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PBDEs and Dechlorane Plus in the environment of Guiyu, Southeast China: A historical location for E-waste recycling (2004, 2014)

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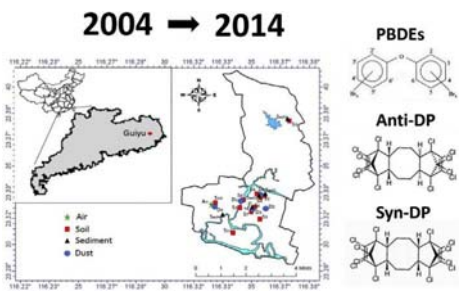
HIGHLIGHTS

- $\Sigma_{3\text{-}7}$PBDEs in soil, road dust, and sediment in 2014 were a little less than or close to those measured in 2004.
- $\Sigma_{3\text{-}7}$PBDEs in road dust were greater than those in soil, sediment and air particles.
- Greater proportions of lesser-brominated BDE, such as BDE-71, were predominant in air particulates.
- Most of the fractions of anti-DP were consistent among samples, ranging from 70% to 80%.
- The health risk assessment showed that HIs of PBDEs or DPs for child and adult were all lower than 0.16.

GRAPHICAL ABSTRACT



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ABSTRACT

Polybrominated diphenyl ethers (PBDEs, tri-to hepta-BDEs) and Dechlorane Plus (DP) in multiple samples (soil, sediment, road dust, and $PM_{2.5}$ particles) in historical locations of our previous work in Guiyu (electronic-waste recycling town) in southeastern China were investigated in 2014. Ten years later, PBDEs and DP were detected in 100% of the samples. $\Sigma_{3\text{-}7}$PBDEs were still relatively great, ranging from 1.2×10^1 to 2.1×10^3 ng/g dry weight (dw) in soil, 2.1 to 3.2×10^3 ng/g dw in sediment, 1.0×10^1 to 1.1×10^4 ng/g dw in road dust, which were a little less than or close to those measured in 2004. However, $\Sigma_{3\text{-}7}$PBDEs in $PM_{2.5}$ (5.0×10^2 to 8.4×10^2 pg/m^3) were significantly lower in 2014. BDE-47, -99, −153, and −183 were predominant congeners, which were also predominant PBDEs reported in Guiyu (2006–2008). Greater proportions of lesser-brominated BDEs were predominant in $PM_{2.5}$ than other samples. DP was detected in 100% of the samples collected with high levels. Total syn-DP and anti-DP concentrations were 3.8 to 2.1×10^3 ng/g dw in soil, 1.1×10^3 to 7.2×10^3 ng/g dw in sediment, 1.4×10^1 to 1.1×10^3 ng/g dw in road dust, and 1.8×10^2 and 1.7×10^2 pg/m^3 in $PM_{2.5}$. Most of the fractions of anti-DP (f_{anti}) (70%–80%) were

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consistent among samples. The health risk assessment showed that hazard indexes (HIs) of PBDEs or DPs for child or adult were all lower than 0.16.

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1. Introduction

Electronic waste (e-waste) has been recycled at Guiyu, a small town located in the east of Guangdong Province in southeastern China, since the 1990s. More than 75% of the 300 individual workshops have been involved in the dismantling or processing of e-waste. Due to the crude recycling activities of e-waste, contamination with polybrominated diphenyl ethers (PBDEs), as well as polychlorinated dibenzo-p-dioxins and dibenzofurans (PCDD/Fs), polychlorinated biphenyls (PCBs), polycyclic aromatic hydrocarbons (PAHs), and some heavy metals, were detected in many matrices in Guiyu. For example, these contaminants were found to be widely distributed in air (Deng et al., 2007), dust (Leung et al., 2011), soil (Wong et al., 2007), and sediment (Luo et al., 2007). The adverse health effect on humans caused by these organic and inorganic contaminants in e-waste recycling area is a growing issue globally (Heacock et al., 2015). A large number of studies have focused on the contaminants with the premature recycling works in Guiyu, and discovered that humans and wildlife have been severely affected in this area (Leung et al., 2007, 2008a, b; Wong et al., 2007, ; Li et al., 2008).

Of the heavy contaminants in Guiyu, PBDEs have received considerable attention in recent years due to their high production and potential toxic effects, such as interfering with endocrine and nervous systems (Herbstman et al., 2010; Kuriyama et al., 2005). PBDEs belongs to the brominated flame retardants (BFRs), which were widely used as additive FRs in paints, textiles, and electronics and have been massively released into the environment (Covaci et al., 2011; Kierkegaard et al., 2004), especially in some countries in Asia such as China and India, where the recycling of discarded concentrations of congeners associated with these electronic devices (e-wastes) is flourishing. Consequently, PBDEs are ubiquitous in water, sediments, atmosphere, plants, and the tissues of animals in e-waste recycling areas (Hites, 2004; Hu et al., 2014; Rodenburg et al., 2014; She et al., 2013). The contamination status of PBDEs in Guiyu is even more serious (Leung et al., 2011), and the health issues related to PBDEs by our previous work have been reported for concentrations observed in soil, dust, water, and sediment in Guiyu (Leung et al., 2006, 2007, 2011). Since the early 2000s, e-waste has been treated in a government-organized recycling center at Guiyu, instead of in traditional family-based private workshops. The implementation of regulations and the establishment of a central facility with modern controls on the release of pollutants by the government to better manage recycling of e-waste have contained releases of PBDEs to the environment. However, the effect was not confirmed.

Because of high toxicity and persistence, PBDEs were phased out by the European Union beginning in 2004 (Schenker et al., 2008). Alternative additive FRs in commercial products are being developed and used (Chen et al., 2009; Hites, 2004). One of the emerging non-PBDE FRs is Dechlorane Plus (DP), which was first identified in the environment in 2006, are widely used in clothing, furniture, as well as electronic device, such as computer and televisions (Hoh et al., 2006). It was reported that concentrations of PBDEs decreased, whereas alternative FRs, especially DP increased in surface soils and river sediments from the e-waste processing sites in northern Vietnam during 2012–2014 (Matsukami et al., 2017). DP has been reported to be persistent, bioavailable, bioaccumulative, long-range transport and might cause subsequent toxicity to wildlife (Tomy et al., 2007; Sverko et al., 2011). Möller et al. analyzed the air water distribution of DP and suggested that DP was susceptible to long-range atmospheric transport (Möller et al., 2010). Sühring et al. reported the distribution and uptake of DP between sediments and benthic fish (Sühring et al., 2016). Although relatively high concentrations of PBDEs and DP have been found in water, sediments, and some wild animals from e-waste recycling locations (Covaci et al., 2011; Luo et al., 2009), few studies have investigated the DP concentrations in many environmental matrices, especially in e-waste recycling areas. In addition to that, most studies have focused on a single matrix, such as the air or sediment in and around the Great Lakes (Hoh et al., 2006; Shoeib et al., 2012; Zhu et al., 2007), serum (Ren et al., 2009), air (Ren et al., 2008), or soils in China (Wang et al., 2010a, b). Data on DP from various compartments of locations where e-waste is being recycled are more limited (Zhu et al., 2007 (Xu et al., 2017);). More research is required to better quantify the contamination of DP in multiple environmental matrices, especially in e-waste recycling areas.

The primary objective of this study was to determine the effects of the regulations instituted in 2004 on the concentrations of PBDEs and DP in soil, road dust, sediment, and particles from the vicinity of Guiyu. Considering that octa-BDE and penta-BDE were banned in China in 2004 and 2007, respectively, the absolute concentrations and the patterns of the relative concentrations of tri-to hepta-BDEs and DP were examined and compared with data from our previous studies conducted in 2004. Presented here are the results of the first study to investigate and compare the occurrence of PBDEs and DPs in various environmental samples at Guiyu over a decade.

2. Materials and methods

Collection of Samples. Soils were collected from 11 locations, including one reference location near Nanyang reservoir (Fig. 1, shown as S_1 to S_{11}) and sediments were collected from 5 locations. Dust was collected from five areas. The mass of each sub-sample was 10–20 g. $PM_{2.5}$ was collected on the roof of a four-story family building during 23–25 September 2014. According to USEPA acceptance criteria (USEPA, 1998). The details of location and collection was in S1 and S6 in supporting information (SI).

Extraction, cleanup, and quantification. The samples and filters were weighed and spiked with internal standard (^{13}C-OctaCDE), and then vortexed. Ultrasonic extraction method was used with hexane/DCM (1:1) for four times. The extracts were combined and concentrated on a rotary evaporator before cleanup on an aluminum oxide open column (9 g Aluminum oxide and 80 mL solvent). The analytes was eluted by Hexane (with 7% DCM) and concentrated to 500 μL before instrument analyses. All of the solvents, standards of BDE-17, 28, 47, 66, 71, 85, 99, 100, 138, 153, 154, 183, 190 and two DP isomers, anti- and syn-DP, of pesticide grade purity were purchased (J.T. Baker, USA). For the analysis of DP, chemical reagents and materials, including dichloromethane, hexane, N-octane, ^{13}C-OctaCDE, and sodium sulfate, were used. Deionized water was generated in house using a Super-Q water generation system. The details was in S6 in SI.

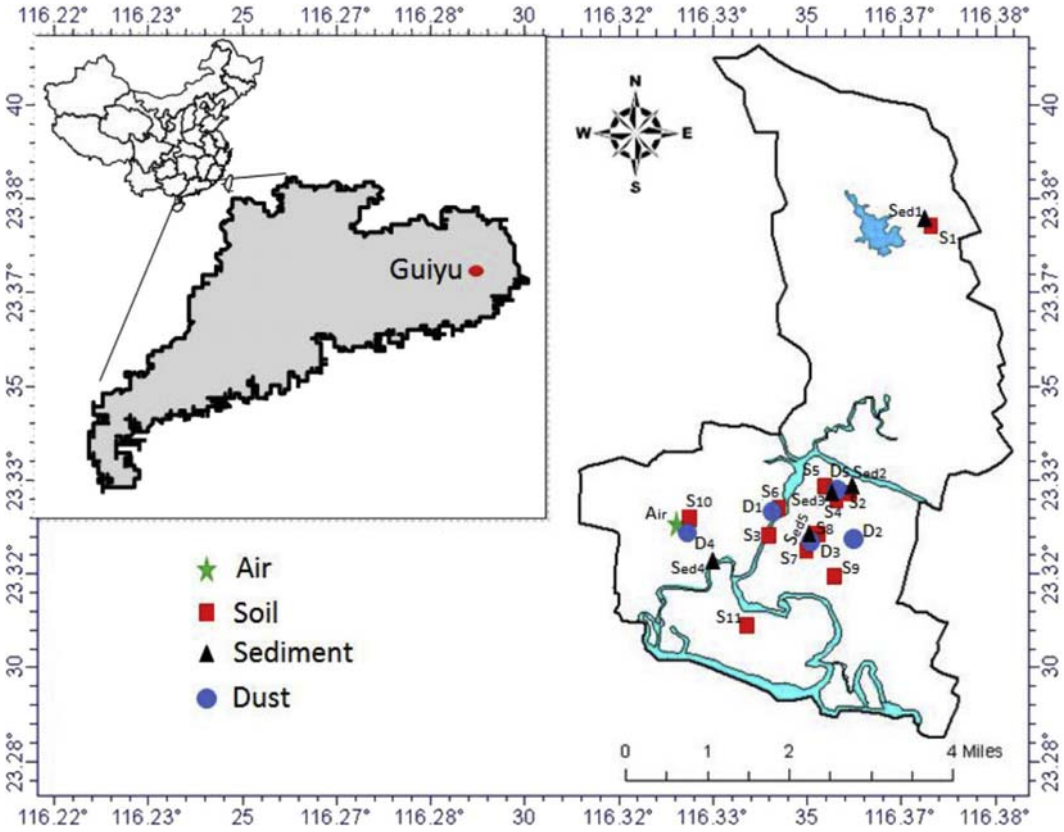

Fig. 1. Locations from which samples of air, soil, sediment, and road dust were collected in Guiyu.

