Atmospheric Concentrations and Dry Deposition of Polybrominated Diphenyl Ethers in Southern Taiwan

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ABSTRACT

While polybrominated diphenyl ethers (PBDEs) have been used extensively for decades as flame retardants in a variety of consumer and commercial products, concerns about these substances have risen due to their adverse effects on human health. To the best of our knowledge, no study has reported on the dry deposition flux and velocity of individual PBDEs. This study was undertaken to investigate the dry deposition characteristics of PBDEs in the ambient air of southern Taiwan. The average atmospheric concentrations for total PBDEs (sum of thirty BDEs) ranged from 24.0 ± 1.83 to 102 ± 13.3 pg/Nm^3. The calculated dry deposition fluxes of total PBDEs were 13.4–60.4 ng/m^2-day, BDE 209 accounted for over 75% of the total PBDEs. The results showed that particle phase deposition dominated the dry deposition processes for PBDEs, and the same trends have been observed in other semi-volatile organic compounds (SOCs). The dry deposition velocities of individual PBDEs increased along with the number of brominated substitutes, ranging from 0.014 to 0.755 cm/s. Together with the results of the author's previous work, the deposition flux of total PBDEs could reach three orders of magnitude higher than those of PCDD/Fs (157–544 pg/m^2-day) and PCBs (89–1010 pg/m^2-day). Since atmosphere deposition is believed to be the main transfer pathway for SOCs entering food chains, its impact on human exposure to PBDEs is of great importance.

Keywords: Polybrominated diphenyl ethers (PBDEs); Gas-Particle partitioning; Dry deposition; Flux; Velocity.

INTRODUCTION

Polybrominated diphenyl ethers (PBDEs) are common additive brominated flame retardants (BFRs) that have been widely used in commercial and industrial products to prevent

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fires (WHO, 1994). PBDEs are commercially produced with three different formulations, i.e., penta-BDE, octa-BDE, and deca-BDE, with deca-BDE (with > 90% BDE209) accounting for 83% of total worldwide production in 2001 (La Guardia *et al.*, 2006). With similar physico-chemical properties to polychlorinated dibenzo-*p*-dioxins/dibenzofurans (PCDD/Fs) and biphenyls (PCBs), they are persistent, toxic, lipophilic, and bioaccumulative, and were highlighted as of particular concern in the Stockholm convention on persistent organic pollutants (POPs) in 2005 (WWF, 2005).

Due to the lack of chemical bonding between PBDEs and products, PBDEs may be released into the environment via manufacturing, usage, and disposal of BFR-containing products (de Wit, 2002; Agrell *et al.*, 2004). As a result of

both the large quantities of PBDEs consumed and their widespread distribution, they have become ubiquitous environmental pollutants, found in air, water, soil, fly ash, sediment, and human tissues (Gill *et al.*, 2004; Hites, 2004; Chao *et al.*, 2007; Artha *et al.*, 2011; Tu *et al.*, 2011; Wang *et al.*, 2011; Tu *et al.*, 2012). Concerns about polybrominated dibenzo-*p*-dioxins and dibenzofurans (PBDD/Fs) have also increased, because PBDD/Fs can be formed during thermal processing of products containing BFRs (Lai *et al.*, 2007).

Atmospheric deposition has been recognized as an important route for the long-range transportation of air pollutants entering the ecosystem. When semi-volatile organic compounds (SOCs), such as PCDD/Fs, PCBs, and PBDEs, are emitted into the air, they can be partitioned between the gas and particulate phases related to their vapor pressures, temperature, and concentrations of total suspended particulates in the atmosphere (Hoff *et al.*, 1996; Lohmann and Jones, 1998; Tasdemir *et al.*, 2004). Gas/particle partitioning is therefore an important factor in understanding the environmental fate of PBDEs and other SOCs. Recently, several studies describing gas-particle partitioning, as well as the atmospheric PCDD/Fs and PCBs deposition from relevant sources in Taiwan, have been published (Lee *et al.*, 1996; Shih *et al.*, 2006; Lee *et al.*, 2009; Wang *et al.*, 2010d; Huang *et al.*, 2011; Mi *et al.*, 2012), however, there is still very little information on the atmospheric deposition of PBDEs.

This study is an extension of our previous research, which focused on the dry deposition characteristics of PBDEs in the ambient air of industrial, urban, and rural areas in southern Taiwan. The simulated values for the gas-particle partitioning of PBDEs were compared with those obtained by direct measurement. Dry deposition fluxes and velocities of PBDEs were then predicted by calculations based on the model.

MATERIALS AND METHODS

PBDEs Sampling

Ambient air samples were collected at four locations, one industrial area, two urban areas, and one rural area located in Tainan city, Taiwan. A total of six ambient samples in each area were collected simultaneously by using a PS-1 sampler (Graseby Andersen, GA) according to the revised EPA Reference Method T09A between November 2010 and May 2011. Particle phase PBDEs were collected on quartz filters, and those in the gas phase were collected in a modified cartridge containing polyurethane foam. A detailed description of sampling sites and procedures, as well as meteorological information for the sampling sites during the periods examined, are given in our previous work (Mi *et al.*, 2012).

Analyses of PBDEs

All the chemical analyses in this study were performed in the Super Micro Mass Research and Technology Center in Cheng Shiu University, certified by the Taiwan EPA for PCDD/Fs sampling and analyses. PBDE analyses were performed following U.S. EPA Method 1614. Each sample was spiked with a known internal standard (10 congeners:

$^{13}C_{12}$ BDE-15, -28,-47, -99, -154, -153, -183, -197, -207, and -209) before Soxhlet extraction to monitor the extraction and cleanup procedures. After extraction, the extract was concentrated and treated with sulfuric acid, followed by a series of cleanup and fraction procedures, including a multi-layered silica column, an alumina column, and an activated carbon column (Wang *et al.*, 2010a). A high resolution gas chromatograph with a mass spectrometer (HRGC/HRMS) was used for PBDE analyses. The HRGC (Hewlett-Packard 6970 Series gas, CA) was equipped with a DB-5HT capillary column (J&W Scientific, CA) and with a splitless injector. The HRMS (Micromass Autospec Ultima, Manchester, UK) had a positive electron impact (EI+) source. Detailed analytical procedures and instrumental parameters of PBDEs are given in our previous works (Wang *et al.*, 2010a, b, c; Wang *et al.*, 2011).

