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# Phase partitioning, concentration variation and risk assessment of polybrominated diphenyl ethers (PBDEs) in the atmosphere of an e-waste recycling site

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# ABSTRACT

The daytime and nighttime air samples were collected from an e-waste recycling site in Guiyu (GY) and its upwind and downwind direction sites in Chendian (CD) and Gurao (GR), respectively, during September and December 2005 to determine the phase partitioning, seasonal and diurnal variation of polybrominated diphenyl ethers (PBDEs). The three sites showed similar congener compositions with BDEs-28, -47, -99, -207 and -209 as the dominant congeners. The gas phase contribution ranged from  $42.1\% \pm 17.1\%$  of the total PBDEs in summer to  $9.0\% \pm 8.6\%$  in winter. Partitioning of PBDEs between gas and particulate phases was well correlated with the subcooled liquid vapor pressure  $(P_1^0)$  for all samples. Both Junge–Pankow adsorption model and  $K_{OA}$ -based absorption model fitted well the PBDEs data in winter but overestimated the sorption of PBDEs in summer. The mean atmospheric concentrations of BDEs-47 (2748 pg m<sup>-3</sup> in summer and 6146 pg m<sup>-3</sup> in winter) and -99 (1656 pg m<sup>-3</sup> in summer and 4911 pg m<sup>-3</sup> in winter) in GY were much higher than those from other places around the world. Additionally, the time series of PBDEs in GY demonstrated a pronounced diurnal variation pattern, but not CD and GR, where PBDEs appeared partly associated with regional accumulation. Source characterization from local emissions may be the major factor controlling the diurnal variation of PBDEs in GY air. The risk assessment showed that the total daily intakes of BDE-99 in GY was 9.0-34.9 ng kg<sup>-1</sup>-bw d<sup>-1</sup>, which is higher than the maximal allowed intake level (0.26 ng kg<sup>-1</sup>-bw d<sup>-1</sup>) with factors of 35–135, indicating possible health risk for GY residents with exposure to PBDEs.

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# 1. Introduction

Polybrominated diphenyl ethers (PBDEs) are a class of brominated flame retardants (BFRs), which are extensively used in electronic appliances, paints and textiles to prevent the propagation of fire. Recently, PBDEs have been testified to be persistent and ubiquitous in the environment (Hites, 2004), organism (Fair et al., 2007; Gallego et al., 2007), and even in human tissues (Bi et al., 2007; Covaci et al., 2008). Three major PBDE commercial mixtures (Penta-BDE, Octa-BDE and Deca-BDE) are commonly used. PBDEs were mainly used in the United States, especially the Penta-products, while the Deca-BDE products were mainly used in Europe and Asia. Due to health risks, technical commercial Penta- and Octa-BDE were banned in the European Union (Cox and Ethymiou, 2003), California and Maine, and industry voluntarily ceased production (Tullo, 2003).

The electronics industry is the world's largest and fastest growing manufacturing industry, and as a consequence of this growth, electrical and electronic waste (e-waste) such as computers, printers, mobile phones, television sets, stereos, and radios is generated in large quantities around the world. Every year, 20-50 million tonnes of e-waste are generated world-wide (DEWA/GRID-Europe, 2005). The e-waste generated throughout the world is being transported to developing countries. It is reported that about 80% of computer e-waste is exported to Asia, and 90% of these exports have been sent to China, illegally, for "recycling" (DEWA/GRID-Europe, 2005). Because the e-waste in China was generally recycled with primitive techniques without facilities to control or decrease pollutants release from e-waste processing, a lot of potential carcinogenic and hazardous substances such as toxic metals (e.g. mercury, lead and cadmium) and persistent organic pollutants (POPs, e.g. dioxins/furans, polychlorinated biphenyls and PBDEs) were released into the environment (Deng et al., 2006, 2007; Leung et al., 2007; Wong et al., 2007; Wen et al., 2009).

In this study, the e-waste recycling site was selected in Guiyu (GY) town, Shantou City, Guangdong Province, southeast China, with a total area of 52 km<sup>2</sup> and a population of 150,000. In this town, 80% of families are engaged in, and nearly 20,000 migrants are employed in, recycling work. Every year, about three millions





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tonnes of e-waste are transported into GY. The methods used to recycle e-waste in this town included chipping and melting plastics, burning coated wire to recover copper, removing electronic components from printed circuit boards and burning unsalvageable materials in the open air (Deng et al., 2006; Leung et al., 2007). Elevated levels of PBDEs have been found in the aerosol, soil, sediment and human body at GY (Deng et al., 2006, 2007; Bi et al., 2007; Leung et al., 2007; Wong et al., 2007).

