JOURNAL OF ENVIRONMENTAL SCIENCES 62 (2017) 3–10

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JES JOURNAL OF ENVIRONMENTAL SCIENCES

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Airborne PCDD/Fs in two e-waste recycling regions after stricter environmental regulations

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ARTICLE INFO

Article history:
Received 31 March 2017
Revised 8 July 2017
Accepted 11 July 2017
Available online 18 July 2017

Keywords:
Polychlorinated dibenzo-*p*-dioxins and furans
E-waste recycling
China
Environmental regulation

ABSTRACT

Since the 2010s, the authorities of Guangdong province and local governments have enhanced law enforcement and environmental regulations to abolish open burning, acid washing, and other uncontrolled e-waste recycling activities. In this study, ambient air and indoor dust near different kinds of e-waste recycling processes were collected in Guiyu and Qingyuan to investigate the pollution status of particles and polychlorinated dibenzo-p-dioxins and furans (PCDD/Fs) after stricter environmental regulations. $PM_{2.5}$ and PCDD/Fs both showed significantly reduced levels in the two regions compared with the documented data. The congener distribution and principal component analysis results also confirmed the significant differences between the current PCDD/Fs pollution characterizations and the historical ones. The estimated total intake doses *via* air inhalation and dust ingestion of children in the recycling region of Guiyu ranged from 10 to 32 pg TEQ/(kg•day), which far exceeded the tolerable daily intake (TDI) limit (1–4 pg TEQ/(kg•day). Although the measurements showed a significant reduction of the release of PCDD/Fs, the pollution status was still considered severe in Guiyu town after stricter regulations were implemented.

Introduction

E-waste is physically and chemically different from industrial or municipal waste (Robinson, 2009). Those types of obsolete products have always included metals, plastics, glass, ceramics and other materials that are valuable and recyclable (Hoffmann, 1992; Robinson, 2009). Driven by economic motives, e-waste recycling was promoted and became a large industry in some developing countries, such as China, India, and Pakistan (Hicks et al., 2005; Hosoda, 2007; Ogunseitan et al., 2009). Unfortunately, most e-wastes were recycled informally and caused severe environmental pollution in these countries (Ha et al., 2009; Hischier et al., 2005; Leung et al., 2006; Li et al., 2007; Robinson, 2009). The pollutants include persistent organic pollutants (POPs), heavy metals, particulate matter, and volatile organic compounds (VOCs) (An et al., 2014; Fu et al., 2012).

Over the last two decades, China appears to have become the largest e-waste dumping site in the world, receiving shipments from the United States, Europe, and neighboring Asian countries such as Japan and South Korea (Chi et al., 2011; Hosoda, 2007). Consequently, e-waste-related pollution has received wide attention in China since the 1990s. Guiyu town and Longtang town (Qingyuan) in Guangdong province are the two best known regions for illegal e-waste recycling activities. Serious environmental pollution has been reported in these areas many times over the last two decades. Due

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http://dx.doi.org/10.1016/j.jes.2017.07.009
1001-0742/

to the high carcinogenic potential to humans and other species and the high concentrations found in different kinds of environmental and biological matrices (Chan and Ming, 2013), polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/Fs) became the most intractable environmental chemicals in such areas. Li et al. (2007) measured the ambient air concentrations of PCDD/Fs in Guiyu and reported the highest documented values in the world, ranging from 0.909 to 48.9 pg WHO-TEQ/m^3 (TEQ: toxic equivalent quantity). In Qingyuan, relatively high PCDD/F levels (0.06–29.28 pg I-TEQ/m^3) in ambient air were also recorded by Xiao et al. (2014). In addition, severely contaminated surface soil, sediment, dust, and biota were observed (Hu et al., 2013; Leung et al., 2006, 2007; Ren et al., 2015; Wong et al., 2007). Open burning, acid washing and open dumping were found to be the major sources of PCDD/Fs in these recycling regions (Duan et al., 2012; Leung et al., 2007; Yu et al., 2006; Zheng et al., 2008; Zhu et al., 2008). Since the 2010s, the authorities of Guangdong province and local governments have enhanced law enforcement and environmental regulations to abolish uncontrolled e-waste recycling activities, especially by the three crude methods described above. At the beginning of 2013, open burning and acid washing were finally abolished in both Guiyu and Qingyuan. Some large centralized industrial parks were also established with more sophisticated treatment techniques. Due to the increasing amounts of e-waste generated in China (Yu et al., 2010) and the economic and social benefits of e-waste recycling, it is more important to standardize the dismantling processes rather than to compulsively prohibit them by laws and regulations. Therefore, the transformations in recent years seem to be successful but have not ended e-waste recycling regulation. It is necessary to reveal the current contamination status and emission sources in these regions to assess the effect of the regulatory actions and provide useful information for further policy-making. As we know, most PCDD/Fs and other POPs are first released into the ambient air and then deposited into other environmental matrices. In this study, PCDD/Fs in $PM_{2.5}$ (fine inhalable particles, with diameters that are generally 2.5 micrometers and smaller), TSP (total suspended particulate) and gas phases and as indoor dust were collected in Guiyu and Qingyuan and quantified after the enhanced regulations were put in place. The changes in the PCDD/Fs pollution status were assessed by comparison with previously reported data. Moreover, the current sources of PCDD/Fs in the two regions were identified and discussed.