Gas-Particle Partitioning

Several researchers have used a temperature-dependent partitioning coefficient (K_p) to describe the partitioning of SOCs between gas and particle phases (Mandalakis *et al.*, 2002; Lin *et al.*, 2010; Wang *et al.*, 2010d; Huang *et al.*, 2011).

$$K_p = \frac{F/TSP}{A} \tag{1}$$

where K_p ($m^3/\mu g$) is a temperature-dependent partitioning constant, TSP ($\mu g/m^3$) is the total suspended particulate concentration, and F (pg/m^3) and A (pg/m^3) are the associated particulate and gaseous concentrations of the analyte, respectively.

When the log K_p is regressed against the logarithm of the subcooled liquid vapor pressure log P_L^o, the partitioning constant can be calculated from the slope m_r and y-intercept of the trend line b_r (Yamassaki *et al.*, 1982), as shown in Eq. (2).

$$\log K_p = m_r \log P_L^o + b_r \tag{2}$$

The y-intercept of the trend line (b_r) depends mainly on the properties associated with SOC. Complete datasets on the gas-particle partitioning of PBDEs at Guangzhou City in southern China have been reported by Chen *et al.* (2006), giving values of $m_r = -0.607$ and $b_r = -5.07$ with $R^2 = 0.632$. Due to the lack of measured data for the gas-particle partitioning of PBDEs, same parameters measured by Chen *et al.* (2006) were adopted in this study.

The sub-cooled liquid vapor pressure P_L^0 (Pa) was evaluated from solid vapor pressure P_S^0 (Pa) using the following equation (Palm *et al.*, 2002):

$$P_L^o = P_S^o/F_r \tag{3}$$

$$log\ F_r = -6.79\ (T_M - T)/(2.303\ T) \tag{4}$$

where F_r is the fugacity ratio, T_M is the melting point of the BDE congener (K), and T is the atmospheric temperature (K).

Atmospheric Dry Deposition

The atmospheric deposition flux of total PBDEs is a combination of both gas- and particle- phase fluxes, which is given by

$$F_T = F_g + F_p \tag{5}$$

$$C_T \times V_{d,T} = C_g \times V_{d,g} + C_p \times V_{d,p} \tag{6}$$

where F_T is the total PBDE deposition fluxes contributed from both gas and particle phases, F_g and F_p are deposition fluxes of PBDEs contributed by the gas phase and particle phase, respectively, C_T is the measured concentrations of total PBDEs in air, $V_{d,T}$ is the dry deposition velocities of total PBDEs, C_g and C_p are the calculated PBDE concentrations in the gas phase and particle phase, respectively, and $V_{d,g}$ and $V_{d,p}$ are the dry deposition velocities of PBDEs in the gas phase and particle phase, respectively.

Dry deposition of gas-phase SOCs is controlled mainly by diffusion, while that of particle-phase is deposited mainly by the gravitational settling. Lee *et al.* (1996) reported that the dry deposition velocity of gaseous SOCs is fairly constant as compared to that of their particle phases. A value of 0.01 cm/s for dry deposition velocity ($V_{d,g}$) of gas-phase polycyclic aromatic hydrocarbon (PAH), proposed by Sheu and Lee (1996) and used by Lee *et al.* (1996), is adopted in this work for the calculation of the PBDE deposition flux contributed by their gas phases. Because of the lack of measured data for the deposition velocities of PBDEs, the average of 0.8 cm/s for particle-bond PBDEs determined by Su *et al.* (2007) was also used here for the approximate calculation of particle deposition flux.

RESULTS AND DISCUSSION

PBDE Concentrations in the Ambient Air

The atmospheric concentration of total PBDEs (sum of thirty BDEs) ranged from 67.3 ± 26.5 to 102 ± 13.3 and 24.0 ± 1.83 to 64.4 ± 24.0 pg/m³ (mean ± standard error) in fall and spring, respectively (Table 1). The highest concentration of total atmospheric PBDEs was found in the urban area among the four sampling areas. Higher atmospheric PBDE concentrations in industrial sites than those in urban and residential areas have been recently reported (Cetin and Odabasi, 2008; Choi *et al.*, 2008), due to leakage from consumer products and industrial facilities that manufacture PBDEs. As noted in our previous work, sampling site I is located near a science-based industrial park dominated by electronics and semiconductor firms (Mi *et al.*, 2012). A much higher level of PBDEs would thus be expected for

site I compared to the other sites, although this was not found in the current study. Therefore, the concentrations of atmospheric PBDEs may represent the usage level of PBDE-containing products (Wang *et al.*, 2011). As shown in Table 1, the higher PBDEs concentration in fall rather than in spring could be attributed to several loss processes, including photolysis, chemical reactivity, wet and dry deposition, and scavenging by vegetation (Duarte-Davidson *et al.*, 1997). Similar trends for other SOCs were also observed in earlier studies (Agree *et al.*, 2004; Huang *et al.*, 2011; Mi *et al.*, 2012).

The results for the atmospheric PBDE levels (the sum of all seven congeners) in the suburban (21–24 pg/m³), urban (32–40 pg/m³), and industrial areas (53–117 pg/m³) of Turkey (Cetin and Odabasi, 2008), in an urban area (100 pg/m³, sum of 26 congeners) of Chicago, USA (Hoh and Hites, 2005), and in an urban area (35.3 ± 15.5 pg/m³, sum of 30 congeners) of Taiwan (Wang *et al.*, 2011), were all comparable with our findings (Table 2). In Guangzhou City, the largest urban center in southern China, with a dense population and heavy traffic, Chen *et al.* (2006) observed very high atmospheric levels of the summed 11 congeners (347–577 pg/m³). These results are higher than those reported here, despite the variation among the number of PBDEs detected in both studies. Additionally, the contribution of BDE-209 to the total PBDEs concentration varies from 37 to 100% in these earlier studies, and this is likely to be due to the increased use of deca-BDE formulations in recent years.

