0.00
BDE-183	11.0	10.7	10.8	2.28	2.25	2.26	6.76	4.30	5.53
BDE-197	1.34	1.58	1.46	0.00	0.00	0.00	3.59	3.17	3.38
BDE-207	8.91	6.65	7.78	0.00	0.00	0.00	24.2	13.4	18.8
BDE-209	7.27	9.09	8.18	0.00	0.00	0.00	13.5	14.3	13.9
$\sum_{15}$PBDE	72.8	57.2	64.9	57.2	42.9	50.1	77.5	61.7	69.6

centrations. All of the BDE congeners ($n = 15$) measured in sediments were also measured in mussels. The highest concentrations in mussels were found in the Western Zone at Lung Kwu Tan (Sta 14) and Dung Chung Bay (Sta 15) in the Pearl River Delta (83.7 ng $g^{-1}$and 80.1 ng g^{-1} dry wt, respectively). Two other stations had $\sum_{15}$PBDE concentrations that were similar to these measurements: Sta 18, Sai Kung (69.6 ng g^{-1} dry wt) and Sta 16, Cheung Chau (64.9 ng g^{-1} dry wt). The stations with the lowest concentrations were located away from the central developed areas; e.g., Sta 2, Chinese University in Tolo Harbour and Sta 13, Lau Fau Shan in Deep Bay, Pearl River Delta, where concentrations were 27 ng g^{-1} and 33.8 ng g^{-1} dry wt, respectively. These two "cleanest" stations exceeded seven of the eight reported concentrations in Singapore by a factor of 10 (Bayen et al., 2003). The three remaining stations had intermediate concentrations to the above-mentioned sites, ranging from Sta 7, Tsim Sha Tsui (46.6 ng g^{-1} dry wt) and Sta 17, Lamma Island (50.1 ng g^{-1} dry wt) to Sta 4, HKUST (67.6 ng g^{-1} dry wt).

Comparisons of our data with other reports of mussel tissue measurements indicate that Hong Kong samples are the highest reported marine samples worldwide to date. Hites (2004) lists two $\sum$PBDE measurements in the Netherlands at 1.7 and 3.2 ng g^{-1} dry wt. Oros et al. (2005) report $\sum$PBDE in mussels from seven San Francisco Estuary sites between 13 and 47 ng g^{-1}dry wt. The previously cited Singapore study (Bayen et al., 2003) reports concentrations of $\sum$PBDEs consistent with the San Francisco Estuary sites, and one order of magnitude higher than sites in the Netherlands.

Other studies using bivalves as biomonitors have reported differing numbers of BDE congeners, ranging from 3 to 7. As is the case with sediment reporting, this probably reflects the selection of analytical techniques and the availability of standards, rather than the actual presence or absence of specific BDE congeners (see Hites, 2004, for a discussion of BDE study comparisons). Three PBDE congeners were reported by Oros et al. (2005) for bay mussels in the San Francisco Estuary, USA: BDE-47, BDE-99 and BDE-100. Christensen et al. (2002) reported four lower brominated PBDEs in blue mussels from Greenland: BDE-47, BDE-99, BDE-100 and BDE-153. Bayen et al. (2003) reported seven PBDE congeners in green-lipped mussels from Singapore: BDE-47, BDE-49, BDE-66, BDE-99, BDE-100, BDE-153 and BDE-154. At this time, there are so few measurements of PBDEs in bivalves, it is difficult to determine the reason(s) for the differences in BDE congeners detected in various studies. One obvious difference amongst studies, however, is the reference congeners used for identification, along with the differences in analytical protocols and instrumentation, as well as possible geographical and species differences.

As in other investigations of bivalves (and, for that matter, other wildlife species), BDE-47, BDE-99 and BDE-100 were commonly found in large concentrations in the Hong Kong samples, but they were not the predominant BDE congeners encountered, except at Sta 4, HKUST. All other sites had different congeners which occurred in the highest proportions. These could be either the higher and/or lower brominated congener species, and showed no consistent patterns amongst sites. The patterns for relative concentrations of major BDE congeners (BDE-47, BDE-99, and BDE-100) do not have obvious consistency with any available data from other parts of Asia, North America and Europe, and the reasons for these apparently major differences in congener distributions awaits further investigation. One accumulation possibility that remains interesting (and provocative) is the potential ability of green-lipped, tropical mussels to retain several more BDE congeners than the northern-latitude mussels, as reported from Europe, Greenland, and the US (Christensen et al., 2002; Christensen and Platz, 2001; Oros et al., 2005), again with the caution cited concerning differences amongst those BDE congeners actually used in analytical measurements.