1. Experimental section

1.1. Two studied e-waste recycling regions

1.1.1. Guiyu

Guiyu is a small town of Shantou city in Guangdong province. It became an e-waste recycling center in 1995. More than 5500 family-run workshops and small-scale factories worked on e-waste recycling in this region. During the 1990s and 2000s, e-waste was recycled in this region by primitive means such as (1) burning wires or cables in wild and even on farmland to recover copper; (2) baking printed circuit board over coal grills to recover lead-tin solder and integrated components; (3) using acid baths to recover gold and dumping the strong acid into nearby rivers and farmlands; and (4) dumping e-waste residues directly into rivers, irrigation ditches and farmlands.

Although regulations have been in place since the 2000s, the reform process was slower than for other recycling centers, largely due to the strong resistance from the people who made a living in this industry. Starting in 2010, to reduce the harmful environmental consequences, the authorities carried out a series of stricter control policies and pushed advanced techniques for recycling industries, which included (1) strict controls on the open burning and acid washing of e-waste; (2) use of wire-stripping machines to recover copper and other metals; and (3) setting up modern plastic industries to sort, melt and granulate recyclable plastic residues. Therefore, plastic recycling and circuit board baking became the two representative industries in Guiyu town in recent years.

1.1.2. Qingyuan

Qingyuan is located in northern Guangdong and has a population of 3.7 million. Since the 1990s, it has become famous for its secondary metal industry and been called the “copper capital of China”. In fact, the secondary metal came mostly from informal e-waste recycling activities. Similar to Guiyu, the recycling was done by many family workshops, and primitive recycling methods were pervasive. The open burning and disposal of e-waste and residuals on farmland, acid washing and the manual dismantling of e-waste were the most common recycling methods in this region before the 2010s. Since the 2000s, legislative approaches and industrial centralization were developed to minimize the negative environmental effects. These regulations had made some significant progress by the beginning of the 2010s. For example, open burning and acid washing were eliminated, most of the family workshops were finally moved into a central recycling park, and several metal recovery factories with advanced technologies and standardized management were established and run well in recent years.

1.2. Sampling location

For a better comparison with the historical reports, the sampling sites were located in the areas that had been studied previously (Li et al., 2007; Xiao et al., 2014). To evaluate the types of e-waste recycling processes and the changes in the recycling modes in these two areas, four and three sampling sites near e-waste workshops were chosen in Guiyu and Qingyuan, respectively. Briefly, the sampling sites in Guiyu were located in the neighborhoods of plastic recycling workshops (GY1# and GY2#) or circuit board baking activities (GY3# and GY4#). Neighborhoods around an informal dismantling family workshop (QY2#) and a green recycling center (QY3#, QY4#) were chosen as typical sampling sites in Qingyuan. In addition, two remote small villages where no e-waste recycling activities were conducted and located upwind of the recycling areas (~7 km away) were investigated as control sites (GY5# and QY1# for Guiyu and Qingyuan, respectively).

1.3. *Sample collection*

TSP, $PM_{2.5}$ and gas phase samples were collected simultaneously on the rooftops of one building from each site in August 2013. The sampling procedure for TSP and gas phase samples was described in our previous study (Zhang et al., 2014). Applying a particle size-selective impactor, $PM_{2.5}$ samples were collected on quartz fiber filters (10.1 cm × 25.4 cm, Whatman, Maidstone, England), and the gas phase was trapped in a polyurethane foam (PUF, thickness: 7.6 cm; diameter: 6.5 cm) using a high-volume air sampler (Tianhong, China). Before sampling, surrogate standards (EPA 23 SSS, Wellington Laboratories, Guelph, Canada) were added in sampling matrix. The prevailing wind in both Guiyu and Qingyuan was northeasterly, and the sampling was conducted in non-rainy weather. The sampling season and strategies were the same as those described by Li et al. (2007) and Xiao et al. (2014).

