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Factors influencing the diurnal atmospheric concentrations and soil-air exchange of PBDEs at an e-waste recycling site in China

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ABSTRACT

Factors influencing the diurnal atmospheric concentrations and soil-air exchange of polybrominated diphenyl ethers (PBDEs) were investigated at an e-waste recycling site in China during winter and summer. Total PBDE concentrations in winter and summer were 11.0–42.4 and 120–431 pg/m³ in the gaseous phase and 13.3–234 and 69.0–208 pg/m³ in the particulate phase, respectively. Total PBDE concentrations in the e-waste contaminated and nearby soils were 869 and 611 ng/g in winter and 2130 and 31 ng/g in summer, respectively. Concentrations in the gaseous phase were significantly higher in summer than in winter, but concentrations in the particle phase were comparable between winter and summer. Because of the influence of occasional accidental inputs, no apparent diurnal variations were found for PBDEs in either the particle or gaseous phase. Both temperatures and local emissions affected air concentrations of PBDEs in this area. The estimated directions of soil-air exchange of PBDEs suggest that the e-waste contaminated soil generally acted as a secondary source of several low brominated PBDEs, while the nearby soil acted as a secondary source of BDE28. The diurnal variation of atmospheric concentrations of PBDEs is not strong enough to alter the soil-air exchange direction.

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1. Introduction

Soil-air exchange has been considered as an important process controlling the atmospheric concentrations and environmental fates of persistent organic pollutants (POPs) in the terrestrial environment (Cabrerizo et al., 2011; Degrendele et al., 2016). Soil acts as a significant reservoir of atmospheric POPs by acting as a sink during acute discharge periods and a source during subsequent light release periods (Dalla Valle et al., 2005; Harner et al., 1995; Meijer et al., 2003; Schuster et al., 2011). Therefore, in addition to the influence of soil and chemical properties, and environmental parameters such as temperature, humidity or wind speed (Cabrerizo et al., 2011), the magnitude and direction of soil-air exchange of POPs can also change temporally depending on air and soil concentrations affected by production restrictions and emissions controls. Understanding factors that affect the soil-air

exchange of POPs is essential for controlling the environmental fate of these chemicals.

E-waste contamination has drawn much attention due to the environmental problems caused by toxic emissions released during crude e-waste recycling activities (Breivik et al., 2016; Cheng et al., 2014; Labunska et al., 2014). Rapid development and increasing personalized demand of electronics have shortened product life-spans, subsequently leading to a major increase in the accumulation of e-waste (Zeng et al., 2016). China is one of the largest recyclers of e-waste, not only because of the largest production and consumption of electronic products, but also the illegal e-waste importation and recycling. Intensive e-waste recycling activities have released many toxins into the environment, such as polybrominated diphenyl ethers (PBDEs), polychlorinated biphenyls, and heavy metals (Luo et al., 2011; Zeng et al., 2013; Zhang et al., 2014a). PBDEs, a group of brominated flame retardants, are conventional additives in many electronic products (Chen et al., 2009). Meanwhile, the more toxic polybrominated dibenzo-*p*-dioxins and dibenzofurans (PBDD/Fs) can also be formed from PBDEs through fly ash-mediated reactions during thermal processes, such as secondary copper and aluminum smelting processes (Wang et al.,

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2016a, 2016b; Zhang et al., 2016). Commercial penta- and octa-PBDEs have been listed as POPs by the Stockholm Convention and have been banned for more than 6 years in over 170 countries, including China (Abbasi et al., 2015). Despite that the Chinese government has taken various measures to control the emission from the recycling processes, such as formalizing and centralizing e-waste recycling and covering contaminated soil with uncontaminated soil, gradual diffusion of pollutants from the contaminated soils into the air at abandoned recycling sites may still occur. However, there is a lack of research on the re-volatilization of organic pollutants via soil-air exchange from e-waste contaminated sites, especially for PBDEs.

The primary objectives of this study were as follows: (i) to investigate the diurnal variation of the atmospheric PBDE concentrations in Qingyuan, Guangdong Province of China, (ii) to determine the direction of soil-air exchange of PBDEs at an abandoned e-waste recycling area, and (iii) to study the influence of diurnal variation of air concentrations and environmental variables on the soil-air exchange of PBDEs at this area.