There is an urgent need to quantify and understand the behaviour of PBDEs in the atmosphere of exposure region, which depends not only on the physicochemical properties of PBDEs but also on the meteorological conditions (Mandalakis et al., 2003; Gouin et al., 2005). Measurements with temporal variation can ascertain if these factors have a significant effect on PBDE levels and the location of possible sources. Additionally, the greatest health concern surrounding PBDEs is their toxic effects in humans (Birnbaum and Staskal, 2004: Costa and Giordano, 2007). In previous paper (Chen et al., 2009), we reported the diurnal variation of PBDEs in summer. This study was performed to help (1) understand the gas-particle partitioning of PBDEs, which is important for assessing their environmental fate and human exposure; (2) investigate and characterize the seasonal and diurnal variation of PBDEs; and (3) assess the risk of PBDEs in this e-waste recycling site.

# 2. Materials and methods

#### 2.1. Sampling sites

Air samples were taken from GY and its upwind and downwind direction sites in Chendian (CD) and Gurao (GR). CD is situated to the southwest of about 9 km away from GY, and GR is located approximately 10 km northeast of GY. The underwear industries are developed in CD and GR. The atmospheric conditions in the three sites are under the strong influence of the Asian monsoon system, with the southwesterly monsoons bringing in relatively clean air from the ocean in summer, and northeasterly winds bringing in air masses moving across northern cities in winter. The winter seasons are characterized by strong winds and dry weather, whereas the summer seasons are hot and humid due to occasional showers and thunderstorms.

#### 2.2. Sample collection

A detailed description of the methods used for sampling, extraction protocol and chemical analysis of air samples has been published elsewhere (Chen et al., 2006, 2009). Briefly, air samples were collected simultaneously from GY and CD during September 13-21 and from GY, CD and GR during December 18-25, 2005. During this period, the diurnal air samples were collected from about 9:00-17:00 as the daytime samples, and 23:00-7:00 as the nighttime samples. The gaps between day and nighttime sampling allowed for better identification of differences in PBDE concentrations. The air samples were drawn at about  $0.3-0.5 \text{ m}^3 \text{ min}^{-1}$ through the quartz fiber filters (QFFs,  $20.3 \text{ cm} \times 25.4 \text{ cm}$ , Whatman), and subsequently through 6.5 cm in diameter, 7.5 cm in thickness polyurethane foam (PUF) plugs by using a high-volume air sampler. Each sample acquisition lasted for about 8 h, and the sampling volume was in the range of 145-214 m<sup>3</sup>. A total of 33 daytime and 33 nighttime samples were collected.

# 2.3. Sample extraction and analysis

The extraction and cleanup procedures have been given elsewhere (Mai et al., 2005). Briefly, the PUF plugs and QFFs were spiked with PCB 209 and <sup>13</sup>C-PCB141, and were Soxhlet extracted with acetone: hexane mixture (1:1) for 72 h. Concentrated extracts were cleaned and fractionated on acid/basic multilayer silica gel columns with 70 mL of dichloromethane: hexane (1:1). The eluant containing PBDEs was concentrated to 200  $\mu$ L, and a known amount (5 ng) of recovery standard (<sup>13</sup>C-PCB208, Cambridge Isotope Laboratories) was added prior to instrumental analysis. The instrumental conditions, and quality assurance/quality control (QA/QC) were provided in Supplementary material.

# 2.4. Organic and elemental carbon determination

A Thermal/Optical Carbon Aerosol Analyzer (Sunset Laboratory, Forest Grove, OR) was used to determine organic carbon (OC) and elemental carbon (EC) contents based on thermal-optical transmittance (TOT) using the National Institute for Occupational Safety and Health (NIOSH) Method 5040 protocol.

# 2.5. Statistical analysis

Correlations between individual compounds, between target compounds and OC/EC or meteorological parameters were tested using the Pearson coefficients. Data were analyzed using the Statistical Product and Service Solutions (SPSS) software package.  $\sum$ PBDEs refer to the sum of all target PBDE congeners. Collection data, OC, total suspended particulate (TSP) and  $\sum$ PBDEs are shown in Table S1 of the Supplementary Material.