Deca-BDE and nona-BDE homologues, the highly brominated-substituted congeners, were the most dominant PBDE congeners in all sampling sites (Fig. 1). BDE-209 was the most abundant congener in all sites (more than 70%), followed by BDE-208, -207, -206, and -184. Similarly, BDE-209 has been shown to be relatively abundant in most ambient air samples in the USA (Hoh and Hites, 2005), Taiwan (Wang *et al.*, 2011), and Turkey (Cetin and Odabasi, 2008). However, the percentage contribution by tetra-BDE and penta-BDE homologues (mainly BDE-47 and BDE-99) in this study was relatively low compared to that found in other works. This is probably due to the change in the formulation of commercial PBDEs from penta-BDE to deca-BDE (Agrell *et al.*, 2004).

Gas-Particle Partitioning of PBDEs

The subcooled liquid vapor pressure (P_L^o) and gas-particle partitioning constant (K_p) for individual PBDEs in the ambient air, as well as gas-particle partitioning, were calculated using the regression models mentioned above. The mean partitioning fractions of particle-phase PBDEs

Table 1. Mean concentrations of PBDEs in the ambient air of four different areas.

PBDEs (pg/m³)	Fall (n = 4)				Spring (n = 2)			
	I	A	B	R	I	A	B	R
Σ_{2-8} BDE	10.7 ± 3.71	14.1 ± 5.97	11.7 ± 2.08	11.5 ± 2.97	7.44 ± 3.41	10.9 ± 0.846	9.60 ± 0.957	5.20 ± 0.446
Σ_{9-10} BDE	56.6 ± 25.9	87.9 ± 14.5	89.6 ± 28.6	60.2 ± 20.6	33.4 ± 6.25	46.7 ± 7.07	54.8 ± 23.1	18.8 ± 2.27
Total PBDEs	67.3 ± 26.5	102 ± 13.3	101 ± 28.6	71.1 ± 21.7	40.8 ± 9.67	57.6 ± 6.23	64.4 ± 24.0	24.0 ± 1.83

n: sample numbers, Σ_{2-8} BDE: sum of di- to octa-BDE, Σ_{9-10} BDE: sum of nona- to deca-BDE.

Table 2. Comparison of PBDE concentrations in the ambient air from different countries.

Country	Location	City	Total PBDEs (pg/m^3)	BDE-209 (%)	Sampling time	References
America	Remote area	Michigan	16.2	37	2002–2003	Hoh and Hites (2005)
	Urban	Chicago	100	100		
	College town	Indiana	19.0	66		
	Agricultural region	Arkansas	30.0	77		
	Remote area	Louisiana	16.4	73		
China	Urban	Guangzhou	347	72	Jun, 2004	Chen *et al.* (2006)
	Industrial		577	70		
Japan	-	Osaka	104–347	96	2001	Ohta *et al.* (2002)
	-	Kyoto	4.4~80	19–74	2000–2001	Hayakawa *et al.* (2002)
Turkey	Suburban	Izmir	24; 21	79; 52	Sep, 2004; Feb,2005	Cetin and Odabasi (2008)
	Urban		32; 40	72; 72	Sep, 2004; Mar,2005	
	Industrial		117; 53	46; 61	Jun, 2005; Mar,2005	
Sweden	Vicinity of MSW	Malmo	18.6	56	2001–2002	Agrell *et al.* (2004)
	Urban		10.9	60		
Taiwan	Heavy steel complex area	Kaohsiung	165 ± 65.0	73	Mar–Apr, 2009	Wang *et al.* (2011)
	Metals complex area		93.9 ± 24.5	-		
	Urban		35.3 ± 15.5	51		
	Industrial	Tainan	40.8 ± 9.67– 67.3 ± 26.5	70–75	Nov, 2010–May 2011	This study
	Urban		57.6 ± 6.23– 102 ± 13.3	70–76		
	Urban		64.4 ± 24.0– 101 ± 28.6	78-80		
	Rural		24.0 ± 1.83– 71.1 ± 21.7	66–75		

in the ambient of four different areas are shown in Tables 3 and 4. Good agreement is found between the experimentally measured and simulated fractions of particulate PBDEs at all sites. The fractions of particulate PBDEs increased along with the number of brominated substitutes, and the highly brominated-substituted congeners (hepta- to deca-BDE) were mainly in the particle phase (> 60%). This is probably because gas-particle partitioning was controlled significantly by the chemical and physical properties of individual compounds, such as vapor pressure. Similar results were also observed elsewhere (Chen *et al.*, 2006; Cetin and Odabasi, 2008). Additionally, individual PBDEs bound to particles increased slightly with decreasing temperature, and the highest level was observed in fall (temperature ranging from 21.3°C to 23.3°C), while the lowest was in spring (temperature ranging from 24.4°C to 26.8°C). Similarly, relatively high levels of PCDD/Fs and PCBs in the particle phase during cold season were observed in the authors' previous work, and this is mainly caused by atmospheric variables, including temperature, wind speed, and wind direction.

The different percentages of PBDEs found by the two methods ranged from –45.2% (deca-BDE) to 78.1% (hexa-BDE) (Tables 4 and 5). The positive values resulted from the adsorption of gas-phase SVOCs onto the filter surface, whereas the negative values occurred due to volatilization (blow-off) of SVOCs from particles on the filter (Bidleman and Harner, 2000) or breakthrough of the compounds from the gas-phase sorbent (Lee and Jones, 1999). The results show

that the influence of the blow-off mechanism was observed mostly from highly brominated-substituted congeners (nona- and deca-BDE) in both seasons. Mandalakis and Stephanou (2007) reported that sampling artifacts (evaporation losses and/or adsorption onto filters) during the collection of particulate PBDEs, as well as the uncertainties inherent in model predictions, may bias the results of both approaches. Slight differences (< 30%) were observed between the measured and simulated particle-bound fractions of tri- and tetra-BDE, and the greatest differences were found for penta- and hexa-BDE. These findings may be attributed to the greater analytical errors found with penta- and hexa-BDE.