2. Materials and methods

2.1. Sampling

Samples were collected at an abandoned e-waste recycling site (Fig. 1) in Qingyuan, Guangdong Province of China [23.57°N, 113.01°E]. This area has the typical monsoon-influenced subtropical climate with a hot, rainy season from March to August and a dry, cool season from September to February. The sampling area is one of the most intensive e-waste recycling regions in China and severely contaminated by crude e-waste recycling activities. According to the laws and regulations of e-waste treatment, a centralized recycling factory has replaced open recycling sites, and these abandoned sites have been covered with ~0.2 m depth uncontaminated soil for over 3 years.

The air was sampled at an abandoned e-waste open-burning site near a village 9–11 December 2011 and 27–29 June 2012. No e-waste recycling activities were observed within 500 m during the sampling periods. Air samples were collected from 12:00 p.m. at 4-h intervals over 2 days using a high-volume active air sampler at a height of 1.5 m

allowed for mixed air to be sampled. The air sample was first passed through a glass fiber filter (GFF, 20.3 × 25.4 cm, prebaked at 450 °C for 4 h) and then a polyurethane foam plug (PUF, 6.5 cm diam. × 7.5 cm, pre-extracted with acetone and dichloromethane) at a flow rate of ~0.3 m³/min to collect the gas and particle phases separately.

Topsoil samples (0–5 cm) were also collected after active air sampling at an abandoned e-waste burning site (BS) and a nearby site (NBS, ~50 m away). Each topsoil sample was a composite of three sub-samples. All samples were wrapped in aluminum foil, put into polyethylene zip-bags, and stored at –20 °C. Meteorological data were monitored simultaneously using a wireless weather station. The wind speed was <3 m/s and no rain event occurred during the sampling periods.

2.2. Sample extraction and analysis

All samples (PUF, GFF, or ~10 g freeze-dried soil) were first spiked with surrogate standards (PCB198 and PCB209), and then Soxhlet extracted with dichloromethane (DCM) and activated copper for 24 h. The extract was solvent-exchanged with hexane and cleaned using a multilayer column containing anhydrous sodium sulfate, 50% (w/w) sulfuric acid-silica gel, neutral silica gel (3% deactivated), and neutral alumina (3% deactivated) from top to bottom with an eluent of 20 mL hexane/DCM (1:1, v/v). After being concentrated to ~50 µL, ¹³C₁₂-PCB141 was added as the internal standard.

Agilent GC7890-5975C MSD operated in electron capture negative ionization with a DB5-MS capillary column (30 m × 0.25 mm i.d. × 0.25 µm film thickness) was used for the determination of 7 BDE congeners (BDEs 28, 47, 99, 100, 153, 154 and 183). Temperatures were set to 150 °C for both the MSD source and quadrupole. BDE209 was not detected in this study because BDE209 was very difficult to evaporate from soil into the air. Total organic carbon (TOC) in the soil sample was detected using a Vario EL-III elemental analyzer.

2.3. QA/QC

A field blank and a procedural blank were analyzed with each batch of 12 samples to identify potential contamination during sampling and analysis. No target compounds were detected in all blanks. The average recoveries of the surrogate PCB198 and PCB209 were 100 ± 7.2% and 101 ± 13% in the air samples and 96.2 ± 6.7% and 111 ± 11% in the soil samples, respectively. The reported results were not corrected for surrogate recovery.

2.4. Calculation of fugacity fraction

Fugacity model (Mackay, 2001) has been commonly used to assess the direction of soil-air exchange of POPs by predicting their soil and air fugacities (Backe et al., 2004; Bidleman and Leone, 2004; Li et al., 2010; Ruzickova et al., 2008). To estimate the equilibrium status, measurements of the chemical's gaseous concentration and its corresponding concentration in surface soil are needed. Fugacity value, which represents the tendency of a chemical to escape from one matrix into another, can be calculated using the fugacity capacity and the concentration in the compartment (Backe et al., 2004; Bidleman and Leone, 2004).