Instrumental Analyses. An Agilent gas chromatograph (GC)–mass spectrometer (MS) system (6890 GC/5973 MSD; Agilent Technologies, Palo Alto, CA) fitted with a DB-1MS column (30 m long × 0.25 mm inner diameter × 0.10-μm film thickness) was used for the analyses. The details was in S6 in SI.

QA/QC. GC-MS peaks were normalized to the peak area of 2,2′,3,3′,4,4′,5,5′-octachlorodi-phenyl-ether-13C12 (^{13}C-OctaCDE) before data reduction. Limit of quantity (LOQ) for all the investigated compounds by GC/MS were estimated based on a signal-to-noise ratio (S/N) of 10 using the lowest concentration standard. LOQ of PBDE was 2.1–12.8 ng/kg, while syn-DP and anti-DP was 17.6 and 27.2 ng/kg, respectively. One procedural blank was included in each analytical batch of six samples. The recovery rate of ^{13}C-OctaCDE was 93% ± 11% and average blank levels (0.11 ± 0.10 ng/g for DP; no PBDEs in blank). The details was in S6 in SI.

Statistical analyses. All statistical tests were performed with SPSS 22.0 software. The normality of the data was checked with the Shapiro-Wilk test. The data were log-transformed to meet the normality assumptions. Levene's test was used to assess the homogeneity of data. Power analysis was conducted using G × Power v3.1.9.2 and the calculated power ($1-\beta$) is at least 0.2. The means of different groups were compared using one-way analysis of variance followed by Duncan's multiple-range post hoc test, with the probability of type I error (α) set at 0.05.

Risk assessment. To assess health risk of child (3–6 years old) and adult exposure to ΣPBDEs and ΣDP in air particle and road dust, three daily exposure models were applied to calculate total estimated daily intake (EDI) amount via three non-dietary ways, namely ingestion, inhalation and dermal contact. Hazard Quotients (HQs) for three exposure pathways ($HQ_{ingestion}$, $HQ_{inhalation}$ and HQ_{dermal}) were calculated using EDI and Reference Dose (RfD) of each contaminant ($HQ_i = EDI_i/RfD_i$), where i is different contaminants. The three HQs were added up and a Hazard Index (HI) was obtain for each contaminant. The exposure factors employed and detailed calculation method were listed in SI.

3. Results and discussion

3.1. Levels and patterns of PBDEs

Soil. The $\sum_{3\text{-}7}$PBDEs levels measured in soil in 2014 in this research ranged from 1.2×10^1 to 2.1×10^3 ng/g, dw (Table 1). Based on the locations with greater $\sum_{3\text{-}7}$PBDEs levels (1.7×10^2 to 2.1×10^3 ng/g, dw), congeners BDE-47, -66, −99, −153, and −183 were commonly detected as the major contributors (Fig. 2).

Compared with our previous work in 2004 (6.4×10^{-1}-6.7×10^2 ng/g, dw, $\sum_{3\text{-}7}$PBDEs in soil) (Leung et al., 2007), the concentrations of $\sum_{3\text{-}7}$PBDEs in 2014 in this research were a little higher. The greatest $\sum_{3\text{-}7}$PBDEs level was observed in the soil collected near the kindergarten (2.1×10^3 and 1.6×10^3 ng/g, Table 1), followed by the living area (1.4×10^3 ng/g, dw, LA_2), the pump station (7.9×10^2 and 9.7×10^2 ng/g, dw), and the road side (1.7×10^2 and 2.3×10^2 ng/g, dw). The sampling sites mentioned above were all near the workshop. Although the levels in rice field (6.2×10^1 and 7.6×10^1 ng/g, dw) and in vegetable field (5.8×10^1 and 4.6×10^1 ng/g, dw) were less than other sampling sites, compared with reference (0.62 and 1.2 ng/g, dw), the concentrations were still relatively great. Recently, PBDEs were also reported to be the most abundant FRs in the soils in Guiyu, with concentrations of 1.3×10^1 to 1.0×10^3 ng/g, dw (di-to deca- BDEs) (Xu et al., 2017).

Table 1
Concentrations of selected PBDE congeners and DPs in soils collected from Guiyu in 2014 (ng/g, dw; n = 69).

Congener	Nanyang reservoir			Duck pond		Rice field		Kindergarten		Pump station		Workshop		Road side		Vegetable field		Huamei middle school		Living area		Meizhou middle school	
	C1	C2	C3	DP1	DP2	RF1	RF2	K1	K2	PS1	PS2	WS1	WS2	RS1	RS2	VF1	VF2	HMSch1	HMSch2	LA1	LA2	MZSch1	MZSch2
BDE-17	0.02	0.23	0.41	1.2	0.65	0.06	0.57	5.6×10^{1}	0.74	0.12	1.9×10^{1}	0.05	0.03	0.20	0.22	0.18	0.15	1.3	0.71	0.93	1.6	0.54	0.29
BDE-28	0.03	0.03	0.72	1.8	2.2	0.94	1.4	7.8×10^{1}	5.70	8.7	2.7×10^{1}	0.32	0.76	2.4	2.6	1.0	1.2	1.1	0.62	6.1	8.9	0.40	0.25
BDE-47	0.13	0.19	4.8	1.7×10^{1}	1.4	1.2×10^{1}	2.1×10^{1}	5.9×10^{2}	1.3×10^{1}	4.3×10^{1}	2.7×10^{2}	1.4	3.5	1.7×10^{1}	3.5×10^{1}	1.2×10^{1}	4.1	5.7	7.8	3.4×10^{1}	3.8×10^{1}	4.0	2.7
BDE-66	0.12	0.18	1.4	4.9	4.6	5.1	3.6	2.0×10^{2}	1.8×10^{1}	1.3×10^{1}	8.2×10^{1}	0.58	0.99	6.6	1.5×10^{1}	2.6	1.8	4.4	1.9	1.2×10^{1}	9.7	0.91	0.76
BDE-71	0.08	0.18	0.35	0.61	0.58	0.69	0.50	2.0×10^{1}	3.03	8.2	6.3	0.18	0.27	3.3	1.9	0.63	0.75	0.49	0.45	1.3	1.5	0.20	0.18
BDE-85	ND	ND	0.66	1.6	1.5	3.5	1.7	5.6×10^{1}	2.6×10^{1}	5.8	3.1×10^{1}	0.50	0.77	1.7×10^{1}	0.64	3.7	2.2	2.4	1.5	3.8×10^{1}	3.5×10^{1}	0.83	0.51
BDE-99	0.14	0.20	6.8	2.4×10^{1}	2.3×10^{1}	1.6×10^{1}	3.4×10^{1}	6.5×10^{2}	1.3×10^{2}	8.3×10^{1}	3.5×10^{2}	2.2	4.7	4.5×10^{1}	5.4×10^{1}	1.0×10^{1}	6.7	1.4×10^{1}	7.8	4.4×10^{1}	4.3×10^{1}	2.9	2.1
BDE-100	0.09	0.07	0.42	1.4	1.7	2.1	5.0	3.3×10^{1}	2.7×10^{1}	1.6×10^{1}	2.2×10^{1}	0.52	0.82	7.5	1.6×10^{1}	2.1	1.2	1.3	1.4	8.2	3.0	0.59	0.49
BDE-138	ND	ND	0.21	0.43	0.26	0.29	0.33	2.8×10^{1}	1.4×10^{2}	4.8	7.1	0.35	0.27	2.8	9.4	0.67	0.19	0.42	0.23	0.75	4.5	0.20	0.17
BDE-153	ND	0.14	2.2	5.7	4.8	9.6	4.8	2.3×10^{2}	2.9×10^{2}	1.5×10^{2}	1.0×10^{2}	1.8	3.2	4.9×10^{1}	3.3×10^{1}	8.5	7.1	5.1	3.8	2.6×0^{1}	1.3×10^{2}	8.5×10^{1}	0.99
BDE-154	ND	ND	0.50	1.5	1.7	2.7	2.5	6.6×10^{1}	1.5×10^{2}	2.2×10^{1}	3.0×10^{1}	0.91	0.95	8.4	5.3	2.1	1.2	2.7	2.1	7.8	1.5×10^{1}	0.65	0.39
BDE-183	ND	ND	0.18	0.72	0.98	8.8	0.38	7.6×10^{1}	7.8×10^{2}	4.3×10^{2}	2.5×10^{1}	3.1	2.3	1.3×10^{1}	5.2×10^{1}	1.4×10^{1}	1.9×10^{1}	1.6	3.7	5.5×10^{1}	1.2×10^{3}	2.1	0.63
BDE-190	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	1.0×10^{1}	ND	3.6
∑PBDE	0.62	1.2	1.9×10^{1}	6.0×10^{1}	4.3×10^{1}	6.2×10^{1}	7.6×10^{1}	2.1×10^{3}	1.6×10^{3}	7.9×10^{2}	9.7×10^{2}	1.2×10^{1}	1.9×10^{1}	1.7×10^{2}	2.3×10^{2}	5.8×10^{1}	4.6×10^{1}	4.0×10^{1}	3.2×10^{1}	2.3×10^{2}	1.4×10^{3}	9.8×10^{1}	1.3×10^{1}
syn-DP	0.32	ND	ND	2.2	9.0×10^{1}	3.0×10^{1}	0.89	1.6×10^{1}	3.0×10^{1}	9.3×10^{1}	1.5×10^{1}	0.84	1.1×10^{1}	2.2×10^{2}	4.3×10^{2}	2.4×10^{1}	1.2×10^{1}	4.8×10^{1}	1.6×10^{1}	3.3×10^{1}	6.6	4.4	2.4
anti-DP	0.94	ND	ND	6.9	4.8×10^{1}	1.1×10^{1}	2.9	5.3×10^{1}	1.3×10^{2}	2.4×10^{2}	4.1×10^{1}	3.0	3.3×10^{1}	6.6×10^{2}	1.7×10^{3}	8.5×10^{1}	4.5×10^{1}	2.0×10^{2}	5.4×10^{1}	1.1×10^{2}	4.1	1.8×10^{1}	7.4
∑DP	1.3	ND	ND	9.1	1.4×10^{2}	4.1×10^{1}	3.8	6.9×10^{1}	1.6×10^{2}	3.3×10^{2}	5.6×10^{1}	3.8	4.4×10^{1}	8.8×10^{2}	2.1×10^{3}	1.1×10^{2}	5.7×10^{1}	2.5×10^{2}	7.0×10^{1}	1.4×10^{2}	1.1×10^{1}	2.2×10^{1}	9.8
IR (%)	75	NA	NA	76	35	27	77	77	81	72	73	78	75	75	80	78	79	81	77	76	38	80	76

ND, not detected; NA, not available.