Settled indoor dust samples were collected by manual brushing in the same buildings or nearby buildings for air sampling. More than 6 samples were collected but were mixed into one pooled sample in each building due to limited sample amounts.

1.4. *Sample preparation and chemical analysis*

All of the organic solvents were pesticide residue grade or ultimately purified grade from Tedia (USA). Silica gel was purchased from Merck (silica gel 60, Darmstadt, Germany). Basic alumina was obtained from Aldrich (Brockmann I, standard grade, Milwaukee, USA). Florisil was from Merck (60–100 mesh, Germany). Calibration standard solutions, PAR stock standards, $^{13}C_{12}$-labeled surrogate standards and $^{13}C_{12}$-labeled injection standards were purchased from Wellington Laboratories (Guelph, Canada) and Cambridge Isotope Laboratories (Andover, MA, USA).

The fiber filter and PUF of each sample were separately transferred into the Soxhlet extractor after the samples were spiked with a mixture of $^{13}C_{12}$-labeled internal standards (EPA 23 IS). For dust samples, EPA 1613 internal standards (EPA 1613 LCS) were added to each sample, which was filtered through a stainless-steel sieve (<0.2 mm) before extraction. After extraction by toluene for 24 hr, the extracts were concentrated to dryness, and the solvent was exchanged to hexane. Applying a cleanup procedure involving acid wash and a multilayer column, PCDD/Fs in each sample were finally purified. The concentrated elutes were added an internal standard (EPA23 RS) for chemical analysis by HRGC/HRMS (high resolution gas chromatography coupled with high resolution mass spectrometry) (Agilent 6890N, Micromass Autospec Ultima, Waters, USA).

1.5. *Quality control*

Field blanks were prepared by loading a PUF plug and QFF filter to the samplers for 48 hr at two sampling sites during the sampling period with no air drawn through. Method blanks and a spike blank were used in each batch of samples ($N \leq 10$). Clean PUF and QFF filters were used as a blank reference matrix for air samples, and clean sand was used for dust samples. No target interference was found in all the blank samples. The recoveries of the surrogate sampling standard and internal standard ranged 85%–113% and 68%–98%, respectively. The recoveries of target compounds ranged from 91% to 108% in the spike blank samples. The method detection limit (MDL) of the target compounds is listed in Table S2, and the data below MDL were recorded as half of MDL.

2. Results and discussion

2.1. *PM and PCDD/F concentrations after stricter regulations*

The TSP and $PM_{2.5}$ concentrations from all sampling sites are illustrated in Table S3. According to the Chinese Ambient Air Quality Standard (GB3095-2012), the PM levels of most sampling sites were below the annual average limit of the second grade standard (TSP: 200 $\mu g/m^3$; $PM_{2.5}$: 35 $\mu g/m^3$) and were much lower than most of the reported data from urban areas in Asia and the previous records of e-waste recycling studies in China (Table S4). According to a study by WHO (WHO, 2003), $PM_{2.5}$ and PM_{10} are produced by mechanical processes such as construction activities and road dust re-suspension, and $PM_{2.5}$ originate primarily from combustion sources and secondarily from aerosols *via* gas-to-particle conversion. The reduction of TSP and $PM_{2.5}$ in Guiyu may result from the disappearance of open burning in recent years. Gullett et al. (2007) investigated the severe particle emissions from open burning of e-waste by conducting simulated rudimentary recycling operations to characterize the air emissions, and they obtained high emission factors of TSP (15,600 for circuit board samples and 17,500 for insulated wire samples, respectively).

Similarly, PCDD/Fs showed significantly decreased levels compared to the reported data in these two areas. The atmospheric total TEQ concentrations (TSP plus gas phase) in the studied recycling sites of Guiyu ((1.21 ± 0.76) pg/m^3, Table 1) were 7 times lower than that in 2005 (average 8.78 pg I-TEQ/m^3) (Li et al., 2007). In Qingyuan, the current data also showed 31 times lower levels for informal or formal recycling sites (0.271 and 0.644 pg I-TEQ/m^3, respectively, Table 1) compared with the values measured in 2009 by Xiao et al. (2014) (summer, average 8.48 pg I-TEQ/m^3). Both the PM and PCDD/F results revealed that the enhanced regulations and centralized dismantling action may have had a positive environmental effect in these two studied regions. Consistent results were also observed in Taizhou, which is another e-waste hot spot in China. Marked decreases in the PCDD/Fs were recorded in Taizhou after the local government closed down nearly 10,000 small-sized informal workshops and stopped open burning activities (Fu et al., 2012).

