Concentrations, profiles and gas-particle partitioning of PCDD/Fs, PCBs and PBDEs in the ambient air of an E-waste dismantling area, southeast China

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Polychlorinated dibenzo-*p*-dioxins (PCDDs), polychlorinated dibenzofurans (PCDFs), polychlorinated biphenyls (PCBs) and polybrominated diphenyl ethers (PBDEs) were monitored in the ambient air of Taizhou, an E-waste dismantling area of southeast China to evaluate their concentrations, profiles and gas-particle partitioning. The Σ PCDD/Fs concentrations ranged from 2.91 to 50.6 pg/m³, with an average of 14.3 pg/m³. The I-TEQs for PCDD/Fs were in the range of 0.20–3.45 pg/m³, with an average of 1.10 pg/m³. The Σ PCBs concentrations and TEQs ranged from 4.23 to 11.35 ng/m³, 0.050 to 0.859 pg(TEQ)/m³, respectively. The concentrations of Σ PBDEs ranged from 92 to 3086 pg/m³, with an average of 894 pg/m³. The pollution levels of PCDD/Fs, PCBs and PBDEs were higher than other urban sites, which may be associated with the E-waste dismantling activities. The PCDD/Fs were found exclusively in the particle phase whereas PCBs distributed dominantly in the gas phase. The gas-particle partitioning was also assessed by correlating the gas-particle partition coefficient (K_p) with the subcooled liquid vapor pressure (P_L^0). The measured particulate sorptions of PCDD/Fs, PCBs and PBDEs were compared with the predictions from Junge-Pankow model and K_{oa} absorption model. The Junge-Pankow model well estimated the particulate fractions of PCBs. However, it underestimated the sorptions of PCDD/Fs and overestimated the fractions of PBDEs. The predicted particulate fractions of PCDD/Fs and PCBs from K_{oa} model fitted well with the measured data.

PCDD/Fs, PCBs, PBDEs, air, gas-particle partitioning, E-waste

Polychlorinated dibenzo-*p*-dioxins (PCDDs), polychlorinated dibenzofurans (PCDFs) and polychlorinated biphenyls (PCBs) are three groups of persistent organic pollutants (POPs) that are stable in the environment, accumulate in the fatty tissue of living organisms and undergo long range transports. PCDD/Fs were not intentionally manufactured. They were formed or released to the environment from various combustion activities and productions of chlorinated chemicals as by-products. PCBs were historically synthesized and used as dielectric fluid transformers and capacitors. Polybrominated diphenyl ethers (PBDEs) are used as flame retardant in plastics, textiles and electronics. The commercial PBDEs products mainly include penta-BDE, octa-

BDE and deca-BDE. Because they show similar physical-chemical properties to some other POPs, PBDEs are recognized as emerging environmental pollutants in recent years. The environmental fate and transport pathways of these chemical pollutants are continually concerned because of their potential exposure to humans.

Atmosphere is a major pathway for these chemical pollutants to transport and redistribute in the environment. PCDD/Fs, PCBs and PBDEs exist between the gas phase and particle phase in the ambient air. It is es-

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essential for us to knowledge about the gas-particle partitioning since it affects their transport pathways, degradation mechanisms, deposition processes and mode of entry into food chains^[1]. However, few studies were carried out for measuring and predicting their levels and partitioning in the ambient air in China^[2,3].

Taizhou situates in the southeast coastline of China. In the 1980s, the dismantling operations of E-wastes emerged and boomed in some towns of Taizhou. The scrap transformers containing PCBs as dielectric fluids were once dismantled in this area. Until now, most of E-wastes are still simply dismantled or incinerated openly to remove PVC coating to retrieve valuable metals. These activities resulted in environmental pollutions of PCDD/Fs and PCBs^[4,5]. The aim of this study was to monitor PCDD/Fs, PCBs and PBDEs in the ambient air of the typical E-waste dismantling area in Taizhou to evaluate their concentrations, profiles and gas-particle distributions. By analyzing the gas-particle partitioning of these pollutants, their environmental fate and transportation were also discussed.