Air (f_a) and soil (f_s) fugacities (Pa) of PBDEs were calculated using the following equations:

$$f_a = C_a R T \quad (1)$$

$$f_s = C_s R T / (0.411 \rho_s \phi_{soc} K_{oa}) \quad (2)$$

where C_a and C_s are the concentrations of PBDEs in the air and soil

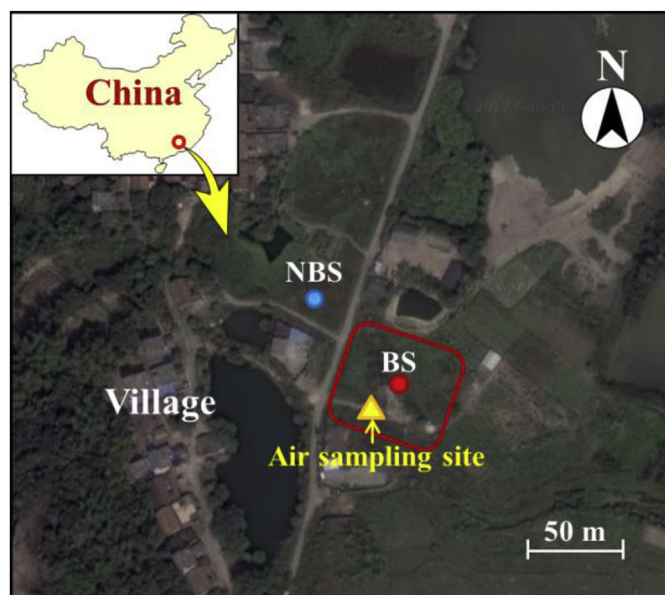


Fig. 1. Sampling site.

(mol/m³), respectively, R is gas constant (8.314 Pa m³/mol/K), T is the absolute air temperature (K), ρ_s is the soil density (assuming 2.5 kg/L), φ_{SOC} is the soil total organic carbon fraction, and K_{oa} is the octanol-air partition coefficient. The K_{oa} for PBDEs were taken from (Harner and Shoeib, 2002) assuming the average temperature is 10 °C in winter and 25 °C in summer, respectively.

Fugacity fraction (ff), defined as a ratio of soil fugacity to the sum of soil and air fugacities (equation (3)), has been used as an indication of the net direction of air-soil exchange.

$$ff = f_s / (f_s + f_a) = C_s / (C_s + 0.411 \rho_s \varphi_{\text{SOC}} K_{\text{oa}} C_a) \quad (3)$$

Theoretically, $ff = 0.5$ indicates equilibrium. For the uncertainties, equilibrium was considered to be reached at a ff of 0.25–0.75 (Wang et al., 2012). Therefore, $ff > 0.75$ indicates net volatilization from soil into air, whereas $ff < 0.25$ indicates net transport from air to soil.

3. Results and discussion

3.1. PBDEs in soil and air

The PBDE concentrations in the surface soils of BS and NBS sites during different seasons are shown in Table 1. The total concentrations of PBDEs in the BS soils were 869 ng/g in winter and 2130 ng/g in summer; whereas they were 611 ng/g in winter and 31 ng/g in summer in the NBS soils. Soil PBDE concentrations varied significantly between winter and summer samples even though they were collected at the same sites during each season; likely, the discrepancy is due to the occasional input and influence of heavy summer rainfall. The soil PBDE concentrations in this study were comparable with those (77.3–249 ng/g) measured at the same area 5 years ago (Wang et al., 2011). This suggests there were still occasional PBDE inputs via e-waste open-burning despite the prohibition of informal recycling. Meanwhile, soil PBDE concentrations in this study were also comparable to those in agricultural soils (median: 170 ng/g) in another e-waste recycling area, Guiyu (Zhang et al., 2014b). BDE153, BDE154, and BDE99 were the dominant congeners in both BS and NBS soils in winter and in NBS soil in summer. However, a significantly high abundance of BDE28 was found in BS soil in summer. Leung et al. (2007) also found relatively high concentrations of BDE47, 99, 100, 153, 154, and 183 in soils from printer roller dump site and acid leaching site and in e-waste combusted residue in Guiyu of China. Taking the relatively elevated levels of PBDEs in BS soil in summer into consideration, significant inputs of PBDEs via illegal recycling likely occurred before summer sampling.