### 3. Results and discussion

# 3.1. Ambient air PBDE concentrations and profiles

The total PBDE concentrations in the gas and particulate phases are reported in Table 1. Average  $\sum$ PBDEs concentrations were in the range from 127 (CD-S) to 4627 pg m<sup>-3</sup> (GY-S) in PUF samples and from 222 (CD-S) to 38 974 pg m<sup>-3</sup> (GY-W) in the filter samples. The gas phase contribution ranged from 42.1 ± 17.1% of the total PBDEs in summer to 9.0 ± 8.6% in winter. The gas phase was dominated by the tri- to penta-BDEs, which accounted for 97.2–99.4% of the total PBDE gas phase concentration. The most abundant congeners for the particulate phase were BDEs-47, -99, -207 and -209, which constituted 65.5–72.3% of the total PBDE particulate phase concentration. These results were similar to those in the atmosphere of PBDEs in Guangzhou (Chen et al., 2006).

The total PBDE concentrations in air in this study ranged from 349 (CD-S) to 41 470 pg m<sup>-3</sup> (GY-W) (Table S2). As compared with the PBDEs data documented in the literature for ambient air around the world, of which the PBDE concentrations ranged from 5 to 580 pg m<sup>-3</sup> (Table S3), the PBDE concentrations in GY were extremely high, especially in winter, which was the highest concentrations yet reported in outdoor atmosphere. The average concentrations of BDE-47 in GY were even about 2-5 times of that in the dismantling hall of the recycling electrical plant of Sweden (Sjödin et al., 2001). However, BDE-209 concentration did not show as high as that in Swedish electrical recycling plant where the commercial Deca-BDE was the most abundant. Compared with another e-waste recycling site Taizhou in China (Li et al., 2008), GY showed much higher PBDE concentrations and lower BDE-209 percentage (Table S3). This result showed different flame retardants used in these e-waste recycling sites. Pearson correlation analysis indicated that PBDE congeners (excluding BDE-47 and -99) have been observed an apparent relationship with meteorological parameters temperature and wind speed at GY (Table S4). This suggests that local sources may have a major contribution to the PBDE concentrations in GY. The good correlation between individual PBDE

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PBDE concentrations in gas and particles (pg  $m^{-3}$ ).

	Summer				Winter					
	GY-S		CD-S		GY-W		CD-W		GR-W	
	Gas	Particle	Gas	Particle	Gas	Particle	Gas	Particle	Gas	Particle
BDE-17					306	365	33.6	9.65	38.0	41.0
BDE-28	788	29.3	22.0	1.22	964	1115	124	32.3	93.0	124
BDE-71					47.3	505	7.91	14.1	3.61	35.8
BDE-47	2365	383	65.2	29.9	863	5282	146	258	52.2	601
BDE-66	536	121	11.3	2.00	217	1850	37.75	91.8	9.62	175
BDE-100	80.7	52.6	3.45	1.97	19.2	992	2.22	37.0	0.79	79.6
BDE-99	806	850	20.8	15.8	63.7	4848	8.62	260	3.97	466
BDE-85	17.4	51.0	0.68	1.00	14.4	228	0.38	15.3	0.40	20.2
BDE-154	16.2	99.1	1.31	3.69	2.92	774	0.29	37.6	0.23	50.4
BDE-153	16.3	320	1.22	9.48	3.66	1030	0.35	64.9	0.47	87.4
BDE-138	0.55	45.7	0.07	0.93	0.10	378	0.08	17.2	0.14	19.8
BDE-183	1.69	334	0.65	14.6	0.02	1188	0.23	61.9	0.03	53.2
BDE-190					0.05	115	0.01	3.40	0.01	4.57
BDE-197		114.0		4.04		660		28.9		20.9
BDE-203		104.4		5.41		685		34.8		25.1
BDE-196		85.5		5.93		512		26.2		18.9
BDE-208		173		5.55		1392		54.8		40.2
BDE-207		399		13.7		3521		129		94.3
BDE-206		192		8.09		1160		60.3		38.8
BDE-209		1949		98.9		12 374		740		362
SUM	4627	5303	127	222	2502	38 974	361	1978	202	2357

congeners in CD and GR suggests similar sources and/or processes of these PBDE congeners (Table S4). Additionally, the concentrations of individual PBDE congeners in CD and GR were 7- to 48-fold lower than those in GY, but still high compared with other areas. The dependence of the concentrations at CD and GR on those of GY in summer and winter ( $r^2 > 0.85$ ) (Fig. S1) and the lack of correlation for PBDE congener concentrations in CD and GR with temperature, wind speed and direction (Table S4) illustrate that the high PBDE concentrations found at CD and GR were partly attributed to long-distance transport from GY.