1 Materials and methods

1.1 Sampling

The climate of Taizhou belongs to typical sub-tropical monsoon, which is characteristic with rich rainfalls (annual volume from 1480 to 1530 mm) and year-round temperatures from 16.9 to 17.3°C. Sampling site A (121°21.731°E, 28°32.724°N) and C (121°21.672°E, 28°32.772°N) are rural areas and in two residential villages of the town. Site B (121°21.675°E, 28°32.755°N) situates in the commercial center of the dismantling town besides the Road 104.

Air samples were concurrently collected in sites A, B and C for an interval of 12 hours by using high volume air samplers (GPS1, Thermo Environmental Instruments, Inc., Franklin, MA). The average temperature and PM10 data during the sampling period were 16.9°C and 90 µg/m³, respectively. The sampling process was conducted according to US EPA Compendium Method TO-9A. Briefly, the sampler was equipped with a quartz fiber filter for sampling particle-bound pollutants, following by a glass cartridge with PUF (6.3 cm diameter, 7.6 cm length) contained in the glass cartridge for sampling gas phase chemicals. Prior to sampling, the quartz fiber filters (Whatman, 10.16 cm diameter) were baked at 450°C for 12 h to remove any organic contaminants.

The PUFs were followed with a 24-h extraction with acetone and another 24-hour extraction with hexane. Then, the sampling cartridges were vacuum-dried in desiccators and stored in sealed bags.

The airflow rate was specified at 0.25 m³/min at the beginning of sampling. After sampling, the GFF and PUF plug were wrapped with hexane-rinsed aluminum foil for protection from light, and stored in sealed bags. After transported to the laboratory, they were stored at -20°C until analysis.

1.2 Sample analysis

All the organic solvents were pesticide residue grade from Fisher (Fair Lawn, NJ, USA). Silica was purchased from Merck (silica gel 60, Darmstadt, Germany). Basic alumina was obtained from Aldrich (Brockmann I, standard grade, Milwaukee, USA). Florisil was from Riedel-deHaën (60 — 100 mesh, Seelze, Germany). Calibration standard solutions, ¹³C₁₂-labeled surrogate standards and ¹³C₁₂-labeled injection standards were purchased from Wellington Laboratories (Guelph, Canada).

Prior to a 24h Soxhlet extraction with toluene, filters or PUFs were spiked with 15 congeners of ¹³C₁₂-labeled surrogate standards for PCDD/Fs. Also 27 PCB congeners of ¹³C₁₂-labeled surrogate standards for PCBs, ¹³C-BDE-47, ¹³C-BDE-99 and ¹³C-BDE-153 were incorporated for the determination of PCBs and PBDES, respectively. The extract was subsequently refluxed in hexane and subjected to a multilayer silica gel column, followed by clean-up on basic alumina column and florisil chromatography columns. The detailed clean-up procedure was described elsewhere^[6]. Prior to HRGC-HRMS analysis, the extract was spiked with the ¹³C₁₂-labeled internal standard (¹³C-1,2,3,4-TCDD and ¹³C-1,2,3,7,8,9-HxCDD for PCDD/Fs; ¹³C-PCB-9, 52, 101, 138 and ¹³C-PCB-194 for PCBs; ¹³C-PCB-138 for PBDEs) for recovery quantification.

The PCDD/Fs, PCBs and PBDEs were quantified on Agilent 6890 gas chromatography coupled with an Autospec Ultima high resolution mass spectrometer with an electron impact (EI) ion source. The HRMS was operated in SIR mode at $R \geq 10000$. Exactly 1 µL of sample solution was injected with a CTC PAL autosampler in splitless mode into a DB-5 fused silica capillary column (60 m for PCDD/Fs and PCBs, 30 m for PBDEs). Helium served as the carrier gas with a constant flow of

1.2 mL/min. The electron emission energy was set to 35 eV, and the source temperature was 270°C. The oven temperature programs were employed as follows: for PCDD/Fs, start 150°C held for 3 min, 150–230°C at 20°C/min held for 18 min, 230–235°C at 5°C/min held for 10 min, 235–320°C at 4°C/min held for 3 min; for PCBs, start 120°C held for 1 min, 120–150°C at 30°C/min, 150–300°C at 2.5°C/min held for 1 min; for PBDEs, start 100°C held for 2 min, 100–230°C at 15°C/min, 230–270°C at 5°C/min, 270–330°C at 10°C/min held for 8 min.