2.2. *PCDD/Fs distribution variation in different recycling sites*

As shown in Table 1, the air samples in four recycling sites in Guiyu had the highest PCDD/F levels of all samples, which were 7 times higher, on average, than the reference site (GY5#), implying the significant impact of the e-waste dismantling activities on the airborne PCDD/F levels in this

Table 1 – Summary of PCDD/F mean concentrations (pg/m^3) in the atmosphere of Guiyu and Qingyuan.

Site	$PM_{2.5}$		TSP		Gas phase		Air concentration (TSP + Gas)	
	∑D/Fs	∑D/Fs (TEQ)	∑D/Fs	∑D/Fs (TEQ)	∑D/Fs	∑D/Fs (TEQ)	∑D/Fs	∑D/Fs (TEQ)
ST1#	13.8	0.291	14.9	0.34	3.38	0.63	18.3	0.97
ST2#	31.3	0.917	37.1	1.35	6.80	1.08	43.9	2.43
ST3#	7.40	0.115	11.83	0.29	2.64	0.44	14.5	0.73
ST4#	7.80	0.146	17.95	0.32	2.79	0.50	20.7	0.82
Mean	15.1	0.367	20.4	0.58	3.90	0.66	24.3	1.24
ST5#	0.59	0.0022	0.54	0.0019	0.60	0.021	1.14	0.023
QY1#	1.33	0.036	2.39	0.081	0.493	0.058	2.9	0.139
QY2#	2.24	0.083	4.18	0.156	0.818	0.116	5.0	0.271
QY3#	7.34	0.091	15	0.191	2.73	0.345	17.8	0.536
QY4#	14.2	0.204	47	0.348	3.144	0.296	50.2	0.644
Mean	6.3	0.104	17.1	0.19	1.80	0.204	18.96	0.40

PCDD/F: polychlorinated dibenzo-p-dioxins and dibenzofuran.
$PM_{2.5}$: fine inhalable particles, with diameters that are generally 2.5 micrometers and smaller; TSP: Total suspended particulate; TEQ: Toxic equivalent quantity.

area. The informal dismantling process for scrap printed circuit boards (SPCBs) has been found to be associated with rapid PCDD/Fs production (workshop air particles:14.5 WHO pg/m^3) (Duan et al., 2012; Ren et al., 2014), which may be due to low temperature pyrolysis (Huang et al., 2009). By contrast, mechanized plastic granulation, which is thought to be an improved approach for replacing the open burning of plastic materials at GY3# and GY4#, seems to cause severe PCDD/Fs contamination during SPCBs dismantling. As far as we know, there is still no study about PCDD/Fs formation in the plastics recycling process. The plastic material is first melted at a low temperature (approximately 200–400°C), which could be a favorable temperature for PCDD/Fs formation. Further investigation about the mechanism is urgently needed.

Compared with Guiyu, the concentrations of PCDD/Fs in Qingyuan were much lower. There could be two reasons for this difference. The first is the variation in the e-waste types between the regions. The e-waste in Qingyuan is mainly from electric equipment, whereas in Guiyu, the material is predominantly electronic items and plastic products. Thus, the predominant dismantling methods in the two sites were different. In Qingyuan, physical separation was the predominant dismantling technique in family workshops. While electronic items contained more precious metals than electrical instruments, the process of dismantling in Guiyu was more complicated and mostly related to thermal processes as mentioned above. Another reason could be the centralized management of the recycling activities by the authorities in Qingyuan in recent years. More than 200 quantified dismantling factories were moved into an e-waste dismantling park that was established and began operation in 2006. In these quantified factories, metals were recovered from seven main kinds of e-wastes (*e.g.*, transformer, cables, wires, electric motors) in the factories that used automotive shredding, separation and metal refining procedures. Since 2012, open burning, which had been used to recover copper and other metals from plastic containing cables and wires, was hardly found in the two regions after stricter prohibition and heavier punishment. Nevertheless, the airborne PCDD/F levels in and around the centralized industrial park (QY3# and QY4#) were nearly at or already exceeded the safety limit (0.6 pg TEQ/m^3) and relatively higher than the family workshop (QY2#) as well as the reference site (QY1#), indicating that better regulation for the recovery factories is still needed.