The compositional differences were minor between the sampling sites (Fig. 1). The similarity in the PBDE profiles also implied similar sources between the three sampling sites. The mass distribution in air (including gas and particulate phases) was dominated by BDEs-28, 47, -99, -207 and -209, which contributed over 70% of the total PBDEs. BDE-209 was present much lower concentration (9.4–23.0% in summer and 29.1–41.0% in winter) relative to the Deca-BDE commercial mixture. The range of BDE-209 contribution found in this study was lower than the other air studies in Asia (over 70%) such as Kyoto, Japan (Hayakawa et al., 2004) and Guangzhou, China (Chen et al., 2006) and in Europe such as Sweden (mean 45%) (Agrell et al., 2004) and the UK (median 59%) (Wilford et al., 2008), but higher than those in samples collected in North America (Hoh and Hites, 2005). The nona-BDE congeners ([BDE-207] > [BDE-208]  $\geq$  [BDE-206]) con-

stituted a mean 8–14% of the total, which were higher than the 2–3% reported in the Deca-BDE commercial mixture. This may be explained by the potential debromination of BDE-209 in the atmosphere. The high BDE-207 contribution may also be derived from the Octa-BDE Bromkal 79-8DE (about 11%) (LA Guardia et al., 2006). The PBDE patterns in this study, if nona- and deca-BDEs were excluded, were similar to that in the technical Penta-BDE mixture Bromkal 70–5DE and DE-71 (Fig. S2), which suggests that both Penta-BDE and Deca-BDE mixture had a great contribution to these sampling sites.

#### 3.2. Atmospheric gas-particle partition of PBDEs

The tri-BDE (BDE-28) was present almost exclusively in the gas phase in the summer (88–98%), whereas the higher brominated PBDEs exhibited greater association with the particles. The octato deca-BDEs were found only in the particulate phase. Meanwhile, the phase distribution shifted to particle at low temperature. The gas phase contribution decreased 33% in winter.

Two different mechanisms have been used to describe the gasparticle partitioning of semivolatile organic compounds (SOCs), that is, adsorption onto the aerosol surface and absorption into the aerosol organic matter. Both mechanisms lead to a linear relationship between logarithm of the gas-particle partition coefficient

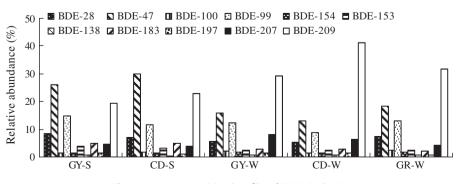


Fig. 1. Average compositional profiles of PBDEs in the air.

(log  $K_p$ ) and logarithm of the subcooled liquid vapor pressure (log  $P_L^0$ ):

$$\log K_{\rm p} = m_{\rm r} \log P_{\rm L}^0 + b_r \tag{1}$$

The slope  $m_r$  should be close to -1 for either adsorption or absorption in true equilibrium state (Pankow, 1994). Therefore, deviations of the slope from -1 have been considered as nonequilibrium conditions or experimental artifacts. Fig. S3 shows the loglog plots of  $K_p$  versus  $P_1^0$  for PBDEs measured in this study. The temperature-dependent vapor pressure was calculated for all PBDEs based on the method of Tittlemier et al. (Tittlemier et al., 2002) using the average ambient temperature during each sampling time. Significant linear correlations between  $\log K_p$  and  $\log P_1^0$  were obtained with  $r^2$  values in the range of 0.585–0.830 and  $m_r$  values from -0.763 to -1.102. The GY samples with the highest correlation coefficient and  $m_r$  close to -1 suggest that PBDEs appeared to be close to equilibrium between the gas and particulate phases. CD and GR sites were further away from emission sources than GY, thus PBDEs should be equilibrium between phases. Goss and Schwarzenbach (Goss and Schwarzenbach, 1998) has drawn that the deviation of slope  $m_r$  from -1 does not necessarily indicate nonequilibrium effect and  $m_r$  could provide insights into the sorption process. Slopes steeper than -1 are characteristic of adsorption, whereas slopes shallower than -0.6 are suggestive of absorption mechanism. The  $m_r$  values in this study suggest that both mechanisms play a role during sorption process.