1.3 Quality assurance and quality control

Three pairs of field blanks and laboratory blanks were incorporated in the analytical procedure for the purpose of quality control. There was no apparent difference between field blanks and laboratory blanks. OCDD, 1,2,3,4,6,7,8-HpCDD, PCB-28 and BDE-47 were the main contaminants in the blanks. However, their concentrations in blanks corresponded to <5% of the concentrations detected in the air samples. A breakthrough test was made by using a second half PUF in series with the first PUF and no breakthrough was found. The sample recoveries for ^{13}C -OCDD were 38–98%. The recoveries for other ^{13}C labeled PCDD/Fs were 55–125%. The recoveries for ^{13}C labeled PCBs were 31–120%. Recoveries for ^{13}C -BDE-47, 99 and ^{13}C -BDE-153 were 41%–109%, 40%–117%, 35%–124%, respectively. All the sample recoveries were complied with US EPA method 1613, method 1668 and method 1614. The de-

tection limits were 0.003–0.014 pg/m³ for PCDD/Fs, 0.010–0.063 pg/m³ for PCBs, and 0.014–0.148 pg/m³ for PBDEs except BDE-209 whose detection limits were 4.55 pg/m³.

2 Results and discussion

2.1 Concentrations and profiles

The concentrations and I-TEQ values of 2,3,7,8-substituted PCDD/Fs during the sampling period are shown in Table 1. The Σ PCDD/Fs concentrations ranged between 2.91 and 50.6 pg/m³, with an average of 14.3 pg/m³. Figure 1(a) shows the PCDD/F profiles for the three sampling sites. The congeners HxCDF, HpCDF and OCDD contributed dominantly to the Σ PCDD/Fs concentrations. The PCDD concentrations increased with the increase of chlorinated level, which was consistent with the general trend reported in previous studies^[7]. However, the PCDFs were contrary to the trend for decreasing concentrations with increasing levels of chlorination.

The TEQ values were calculated according to the International Toxicity Equivalency Factor (I-TEF) [International toxicity equivalency (I-TEF) method of risk assessment for complex mixtures of dioxins and related compounds. Pilot study on international information exchange on dioxins and related compounds. Report No. 176, 1988]. The I-TEQs were in the range 0.20–3.45 pg/m³, with an average of 1.10 pg/m³. The congener 2,3,4,7,8-PeCDF contributed dominantly to the total I-TEQs with an average of 46.2%. Other significant