As we know, 2378-PCDD/Fs congeners and 4–8 chlorinated homologue profiles are always used to identify the source differences of environmental PCDD/Fs pollution (Kishida, 2013; Ogura et al., 2001). The profiles shown in Fig. 1 reveal that relatively higher abundances of low chlorinated PCDFs (4–6 Cl) and lower abundances of high chlorinated 2378-PCDFs and 2378-PCDDs (7–8 Cl) were found at e-waste recycling sites in Guiyu, compared with control sites. These also illustrate some differences between the studied regions. For example, the higher contribution of four and five chlorinated 2378-PCDFs but lower OCDF was observed in Guiyu in contrast to Qingyuan areas. The comparison of the current data and the former reports shows a drastic reduction of low chlorinated 2378-PCDFs and high chlorinated 2378-PCDDs. These consistent results may indicate the important contribution of these two groups of 2378-PCDD/Fs to the severe PCDD/Fs pollution noted in previous reports and currently found in Guiyu. Moreover, principal component analysis (PCA) was performed based on homologous data for samples from different recycling sites in this study and previous reports. As shown in Fig. 2, the factor loading plot of PCA shows that the deposition samples from the two recycling regions were divided into different discrete clusters. All samples from the recycling sites in Guiyu fell into one group and were distinct from those from the rural site (GY5#) and the sites of Qingyuan and from the representative points of the former reports (GY2005). The sample points from Qingyuan were also distinct from those described in the former reports (QY2009). The results imply that there are some differences between the air samples collected from different recycling districts and previous reports, which indicates different dominant PCDD/F emission sources in these two recycling regions after stricter environmental regulation.

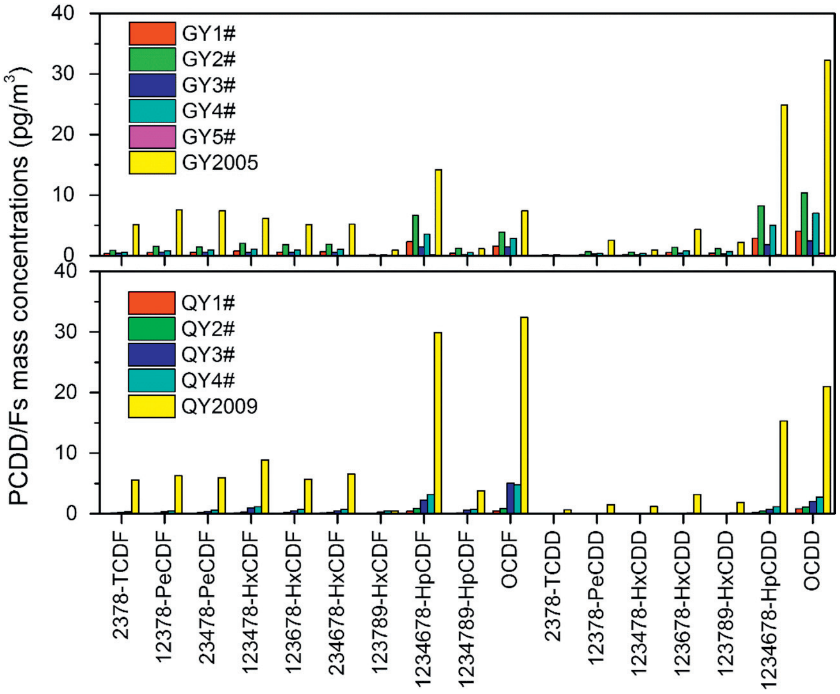

Fig. 1 – PCDD/F congener distributions (mass concentration) in Guiyu and Qingyuan before and after stricter regulations (data source: QY2009, from Xiao et al. (2014); GY2005, from Li et al. (2007)).