Dry Deposition Fluxes of PBDEs

The mean dry deposition fluxes of total PBDEs measured at four sites ranged from 39.0 to 60.4 ng/m^2-day and 13.4 to 37.6 ng/m^2-day in fall and spring, respectively (Tables 5 and 6). The results reveal that particle bond deposition contributed more than 99% of total dry deposition flux at all sites, which is similar to the observations of dry deposition flux of PCDD/Fs and PCBs reported in the authors' previous work. This may be due to the higher amounts of particles in the atmosphere of the investigated areas. The dry deposition fluxes of total PBDEs determined from this study were approximately two times higher than those observed at Lake Maggiore in northern Italy (17.6 ng/m^2-day, sum of eight congeners) (Mariani *et al.*, 2008), and at an industrial urban reference site in southern Sweden (18.2 ng/m^2-day, sum of nine congeners) (ter Schure *et al.*, 2004). The differences
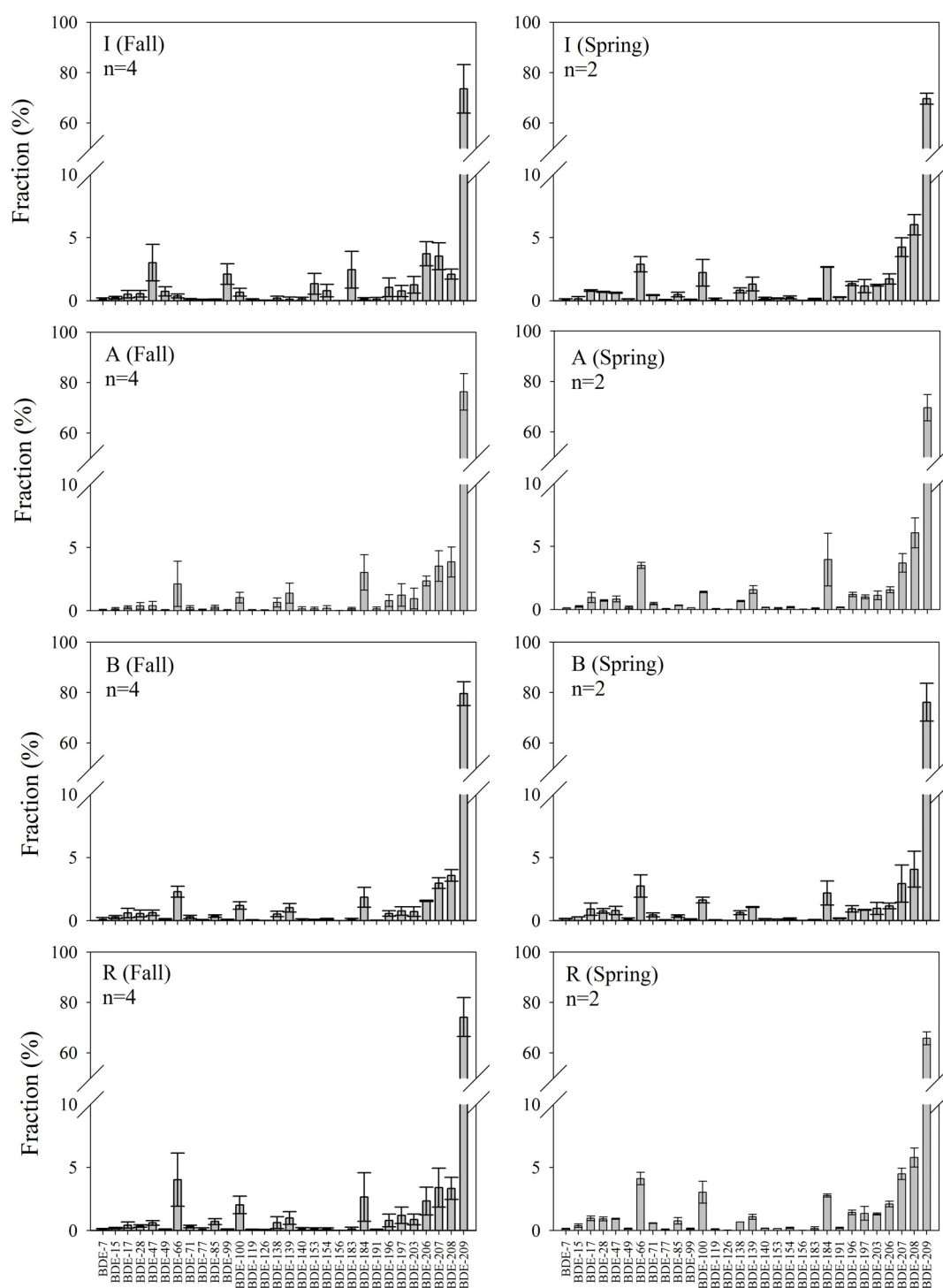
Fig. 1. Congener profiles of PBDEs in the ambient air of four different areas.

in the fluxes can be attributed to variations in ambient concentrations, sampling sites, durations, and the number of PBDEs detected. With regard to the relative contribution of individual congeners, BDE-209 was predominant among 30 PBDEs, accounting for 76.6% to 87.1% of the total dry flux, followed by nona-BDE (BDE-207 and BDE-206). The high contribution of BDE-209 to total dry flux may reflect the changes in the production and usage pattern from the commercial penta- to deca-BDE (ter Schure *et al.*, 2004).

The estimated monthly fluctuations of dry deposition fluxes of PBDEs in the ambient air of the four sampling sites are given in Fig. 2. The total dry deposition fluxes for all sampling sites had the highest level in cold season (December) and the lowest level in warm season (May), and these decreased as the temperature increased. The same trend in atmospheric deposition fluxes of PCDD/Fs and PCBs was previously observed in Mi *et al.* (2012). The seasonal fluctuation of PBDEs flux can be explained by

Table 3. Mean partitioning fractions of particle-phase PBDEs in the ambient air of four different areas in fall.