Table 1 Concentrations (pg/m³), TEQs (pg(TEQ)/m³) of PCDD/Fs, PCBs and PBDEs

Sample No.	A1	A2	A3	A4	B1	B2	B3	B4	C1	C2	C3	C4
Date	28/10/05	28/10/05	29/10/05	29/10/05	28/10/05	28/10/05	29/10/05	29/10/05	28/10/05	28/10/05	29/10/05	29/10/05
Day or night	day	night	day	night	day	night	day	night	day	night	day	night
Volume (m ³)	103	184	109	228	90	197	114	201	142	184	121	201
Σ PCDDs	5.55	0.90	1.55	8.86	1.99	3.02	4.07	2.84	2.26	0.77	1.20	24.3
Σ PCDFs	11.8	2.02	9.48	19.3	3.20	13.1	1.85	17.8	3.55	2.98	2.59	26.2
Σ PCDDs/ Σ PCDFs	0.47	0.44	0.16	0.46	0.62	0.23	2.20	0.16	0.64	0.26	0.46	0.93
Σ PCDD/Fs	17.3	2.91	11.0	28.2	5.19	16.1	5.91	20.6	5.81	3.75	3.80	50.6
I-TEQ for PCDD/Fs	1.33	0.22	0.96	2.59	0.38	1.28	0.20	1.71	0.42	0.24	0.39	3.45
Σ PCBs	9906	5758	7603	6493	11352	6129	6459	7160	9452	4232	4762	7321
TEQ for PCBs	0.195	0.064	0.116	0.347	0.178	0.206	0.078	0.243	0.108	0.05	0.069	0.859
TEQs for PCDD/Fs+PCBs	1.53	0.29	1.08	2.93	0.56	1.48	0.28	1.95	0.52	0.29	0.46	4.31
Σ PBDEs	3086	224	92	702	728	243	445	2543	925	282	254	1201
BDE-209	3009	3079	415	1228	1308	59	1639	341	205	165	536	1231

Figure 1 Profiles of PCDD/Fs, PCBs and PBDEs.

contributors to the total I-TEQ were as follows: 2,3,4,6,7,8-HxCDF (~9.7%), 1,2,3,6,7,8-HxCDF (~7.5%), 1,2,3,4,7,8-HxCDF (~6.5%), 1,2,3,7,8-PeCDD (~5.0%). The I-TEQs in Taizhou air was higher than the general PCDD/Fs levels in the ambient air with Σ TEQ <10 fg/m³ for remote area, ~20–50 fg(TEQ)/m³ for rural area and ~100–400 fg (TEQ)/m³ for urban/industry area^[8].

The Σ PCBs ranged from 4.23 to 11.35 ng/m³, with an average of 7.22 ng/m³. The TEQs contributed by coplanar PCBs were calculated using World Health Or-

ganizations Toxic Equivalent Factors^[9] and the values were in the range 0.050–0.859 pg(TEQ)/m³ (average 0.209 pg(TEQ)/m³). The results were higher than the atmospheric PCBs in other locations. For instance, the concentration of total PCB and TEQ ranged from 62 to 250 pg/m³ and from 2 to 14 fg(TEQ)/m³, for Yokohama City, Japan^[10]. Tasdemir reported the mean concentrations of Σ_{50} PCBs ranged between 0.4 and 8.3 ng/m³ in the ambient air of Chicago^[11]. The PCB profiles were shown in Figure 1(b) with the lighter congener PCB-28 dominating the profile patterns. Among coplanar PCBs, the congener PCB-118 presented the highest concentrations. However, the main contributor to Σ TEQ was PCB-126, which was responsible for an average contribution of 85.5%.

The Σ PBDEs and BDE-209 concentrations are shown in Table 1. Σ PBDEs refers to all the target PBDE congeners detected in the study except BDE-209. The concentrations of Σ PBDEs ranged between 92 and 3086 pg/m³, with an average of 894 pg/m³. BDE-209 was in the range 59–3079 pg/m³ (average 1101 pg/m³). The Σ PBDEs and BDE-209 concentrations were comparable to a large industrial zone in Guangzhou air, but higher than those from East-Central U.S.^[3,12]. PBDEs were generally used as flame retardants for electric products. The activity of dismantling E-wastes increased or released PBDEs to the ambient air. BDE-209 was the dominant congener for the total PBDEs. The other main contributors were BDE-47 and BDE-99. The result was consistent with other reports^[3].

2.2 Distribution in gas and particle phases

The Σ PCDD/Fs appeared to present exclusively in the particle phase, with average concentrations of 13.67 pg/m³ in the aerosols and 0.94 pg/m³ in the gas phase. Figure 2 shows the relative gas-particle distribution of individual congeners. The heavier congeners were more associated with the particle phase, while the lighter congeners of TCDD/Fs tended to distribute in the gas phase. The results were consistent with the data in the ambient air of Huston^[13].