One similar pattern in southeast Asia, shown in the Singaporean mussel study (Bayen et al., 2003), was trace accumulation of BDE-153 and BDE-154, although in our samples, some stations showed substantially larger contributions from these two congeners (15% at Sta 4, 24% at Sta 7, 18% at Sta 14, 29% at Sta 16). Another difference in our results is the apparently higher concentrations of the heavily brominated BDEs (BDE-183, BDE-197, BDE-207, BDE-209), which have not been previously reported in significant concentrations in marine biota (see Ueno et al., 2004, for fish) or mussel samples. Although the exact reasons for this pattern were not investigated, retention of sediment particles with sequestered higher brominated BDEs could be a causative factor. Another possibility is that these congeners are not routinely or commonly analysed (e.g. see Hites, 2004, caution regarding BDE-209). In summary, we conclude that it is not possible, at this time, to quantitatively compare BDE congener profiles between studies, because of differences in methods amongst laboratories, as well as obvious site-specific differences, depositional patterns, sources of PBDEs to the environment, and other potential unexplained characteristics of sites. Even within a single study, the varied patterns of congener distributions, as shown in the Hong Kong data, are very difficult to interpret and explain (see below).

Victoria Harbour. Only one station (Tsim Sha Tui, Sta 7) was sampled for mussels from Victoria Harbour. The $\sum_{15}$PBDE is intermediate in concentration (46.6 ng g^{-1} dry wt) between the most and least contaminated sites in Hong Kong waters. All of the analysed congeners were present in the sample. The BDE

congener profile shows eight congeners (BDE-3, BDE-47, BDE-99, BDE-153, BDE-183, BDE-197, BDE-207, and BDE-209) ranging in contribution from 5% (BDE-3) to 17% (BDE-99) of the $\sum_{15}$PBDE concentration for the site. The remaining congeners ($n = 7$) each contributed less than 2% to the $\sum_{15}$PBDE.

Eastern Hong Kong Zone. The three mussel samples from this zone (Sta 2, 4, and 18) had different patterns of $\sum_{15}$PBDE concentrations. The least contaminated site in the study, Sta 2, Chinese University, had a $\sum_{15}$PBDE concentration of 27 ng g^{-1} dry wt, whilst Sta 4, HKUST and Sta 18, Sai Kung had higher concentrations at 67.6 and 69.6 ng g^{-1} dry wt, respectively. The predominant congener at all sites was different: at Chinese University, BDE-197 (28% of $\sum_{15}$PBDE); Sai Kung, BDE-28 (10%); and HKUST, BDE-99 (32%). The ratios and the order of decreasing percentages of the congeners at each site were also dissimilar. All congeners were present at the HKUST site, but BDE-153 and BDE-154 congeners were missing at Chinese University, and BDE-154 was not present in the Sai Kung sample. In summary, there were no consistent, obvious, patterns of BDE congener distribution in the mussels from the Eastern Zone. The unique BDE congener pattern at Chinese University could be the result of a number of factors, including different local sources or distributional patterns caused by the semi-enclosed harbour area.

Pearl River Delta—Western Hong Kong Zone. The three mussel samples from this zone (Sta 13, 14, and 15) had different patterns of $\sum_{15}$PBDE concentrations. As with the Eastern Zone, two stations had the highest concentrations of $\sum_{15}$PBDEs: Sta 14, Lung Kwu Tan (83.7 ng g^{-1}dry wt) and Sta 15, Dung Chung Bay (80.1 ng g^{-1}dry wt); one station had lower concentrations: Sta 13, Lau Fau Shan (33.8 ng g^{-1} dry wt). One of the few obvious differences amongst the stations was the presence of higher brominated congeners (BDE-183, BDE-197, BDE-207, and BDE-209), and at Sta 14 and Sta 15, almost 50% of $\sum_{15}$PBDE was composed of these congeners. However, at Sta 13, these congeners made up less than 10% of the total $\sum_{15}$PBDE. As with the Eastern Zone, there were no obvious similarities in the patterns of BDE congener distributions in mussel samples amongst the Pearl River Delta—Western Hong Kong Zone.