Site	I			A			B			R		
Phase	P^m (%)	P^s (%)	D (%)	P^m (%)	P^s (%)	D (%)	P^m (%)	P^s (%)	D (%)	P^m (%)	P^s (%)	D (%)
BDE-7	2.02	0.862	1.16	3.64	0.783	2.86	22.3	0.812	21.5	1.00	0.858	0.145
BDE-15	1.54	0.372	1.17	2.20	0.340	1.85	11.4	0.334	11.0	1.23	0.366	0.864
BDE-17	3.02	2.46	0.554	4.04	2.18	1.86	14.6	2.19	12.4	2.44	2.43	0.006
BDE-28	2.85	2.49	0.368	6.44	2.31	4.14	11.8	2.21	9.63	3.11	2.43	0.684
BDE-49	11.1	6.93	4.21	13.3	6.56	6.70	19.3	6.27	13.0	7.38	6.77	0.613
BDE-71	10.6	6.91	3.74	17.5	6.45	11.1	21.3	6.22	15.0	6.29	6.81	–0.519
BDE-47	12.5	7.01	5.51	15.7	6.57	9.16	19.2	6.29	12.9	11.3	6.86	4.40
BDE-66	15.4	6.95	8.41	18.7	6.51	12.2	23.7	6.22	17.4	16.9	6.80	10.1
BDE-77	34.6	6.85	27.8	36.7	6.56	30.1	43.4	6.23	37.2	11.0	6.92	4.07
BDE-100	40.6	2.19	38.4	42.2	2.03	40.1	43.8	1.98	41.8	28.0	2.12	25.9
BDE-119	51.4	2.13	49.3	52.4	2.00	50.4	61.0	1.95	59.0	33.4	2.14	31.2
BDE-99	53.1	2.18	50.9	50.4	2.01	48.4	55.1	1.97	53.2	43.9	2.12	41.7
BDE-85	75.2	2.16	73.1	61.8	2.03	59.8	72.8	1.98	70.8	62.9	2.12	60.7
BDE-126	86.1	2.15	84.0	81.2	2.10	79.1	84.0	1.96	82.1	54.7	2.17	52.6
BDE-154	84.4	23.0	61.4	67.0	21.7	45.2	79.8	21.5	58.4	63.9	23.0	41.0
BDE-153	93.3	22.9	70.3	71.7	21.7	50.0	87.5	21.3	66.2	87.9	22.5	65.4
BDE-139	90.3	23.0	67.4	61.1	21.7	39.4	84.0	20.9	63.1	84.2	22.8	61.4
BDE-140	94.2	22.9	71.2	67.8	21.7	46.1	91.1	21.4	69.7	88.7	22.9	65.9
BDE-138	96.3	23.0	73.3	69.9	21.7	48.1	94.5	21.5	73.0	91.3	22.7	68.6
BDE-156	96.5	23.0	73.5	4.08	21.5	–17.4	50.0	21.3	28.7	98.3	23.4	74.9
BDE-184	97.1	66.7	30.4	75.6	64.8	10.8	95.9	65.7	30.2	93.6	67.2	26.4
BDE-183	98.1	66.5	31.6	81.0	65.1	15.9	94.1	64.7	29.4	97.9	66.2	31.7
BDE-191	98.0	66.5	31.5	88.2	65.1	23.2	76.4	64.6	11.8	93.9	65.6	28.3
BDE-197	98.1	86.1	12.0	77.3	85.2	–7.92	93.7	85.0	8.75	97.2	85.8	11.4
BDE-203	95.8	86.0	9.83	77.3	85.1	–7.74	96.1	84.9	11.2	89.1	85.7	3.37
BDE-196	96.7	86.0	10.6	70.5	85.1	–14.6	95.5	84.9	10.6	88.5	85.7	2.82
BDE-208	80.3	95.1	–14.8	78.3	94.6	–16.2	87.4	94.6	–7.19	79.6	95.1	–15.4
BDE-207	83.7	95.1	–11.4	74.9	94.6	–19.7	90.0	94.6	–4.58	81.3	95.0	–13.7
BDE-206	74.9	95.1	–20.3	72.7	94.4	–21.7	89.9	94.6	–4.79	70.1	95.0	–24.9
BDE-209	54.5	93.6	–39.1	68.2	92.7	–24.6	77.6	92.9	–15.4	48.2	93.4	–45.2

P^m: measured partitioning percentages of particle-phase PBDEs.
P^s: simulated partitioning percentages of particle-phase PBDEs.
D: percentages of difference evaluated by $P^m - P^s$.

Table 4. Mean partitioning fractions of particle-phase PBDEs in the ambient air of four different areas in spring.

Site	I			A			B			R		
Phase	P^m (%)	P^s (%)	D (%)	P^m (%)	P^s (%)	D (%)	P^m (%)	P^s (%)	D (%)	P^m (%)	P^s (%)	D (%)
BDE-7	3.53	0.940	2.59	0.736	0.826	–0.090	0.471	0.824	–0.353	1.74	0.962	0.778
BDE-15	5.13	0.405	4.73	1.76	0.356	1.40	1.40	0.359	1.04	1.84	0.413	1.42
BDE-17	3.13	2.63	0.500	1.50	2.33	–0.823	2.13	2.36	–0.230	2.28	2.73	–0.446
BDE-28	3.45	2.65	0.796	1.00	2.36	–1.35	2.31	2.37	–0.053	2.27	2.72	–0.456
BDE-49	8.76	7.47	1.29	5.29	6.61	–1.32	4.90	6.66	–1.77	8.05	7.64	0.417
BDE-71	4.67	7.40	–2.73	5.20	6.56	–1.36	5.08	6.68	–1.61	5.74	7.62	–1.88
BDE-47	19.3	7.47	11.9	10.1	6.64	3.50	7.83	6.68	1.16	7.38	7.67	–0.283
BDE-66	13.3	7.42	5.87	10.0	6.62	3.40	10.4	6.63	3.78	9.95	7.62	2.33
BDE-77	17.2	7.41	9.77	18.0	6.71	11.3	19.7	6.68	13.0	18.5	7.67	10.8
BDE-100	36.3	2.38	33.9	23.8	2.11	21.6	21.6	2.11	19.4	19.6	2.43	17.2
BDE-119	30.0	2.36	27.6	29.3	2.08	27.2	25.1	2.10	23.0	19.0	2.42	16.6
BDE-99	49.9	2.37	47.5	37.0	2.11	34.9	33.4	2.10	31.3	28.0	2.43	25.6
BDE-85	72.9	2.36	70.5	51.5	2.12	49.4	52.4	2.10	50.3	43.6	2.43	41.1
BDE-126	72.5	2.37	70.1	60.8	2.16	58.6	68.5	2.12	66.4	51.8	2.45	49.3
BDE-154	61.5	24.8	36.6	56.4	22.8	33.6	63.9	22.7	41.2	45.3	25.4	19.9
BDE-153	77.4	24.9	52.6	75.0	22.8	52.2	84.9	22.7	62.2	71.4	25.4	46.0
BDE-139	78.3	24.7	53.6	63.9	22.8	41.1	81.0	22.4	58.5	67.8	25.4	42.5