Road dust. The $\sum_{3\text{-}7}$PBDEs levels measured in road dust in 2014 in this research were ranged from 1.0×10^{1} to 1.1×10^{4} ng/g, dw (Table 2). The greatest $\sum_{3\text{-}7}$PBDEs level was observed in dust collected from along a road in Nan'an village (1.1×10^{4} ng/g, dw NAV_2; Table 2), followed by Yujiao village (6.8×10^{3} and 5.3×10^{3} ng/g, dw, for YJV_1 and YJV_2, respectively) and NAV_3 and NAV_1 (3.3×10^{3} and 2.5×10^{3} ng/g, dw, respectively) in Nan'an village. The lowest concentration of $\sum_{3\text{-}7}$PBDEs was measured in dust collected from a road near Guiyu middle school (1.0×10^{1} ng/g, dw $GYMSch_2$), followed by the living area (4.3×10^{1} ng/g, dw LA_1).

In most cases, greater proportions were contributed by BDE-183 (e.g., YJV_2: 81.9%, YJV_1: 58.2%, WS_2: 57.5%), followed by BDE-47 (e.g., $GYMSch_2$: 32.1%, LA_3: 32.66%) and BDE-99 (e.g., NAV_1: 33.96%, LA_3: 29.18%) (Fig. 2). In all cases, BDE-17, -28, −66, −71, −85, −100, −138, −154, and −190 contributed less than 10% of the $\sum_{3\text{-}7}$PBDE, except for BDE-66 in LA_3 (10.25%) and BDE-190 in LA_2 (17.57%).

Compared with our previous work in 2004 ($\sum_{3\text{-}7}$PBDEs, 8.1×10^{2}–2.0×10^{4} ng/g, dw, in dust), the concentrations of $\sum_{3\text{-}7}$PBDEs in 2014 were similar (Leung et al., 2011). The concentrations of $\Sigma_{3\text{-}7}$PBDEs were reported to range from 5.1 to 6.4×10^{1} ng/g in house dust and office dust in Beijing which were far less than the levels surveyed in this research, and among tri-to hepta-BDE, BDE-28, 47, 99 and 153 were predominant in office dust, whereas BDE-99 and BDE-47 were the predominant congener in house dust (Wang et al., 2018). PBDEs were also been reported to be detected in indoor dust from e-waste recycling, rural, and urban areas in South China with median concentrations ranging from 2.4×10^{3} to 3.0×10^{4} ng/g, dw, and the EDIs were reported to be 4.0–1.5×10^{3} ng/kg/day for toddlers and adults (He et al., 2018).

Historically, Nan'an village had many private household e-waste recycling workshops (Deng et al., 2006), and disassembly workshops were located in Yujiao village. Greater concentrations of ΣPBDEs would be expected in these locations (Table 2). Numbers of studies have investigated the concentrations of PBDEs in house dust and have demonstrated associations between PBDEs in house dust and human health (Dodson et al., 2012; Fromme et al., 2009; Jones-Otazo et al., 2005; Kang et al., 2011; Meeker et al., 2009; Wu et al., 2007). In this study, the concentrations of most congeners in road dust were significantly greater than those in soil (Table S2), except for BDE-85, -99, −153, and −154. Because road dust is lighter and more easily disturbed by vehicles and other human activities than soil and sediment, it is more mobile and residents, especially children, have a greater probability of exposure, through inhalation and dermal contact, and thus the health of these individuals is at greater risk. HIs of $\sum_{3\text{-}7}$PBDE derived from road dust in this study ranged from 1.7×10^{-4} to 1.6×10^{-1} for children and from 2.8×10^{-5} to 2.5×10^{-2} for adult (Table S4 and Table S5).

Sediment. The $\sum_{3\text{-}7}$PBDEs levels measured in sediment in 2014 in this research were ranged from 2.1 to 3.2×10^{3} ng/g, dw. The greatest concentration of $\sum_{3\text{-}7}$PBDEs was observed in sediments collected from a branch of the Lianjiang River, which is near Houwang village (2.1×10^{3} ng/g, dw HWV_2; Table 3), followed by the Lianjiang branch (1.3×10^{3} and 2.9×10^{2} ng/g, dw, for LJB_2 and LJB_1, respectively), the duck pond (2.5×10^{2} ng/g, dw DP_1), and the pump station (9.0×10^{1} and 6.6×10^{1} ng/g, dw, for PS_2 and PS_1, respectively). Besides reference (0.72 and 0.77 ng/g, dw), the lowest concentration of $\sum_{3\text{-}7}$PBDE was measured in sediment collected from the duck pond (2.1 ng/g, dw DP_2). Compared with our previous work in 2004 ($\sum_{3\text{-}7}$PBDE, 2.9×10^{1}–1.2×10^{3} ng/g, dw, in sediment), the concentrations of $\sum_{3\text{-}7}$PBDE in 2014 were similar to those from the sediment samples taken in 2004 (Leung et al., 2006).

BDE-99 was the dominant congener and contributed 22.1%–38.5% of the $\sum_{3\text{-}7}$PBDEs for all locations, except locations C1 (15.3%) and PS_2 (4.49%), followed by BDE-47 and -153 (Fig. 2). For HWV_1,

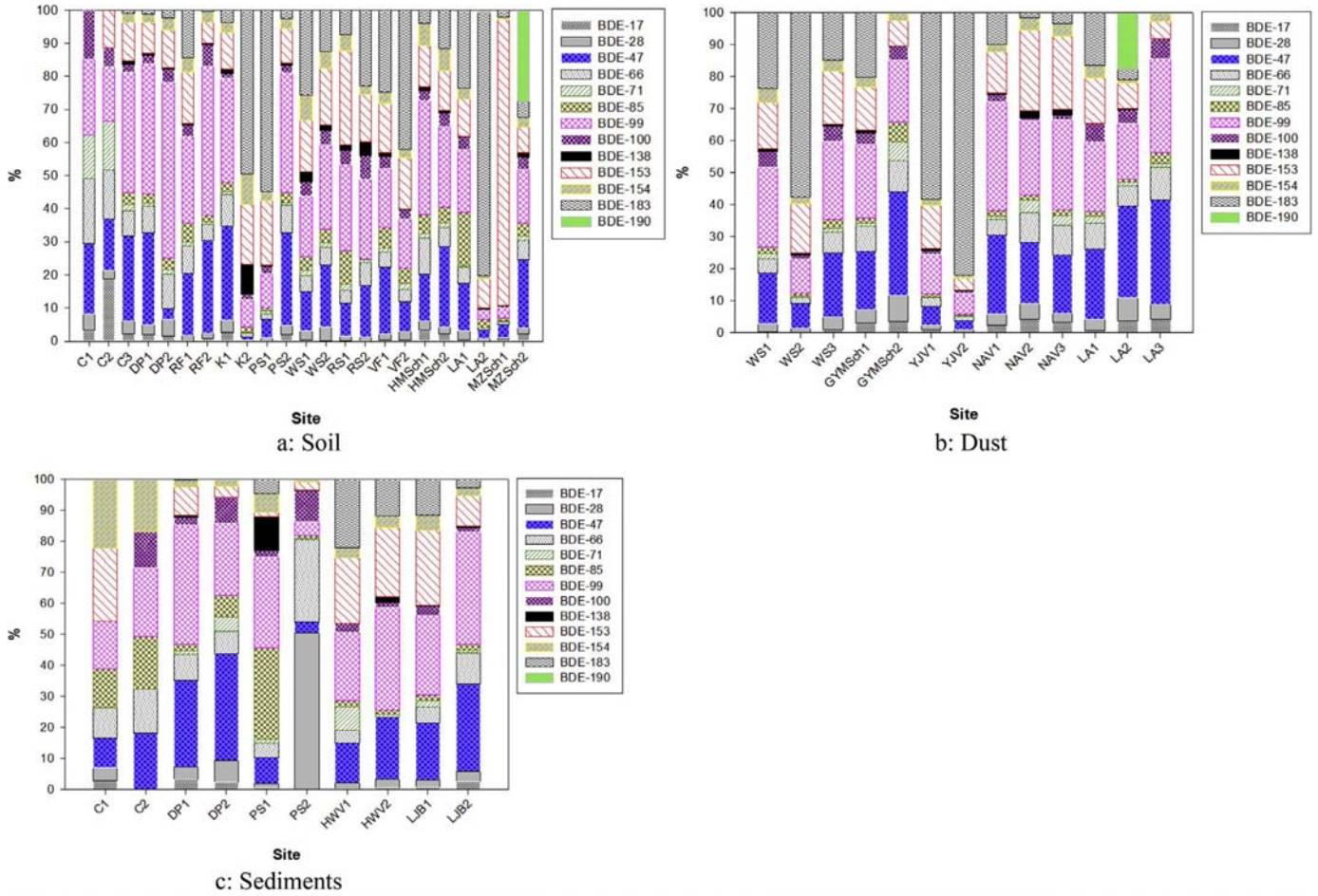

Fig. 2. Profiles of relative concentrations of BDE congeners in various environment (a: Soil; b: Dust; c: Sediment).

Table 2
Concentrations of selected PBED congeners and DPs in road dust collected at various locations in Guiyu, 2014 (ng/g, dw; n = 39).