The total concentrations of PBDEs in the gaseous phase were 11.0–42.4 pg/m³ (mean: 29.7 ± 9.46 pg/m³) in winter and 120–431 pg/m³ (mean: 207 ± 82.1 pg/m³) in summer, whereas PBDEs in the particle phase were 13.3–234 pg/m³ (mean:

112 ± 78.8 pg/m³) in winter and 69.0–208 pg/m³ (mean: 118 ± 45.7 pg/m³) in summer (Fig. 2). Seasonal trends were observed only for gaseous PBDEs, with relatively higher concentrations in summer than in winter, likely due to enhanced volatilization with increased temperatures in summer. However, seasonal variation was insignificant for the particle PBDEs. These results are consistent with those of a previous study conducted at a nearby in-use e-waste recycling site (Tian et al., 2011). That is, relatively higher concentrations of PBDEs and other brominated flame retardants (BFRs) were higher in summer than in winter for the gaseous phase, and BFRs in particulate phase did not exhibit significant seasonal variability. Contaminant concentrations at the abandoned e-waste recycling site were slightly lower than those measured at the nearby in-use site but higher than those in the rural area 25 km away, especially in summer (Tian et al., 2011). This difference implies that e-waste recycling activities and subsequent emissions from contaminated soils are still the most significant sources of PBDEs in this area. The most dominant congener in the gaseous phase was BDE28, whereas BDE99 and BDE183 dominated in the winter and summer particle phases, respectively.

3.2. Diurnal variations of PBDEs in the air

In theory, a significant temperature-driven diurnal variation of

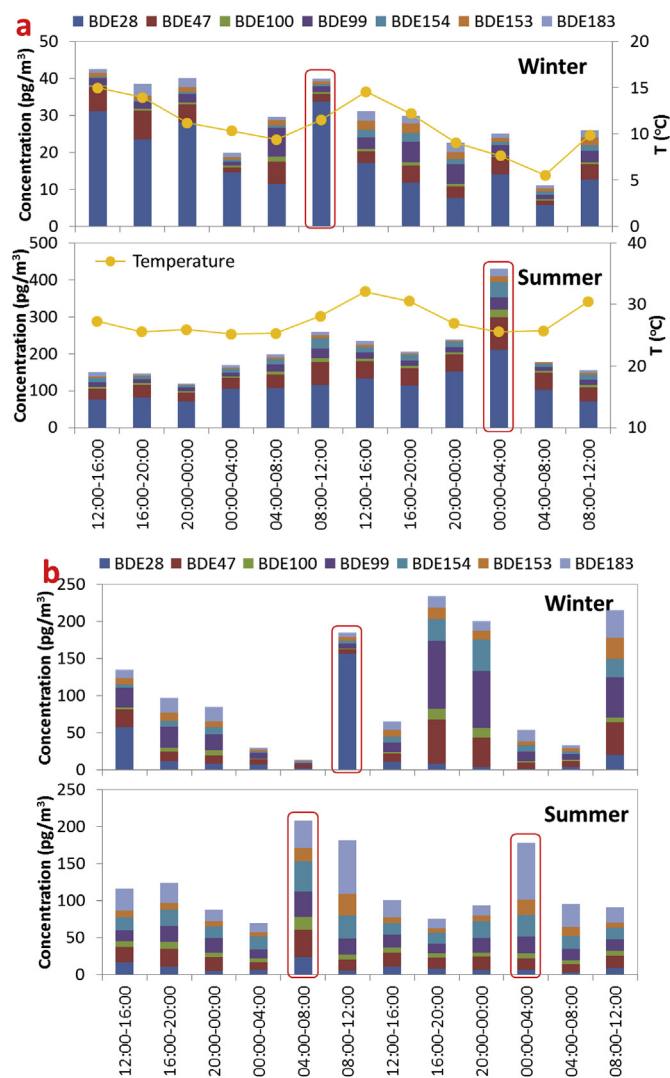


Fig. 2. Diurnal variations of gaseous (a) and particulate (b) PBDEs in the atmosphere.

Table 1
PBDE concentrations in the surface soils of BS and NBS sites.