Table 4. *(continued).*

Site	I			A			B			R		
Phase	P^m (%)	P^s (%)	D (%)	P^m (%)	P^s (%)	D (%)	P^m (%)	P^s (%)	D (%)	P^m (%)	P^s (%)	D (%)
BDE-140	85.8	24.7	61.1	73.5	22.9	50.6	82.4	22.5	59.9	80.6	25.4	55.2
BDE-138	89.3	24.8	64.6	88.8	22.8	65.9	94.6	22.6	72.0	88.7	25.3	63.4
BDE-156	100	24.6	75.4	100	22.6	77.4	100	21.9	78.1	100	25.5	74.5
BDE-184	71.5	69.0	2.51	80.0	66.7	13.3	86.4	66.5	19.9	83.4	69.5	13.9
BDE-183	94.6	68.9	25.7	95.5	66.3	29.1	97.9	66.4	31.5	92.2	69.6	22.7
BDE-191	97.8	68.5	29.3	97.0	66.3	30.7	98.1	65.4	32.7	97.3	69.3	27.9
BDE-197	97.9	87.2	10.7	97.0	85.9	11.1	98.7	85.8	12.9	93.8	87.6	6.27
BDE-203	97.9	87.3	10.5	97.1	86.0	11.1	98.8	85.9	12.9	94.6	87.6	6.98
BDE-196	97.9	87.3	10.7	97.5	86.0	11.5	98.9	85.9	13.0	93.0	87.6	5.41
BDE-208	93.1	95.6	−2.55	94.2	95.1	−0.942	96.1	95.1	1.0	82.2	95.7	−13.5
BDE-207	92.9	95.6	−2.67	93.7	95.1	−1.33	96.0	95.1	0.9	79.1	95.7	−16.6
BDE-206	92.8	95.5	−2.79	93.8	95.0	−1.24	96.2	95.1	1.1	77.2	95.7	−18.5
BDE-209	87.3	94.2	−6.91	91.0	93.5	−2.55	95.6	93.5	2.1	67.7	94.4	−26.6

P^m: measured partitioning percentages of particle-phase PBDEs.
P^s: simulated partitioning percentages of particle-phase PBDEs.
D: percentages of difference evaluated by $P^m - P^s$.

Table 5. Dry deposition fluxes of PBDEs in the ambient air of four different areas in fall (pg/m^2-day).

Site	I		A		B		R	
Phase	$F_{d,T}$[a]	P %[b]	$F_{d,T}$[a]	P %[b]	$F_{d,T}$[a]	P %[b]	$F_{d,T}$[a]	P %[b]
BDE-7	1.02	41.0	0.864	38.7	1.19	39.6	1.01	40.9
BDE-15	1.63	23.0	1.52	21.5	2.74	21.2	1.43	22.7
BDE-17	8.01	66.9	6.11	64.0	12.5	64.2	7.45	66.6
BDE-28	9.17	67.1	8.99	65.4	12.3	64.4	6.10	66.5
BDE-49	26.4	85.6	21.4	84.9	30.0	84.2	22.6	85.3
BDE-71	3.59	85.6	2.83	84.7	4.65	84.1	2.89	85.4
BDE-47	118	85.8	113	84.9	116	84.3	156	85.5
BDE-66	13.7	85.7	12.5	84.8	13.3	84.1	11.3	85.4
BDE-77	2.15	85.5	3.37	84.9	2.60	84.2	3.51	85.6
BDE-100	10.3	64.1	6.21	62.3	7.61	61.8	11.2	63.4
BDE-119	1.18	63.5	1.18	62.0	1.44	61.4	1.03	63.7
BDE-99	32.4	64.1	23.4	62.2	25.8	61.7	31.9	63.4
BDE-85	1.36	63.9	1.31	62.3	1.07	61.7	1.17	63.4
BDE-126	0.307	63.7	0.728	63.2	0.279	61.5	0.583	64.0
BDE-154	77.7	96.0	104	95.7	77.4	95.6	70.0	96.0
BDE-153	130	96.0	219	95.7	150	95.6	108	95.9
BDE-139	11.2	96.0	21.8	95.7	8.23	95.5	13.6	95.9
BDE-140	13.6	96.0	22.3	95.7	10.3	95.6	12.5	96.0
BDE-138	20.0	96.0	30.9	95.7	18.1	95.6	14.9	95.9
BDE-156	0.476	96.0	0.381	95.6	0.031	95.6	0.991	96.1
BDE-184	39.0	99.4	71.9	99.3	42.8	99.4	37.3	99.4
BDE-183	671	99.4	1,375	99.3	806	99.3	826	99.4
BDE-191	26.9	99.4	57.9	99.3	3.33	99.3	19.3	99.3
BDE-197	275	99.8	476	99.8	311	99.8	323	99.8
BDE-203	446	99.8	738	99.8	415	99.8	490	99.8
BDE-196	375	99.8	576	99.8	381	99.8	357	99.8
BDE-208	897	99.9	1,559	99.9	1,033	99.9	1,074	99.9
BDE-207	1,476	99.9	2,372	99.9	1,946	99.9	1,587	99.9
BDE-206	1,600	99.9	2,613	99.9	2,352	99.9	1,561	99.9
BDE-209	32,754	99.9	49,980	99.9	52,365	99.9	34,736	99.9
Σ_{2-8} BDE	2,315	97.2	3,898	98.1	2,454	97.1	2,531	97.3
Σ_{9-10} BDE	36,727	99.9	56,524	99.9	57,696	99.9	38,958	99.9
Total PBDEs	39,042	99.8	60,421	99.8	60,151	99.8	41,489	99.8

[a] $F_{d,T}$: Total deposition flux of PBDEs, [b] P %: The ratio of dry deposition flux contributed by the particle-phase of PBDEs.