The partitioning trend of PCBs was contrary to that of PCDD/Fs. The Σ PCBs concentrations (average 6932 pg/m³) in the gas phase were dominantly higher than that in the particle phases (average 287 pg/m³). However, the gas-particle distribution of PCB congeners was also based on molecular weights, which was similar to that of PCDD/Fs. For instance, tri-CB (PCB-28) and tetra-CB

Figure 2 Relative distribution of PCDD/Fs, PCBs and PBDEs between gas phase and particle phase.

(PCB-52) presented dominantly in the gas phase whereas the deca-CB (PCB-209) was mainly found in the particle phase.

For Σ PBDEs and BDE-209, the average concentrations were 505 pg/m³, 51 pg/m³ in the gas phase and 389 pg/m³, 1054 pg/m³ in the particle phase, respectively. Similar to the distribution pattern of PCDD/Fs and PCBs, the lighter congener BDE-28, 47 was mainly in the gas phase while the heavier congener BDE-209 existed dominantly in the particle phase.

The gas-particle partition coefficient (K_p) is common used to describe gas-particle partitioning of SVOCs by using $K_p = (F/A)/TSP$, where F and A (ng/m³) are particle-associated and gaseous concentrations, respectively, and TSP (μ g/m³) represents the total suspended particulate matter concentration^[14]. Two mechanisms exist for the partitioning process i.e. adsorption into the surface area of aerosols and absorption into the organic matter. For both mechanisms, the partitioning information can be extracted from the regression of $\log K_p$ against logarithm of the subcooled liquid vapor pressure (P_L^0), i.e. $\log K_p = m_r \log P_L^0 + b_r$. The slope should be close to -1 for either adsorption or absorption mechanism in true equilibrium state^[15]. The intercept was mainly dependent on the type and properties of the aerosols.

Table 2 shows the regression data of $\log K_p$ versus $\log P_L^0$ for PCDD/Fs, PCBs and PBDEs. Previous studies

Table 2 Regression data of $\log K_p$ versus $\log P_L^0$

	Slope m_r	Intercept b_r	R^2	P
PCDD/Fs				
Site A	-0.62	-5.02	0.60	< 0.0001
Site B	-0.46	-4.73	0.36	< 0.0001
Site C	-0.64	-5.11	0.45	< 0.0001
PCBs				
Site A	-0.97	-6.05	0.81	< 0.0001
Site B	-0.76	-5.28	0.74	< 0.0001
Site C	-0.75	-5.51	0.74	< 0.0001
PBDEs				
Site A	-0.56	-6.14	0.36	2.86×10^{-4}
Site B	-0.44	-5.99	0.17	0.019
Site C	-0.64	-6.53	0.39	1.34×10^{-4}

reported the temperature dependent P_L^0 of PCDD/Fs by correlating the P_L^0 with gas chromatographic retention indexes (RI)^[16–18], i.e. $\log P_L^0 = -1.34RI/T + 1.67 \times 10^{-3}RI - 1320/T$. The changes in vapor pressures of PBDEs with temperature were described by an integrated Clausius-Clapeyron equation in the form of $\log P_L^0 = B/T + A$ ^[19]. The vapor pressures of PCBs can be correlated with temperature in a similar equation to PBDEs^[20]. In this study, we also use these methods to obtain the subcooled vapor pressures of PCDD/Fs, PCBs and PCDD/Fs at the average temperature of 17°C during the sampling period. The average PM10 data of 90 μ g/m³ was used because TSP was not routinely measured. PCBs in site A ap-

peared approach equilibrium. However, other slopes were shallower than -1 , indicating disequilibrium for the three groups of chemicals. PCBs were more close to equilibrium than PCDD/Fs and PBDEs, which can be explained by the fact that PCBs were mainly associated with gas phases as discussed earlier. Different to PCBs, PCDD/Fs and PBDEs in sites A and C were more close to equilibrium than that at site B due to less traffic influences.