Soil (ng/g)	Winter		Summer	
	BS	NBS	BS	NBS
BDE28	15.4	55.1	568	2.11
BDE47	84.0	29.3	411	2.99
BDE100	37.9	26.5	175	2.55
BDE99	158	84.0	250	3.32
BDE154	212	203	474	9.72
BDE153	236	183	157	4.02
BDE183	125	30.0	92.1	6.59
PBDEs	869	611	2130	31.3

POPs in the air should be observed in the absence of external inputs. However, PBDE concentrations in the gaseous and particulate phases varied widely without apparent diurnal trends (Fig. 2). Elevated levels of gaseous PBDEs were mostly found around noon and low levels around midnight or early in the morning. High particulate PBDE concentrations were mostly found in the morning and low levels between midnight and dawn. Fig. 2 suggests that there were some occasional inputs of PBDEs (see red squares), which may be due to illegal e-waste recycling such as open-burning in the nearby area. If we eliminate these samples, which may be influenced by inputs from accidental releases, significant diurnal variations in high and low concentrations around noon and midnight respectively can be observed for both particle and gaseous PBDEs during both summer and winter sampling periods. This result suggests a diurnal variation of PBDEs in the air is affected by both temperature and local emissions. Chen et al. (2009) also found significant diurnal variations of atmospheric BDE47 and BDE99 concentrations at the e-waste recycling site of Guiyu, but no apparent diurnal trend for BDE153 and BDE209. The unique behaviors of the PBDE congeners were due to differing types of e-waste or recycling processes. Despite laws, regulations, and management carried out by the Chinese governments to ban primitive e-waste recycling activities, evidence suggests legal or illegal, profit-driven e-waste recycling may still occur in this area.

3.3. Factors influencing atmospheric PBDEs

Both ambient temperatures and local emissions may influence atmospheric concentrations of PBDEs in this area. Significant correlations ($r > 0.605$, $p < 0.05$) were found between ambient temperatures and gaseous concentrations of 7 individual PBDE congeners, and none ($p > 0.1$) for temperature and PBDE particle concentrations, except for BDE183 ($r = 0.449$, $p = 0.029$). This result suggests that temperature was the key factor controlling PBDE concentrations in the gaseous phase and was the insignificant factor in the particle phase. Meanwhile, PBDE concentrations may be also influenced by atmospheric photodegradation (which is the major loss process of gaseous PBDEs) and deposition (which is major loss process of particle PBDEs) (Wania and Dugani, 2003). Moreover, the correlation coefficients between temperature and gaseous PBDEs decreased linearly with increasing molecular weight ($r = -0.832$, $p = 0.02$), implying the gaseous emissions of low brominated PBDEs were more sensitive to the temperature variation.

The Clausius-Clapeyron (CC) equation, the natural logarithm of the partial pressure of gas (P, atm) regressed with the reciprocal absolute temperature (T, K), can be used to identify which influence is more important for controlling air concentrations, temperature-driven evaporation from local surfaces or inputs via long-range atmospheric transport (Hoff et al., 1998; Tian et al., 2011; Wania et al., 1998). The CC plot of gaseous PBDEs during summer and winter are shown in Fig. 3. Significant linear correlations (r^2 : 0.663–0.882, $p < 1.3 \times 10^{-6}$, Table S1) between $\ln P$ and $1/T$ were observed for individual PBDE congeners with the seasons combined, but no correlations ($p > 0.084$) for winter and summer separately, except for BDE28 ($p = 0.007$) in winter. This trend suggests that annual variations of gaseous PBDEs were strongly controlled by temperature-driven air-surface emissions from local sources (e.g., contaminated soils or e-waste), and short-term variations by random inputs such as accidental releases from e-waste recycling activities. The mean slope of the CC plots for PBDEs in this study was $-10\,008 \pm 2544$ (Table S1). This slope is comparable to values reported for North America (-6580 ± 601 for BDE47 and -5290 ± 962 for BDE99) (Venier and Hites, 2008) and rural areas of South China (-7185 ± 2500) (Tian et al., 2011). However, the slope

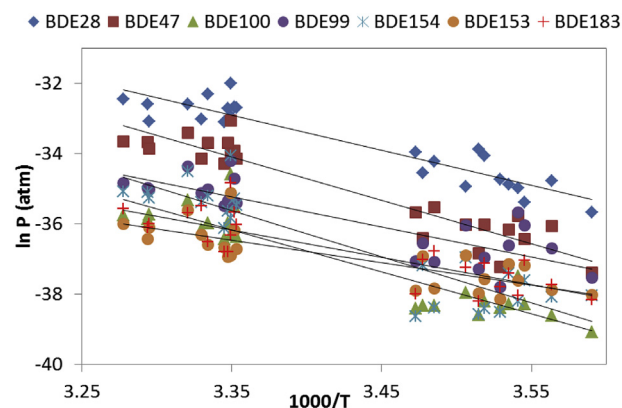


Fig. 3. Clausius-Clapeyron plot of PBDEs at the abandoned e-waste site.