The Junge–Pankow adsorption and  $K_{OA}$ -based absorption models have been conventionally used to predict the fraction of SOCs in the particles ( $\varphi$ ). Based on Junge–Pankow model,  $\varphi$  is related to the  $P_L^0$  of the compound and the particle surface area per unit volume of air ( $\theta$ ) by

$$\varphi = c\theta / (P_{\rm L} + c\theta) \tag{2}$$

where *c* is a constant. Values of c = 17.2 Pa cm and  $\theta = 1.1 \times 10^{-5}$  cm<sup>-3</sup> for urban air are often used, although they might vary with the class of compound and sampling sites (Pankow, 1987; Bidleman, 1988). The *K*<sub>OA</sub>-based absorption model predicts the values of *K*<sub>p</sub> by

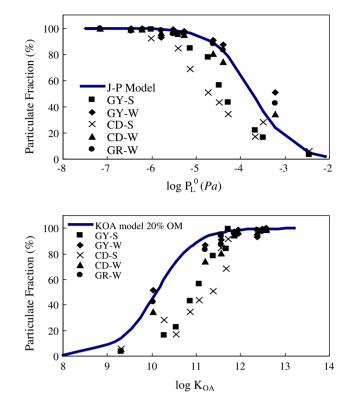
$$\log K_{\rm p} = \log K_{\rm OA} + \log f_{\rm om} - 11.9 \tag{3}$$

$$\varphi = K_{\rm p} TSP / (1 + K_{\rm p} TSP) \tag{4}$$

Values of  $K_{OA}$  have been reported as a function of temperature for PBDE congeners (Harner and Shoeib, 2002). The OC contents of particles in this study were in the range of 7.1–29.9% with an average of 15.2%, which was equivalent to approximately 20% of organic matter. Fig. 2 compares the percent of PBDEs in the particulate phase predicted by the Junge–Pankow adsorption model and  $K_{OA}$ -based absorption model with the average measured values. A good agreement was obtained between the predicted and measured for all samples collected in winter. However, both models overestimated the sorption of PBDEs in summer, particularly for the light PBDEs (Fig. 2). The derivation between the modeling and the measured data in this study was possible due to the different parameter values for different compounds and aerosol types in summer. It should be further investigated.

# 3.3. Seasonal and diurnal variation

Seasonal variation of PBDE concentrations was noted during this study, which may be governed by meteorological conditions such as atmospheric stability and variations in the strength of potential emission sources. The mean individual PBDE concentrations in winter increased by a factor of 2.2–8.8 for GY and 4.1–17.4 for CD, respectively. Frequent inversion temperature would result in



**Fig. 2.** Comparison of the measured particulate fractions of PBDEs with theoretical predictions of the Junge–Pankow adsorption model and  $K_{OA}$ -based absorption model with 20% of organic matter.

higher concentrations of PBDEs during winter months, while higher rainfall in summer would wash more PBDEs in the atmosphere resulting in lower concentrations (ter Schure and Larsson, 2002).

The PBDE homologue profiles showed seasonal variations. Low brominated PBDEs had a greater contribution in summer than in winter, and the opposite was true for highly brominated PBDEs. In addition to the possible variation in emission sources, the difference could be interpreted by following reasons: (1) low brominated PBDE congeners could be accelerated emission from ewaste due to higher temperature in summer (Gouin and Harner, 2003). (2) Photolytic degradation of highly brominated PBDEs to lower brominated BDEs in the atmosphere readily occurred due to the strong sunlight in summer (Strandberg et al., 2001; Lee et al., 2004). Therefore, it is logical to observe relatively higher low brominated PBDEs in the air in summer.

The diurnal variation is a dominant process controlling the concentration of POPs. Fig. 3 indicates the diurnal variation of PBDE congeners in the atmosphere of GY. BDE-47 and -99 concentrations were significantly higher in the daytime when the mixing layer depth was relatively high and primary emissions from e-waste dismantling were intense, and declined at night. BDE-183 showed the opposite trend with higher concentration occurring after the boundary layer collapsed in the night. Such diurnal behaviour is characteristics for air pollutants from local emissions. However, no concentration trend was observed for BDE-209. BDE-209 showed a lower contribution to  $\sum$ PBDEs during daytime than during the night (16.1% versus 24.1% in summer, and 23.5% versus 34.0% in winter). Therefore, the lack of strong diurnal variations of BDE-209 might be the result of its photochemical procedure, which is usually most intense in the afternoon, tends to be dwarfed by the much strong variations in mass concentrations associated with source emission. Meanwhile, these diurnal variations may be skewed by abnormally low/high loading events. The dip and