Southern Hong Kong Island Zone. Mussels from two stations (Cheung Chau, Sta 16 and Lamma Island, Sta 17) had relatively high concentrations of $\sum_{15}$PBDEs in mussel tissues (64.9 and 50.1 ng g^{-1}dry wt, respectively). The predominant BDE congeners at both sites were different (Sta 16, BDE-153 and Sta 17, BDE-28), as well as the percentages of the higher substituted congeners (BDE-197, BDE-207, BDE-209). Cheung Chau had 27% of $\sum_{15}$PBDEs as these three highly brominated congeners, whilst they were not present in the Lamma Island sample. As with the three previously described areas, there were no consistent similarities in patterns of BDE congener distributions in the mussels.

3.3. Comparisons of mussel and sediment concentrations and congener distributions

Mussels and sediments were co-collected at five sites (Sta 4, 7, 13, 15, 18) and the $\sum_{15}$PBDEs concentrations are shown in Fig. 3. Concentrations of $\sum_{15}$PBDEs in the two media were generally correlated, but were not directly proportional. The ratios of mussel/sediment $\sum_{15}$PBDE concentrations ranged from 1.64 (Sta 18) to 4.66 (Sta 7).

With respect to the distributions of individual congeners in mussel tissues and sediment, Fig. 4 compares the distributions of the individual congeners at the Sai Kung site (Sta 18). There are general similarities in the concentrations of the individual congeners in the two media, with higher concentrations generally found in mussel tissues. For example, BDE-207 is the most predominant congener in both the sediment and mussel tissue samples, although the BDE-207 concentration in mussel tissues was approximately 1.5 times the concentration found in sediment. An exception to the pattern of higher concentrations in mussel tissue occurred with BDE-197 at this site, where the sediment concentration was almost 3 times that found in mussel tissues. Another interesting pattern of the mussel-sediment distribution occurred at Tsim Sha Tsui (Sta 7), which had a relatively low $\sum_{15}$PBDEs sediment concentration (10 ng g^{-1} dry wt) and a relatively high mussel concentration (46.6 ng g^{-1} dry wt; Fig. 5). BDE-99 and BDE-153 were prominent in the mussel profile, along with BDE-47, BDE-183, BDE-197, BDE-207 and BDE-209, occurring in concentrations between 3 and 10.9 ng g^{-1} dry wt. In

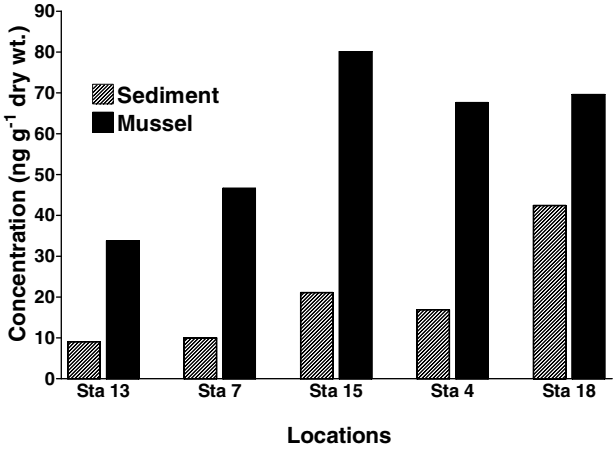

Fig. 3. Comparison of $\sum_{15}$PBDEs concentrations in both sediments and mussels collected from the same locations: Sta 13, Lau Fau Shan; Sta 7, Tsim Sha Tsui; Sta 15, Dung Chung Bay; Sta 4, Hong Kong University of Science and Technology, HKUST; and Sta 18, Sai Kung.