in this study is flatter than values ($-20\,840 \pm 3250$) for the nearby e-waste recycling area (Tian et al., 2011). A relatively flat slope may indicate that, in addition to the influence of temperature-driven emissions from local surfaces, atmospheric levels are influenced by regional or long-range atmospheric transport (Hoff et al., 1998; Wania et al., 1998).

Gas-particle (G-P) partitioning is another important process that influences concentrations of atmospheric POPs (Chen et al., 2006; Yang et al., 2013). The ratios of gaseous-to-total PBDEs are shown in Fig. S1. Lower-brominated PBDEs, such as BDE28, mostly volatilize, while higher-brominated PBDEs, e. g., penta-, hexa-, and hepta-BDEs, are mainly bound to the particle phase. However, the gas-particle partitioning of BDE47 varies with seasons and is mostly present in the particle phase in winter ($G/(G + P) = 24\%$) and in the gaseous phase in summer ($G/(G + P) = 70\%$). High temperatures can lead to evaporation of PBDEs from particle to gaseous phase and subsequently a relatively large proportion of gaseous PBDEs in summer. The partitioning ratio also varied widely on a daily basis, especially in winter. The sudden increase or decrease of this ratio may suggest an additional input of POPs from local sources.

3.4. Soil-air exchange of PBDEs

The direction of soil-air exchange of PBDEs at BS and NBS sites of the abandoned e-waste recycling area was assessed using air (f_a) and soil fugacities (f_s). The values of f_a were significantly correlated with those of f_s (winter, $p < 0.012$; summer, $p < 0.002$), indicating atmospheric levels of PBDEs were mostly controlled by re-volatilization from contaminated soil (Degrendele et al., 2016).

Variation among fugacity fractions of individual PBDE congeners at different sampling sites is shown in Fig. 4. Due to uncertainties and error propagation, only a ff between 0.25 and 0.75 was considered in equilibrium. In winter at the BS site, lower-brominated PBDEs (BDE28 and BDE47) generally evaporated from soil into air, whereas higher-brominated PBDEs were in equilibrium between soil and air. At the NBS site, however, only BDE28 and BDE183 were significantly volatilized into air and deposited into soil, respectively; other PBDEs were mainly in equilibrium between soil and air. In summer at the BS site, lower-brominated PBDEs (BDE28, 47, and 100) significantly evaporated, while higher-brominated PBDEs in the soil were still in equilibrium with those in air. At the NBS site, only BDE28 showed net volatilization from soil into air, while other PBDEs were mainly considered to be in equilibrium. Significant differences were found between the ff values in summer and those in winter for lower-brominated PBDEs, i.e., values were higher in summer than in winter. The increased evaporation tendency of PBDEs in summer is caused by elevated