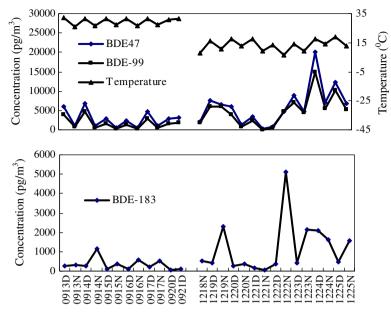


Fig. 3. Diurnal variations of PBDEs in GY atmosphere.

then rise during December 20–22 on the diurnal curve of PBDEs, for example, were mainly caused by the abrupt change in the mass concentration and wind speed.

Diurnal variation of POPs was possibly governed by source characterization, temperature, atmospheric mixing layer height and stability, and hydroxyl radical concentration (Macleod et al., 2007). The day-to-night changes at GY could not be attributed to changes in the mixing layer height and temperature which would equally affect the concentrations of all PBDE congeners. Limited studies showed that the effects from OH reaction and photolysis were not important for the short-term variation of PBDEs in the atmosphere (Iacovidou et al., 2009). Based on the results, source characterization might the major factor controlling the short-term variability of PBDEs in GY air. That is, different e-waste types and/or different recycling processes were applied during day and night. In CD and GR, the diurnal variations were different from those in GY. We did not observe the distinctively diurnal variation of BDEs-47, -99, -183 and -209. From the above discussion, a significant fraction of the PBDEs in CD and GR were from long-distance transport. During atmospheric transport, some processes that can change PBDE composition such as deposition and degradation could occur. Consequently, the rather weak diurnal variation of the ambient concentrations of PBDEs was found in CD and GR.

#### 3.4. Implications for human exposure

The high PBDE concentrations observed in the air of the e-waste recycling sites are consistent with elevated concentrations of these compounds observed in the population compared with the other cities and countries, as shown in human serum (Jakobsson et al., 2002; Thuresson et al., 2006; Bi et al., 2007; Qu et al., 2007). Dietary intake is generally accepted to be the main exposure route for persistent organohalogen compounds. However, diet was found only a small contribution to PBDE exposures, and air inhalation, dust ingestion and dermal absorption may be important contributors of human exposure to PBDEs (Allen et al., 2008; Johnson-Restrepo and Kannan, 2009). In this study, the risk of potential adverse health effects was calculated using the mean measured concentrations. The daily exposure dose by air inhalation (DED<sub>air</sub>)

of PBDEs were estimated using the following equation (Nouwen et al., 2001):

$$\mathsf{DED}_{\mathrm{air}} = C \cdot IR \cdot f_{\mathrm{r}} / BW \tag{5}$$

where *C* is the average PBDE concentration in air  $(pg m^{-3})$ ; IR is inhalation rate (20 m<sup>3</sup> d<sup>-1</sup> for adults and 10.9 m<sup>3</sup> d<sup>-1</sup> for children);  $f_r$  is the alveolar fraction retained in the lungs (a value of 0.75 was used for both adults and children); and BW is the body weight (60 kg for adults and 25 kg for children were used in this study). The estimated DED for GY children and adults from air inhalation (gas + particles) varied between 2.48 and 13.6 ng kg<sup>-1</sup>-bw d<sup>-1</sup> (Table 2), which was 14-74-fold higher than those observed in the United States (Johnson-Restrepo and Kannan, 2009). The inhalation intake doses were reported, contributing approximately 4.6% of the total daily intake (TDI) in USA (Johnson-Restrepo and Kannan, 2009). Because data about other exposure pathways other than inhalation in GY are scarce, we tentatively estimated TDI of PBDEs using a value of 4.6%. The results of TDI for summer were 54.0 and 70.6 ng kg<sup>-1</sup>-bw d<sup>-1</sup> for adults and children, and in winter they were 225 and 295 ng kg<sup>-1</sup>-bw d<sup>-1</sup>, respectively (Table 2). The calculated exposure doses to PBDEs for children and adults in GY were much higher than those for US and Germany populations (Zuurbier et al., 2006; Johnson-Restrepo and Kannan, 2009) and other population in China (Meng et al., 2007). The total intake of PBDEs was far below the lowest observed adverse effect level of 1 mg kg<sup>-1</sup>-bw d<sup>-1</sup> recommended by Darnerud et al. (Darnerud et al., 2001). However, the total intakes of BDE-99 in GY was 9.0–34.9 ng  $kg^{-1}$ -bw  $d^{-1}$ , which was higher than the maximal allowed intake level  $(0.26 \text{ ng kg}^{-1}\text{-bw d}^{-1})$  recommended by Winter-Sorkina et al. (Winter-Sorkina et al., 2006) with factors of 35–135, indicating possible health risk for GY residents with exposure to PBDEs. Furthermore, long-distance transport of PBDEs from GY has turned CD and GR into high-risk sites, with average daily intake of BDE-99 of 0.20–3.34 ng kg<sup>-1</sup>-bw d<sup>-1</sup> (Table 2), which are also close to or exceed the maximal allowed intake level. The international bans and voluntary phase-out of Penta-BDE and Octa-BDE commercial use are an essential step toward overall lowering of exposure and body burdens of PBDEs. However, effective management policies and remediation techniques are urgently