Table 6. Dry deposition fluxes of PBDEs in the ambient air of four different areas in spring (pg/m²-day).

Site	I		A		B		R	
Phase	$F_{d,T}$[a]	P %[b]	$F_{d,T}$[a]	P %[b]	$F_{d,T}$[a]	P %[b]	$F_{d,T}$[a]	P %[b]
BDE-7	0.415	41.6	0.872	39.9	1.37	40.2	0.479	43.6
BDE-15	0.679	23.4	1.55	22.4	2.06	22.4	1.03	24.9
BDE-17	8.68	68.3	13.1	65.7	13.0	65.8	6.21	69.1
BDE-28	7.49	68.4	10.3	66.1	11.8	66.0	5.95	69.1
BDE-49	15.0	86.4	25.4	85.0	25.6	85.0	13.4	86.8
BDE-71	3.34	86.4	5.19	84.9	3.42	84.9	2.15	86.7
BDE-47	69.3	86.5	108	85.1	89.2	85.1	59.2	86.8
BDE-66	10.7	86.4	14.3	85.0	14.6	85.1	8.34	86.8
BDE-77	1.83	86.5	1.99	85.1	2.07	85.2	1.33	86.8
BDE-100	4.77	66.0	4.40	63.2	5.11	63.2	4.54	66.5
BDE-119	0.903	65.8	1.62	63.1	1.33	63.1	0.868	66.2
BDE-99	22.0	66.0	18.3	63.2	23.5	63.3	18.1	66.5
BDE-85	1.12	66.3	0.663	63.4	0.982	63.4	0.664	66.5
BDE-126	0.199	66.0	0.120	64.1	0.167	63.8	0.107	66.6
BDE-154	58.7	96.3	62.0	95.9	62.2	95.9	29.5	96.4
BDE-153	93.6	96.4	145	95.9	113	95.9	46.6	96.4
BDE-139	13.3	96.4	14.2	95.9	14.4	95.9	7.36	96.4
BDE-140	13.4	96.3	10.2	96.0	12.5	95.9	6.60	96.4
BDE-138	18.3	96.4	17.1	95.9	17.1	96.0	8.98	96.4
BDE-156	0.640	96.5	0.462	96.1	0.705	96.1	0.303	96.5
BDE-184	24.6	99.4	23.8	99.4	23.8	99.4	17.9	99.5
BDE-183	521	99.4	1,015	99.4	594	99.4	319	99.5
BDE-191	53.3	99.4	46.3	99.4	53.2	99.4	23.8	99.5
BDE-197	331	99.8	403	99.8	337	99.8	207	99.8
BDE-203	279	99.8	345	99.8	323	99.8	194	99.8
BDE-196	303	99.8	377	99.8	335	99.8	189	99.8
BDE-208	460	99.9	591	99.9	474	99.9	332	99.9
BDE-207	1,152	99.9	1,382	99.9	1,123	99.9	715	99.9
BDE-206	1,633	99.9	2,276	99.9	1,610	99.9	923	99.9
BDE-209	18,548	99.9	26,033	99.9	32,324	99.9	10,299	99.9
Σ_{2-8} BDE	1,856	97.8	2,665	97.7	2,080	97.2	1,173	97.4
Σ_{9-10} BDE	21,793	99.9	30,282	99.9	35,530	99.9	12,268	99.9
Total PBDEs	23,649	99.8	32,947	99.7	37,610	99.8	13,442	99.7

[a] $F_{d,T}$: Total deposition flux of PBDEs, [b] P %: The ratio of dry deposition flux contributed by the particle-phase of PBDEs.

the different ambient concentrations and meteorological conditions (i.e., temperature, relative humidity, wind speed, and atmospheric stability) (Tasdemir and Holsen, 2005). Additionally, the deposition flux of total PBDEs can be three orders of magnitude higher than those of PCDD/Fs (157 to 544 pg/m²-day) and PCBs (89 to 1010 pg/m²-day). Since atmospheric deposition is believed to be the main transfer pathway of SOCs into food chains, its impact on human exposure to PBDEs is of great importance.

Dry Deposition Velocities of Individual PBDEs

In order to better understand the dry deposition process, the total deposition velocities of individual PBDEs were calculated, as shown in Fig. 3. The estimated deposition velocities of individual PBDEs ranged from 0.014 cm/s (di-BDE) to 0.748 cm/s (deca-BDE), with a mean of 0.679 cm/s in fall, and from 0.015 cm/s (di-BDE) to 0.755 cm/s (deca-BDE) with a mean of 0.664 cm/s in spring. The calculated deposition velocities of PBDEs increased along with the number of brominated substitutes, and similar

patterns of deposition velocities were observed for PCDD/Fs and PCBs in our previous work (Mi *et al.*, 2012). However, no significant seasonal variation was found for the total (gas + particulate phases) deposition velocity of PBDEs.