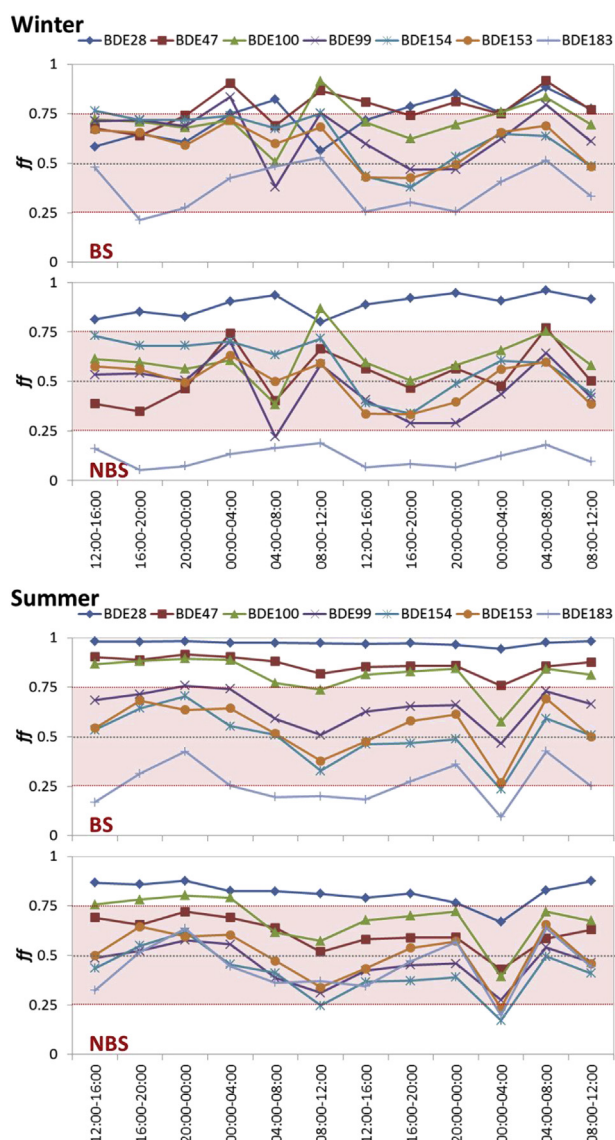


Fig. 4. Temporal variation of PBDE fugacity fractions (ff) at BS site and NBS sites in the abandoned e-waste recycling area. (For the uncertainties, equilibrium was considered to be reached at a ff of 0.25–0.75).

temperatures. Although the ff values varied between seasons, to some extent, the soil-air exchange tendencies were similar between winter and summer. This similarity implies the influence of local emissions was more significant than that of temperature on the direction of soil-air exchange of PBDEs in this area. A previous study suggested that net volatilization from soils was found for both lower- and higher-brominated PBDEs at two background sites in the Great Hungarian Plain (Degrendele et al., 2016). Conversely, net depositions were found for all PBDE congeners at suburban and urban areas of Izmir, Turkey (Cetin and Odabasi, 2007).

The soil-air exchange of PBDEs also exhibited a temporal variation, but no significant diurnal trend was found. This variation was a consequence of different PBDE concentrations in the atmosphere but not strong enough to alter the direction of soil-air exchange of most PBDEs (Fig. 4). The exception to this temporal variation was for the lower-brominated PBDEs (BDE28, 47, and 100) at the BS site in winter; in this case, these PBDEs oscillated between evaporation and equilibrium even within one day.

Overall results suggest the soil at the BS site acted as a secondary

source of lower-brominated PBDEs (BDE28, 47, and 100). At the NBS site, soil served as a secondary source of BDE28 on an annual basis and as a sink of BDE183 only in winter. Longtang Town of Qingyuan City has become one of the largest e-waste recycling areas since the early 1980s. The local government has taken steps to control e-waste dismantling activities and mitigate heavy contamination, including the prohibition of open-burning and covering contaminated soil with outside soil. However, abandoned recycling sites still act as secondary sources of many POPs and e-waste contamination has a far-reaching impact. Moreover, profit-driven, illegal e-waste dismantling still occasionally occurs in this area, highlighting the importance of managing e-waste recycling sites even long after they are abandoned.

4. Conclusions

This study investigated the diurnal variation of PBDEs in the air and their potential exchange trends between air and soil at an abandoned e-waste dismantling site in South China. Driven by the temperature, PBDE concentrations were relatively higher in summer than in winter; however, no significant diurnal variation trend was found because of accidental inputs via illegal e-waste recycling activities. The estimated soil-air exchange trend suggests e-waste contaminated soil acted as a secondary source of lower-brominated PBDEs to the atmosphere and that nearby soil served as a secondary source of BDE28 on an annual basis and a sink of BDE183 in winter. The directions of soil-air exchange were generally consistent on a daily basis, except where there were possible accidental inputs. Despite laws and regulations imposed by the Chinese government to reduce e-waste pollution, the investigated abandoned e-waste recycling site continues to act as a secondary source of lower-brominated PBDEs. Moreover, occasional illegal e-waste recycling activities may still be occurring in this area. Therefore, more strict rules and regulations should be established to prohibit illegal recycling activities and manage abandoned recycling sites.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at <https://doi.org/10.1016/j.apr.2017.09.003>.

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