Congener	Workshop			Guiyu middle school		Yujiao village		Nan'an village			Living area		
	WS1	WS2	WS3	Sch1	Sch2	YJV1	YJV2	NAV1	NAV2	NAV3	LA1	LA2	LA3
BDE-17	0.79	2.9	1.2	1.0×10^1	0.36	7.3×10^1	5.0	5.6×10^1	4.6×10^2	1.1×10^2	0.34	7.7	2.0
BDE-28	4.8	2.5×10^1	5.3	1.3×10^1	0.86	9.6×10^1	4.5×10^1	8.9×10^1	5.0×10^2	9.7×10^1	1.5	1.5×10^1	2.1
BDE-47	3.0×10^1	1.4×10^2	2.6×10^1	5.9×10^1	3.3	3.9×10^2	1.5×10^2	6.0×10^2	2.0×10^3	6.0×10^2	9.4	5.8×10^1	1.5×10^1
BDE-66	8.5	3.7×10^1	8.5	2.6×10^1	1.0	1.8×10^2	7.9×10^1	1.3×10^2	1.0×10^3	3.1×10^2	3.4	1.3×10^1	4.8
BDE-71	3.0	5.3	1.5	3.2	0.60	2.3×10^1	8.7	1.9×10^1	4.1×10^2	1.0×10^2	0.91	2.3	0.57
BDE-85	4.0	2.0×10^1	3.4	5.1	0.66	6.7×10^1	3.1×10^1	4.5×10^1	1.9×10^2	6.1×10^1	0.73	2.0	1.6
BDE-99	4.8×10^1	2.0×10^2	3.2×10^1	7.5×10^1	2.0	8.3×10^2	3.4×10^2	8.3×10^2	2.5×10^3	9.3×10^2	9.3	3.6×10^1	1.4×10^1
BDE-100	9.7	2.2×10^1	5.9	1.2×10^1	0.46	8.4×10^1	3.7×10^1	5.9×10^1	7.5×10^1	4.2×10^1	2.4	8.5	2.9
BDE-138	1.6	1.5×10^1	1.0	2.6	ND	5.1×10^1	1.8×10^1	1.3×10^1	2.4×10^2	6.6×10^1	ND	1.1	ND
BDE-153	2.8×10^1	2.9×10^2	2.1×10^1	4.4×10^1	0.83	9.2×10^2	2.0×10^2	3.1×10^2	2.7×10^3	7.5×10^2	6.1	1.6×10^1	2.6
BDE-154	8.4	4.4×10^1	4.4	1.0×10^1	0.25	1.1×10^2	4.7×10^1	5.7×10^1	4.2×10^2	1.3×10^2	1.7	2.4	1.3
BDE-183	4.6×10^1	1.1×10^3	1.9×10^1	6.6×10^1	ND	4.0×10^3	4.4×10^3	2.4×10^2	1.8×10^2	1.1×10^2	7.1	6.5	ND
BDE-190	ND	ND	ND	ND	ND	1.7×10^1	3.2	ND	ND	ND	ND	3.6×10^1	ND
∑PBDE	1.9×10^2	1.9×10^3	1.3×10^2	3.3×10^2	1.0×10^1	6.8×10^3	5.3×10^3	2.5×10^3	1.1×10^4	3.3×10^3	4.3×10^1	2.0×10^2	4.7×10^1
syn-DP	4.0×10^1	4.9×10^1	2.8×10^1	3.8×10^1	1.5×10^1	3.1×10^2	2.7×10^2	1.1×10^2	3.2	3.6	4.4	1.0×10^1	5.5
anti-DP	1.3×10^2	1.7×10^2	1.0×10^2	1.3×10^2	4.3×10^1	8.1×10^2	8.7×10^2	4.3×10^1	1.1×10^1	1.1×10^1	1.2×10^1	2.4×10^1	1.7×10^1
∑DP	1.7×10^2	2.2×10^2	1.3×10^2	1.7×10^2	5.8×10^1	1.1×10^3	1.1×10^3	1.5×10^2	1.4×10^1	1.5×10^1	1.6×10^1	3.4×10^1	2.3×10^1
IR (%)	76	78	78	77	74	72	76	28	77	75	73	71	76

ND, not detected.

the predominant congeners were BDE-99 (22.2%), −153 (21.1%), −183 (22.0%), and −47 (12.9%), whereas HWV_2 was similar except that the dominant congeners were BDE-99 (33.2%), −153 (22.4%), −183 (11.7%), and −47 (20.0%). However, for PS_2, the dominant congeners were BDE-28 (50.5%) and −66 (26.5%). The concentration of BDE-99, whose likely source was penta-BDE mixtures, exceeded the guideline concentration for sediment (0.40 ng/g, dw). The concentration of BDE-47, whose likely source

Table 3
Concentrations of selected PBDE and DP congeners in the sediments collected from Nanyang reservoir (reference) and Lianjiang River (ng/g, dw; n = 20) and in air particles (pg/g, dw; n = 6) in Guiyu, 2014.

Congener	Nanyang reservoir		Duck pong		Pump station		Houwang village		Lianjiang branch		Air particles Nan'an District	
	C1	C2	DP1	DP2	PS1	PS2	HWV1	HWV2	LJB1	LJB2	NAV1a	NAV2a
BDE-17	0.02	ND	8.0	0.05	0.43	ND	0.08	1.6×10^1	2.1	3.4×10^1	3.3	7.2×10^1
BDE-28	0.03	ND	1.0×10^1	0.15	0.87	4.6	0.56	8.7×10^1	6.1	4.2×10^1	ND	ND
BDE-47	0.07	0.14	6.9×10^1	0.73	5.5	3.2	3.9	6.3×10^2	4.9×10^1	3.6×10^2	8.2×10^1	5.7×10^1
BDE-66	0.07	0.11	2.1×10^1	0.15	3.2	2.4×10^1	1.2	1.9	1.4×10^1	1.3×10^2	1.7×10^1	$1.8. \times 10^1$
BDE-71	ND	ND	3.0	0.10	0.69	0.47	2.2	3.0×10^1	5.5	1.4×10^1	8.0×10^1	1.7×10^2
BDE-85	0.09	0.13	5.1	0.15	2.0×10^1	0.88	0.63	3.8×10^1	5.5	2.4×10^1	ND	2.5×10^1
BDE-99	0.11	0.17	9.5×10^1	0.49	2.0×10^1	4.1	6.7	1.0×10^3	6.8×10^1	4.6×10^2	ND	1.3×10^1
BDE-100	ND	0.09	5.9	0.18	1.3	8.9	0.80	5.0×10^1	8.3	2.0×10^1	ND	8.0×10^1
BDE-138	ND	ND	1.4	ND	7.3	0.36	ND	5.9×10^1	0.60	4.5	1.8×10^1	2.3×10^1
BDE-153	0.17	ND	2.3×10^1	0.07	0.94	2.3	6.4	7.1×10^2	6.5×10^1	1.3×10^2	1.2×10^2	2.6×10^2
BDE-154	0.16	0.13	5.5	0.05	4.0	0.76	1.0	1.2×10^2	1.3×10^1	3.1×10^1	8.3	ND
BDE-183	ND	ND	0.64	ND	3.0	ND	6.7	3.7×10^2	3.1×10^1	3.5×10^1	1.7×10^2	1.2×10^2
BDE-190	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
∑PBDE	0.72	0.77	2.5×10^2	2.1	6.6×10^1	5.0×10^1	3.0×10^1	3.2×10^3	2.7×10^2	1.3×10^3	5.0×10^2	8.4×10^2
syn-DP	0.13	ND	0.25	0.42	8.6	1.9×10^1	7.4	2.0×10^3	6.6×10^1	6.0×10^1	4.8×10^1	3.8×10^1
anti-DP	0.58	ND	0.81	1.2	3.2×10^2	8.4×10^1	2.1×10^1	5.2×10^3	9.9×10^1	1.9×10^2	1.3×10^2	1.3×10^2
∑DP	0.71	ND	1.1	1.6	3.3×10^2	1.0×10^2	2.8×10^1	7.2×10^3	1.7×10^2	2.5×10^2	1.8×10^2	1.7×10^2
IR	82	NA	76	74	97	82	74	72	60	76	73	77

ND, not detected; NA, not available.

was tetra-BDE, exceeded the guideline for tetra-BDE in sediments (3.9×10^1 ng/g, dw) (Government of Canada, 2011).

Air particles. $PM_{2.5}$ particles were sampled on the roof of a resident's 3 layers building, of which the ground floor is used for e-waste recycling. The concentration of $\sum_{3\text{-}7}$PBDEs observed in 2004 (1.4×10^4 pg/m^3) (Deng et al., 2007) was much greater than those measured in this study in 2014 (5.0×10^2 and 8.4×10^2 pg/m^3; Table 3). Of the control measures, the restriction of open combustion might reduces the levels of PBDEs in air particles in recent years. The HIs of $\sum_{3\text{-}7}$PBDE derived from air particle were 3.1×10^{-2} to 5.8×10^{-2} for children and 2.4×10^{-2} to 4.4×10^{-2} for adult (Table S3).

The $\sum_{3\text{-}7}$PBDE levels in this study were a little higher than the previous observations made in $PM_{2.5}$ particle in Hong Kong (9.2×10^1–4.2×10^2 pg/m^3) by Deng et al. (2007). It was reported that the concentrations of Σ_9PBDEs in $PM_{2.5}$ particles ranged from <0.01 to 1.0×10^3 pg/m^3 in Chinese cities (Liu et al., 2016).

Brief summary. Although octa-BDE and penta-BDE were banned in China in 2004 and 2007, respectively, concentrations of $\sum_{3\text{-}7}$PBDEs in soil, road dust, and sediment in 2014 were a little less than or close to those measured in 2004. It was reported that, in surface soils and river sediments in northern Vietnam, although levels of PBDEs decreased, but remained high with total concentrations up to 2900 ng/g dw in soils and 2200 ng/g dw in river sediments (Matsukami et al., 2017). Of the environmental matrixes surveyed in the present study, greater concentrations of PBDEs were found in road dust, especially less-brominated congeners. A potential reason for this finding is that road dust is subjected to the influence of sunlight more than soil and sediment, of which only the surface layer is affected by sunlight. It has been reported that more-brominated PBDE can be degraded to lesser brominated congeners by photolysis (Schenker et al., 2008; Watanabe and Tatsukawa, 1987; Eriksson et al., 2004; Söderström et al., 2004).

The patterns of the relative concentrations of congeners varied among matrices in this study and we found no difference in the concentrations of $\sum_{3\text{-}7}$PBDE among the matrices of soil, road dust, and sediment in the present study (Table S2). In soil, road dust, and sediment, the most abundant PBDE congeners were BDE-47, -99, −153, and −183. However, BDE-99 was not the dominant congener in $PM_{2.5}$ particulates. This result is consistent with those observed in soil and sediment in 2004 in our previous work (Leung et al., 2006, 2007; Luo et al., 2007). These congeners were also the predominant PBDEs reported in Guiyu in 2006–2008 and suggested to cause adverse health effects in residents in the vicinity of Guiyu (Wu et al., 2010; Xu et al., 2012). These BDE congeners, especially −47, −99, and −183, have been found to account for the majority of ΣPBDE in indoor dust in Hong Kong (Kang et al., 2011). Recently, eight PBDEs (BDE-28, -47, −66, −99, −100, −153, −154, −183) were analyzed in sediments in North Sea in the in Europe (total up to 205 pg/g) and BDE-47 accounted for over 60% of the total PBDE contamination (Sühring et al., 2016). Of these high detected congeners in the present study, BDE-47 and -99 are the major congeners of the commercial product penta-BDE, whereas BDE-153 occurs in penta-BDE and octa-BDE products. A large proportion of BDE-183 (13%–42%) can be found in octa-BDE. The formulations of penta-BDE and octa-BDE products are considered to have greater toxic potency than deca-BDE (Ikonomou et al., 2002; Meironyté et al., 1999). It was shown that the most persistent PBDE congener (BDE-99) disrupts neurobehavioral development and causes permanent effects on the rat male reproductive system that become apparent in adulthood (Kuriyama et al., 2005), and the concentrations of BDE-47 and -183 in human hair were positively correlated with those in indoor dust (Kang et al., 2011).

3.2. Levels and patterns of DP

Soil. The concentrations of DP in soil ranged from 3.8 to 2.1×10^3 ng/g, dw (Table 1). The greatest concentrations of the sum of congeners of DP (ΣDP, anti-DP and syn-DP) were observed in the soil samples taken along the road (8.8×10^2 and 2.1×10^3 ng/g, dw), followed by those from the pump station (3.3×10^2 ng/g, dw PS_1), Huamei middle school (2.5×10^2 ng/g, dw HMSch1), kindergarten (1.6×10^2 ng/g, dw K_2), duck pond (1.4×10^2 ng/g, dw DP_2), and living area (1.4×10^2 ng/g, dw).