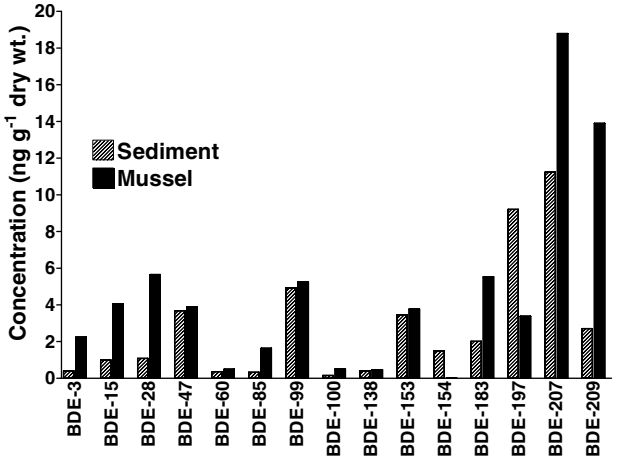

Fig. 4. Comparison of individual BDE congeners in mussels and sediments at Sta 18, Sai Kung. Concentrations in ng g^{-1} dry wt.

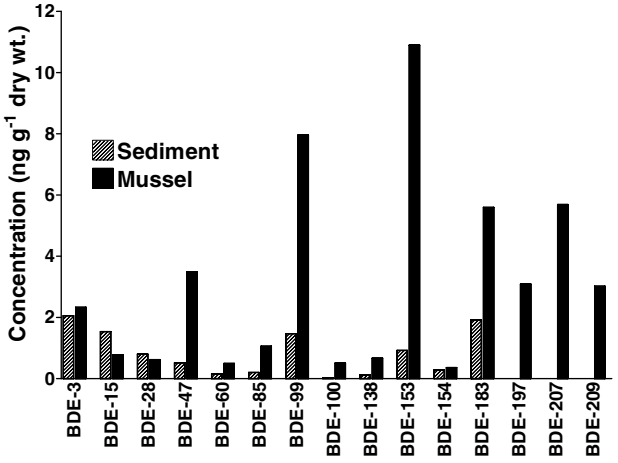

Fig. 5. Comparison of individual BDE congeners in mussels and sediments at Sta 7, Tsim Sha Tsui. Concentrations in ng g^{-1} dry wt.

sediments, these congeners were between non-detect to <2.0 ng g^{-1} dry wt, and the most prominent congeners in the sediment shifted toward the lower brominated species.

In conclusion, concentrations of PBDEs in sediments and mussels in Hong Kong coastal waters are high in comparison to those measured to date in other environments. The general distribution pattern of PBDEs in Hong Kong sediment and mussels reflects a gradient of locations from potential sources (such as industrialized areas, shipping, densely population areas) to less impacted areas (such as some of the Western and Eastern Zone sites). Some exceptions to this general pattern were found with a few highly PBDE-influenced sites in the Western (Dung Chung Bay, Sta 15) and Eastern Zone (Sai Kung, Sta 18). Whilst mussels and sediments showed generally similar patterns of $\sum_{15}$PBDE distributions, there were anomalies in the patterns of specific congeners. The presence of high concentrations of PBDEs in sediments and mussels, as well as the presence of congeners not reported in high concentrations in other areas of the globe, is of potential concern for Hong Kong and the Pearl River Delta.

3.4. Research recommendations

It is clear that PBDEs are ubiquitous environmental contaminants and occur in relatively high concentrations in surficial sediments and mussels collected from Hong Kong waters. The patterns of distribution, the mechanisms of their entry into, and their distribution throughout Hong Kong and the Pearl River Delta are unclear at this time. The risks of exposure to wildlife species and humans via ingestion, inhalation, consumption of contaminated food, and other potential routes of exposure are strongly suggested by our research findings. Research on PBDE distributions in various environmental compartments is urgently needed. More high quality, congener-specific data are required to determine sources, and to develop models and estimates of distribution. There is a need to compare PBDE data within and beyond the Hong Kong-Pearl River Delta system, and to determine time and geographical trends, along with mass quantities in humans and the environment. Certainly, the development of a consensus-based standard or reference material might further assist in clarifying the apparent differences in BDE detection and reporting.

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