The total (gas + particulate phases) deposition velocity for BDE-209 calculated in the present work was comparable to that for particulate phases measured in Hong Kong (Li *et al.*, 2010) and urban Guangzhou in China (Zhang *et al.*, 2012), but significantly lower than the values detected for a site near a municipal solid waste incineration and electronics recycling plant in Sweden (ter Schure *et al.*, 2004) and in urban and suburban Izmir in Turkey (Cetin and Odabasi, 2007). The differences among deposition velocities could be attributed to the particle size distribution, sampling site, meteorological conditions, and different measurement techniques (ter Schure *et al.*, 2004). Finally, when compared with the total deposition velocity of other SOCs, the calculated values of PBDEs were in line with those reported for PCDD/Fs (0.031–0.546 cm/s) and PCBs (0.069–3.38 cm/s), as shown in our previous work (Mi *et al.*, 2012).
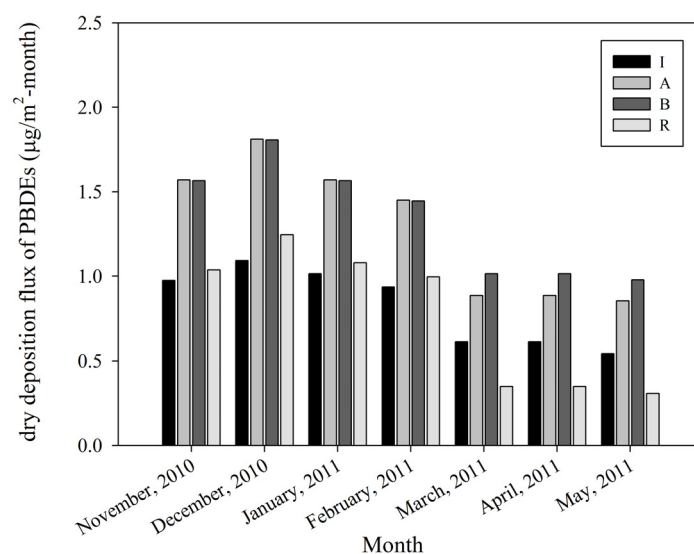
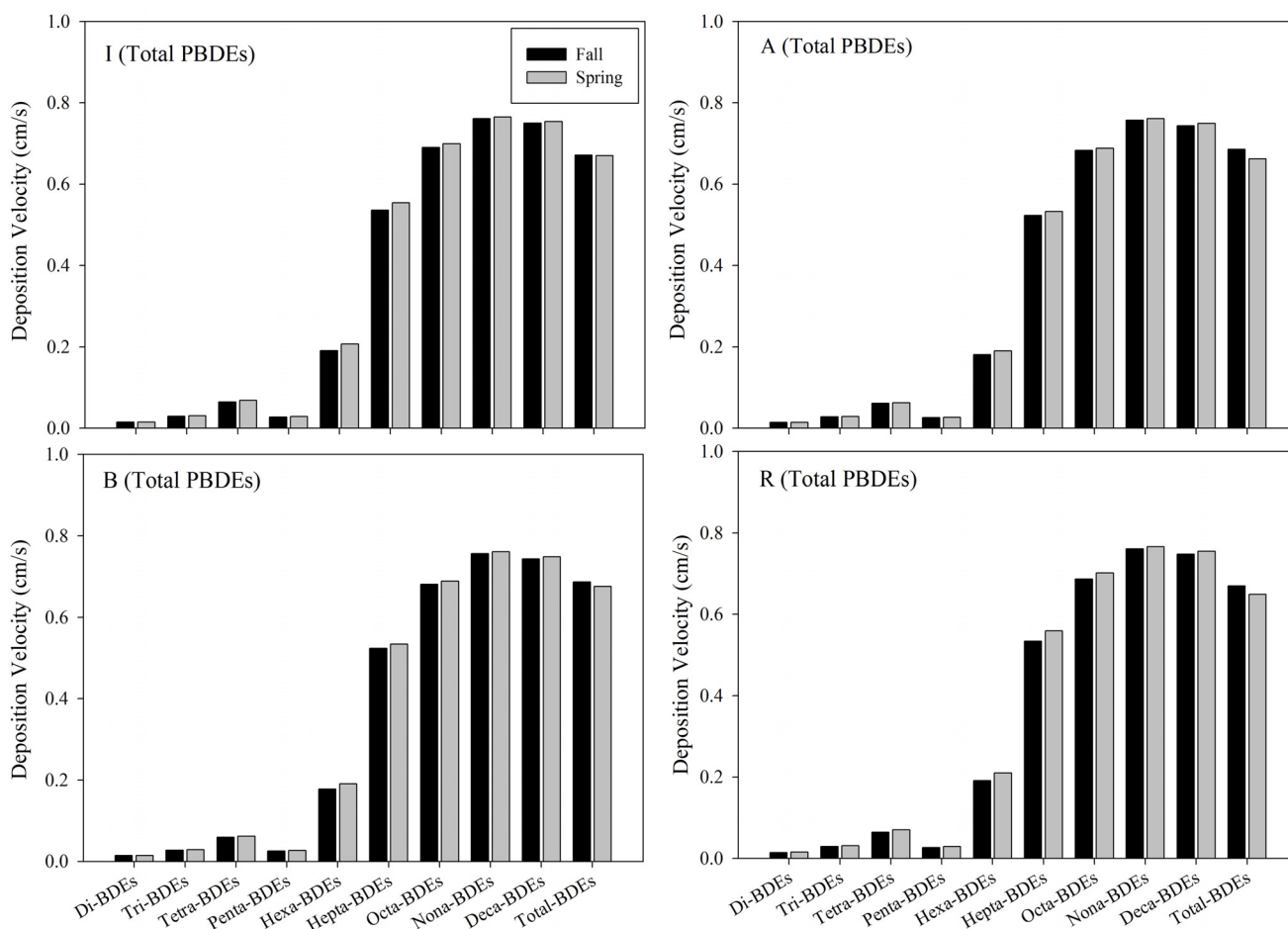**Fig. 2.** Estimated monthly dry deposition fluxes of PBDEs of four different areas.

Fig. 3. Mean dry deposition velocities of individual PBDEs of four different areas.

CONCLUSIONS

This study is an extension of our previous research, which focused on the dry deposition characteristics of PBDEs in the ambient air of southern Taiwan. Higher atmospheric

PBDE concentrations in urban sites than those in rural and industrial areas were observed, and these may indicate different usage levels with regard to PBDE-containing products. The fractions of particulate PBDEs increased along with the number of brominated substitutes, and the

highly brominated-substituted congeners were found to be predominantly associated with particles at all sites. The calculated deposition flux of total PBDEs was found to decrease as the temperature increased between 13.4 and 60.4 ng/m^2-day, and BDE 209 accounted for over 75% of the total PBDEs. The high contribution of BDE-209 to total flux may reflect the changes in production and usage patterns from the commercial penta- to deca-BDE mixtures. The deposition velocities of individual PBDEs increased along with the number of brominated substitutes, and ranged from 0.014 to 0.755 cm/s. Similar patterns of velocities have also been observed in our previous work (Mi *et al.*, 2012) for PCDD/Fs and PCBs.

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