The concentration of DP in this research was similar to the soil samples around a DP manufacturing plant in East China (5×10^{-1} to 2.3×10^3 ng/g dw) (Zhang et al., 2015). Recently, DP was reported to be a primary FR, with concentrations of 0.6–1.5×10^2 ng/g, dw in soil samples in Guiyu (Xu et al., 2017), which was a little lower than those in our study.

Accordance with PBDEs, the sampling sites near the workshop had high levels of DP. The sampling sites of workshop (3.8 and 4.4 × 101 ng/g, dw WS1 and WS2), which located near workshop, with electronic wastes stored in the open area, did not have high levels of DP or PBDEs compared with other sampling sites. The probable reason may be that, stored procedure made less contamination than electronic waste discharge.

The isomer ratio (IR) of DP was used in the study. The IR is calculated as the fraction of anti-DP or syn-DP in ΣDP. To compare with other studies, the IR in this study was the fraction of anti-DP (f_{anti}). The fanti values of commercial DP were reported to be in the range of 59%–80% (Hoh et al., 2006; Guerra et al., 2011; Wang et al., 2010a, b). The f_{anti} of the soil samples were ranged from 72% to 81% except for the soil samples taken from DP_2, LA_2 and RF1, which was nearly in the range of commercial DP. Anti-DP has been reported to accumulate in certain abiotic media, but in biota (especially aquatic biota), syn-DP has been reported to accumulate preferably (Sühring et al., 2016; Shen et al., 2011a,b).

Road dust. The concentrations of DP in dust ranged from 1.4×10^1 to 1.1×10^3 ng/g, dw (Table 2). The greatest concentration of ΣDP was found in road dust collected from Yujiao village (1.1×10^3 ng/g, dw), followed by that from the workshop (1.7×10^2, 2.2×10^2, and 3.3×10^2 ng/g, dw, for WS_2, WS_1, and WS_3, respectively), Guiyu middle school (1.7×10^2 ng/g, $GYMSch_1$) and Nan'an village (1.5×10^2 ng/g, dw; NAV_1). Anti-DP contributed 72%–78% to ΣDP, except NAV_1, which contributed 28% of the ΣDP. The f_{anti} values of the road dust were also in the range of commercial DP. HIs of DP derived from road dust were 2.3×10^{-9} to 2.1×10^{-7} for children and for 3.6×10^{-10} to 3.4×10^{-8} for adult (Table S3 and Table S4).

DP has been detected in indoor dust from e-waste recycling, rural, and urban areas in South China with median concentrations ranging from 2.0×10^1–1.9×10^3 ng/g dw, with their EDIs were 0.1–1.3×10^2 ng/kg/day for toddlers and adults (He et al., 2017), which were similar to our study. The concentrations of DP in dust in this research were far above the dust samples taken in five kindergartens in Hong Kong (1.3–8.7 ng/g dw in dust) in our previous work (Deng et al., 2016).

Sediment. The concentrations of DP in sediments ranged from 1.1×10^3 to 7.2×10^3 ng/g, dw (Table 3). The greatest concentration of ΣDPs was found in sediment collected from Lianjiang River near Houwang village (7.2×10^3 ng/g, dw, HWV_2; Table 3), followed by the pump station (3.3×10^2 ng/g, dw PS_1) and other locations on the Lianjiang River (1.7×10^2 and 2.5×10^2 ng/g for LJB_1 and LJB_2, respectively). Anti-DP was reported to be enriched in the sediment (Sverko et al., 2011). In the present study, greater proportions of anti-DP than syn-DP were measured in all samples. The f_{anti} of the sediment samples ranged from 60% to 82%, except PS1 on the Pump station (97%).

Because of a large $logK_{OW}$ value, DP is easily partitioned into sediments. The concentrations of DP in sediment in this research were much higher than the concentrations in sediment from many cities in China (mean DP concentration, 0.05–8 ng/g, dw) (Jia et al., 2011; Ma et al., 2011; Wang et al., 2011; Zhao et al., 2011; Zhang et al., 2015) and in the North American Great Lakes area (mean DP concentration, 0.33 ng/g –26 ng/g, dw) (Sverko et al., 2008; Shen et al., 2011a,b). However, they were similar to a reservoir sediment near e-waste recycling plant which located about 400 km far from Guiyu (mean DP concentration 7590 ng/g, dw) (Zhang et al., 2010b).

Air particles. In this study, the concentrations of DP in $PM_{2.5}$ particles were 1.8×10^2 and 1.7×10^2 pg/m^3, respectively (Table 3). Greater proportions of anti-DP than syn-DP were measured in all $PM_{2.5}$ particles. The f_{anti} of the air particle samples were 73% and 77%. HIs of DP derived from $PM_{2.5}$ air particles were 1.7×10^{-7} and 1.3×10^{-7} for children, as well as 1.3×10^{-7} and 1.0×10^{-7} for adult (Table S3), which were far less to exhibit health risk to children or adult. The HQs of DP in the manufacturing facility area and e-waste recycling area in China were reported to range from 1.1×10^{-3} to 8.8×10^{-3} and 1.0×10^{-6} to 5.3×10^{-5} through oral, dermal and inhalation pathways, respectively (Wang et al., 2013).

The concentrations of DP in $PM_{2.5}$ particles in this research were higher than those in the outdoor air in Kunming City, in the Southern China (6.6×10^1 pg/m^3) (Ren et al., 2008), and similar to those in the active air samples around a DP manufacturing plant in East China (5.5–3.3×10^3 pg/m3) (Zhang et al., 2015).

Brief summary. In this study, we detected DP in 100% of the samples collected. The f_{anti} was similar, with values (70%–80%) in most matrices and locations, and in the range of commercial DP (59%–80%), which indicated that the efficiency of transforming DPs in different environmental matrices may be similar in this survey. The results were in accord with those in the study of Xu et al. in which syn-DP and anti-DP isomers were found not stereo selectively decomposed during the e-waste processing in the soils of Guiyu (Xu et al., 2017). Several studies have focused on samples of humans and wildlife (Ren et al., 2009; Muñoz-Arnanz et al., 2011; Ben et al., 2013), with only a few reports of abiotic environmental matrices (Ren et al., 2008; Yu et al., 2010). Some studies have attempted to correlate concentrations of DP in biota/human and specific environmental matrices (Tomy et al., 2007; Wu et al., 2009; Zhang et al., 2010a; Zheng et al., 2010; Chen et al., 2011). However, these studies did not consider all potential sources of DP that might result in exposure to humans or wildlife that could have adverse effects. This study provides baseline concentrations of DP in a range of environmental matrices that can be used in the future to determine whether concentrations are increasing or whether there has been bioaccumulation/biomagnification or trophic magnification.

4. Conclusions

Even though open combustion and dumping of processed materials have been restricted by the local government for several years, concentrations of $\sum_{3\text{-}7}$PBDEs in soil, road dust, and sediment in 2014 were a little less than or close to those measured in 2004. The concentrations of $\Sigma_{3\text{-}7}$PBDEs in road dust were greater than those in soils or sediments. The concentrations of $\Sigma_{3\text{-}7}$PBDEs in $PM_{2.5}$ particulates were approximately 100-fold less than they were in 2004. DP was detected in 100% of the samples collected with high levels, and the fractions of anti-DP (f_{anti}) were consistent among samples, most of which ranging from 70% to 80% and was in the range of commercial DPs (59–80%). The health risk assessment showed that HIs of PBDEs or DPs for child and adult were all lower than 0.16.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at https://doi.org/10.1016/j.chemosphere.2018.02.041.

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PBDEs and Dechlorane Plus in the Environment of Guiyu, Southeast China: A Historical Location for E-Waste Recycling (2004 and 2014)

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Table S1. Descriptions of sampling locations.

	Location	**Location no.**	**GPS coordinates**	**Description**
Soil	Reservoir (reference)	S_1	N23.378225 E116.372927	Nanyang reservoir.
	Duck pond	S_2	N23.331667 E116.356944	A location within the dried Lianjiang river floor.
	Rice field	S_3	N23.327778 E116.346111	A rice field irrigated by water from Lianjiang River.
	Kindergarten	S_4	N23.33172 E116.355073	A kindergarten near workshops.
	Pump station	S_5	N23.331519 E116.355697	A pump station pumping water from Lianjiang river, next to workshop and duck raising area.
	Workshop	S_6	N23.330277 E116.346389	A location near workshop, with electronic wastes stored in the open area.
	Road side	S_7	N23.322675 E116.352082	Near Yujiao village.
	Vegetable field	S_8	N23.32336 E116.352409	Near Yujiao village and next to the branch of Lianjiang river.
	Huamei middle school	S_9	N23.316586 E116.35528	A road side near Huamei middle school.
	Living area	S_{10}	N23.325833 E116.329444	A location with historically intensive e-waste recycling activities within a living area.
	Meizhou middle	S_{11}	N23.308334	At the bank of Lianjiang river, next to Meizhou middle

	school		E116.339722	school.
Road dust	Workshop	D_1	N23.330277 E116.346389	A location near workshop, with electronic wastes stored in the open area.
	Guiyu middle school	D_2	N23.322169 E116.357871	A road side in front of Guiyu middle school, 50 m away from the main market street.
	Yujiao village	D_3	N23.321671 E116.350972	A road side in front of a popular restaurant, next to one branch of Lianjiang river.
	Nan'an district	D_4	N23.325845 E116.329465	A location with historically intensive e-waste recycling activities within a living area.
	Living area	D_5	N23.328143 E116.356074	A living area with great density of houses, next to one branch of Lianjiang river.
Sediment	Reservoir (reference)	Sed_1	N23.378225 E116.372927	Nanyang reservoir
	Duck pond	Sed_2	N23.331667 E116.356944	A location within the dried Lianjiang River floor.
	Pump station	Sed_3	N23.331519 E116.355697	A pump station pumping water from Lianjiang River, next to workshop and duck raising area
	Houwang village	Sed_4	N23.318889 E116.333889	A river side near living area. Residents were used to do laundry.
	Lianjiang River branch	Sed_5	N23.326944 E116.353611	A newly constructed branch of Lianjiang River, next to a main road for vehicles.
Air	Nan'an 4^{th} Road	Air	N23.325833 E116.329444	On the roof of a resident's 3 layers building, the ground floor is used for e-waste recycling.

Table S2. Comparison of congeners in various environmental matrices (ANOVA followed by Duncan's test).

Congener	**Soil (n=20)**	**Road dust (n=13)**	**Sediment (n=8)**
BDE-17	a	b	ab
BDE-28	a	b	ab
BDE-47	a	b	ab
BDE-66	a	b	a
BDE-71	a	b	ab
BDE-85	a	a	a
BDE-99	a	a	a
BDE-100	a	b	ab
BDE-138	a	b	ab
BDE-153	a	a	a
BDE-154	a	a	a
BDE-183	a	b	a
BDE-190	NA	NA	NA
∑PBDE	a	a	a
syn-DP	a	a	a
anti-DP	a	a	a
∑DP	a	a	a

Different letters ("a", "b" and "ab") in the same row indicate significant different (P<0.05). NA, not available.