#### Table 2

Calculated human total daily intake (TDI) of PBDEs for children and adults.

Factors		Parameters	
		Children	Adults
Body weight (BW)(kg) Inhalation rate (IR)( $m^3 d^{-1}$ ) <sup>a</sup> Alveolar fraction retained in the lungs ( $f_r$ )		25 10.9 0.75	60 20 0.75
$\sum$ PBDEs exposure dose C (pg m <sup>-3</sup> ) <sup>b</sup>	GY-S	9930	9930
	CD-S	349	349
	GY-W	41 476	41 476
	CD-W	2339	2339
	GR-W	2560	2560
$DED_{\mathrm{air}}^{\mathbf{c}}(\mathrm{ng}\mathrm{kg}^{-1}\text{-}\mathrm{bw}\mathrm{d}^{-1})$	GY-S	3.25	2.48
	CD-S	0.11	0.09
	GY-W	13.6	10.4
	CD-W	0.76	0.58
	GR-W	0.84	0.64
TDI (ng kg <sup>-1</sup> -bw d <sup>-1</sup> ) <sup>d</sup>	GX-W GY-S CD-S GY-W CD-W GR-W	0.04 70.6 2.48 295 16.6 18.2	54.0 1.90 225 12.7 13.9
BDE-99 exposure dose C	GK-W	10.2	15.9
(pg m <sup>-3</sup> )	GY-S	1656	1656
	CD-S	36.6	36.60
	GY-W	4911	4911
	CD-W	269	269
	GR-W	470	470
DED <sub>air</sub> (ng kg <sup>-1</sup> -bw d <sup>-1</sup> )	GY-S	0.54	0.41
	CD-S	0.01	0.01
	GY-W	1.61	1.23
	CD-W	0.09	0.07
	GR-W	0.15	0.12
TDI (ng kg <sup>-1</sup> -bw d <sup>-1</sup> )	GY-S	11.8	9.00
	CD-S	0.26	0.20
	GY-W	34.9	26.7
	CD-W	1.91	1.46
	GR-W	3.34	2.55

<sup>a</sup> Based on USEPA (1997).

<sup>b</sup> C: average PBDE concentration in air, the values from Table 1.

<sup>c</sup> DED<sub>air</sub>: daily exposure dose by air inhalation, DED<sub>air</sub> =  $C \cdot IR \cdot f_r / BW$ .

<sup>d</sup> TDI = DED<sub>air</sub>/4.6%.

needed to prevent the deterioration of environmental quality in the e-waste recycling sites.

### 4. Conclusion

As a typical e-waste processing site, GY may serve as an ongoing strong source of PBDEs and affect the nearby regions by longdistance transport. The percentages of BDE 209 compared to the total PBDE concentration in GY were much lower than most other places. This was attributed to "old" waste in GY that probably contained a higher portion of Penta- and Octa- mixtures than products produced today. Since Penta- and Octa-mixtures are not used in new produces and the demand for Deca-BDE is increasing, there would be a lot of waste containing Deca-BDE in GY. Further work should focus on the atmospheric chemical processes of PBDEs and the highly brominated congeners and their health effects.

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# Appendix A. Supplementary material

Supplementary data associated with this article can be found, in the online version, at doi:10.1016/j.chemosphere.2010.12.035.

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