2.3 Comparison of partitioning models

Two different sorption models have been proposed and successfully used to describe the atmospheric gas-particle partitioning of SVOCs. One is Junge-Pankow model^[21,22], which is based on linear Langmuir isotherm. It relates the particulate fraction of chemicals with the subcooled liquid vapor pressure of the compound and the particle surface area, as shown in the following equation:

$$\varphi = \frac{c\theta}{P_L^0 + c\theta}, \quad (1)$$

where φ is the fraction of the total atmospheric concentration that is sorbed on the particulate matter. The parameter c is a constant that based on the heat of desorption from the particle surface, the heat of vaporization of the compound, and the moles of adsorption sites on the aerosol. An empirical value of $0.172 \text{ Pa}\cdot\text{m}$ is typically used although it may vary with the class of compound. θ is the aerosol surface area and ranges from 4.2×10^{-5} for clean continental background, 1.5×10^{-4} for rural area, and $1.1 \times 10^{-3} \text{ cm}^2 \text{ cm}^{-3}$ for urban area^[23].

Figure 3 compares the particulate fractions (φ) of PCDD/Fs, PCBs and PBDEs predicted by Junge-Pankow model with the average measured data. The measured φ was calculated using the ambient concentrations in the particle phase divided by the total ambient concentrations (gas + particle). Although the measured φ was close to the urban partitioning curve, the Junge-Pankow model tended to underestimate the particulate sorption of PCDD/F congeners. This result was contradicted with data reported in the literature^[7]. The predicted urban curve fitted well with the measured PCBs in site A and C whereas it slightly underestimated the particulate sorption in site B due to the traffic perturbations. As seen from Figure 3(c), Junge-Pankow model overestimated the particulate fractions of PBDEs. The derivation between the modeling and the measured data was due to the different c values for different com-

Figure 3 Comparison between predictions from Junge-Pankow model and measurements.

pounds and aerosol types^[7]. The parameter value used in literature may not be appropriate for the E-waste site.

Another partitioning model was K_{oa} absorption model^[24] that requires knowledge of K_{oa} and the organic matter fraction (f_{om}) of the aerosol which are more easily measurable than the parameters of the Junge-Pankow model, i.e. $\log K_p = \log K_{oa} + \log f_{om} - 11.91$. The fraction on particles is then calculated from K_p and TSP by

$$\varphi = \frac{K_p \text{PM}_{10}}{1 + K_p \text{PM}_{10}}. \quad (2)$$

Figure 4 compares the predicted and measured φ by

Figure 4 Comparison between predictions from K_{oa} absorption model and measurements. om, Organic matter.

model consisting of 10%, 20%, and 30% organic matter in the particulates. The K_{oa} values for PCDD/Fs were correlated with published retention time indices at the average temperature of 17°C, i.e. $\log K_{oa} = a + b (RTI)^{[25]}$. The temperature dependent values for PCBs were calculated based on the method that correlated $\log K_{oa}$ with $\log P_L^0$ [26]. The K_{oa} values for PBDEs were obtained by correlating $\log K_{oa}$ with reversal temperatures [27]. The K_{oa} model well estimated the particulate sorptions of PCDD/Fs. The lower chlorinated PCDD/Fs ($\log K_{oa} < 10.5$) were generally close to the partitioning predicted by 10% organic matter in the aerosols, while the higher chlorinated congeners ($\log K_{oa} > 10.5$) were close to those consisting of 20–30% organic matters. The measured PCBs in sites A and C presented between the predicted partitioning curves consisting 10% and 20% organic matter, whereas PCBs in site B were close to the prediction curve consisting 10% organic matter. The K_{oa} model overestimated the particulate fractions of PBDEs, which should be further investigated.

3 Conclusions

In this study, PCDD/Fs, PCBs and PBDEs in the ambient air of Taizhou were measured. The concentrations of the three groups of pollutants were higher than other urban sites. The lighter congeners of these pollutants were found mainly in the gas phase, whereas the heavier congeners were dominantly presented in the particle phase. For the three sampling sites, the atmospheric pollutants did not reach gas-particle equilibrium. The pollution sources for the three groups of POPs should be further investigated.

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