Table S3 Non-cancer risk values of PBDEs and DP in air particles for child and adult.

Congener	Children		Adults	
	NAV1	NAV2	NAV1	NAV2
BDE-17	2.99E-04	6.53E-03	2.28E-04	4.97E-03
BDE-28	0	0	0	0
BDE-71	7.25E-03	1.54E-02	5.52E-03	1.17E-02
BDE-47	7.43E-03	5.17E-03	5.66E-03	3.93E-03
BDE-66	1.54E-03	1.63E-03	1.17E-03	1.24E-03
BDE100	0	7.25E-03	0	5.52E-03
BDE99	0	1.18E-03	0	8.97E-04
BDE85	0	2.27E-03	0	1.73E-03
BDE154	3.76E-04	0	2.86E-04	0
BDE153	5.44E-03	1.18E-02	4.14E-03	8.97E-03
BDE138	8.16E-04	1.04E-03	6.21E-04	7.94E-04
BDE183	7.70E-03	5.44E-03	5.87E-03	4.14E-03
BDE-190	0	0	0	0
∑PBDE	3.09E-02	5.77E-02	2.35E-02	4.39E-02
syn-DP	4.27E-08	3.38E-08	3.25E-08	2.57E-08
anti-DP	1.16E-07	1.16E-07	8.80E-08	8.80E-08
∑DP	1.69E-07	1.33E-07	1.29E-07	1.01E-07

Table S4 Non-cancer risk values of PBDEs and DP in dust for child

Congener	WS1			WS2			WS3		
	Non-dietary ingestion	Intake through dermal contact	Total	Non-dietary ingestion	Intake through dermal contact	Total	Non-dietary ingestion	Intake through dermal contact	Total
BDE-17	1.30E-05	9.60E-07	1.40E-05	4.77E-05	3.52E-06	5.13E-05	1.98E-05	1.46E-06	2.12E-05
BDE-28	7.90E-05	5.83E-06	8.48E-05	4.11E-04	3.04E-05	4.42E-04	8.72E-05	6.44E-06	9.37E-05
BDE-71	4.94E-05	3.65E-06	5.30E-05	8.72E-05	6.44E-06	9.37E-05	2.47E-05	1.82E-06	2.65E-05
BDE-47	4.94E-04	3.65E-05	5.30E-04	2.30E-03	1.70E-04	2.47E-03	4.28E-04	3.16E-05	4.60E-04
BDE-66	1.40E-04	1.03E-05	1.50E-04	6.09E-04	4.50E-05	6.54E-04	1.40E-04	1.03E-05	1.50E-04
BDE100	1.60E-04	1.18E-05	1.71E-04	3.62E-04	2.67E-05	3.89E-04	9.71E-05	7.17E-06	1.04E-04
BDE99	7.90E-04	5.83E-05	8.48E-04	3.29E-03	2.43E-04	3.53E-03	5.27E-04	3.89E-05	5.66E-04
BDE85	6.58E-05	4.86E-06	7.07E-05	3.29E-04	2.43E-05	3.53E-04	5.60E-05	4.13E-06	6.01E-05
BDE154	6.91E-05	5.10E-06	7.42E-05	3.62E-04	2.67E-05	3.89E-04	3.62E-05	2.67E-06	3.89E-05
BDE153	2.30E-04	1.70E-05	2.47E-04	2.39E-03	1.76E-04	2.56E-03	1.73E-04	1.28E-05	1.86E-04
BDE138	1.32E-05	9.72E-07	1.41E-05	1.23E-04	9.12E-06	1.33E-04	8.23E-06	6.08E-07	8.84E-06
BDE183	3.79E-04	2.80E-05	4.06E-04	9.05E-03	6.68E-04	9.72E-03	1.56E-04	1.15E-05	1.68E-04
BDE-190	0	0	0.00E+00	0	0	0.00E+00	0	0	0.00E+00
∑PBDE	2.48E-03	1.83E-04	2.67E-03	1.94E-02	1.43E-03	2.08E-02	1.75E-03	1.29E-04	1.88E-03
syn-DP	6.45E-09	4.77E-10	6.93E-09	7.91E-09	5.84E-10	8.49E-09	4.52E-09	3.34E-10	4.85E-09
anti-DP	2.10E-08	1.55E-09	2.25E-08	2.74E-08	2.03E-09	2.95E-08	1.61E-08	1.19E-09	1.73E-08
∑DP	2.58E-08	1.91E-09	2.77E-08	3.07E-08	2.26E-09	3.29E-08	1.77E-08	1.31E-09	1.91E-08

Congener	Sch1			Sch2			YJV1		
	Non-dietary ingestion	Intake through dermal contact	Total	Non-dietary ingestion	Intake through dermal contact	Total	Non-dietary ingestion	Intake through dermal contact	Total
BDE-17	1.65E-04	1.22E-05	1.77E-04	5.93E-06	4.38E-07	6.36E-06	1.20E-03	8.87E-05	1.29E-03
BDE-28	2.14E-04	1.58E-05	2.30E-04	1.42E-05	1.05E-06	1.52E-05	1.58E-03	1.17E-04	1.70E-03
BDE-71	5.27E-05	3.89E-06	5.66E-05	9.88E-06	7.29E-07	1.06E-05	3.79E-04	2.80E-05	4.06E-04
BDE-47	9.71E-04	7.17E-05	1.04E-03	5.43E-05	4.01E-06	5.83E-05	6.42E-03	4.74E-04	6.89E-03
BDE-66	4.28E-04	3.16E-05	4.60E-04	1.65E-05	1.22E-06	1.77E-05	2.96E-03	2.19E-04	3.18E-03
BDE100	1.98E-04	1.46E-05	2.12E-04	7.57E-06	5.59E-07	8.13E-06	1.38E-03	1.02E-04	1.48E-03
BDE99	1.23E-03	9.12E-05	1.33E-03	3.29E-05	2.43E-06	3.53E-05	1.37E-02	1.01E-03	1.47E-02
BDE85	8.39E-05	6.20E-06	9.01E-05	1.09E-05	8.02E-07	1.17E-05	1.10E-03	8.14E-05	1.18E-03
BDE154	8.23E-05	6.08E-06	8.84E-05	2.06E-06	1.52E-07	2.21E-06	9.05E-04	6.68E-05	9.72E-04
BDE153	3.62E-04	2.67E-05	3.89E-04	6.83E-06	5.04E-07	7.33E-06	7.57E-03	5.59E-04	8.13E-03
BDE138	2.14E-05	1.58E-06	2.30E-05	0	0	0.00E+00	4.20E-04	3.10E-05	4.51E-04
BDE183	5.43E-04	4.01E-05	5.83E-04	0	0	0.00E+00	3.29E-02	2.43E-03	3.53E-02
BDE-190	0	0	0.00E+00	0	0	0.00E+00	1.40E-04	1.03E-05	1.50E-04
∑PBDE	4.35E-03	3.22E-04	4.68E-03	1.61E-04	1.19E-05	1.73E-04	7.06E-02	5.22E-03	7.59E-02
syn-DP	6.13E-09	4.53E-10	6.58E-09	2.42E-09	1.79E-10	2.60E-09	5.00E-08	3.69E-09	5.37E-08
anti-DP	2.10E-08	1.55E-09	2.25E-08	6.94E-09	5.12E-10	7.45E-09	1.31E-07	9.65E-09	1.40E-07
∑DP	2.42E-08	1.79E-09	2.60E-08	9.84E-09	7.27E-10	1.06E-08	1.94E-07	1.43E-08	2.08E-07

Congener	YJV2			NAV1			NAV2		
	Non-dietary ingestion	Intake through dermal contact	Total	Non-dietary ingestion	Intake through dermal contact	Total	Non-dietary ingestion	Intake through dermal contact	Total
BDE-17	8.23E-05	6.08E-06	8.84E-05	9.22E-04	6.81E-05	9.90E-04	7.57E-03	5.59E-04	8.13E-03
BDE-28	7.41E-04	5.47E-05	7.95E-04	1.46E-03	1.08E-04	1.57E-03	8.23E-03	6.08E-04	8.84E-03
BDE-71	1.43E-04	1.06E-05	1.54E-04	3.13E-04	2.31E-05	3.36E-04	6.75E-03	4.98E-04	7.25E-03
BDE-47	2.47E-03	1.82E-04	2.65E-03	9.88E-03	7.29E-04	1.06E-02	3.29E-02	2.43E-03	3.53E-02
BDE-66	1.30E-03	9.60E-05	1.40E-03	2.14E-03	1.58E-04	2.30E-03	1.65E-02	1.22E-03	1.77E-02
BDE100	6.09E-04	4.50E-05	6.54E-04	9.71E-04	7.17E-05	1.04E-03	1.23E-03	9.12E-05	1.33E-03
BDE99	5.60E-03	4.13E-04	6.01E-03	1.37E-02	1.01E-03	1.47E-02	4.11E-02	3.04E-03	4.42E-02
BDE85	5.10E-04	3.77E-05	5.48E-04	7.41E-04	5.47E-05	7.95E-04	3.13E-03	2.31E-04	3.36E-03
BDE154	3.87E-04	2.86E-05	4.15E-04	4.69E-04	3.46E-05	5.04E-04	3.46E-03	2.55E-04	3.71E-03
BDE153	1.65E-03	1.22E-04	1.77E-03	2.55E-03	1.88E-04	2.74E-03	2.22E-02	1.64E-03	2.39E-02
BDE138	1.48E-04	1.09E-05	1.59E-04	1.07E-04	7.90E-06	1.15E-04	1.98E-03	1.46E-04	2.12E-03
BDE183	3.62E-02	2.67E-03	3.89E-02	1.98E-03	1.46E-04	2.12E-03	1.48E-03	1.09E-04	1.59E-03
BDE-190	2.63E-05	1.94E-06	2.83E-05	0	0	0.00E+00	0	0	0.00E+00
∑PBDE	4.99E-02	3.68E-03	5.35E-02	3.52E-02	2.60E-03	3.78E-02	1.47E-01	1.08E-02	1.57E-01
syn-DP	4.36E-08	3.22E-09	4.68E-08	1.77E-08	1.31E-09	1.91E-08	5.16E-10	3.81E-11	5.54E-10
anti-DP	1.40E-07	1.04E-08	1.51E-07	6.94E-09	5.12E-10	7.45E-09	1.77E-09	1.31E-10	1.91E-09
∑DP	1.77E-07	1.31E-08	1.91E-07	7.10E-08	5.24E-09	7.62E-08	2.10E-09	1.55E-10	2.25E-09

Congener	NAV3			LA1			LA2		
	Non-dietary ingestion	Intake through dermal contact	Total	Non-dietary ingestion	Intake through dermal contact	Total	Non-dietary ingestion	Intake through dermal contact	Total
BDE-17	1.81E-03	1.34E-04	1.94E-03	5.60E-06	4.13E-07	6.01E-06	1.27E-04	9.36E-06	1.36E-04
BDE-28	1.60E-03	1.18E-04	1.71E-03	2.47E-05	1.82E-06	2.65E-05	2.47E-04	1.82E-05	2.65E-04
BDE-71	1.65E-03	1.22E-04	1.77E-03	1.50E-05	1.11E-06	1.61E-05	3.79E-05	2.80E-06	4.06E-05
BDE-47	9.88E-03	7.29E-04	1.06E-02	1.55E-04	1.14E-05	1.66E-04	9.55E-04	7.05E-05	1.03E-03
BDE-66	5.10E-03	3.77E-04	5.48E-03	5.60E-05	4.13E-06	6.01E-05	2.14E-04	1.58E-05	2.30E-04
BDE100	6.91E-04	5.10E-05	7.42E-04	3.95E-05	2.92E-06	4.24E-05	1.40E-04	1.03E-05	1.50E-04
BDE99	1.53E-02	1.13E-03	1.64E-02	1.53E-04	1.13E-05	1.64E-04	5.93E-04	4.38E-05	6.36E-04
BDE85	1.00E-03	7.41E-05	1.08E-03	1.20E-05	8.87E-07	1.29E-05	3.29E-05	2.43E-06	3.53E-05
BDE154	1.07E-03	7.90E-05	1.15E-03	1.40E-05	1.03E-06	1.50E-05	1.98E-05	1.46E-06	2.12E-05
BDE153	6.17E-03	4.56E-04	6.63E-03	5.02E-05	3.71E-06	5.39E-05	1.32E-04	9.72E-06	1.41E-04
BDE138	5.43E-04	4.01E-05	5.83E-04	0	0	0.00E+00	9.05E-06	6.68E-07	9.72E-06
BDE183	9.05E-04	6.68E-05	9.72E-04	5.84E-05	4.31E-06	6.27E-05	5.35E-05	3.95E-06	5.74E-05
BDE-190	0	0	0.00E+00	0	0	0.00E+00	2.96E-04	2.19E-05	3.18E-04
∑PBDE	4.57E-02	3.38E-03	4.91E-02	5.83E-04	4.31E-05	6.26E-04	2.86E-03	2.11E-04	3.07E-03
syn-DP	5.81E-10	4.29E-11	6.24E-10	7.10E-10	5.24E-11	7.62E-10	1.61E-09	1.19E-10	1.73E-09
anti-DP	1.77E-09	1.31E-10	1.91E-09	1.94E-09	1.43E-10	2.08E-09	3.87E-09	2.86E-10	4.16E-09
∑DP	2.26E-09	1.67E-10	2.43E-09	2.90E-09	2.14E-10	3.12E-09	6.45E-09	4.77E-10	6.93E-09

Congener	LA3		
	Non-dietary ingestion	Intake through dermal contact	Total
BDE-17	3.29E-05	2.43E-06	3.53E-05
BDE-28	3.46E-05	2.55E-06	3.71E-05
BDE-71	9.38E-06	6.93E-07	1.01E-05
BDE-47	2.47E-04	1.82E-05	2.65E-04
BDE-66	7.90E-05	5.83E-06	8.48E-05
BDE100	4.77E-05	3.52E-06	5.13E-05
BDE99	2.30E-04	1.70E-05	2.47E-04
BDE85	2.63E-05	1.94E-06	2.83E-05
BDE154	1.07E-05	7.90E-07	1.15E-05
BDE153	2.14E-05	1.58E-06	2.30E-05
BDE138	0	0	0.00E+00
BDE183	0	0	0.00E+00
BDE-190	0	0	0.00E+00
∑PBDE	7.39E-04	5.46E-05	7.94E-04
syn-DP	8.87E-10	6.55E-11	9.53E-10
anti-DP	2.74E-09	2.03E-10	2.95E-09
∑DP	3.55E-09	2.62E-10	3.81E-09

Table S5 Non-cancer risk values of PBDEs and DP in dust for adult

Congener	WS1			WS2			WS3		
	Non-dietary ingestion	Intake through dermal contact	Total	Non-dietary ingestion	Intake through dermal contact	Total	Non-dietary ingestion	Intake through dermal contact	Total
BDE-17	1.78E-06	4.74E-07	2.26E-06	6.54E-06	1.74E-06	8.28E-06	2.71E-06	7.20E-07	3.43E-06
BDE-28	1.08E-05	2.88E-06	1.37E-05	5.64E-05	1.50E-05	7.14E-05	1.20E-05	3.18E-06	1.51E-05
BDE-71	6.77E-06	1.80E-06	8.57E-06	1.20E-05	3.18E-06	1.51E-05	3.38E-06	9.00E-07	4.28E-06
BDE-47	6.77E-05	1.80E-05	8.57E-05	3.16E-04	8.40E-05	4.00E-04	5.87E-05	1.56E-05	7.43E-05
BDE-66	1.92E-05	5.10E-06	2.43E-05	8.35E-05	2.22E-05	1.06E-04	1.92E-05	5.10E-06	2.43E-05
BDE100	2.19E-05	5.82E-06	2.77E-05	4.96E-05	1.32E-05	6.28E-05	1.33E-05	3.54E-06	1.69E-05
BDE99	1.08E-04	2.88E-05	1.37E-04	4.51E-04	1.20E-04	5.71E-04	7.22E-05	1.92E-05	9.14E-05
BDE85	9.03E-06	2.40E-06	1.14E-05	4.51E-05	1.20E-05	5.71E-05	7.67E-06	2.04E-06	9.71E-06
BDE154	9.48E-06	2.52E-06	1.20E-05	4.96E-05	1.32E-05	6.28E-05	4.96E-06	1.32E-06	6.28E-06
BDE153	3.16E-05	8.40E-06	4.00E-05	3.27E-04	8.70E-05	4.14E-04	2.37E-05	6.30E-06	3.00E-05
BDE138	1.81E-06	4.80E-07	2.28E-06	1.69E-05	4.50E-06	2.14E-05	1.13E-06	3.00E-07	1.43E-06
BDE183	5.19E-05	1.38E-05	6.57E-05	1.24E-03	3.30E-04	1.57E-03	2.14E-05	5.70E-06	2.71E-05
BDE-190	0	0	0.00E+00	0	0	0.00E+00	0	0	0.00E+00
∑PBDE	3.40E-04	9.04E-05	4.31E-04	2.66E-03	7.06E-04	3.36E-03	2.40E-04	6.39E-05	3.04E-04
syn-DP	8.85E-10	2.35E-10	1.12E-09	1.08E-09	2.88E-10	1.37E-09	6.19E-10	1.65E-10	7.84E-10
anti-DP	2.88E-09	7.64E-10	3.64E-09	3.76E-09	1.00E-09	4.76E-09	2.21E-09	5.88E-10	2.80E-09
∑DP	3.54E-09	9.41E-10	4.48E-09	4.20E-09	1.12E-09	5.32E-09	2.43E-09	6.47E-10	3.08E-09

Congener	Sch1	Sch2	YJV1

	Non-dietary ingestion	Intake through dermal contact	Total	Non-dietary ingestion	Intake through dermal contact	Total	Non-dietary ingestion	Intake through dermal contact	Total
BDE-17	2.26E-05	6.00E-06	2.86E-05	8.12E-07	2.16E-07	1.03E-06	1.65E-04	4.38E-05	2.09E-04
BDE-28	2.93E-05	7.80E-06	3.71E-05	1.94E-06	5.16E-07	2.46E-06	2.17E-04	5.76E-05	2.74E-04
BDE-71	7.22E-06	1.92E-06	9.14E-06	1.35E-06	3.60E-07	1.71E-06	5.19E-05	1.38E-05	6.57E-05
BDE-47	1.33E-04	3.54E-05	1.69E-04	7.45E-06	1.98E-06	9.43E-06	8.80E-04	2.34E-04	1.11E-03
BDE-66	5.87E-05	1.56E-05	7.43E-05	2.26E-06	6.00E-07	2.86E-06	4.06E-04	1.08E-04	5.14E-04
BDE100	2.71E-05	7.20E-06	3.43E-05	1.04E-06	2.76E-07	1.31E-06	1.90E-04	5.04E-05	2.40E-04
BDE99	1.69E-04	4.50E-05	2.14E-04	4.51E-06	1.20E-06	5.71E-06	1.87E-03	4.98E-04	2.37E-03
BDE85	1.15E-05	3.06E-06	1.46E-05	1.49E-06	3.96E-07	1.89E-06	1.51E-04	4.02E-05	1.91E-04
BDE154	1.13E-05	3.00E-06	1.43E-05	2.82E-07	7.50E-08	3.57E-07	1.24E-04	3.30E-05	1.57E-04
BDE153	4.96E-05	1.32E-05	6.28E-05	9.36E-07	2.49E-07	1.19E-06	1.04E-03	2.76E-04	1.31E-03
BDE138	2.93E-06	7.80E-07	3.71E-06	0	0	0.00E+00	5.75E-05	1.53E-05	7.28E-05
BDE183	7.45E-05	1.98E-05	9.43E-05	0	0	0.00E+00	4.51E-03	1.20E-03	5.71E-03
BDE-190	0	0	0.00E+00	0	0	0.00E+00	1.92E-05	5.10E-06	2.43E-05
∑PBDE	5.97E-04	1.59E-04	7.56E-04	2.21E-05	5.87E-06	2.79E-05	9.68E-03	2.57E-03	1.23E-02
syn-DP	8.41E-10	2.23E-10	1.06E-09	3.32E-10	8.82E-11	4.20E-10	6.86E-09	1.82E-09	8.68E-09
anti-DP	2.88E-09	7.64E-10	3.64E-09	9.51E-10	2.53E-10	1.20E-09	1.79E-08	4.76E-09	2.27E-08
∑DP	3.32E-09	8.82E-10	4.20E-09	1.35E-09	3.59E-10	1.71E-09	2.65E-08	7.06E-09	3.36E-08

Congener	NAV3			LA1			LA2		
	Non-dietary ingestion	Intake through dermal contact	Total	Non-dietary ingestion	Intake through dermal contact	Total	Non-dietary ingestion	Intake through dermal contact	Total
BDE-17	1.13E-05	3.00E-06	1.43E-05	1.26E-04	3.36E-05	1.60E-04	1.04E-03	2.76E-04	1.31E-03
BDE-28	1.02E-04	2.70E-05	1.29E-04	2.01E-04	5.34E-05	2.54E-04	1.13E-03	3.00E-04	1.43E-03
BDE-71	1.96E-05	5.22E-06	2.48E-05	4.29E-05	1.14E-05	5.43E-05	9.25E-04	2.46E-04	1.17E-03
BDE-47	3.38E-04	9.00E-05	4.28E-04	1.35E-03	3.60E-04	1.71E-03	4.51E-03	1.20E-03	5.71E-03
BDE-66	1.78E-04	4.74E-05	2.26E-04	2.93E-04	7.80E-05	3.71E-04	2.26E-03	6.00E-04	2.86E-03
BDE100	8.35E-05	2.22E-05	1.06E-04	1.33E-04	3.54E-05	1.69E-04	1.69E-04	4.50E-05	2.14E-04
BDE99	7.67E-04	2.04E-04	9.71E-04	1.87E-03	4.98E-04	2.37E-03	5.64E-03	1.50E-03	7.14E-03
BDE85	6.99E-05	1.86E-05	8.85E-05	1.02E-04	2.70E-05	1.29E-04	4.29E-04	1.14E-04	5.43E-04
BDE154	5.30E-05	1.41E-05	6.71E-05	6.43E-05	1.71E-05	8.14E-05	4.74E-04	1.26E-04	6.00E-04
BDE153	2.26E-04	6.00E-05	2.86E-04	3.50E-04	9.30E-05	4.43E-04	3.05E-03	8.10E-04	3.86E-03
BDE138	2.03E-05	5.40E-06	2.57E-05	1.47E-05	3.90E-06	1.86E-05	2.71E-04	7.20E-05	3.43E-04
BDE183	4.96E-03	1.32E-03	6.28E-03	2.71E-04	7.20E-05	3.43E-04	2.03E-04	5.40E-05	2.57E-04
BDE-190	3.61E-06	9.60E-07	4.57E-06	0	0	0.00E+00	0	0	0.00E+00
∑PBDE	6.84E-03	1.82E-03	8.65E-03	4.82E-03	1.28E-03	6.11E-03	2.01E-02	5.34E-03	2.54E-02
syn-DP	5.97E-09	1.59E-09	7.56E-09	2.43E-09	6.47E-10	3.08E-09	7.08E-11	1.88E-11	8.96E-11
anti-DP	1.92E-08	5.12E-09	2.44E-08	9.51E-10	2.53E-10	1.20E-09	2.43E-10	6.47E-11	3.08E-10
∑DP	2.43E-08	6.47E-09	3.08E-08	9.73E-09	2.59E-09	1.23E-08	2.88E-10	7.64E-11	3.64E-10

Congener	YJV2			NAV1			NAV2		
	Non-dietary ingestion	Intake through dermal contact	Total	Non-dietary ingestion	Intake through dermal contact	Total	Non-dietary ingestion	Intake through dermal contact	Total
BDE-17	2.48E-04	6.60E-05	3.14E-04	7.67E-07	2.04E-07	9.71E-07	1.74E-05	4.62E-06	2.20E-05
BDE-28	2.19E-04	5.82E-05	2.77E-04	3.38E-06	9.00E-07	4.28E-06	3.38E-05	9.00E-06	4.28E-05
BDE-71	2.26E-04	6.00E-05	2.86E-04	2.05E-06	5.46E-07	2.60E-06	5.19E-06	1.38E-06	6.57E-06
BDE-47	1.35E-03	3.60E-04	1.71E-03	2.12E-05	5.64E-06	2.68E-05	1.31E-04	3.48E-05	1.66E-04
BDE-66	6.99E-04	1.86E-04	8.85E-04	7.67E-06	2.04E-06	9.71E-06	2.93E-05	7.80E-06	3.71E-05
BDE100	9.48E-05	2.52E-05	1.20E-04	5.42E-06	1.44E-06	6.85E-06	1.92E-05	5.10E-06	2.43E-05
BDE99	2.10E-03	5.58E-04	2.66E-03	2.10E-05	5.58E-06	2.66E-05	8.12E-05	2.16E-05	1.03E-04
BDE85	1.38E-04	3.66E-05	1.74E-04	1.65E-06	4.38E-07	2.09E-06	4.51E-06	1.20E-06	5.71E-06
BDE154	1.47E-04	3.90E-05	1.86E-04	1.92E-06	5.10E-07	2.43E-06	2.71E-06	7.20E-07	3.43E-06
BDE153	8.46E-04	2.25E-04	1.07E-03	6.88E-06	1.83E-06	8.71E-06	1.81E-05	4.80E-06	2.28E-05
BDE138	7.45E-05	1.98E-05	9.43E-05	0	0	0.00E+00	1.24E-06	3.30E-07	1.57E-06
BDE183	1.24E-04	3.30E-05	1.57E-04	8.01E-06	2.13E-06	1.01E-05	7.33E-06	1.95E-06	9.28E-06
BDE-190	0	0	0.00E+00	0	0	0.00E+00	4.06E-05	1.08E-05	5.14E-05
∑PBDE	6.27E-03	1.67E-03	7.93E-03	7.99E-05	2.13E-05	1.01E-04	3.91E-04	1.04E-04	4.96E-04
syn-DP	7.96E-11	2.12E-11	1.01E-10	9.73E-11	2.59E-11	1.23E-10	2.21E-10	5.88E-11	2.80E-10
anti-DP	2.43E-10	6.47E-11	3.08E-10	2.65E-10	7.06E-11	3.36E-10	5.31E-10	1.41E-10	6.72E-10
∑DP	3.10E-10	8.23E-11	3.92E-10	3.98E-10	1.06E-10	5.04E-10	8.85E-10	2.35E-10	1.12E-09

Congener	LA3		
	Non-dietary ingestion	Intake through dermal contact	Total
BDE-17	4.51E-06	1.20E-06	5.71E-06
BDE-28	4.74E-06	1.26E-06	6.00E-06
BDE-71	1.29E-06	3.42E-07	1.63E-06
BDE-47	3.38E-05	9.00E-06	4.28E-05
BDE-66	1.08E-05	2.88E-06	1.37E-05
BDE100	6.54E-06	1.74E-06	8.28E-06
BDE99	3.16E-05	8.40E-06	4.00E-05
BDE85	3.61E-06	9.60E-07	4.57E-06
BDE154	1.47E-06	3.90E-07	1.86E-06
BDE153	2.93E-06	7.80E-07	3.71E-06
BDE138	0	0	0.00E+00
BDE183	0	0	0.00E+00
BDE-190	0	0	0.00E+00
∑PBDE	1.01E-04	2.69E-05	1.28E-04
syn-DP	1.22E-10	3.23E-11	1.54E-10
anti-DP	3.76E-10	1.00E-10	4.76E-10
∑DP	4.87E-10	1.29E-10	6.16E-10

S6 Details of Method

Collection of Samples. Soils were collected from 11 locations, including one reference location near Nanyang reservoir (Figure 1, shown as S_1 to S_{11}) and sediments were collected from 5 locations. In addition to this regional reference location, the sampling locations included a duck pond, a rice field, a kindergarten, a pump station, a workshop, a road side, a vegetable field, a living area, and two middle schools (Huamei middle school and Meizhou middle school; for descriptions of the locations see Table S1). Two samples were collected at each location (and three samples were collected from the reference location). Each sample was divided into six aliquots of approximately 50 g each, mixed well, and stored in a plastic zip bag and transported to the laboratory within 2 days.

Dust was collected from five areas, including a workshop, Guiyu middle school, Yujiao village, Nan'an district (a historical intensive workshop area), and a living area (Table 2, shown as D_1 to D_5). There were two or three sampling locations for each area. At each location, six subsamples were collected and mixed well to form a composite sample. The mass of each sub-sample was 10 to 20 g. The composite samples were stored in plastic zip bags and transported to the laboratory within 2 days.

Sediments were collected from five locations, including the reference location in Nanyang reservoir (Figure 1, shown as Sed_1 to Sed_5). Triplicate samples were collected at each location. For each location, two samples (each combining six subsamples) were collected (approximately 50 each) stored in a plastic zip bag and transported to the laboratory within 2 days.

$PM_{2.5}$ was collected on the roof of a four-story family building during 23-25 September, 2014. The samplers were placed between 1 and 1.2 m above the roof. Ambient air was taken at an average flow rate of 5 L/min with a Mini Vol^{TM} air sampler (Airmetrics, USA) equipped with an impaction type $PM_{2.5}$ inlet and a Whatman quartz 47 mm fiber filter for approximately 20 h. Blank samples were collected by placing a sterilized filter inside the sampler for 20 h while not in operation before every sampling period. After sampling, the filters were wrapped in aluminum foil and stored in zip bags at −20°C.

Concentrations of $PM_{2.5}$ were determined by weighing the filters before and after exposure. Before and after the sampling periods, the filters were conditioned in a desiccator for 24 to 48 h and then weighed in an air-conditioned weighing room (approximately 22°C ± 2°C; relative humidity, 40% ± 5%) according to USEPA acceptance criteria (USEPA, 1998). The mass of the particulates was determined gravimetrically by use of a micro balance (Sartorius) with a sensitivity of ±0.001 mg for filters. All treatments were carefully handled using a pair of stainless steel scissors.

Extraction, cleanup, and quantification. The masses of aliquots of soil, dust, and sediment with sodium sulfate were determined and placed into centrifuge tubes. The samples and filters were then spiked with internal standard (^{13}C-OctaCDE), and then vortexed. After that, the samples were left in the dark overnight at room temperature and extracted four times with hexane/DCM (1:1). Ultrasonic extraction method was used. Between each extraction, the tubes were centrifuged to better separate solvents from solids in the samples. The extracts were combined and concentrated on a rotary evaporator before cleanup on an aluminum oxide open column (9 g Aluminum oxide and 80 mL solvent). The analytes was eluted by Hexane (with 7% DCM). The eluents were concentrated to 500 μL before instrument analyses. All of the solvents, standards of BDE-17, 28, 47, 66, 71, 85, 99, 100, 138, 153, 154, 183, 190 and two DP isomers, anti- and syn-DP, of pesticide grade purity were purchased (J.T. Baker, USA). For the analysis of DP, chemical reagents and materials, including dichloromethane, hexane, N-octane, ^{13}C-OctaCDE, and sodium sulfate, were used. Deionized water was generated in house using a Super-Q water generation system.

Instrumental Analyses. An Agilent gas chromatograph (GC)–mass spectrometer (MS) system (6890 GC/5973 MSD; Agilent Technologies, Palo Alto, CA) fitted with a DB-1MS column (30 m long × 0.25 mm inner diameter × 0.10-μm film thickness) was used for the analyses. The oven temperature started at 70°C (last for 1.5 min) and was increased to 210°C (15°C/min) then to 250°C (2°C/min), to 290°C (15°C/min), and then kept at 290°C (15 min). The carrier gas was Helium. MS system was operated in methane negative chemical ionization (NCI) mode. Selected ion monitoring mode was

used for MS operation.

QA/QC. All of the spiking solutions and calibration standards were prepared by serial dilution. GC-MS peaks were normalized to the peak area of 2,2′,3,3′,4,4′,5,5′-octachlorodi-phenyl-ether-13 C12 (^{13}C-OctaCDE) before data reduction. Limit of quantity (LOQ) for all the investigated compounds by GC/MS were estimated based on a signal-to-noise ratio (S/N) of 10 using the lowest concentration standard. LOQ of PBDE was 2.1-12.8 ng/kg, while syn-DP and anti-DP was 17.6 and 27.2 ng/kg, respectively. The peaks of the anti- and syn-isomers of DP were combined together for reporting. Daily multiple-concentration calibration was conducted. Positive identification of DP peaks was assured by comparing both retention times and their mass ratios referenced to those of authentic standards. All laboratory glassware, gloves, and polyethylene bags used in the sample preparation were determined to be free of PBDE and DP. One duplicate sample analysis was performed with every second batch. One laboratory reference sample and one laboratory blank were included in each analytical batch of six samples. One procedural blank was included in each analytical batch of six samples. The recovery rate of ^{13}C-OctaCDE was 93%±11% and average blank levels (0.11±0.10 ng/g for DP; no PBDEs in blank).