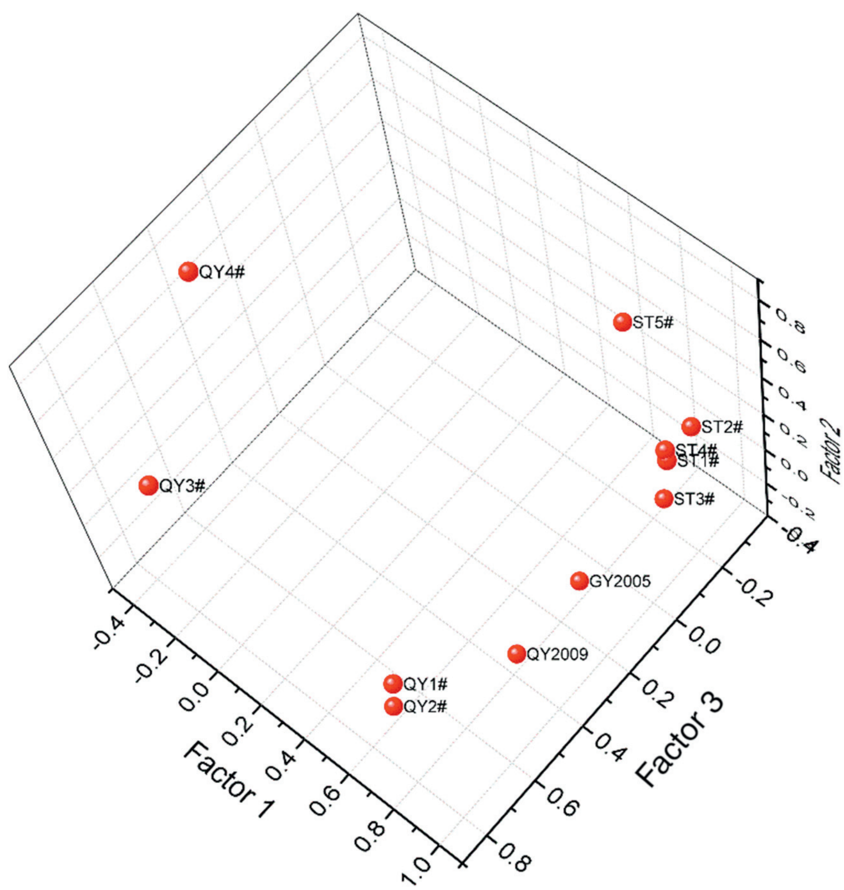

Fig. 2 – Factor loading plot of principal component analysis (PCA) for the deposition samples from the e-waste recycling sites, rural sites and former studies (data source: QY2009, from Xiao et al. (2014); GY2005, from Li et al. (2007)).

2.3. Size distribution of particle-bound PCDD/Fs

High correlations of PCDD/Fs congener distribution in $PM_{2.5}$ and TSP were obtained for all e-waste recycling sites (r > 0.97). The average $PM_{2.5}$/TSP ratio of the total 2378-PCDD/Fs at four recycling sites in Guiyu (GY1#, GY2#, GY3# and GY4#) and central industry park in Qingyuan (QY4#) were 92%, 84%, 62%, 43%, and 30.2%, which were consistent with the mass ratios of $PM_{2.5}$/TSP (Appendix A Table S1). No size effect between $PM_{2.5}$ and TSP could be found for the total PCDD/Fs in the recycling sites. Similar results for the PCBs and PBDEs in another recycling region, Taizhou, were reported several years ago (Han et al., 2009, 2010).

2.4. Transfer of PCDD/Fs in e-waste recycling site

The TEQ concentration of PCDD/Fs in indoor dust around e-waste recycling areas in Guiyu ranged from 1020 to 3637 pg/g (average 2662 pg/g) and was 59 times higher than the reference site (ST5#, 45.4 pg/g) and even higher than any other reported data from e-waste areas (Leung et al., 2007; Ma et al., 2008; Ogunseitan et al., 2009). In Qingyuan, dust concentration was about ten times lower than that from Guiyu, ranging from 49 to 446 TEQ pg/g, and the highest level was found at an office building indoor environment near the e-waste industrial park.

High correlations (Pearson r, 0.808) of PCDD/Fs for congener composition were found between total air samples (TSP plus gas) and dust samples. In fact, dust samples were collected from classrooms and subdistrict offices where no indoor e-waste activities were related. Therefore, such high PCDD/F values in these dust samples may come from the ambient air deposition around the e-waste activities. To further investigate the transfer of PCDD/Fs in e-waste recycling sites, the relationships among $PM_{2.5}$, TSP, gas phase, and dust samples were also evaluated. A relatively higher averaged correlation coefficient (Pearson, r) between dust and particles (0.759) was obtained than found for gas phase samples (0.416), indicating that the PCDD/Fs in dust were mostly attributed to day-to-day particle deposition from the ambient contaminated environment. In addition, no difference could be found between the $PM_{2.5}$ and TSP evaluation, which suggested that PCDD/Fs on fine particles in this recycling site may ultimately transfer to indoor environment in these regions.

2.5. Implication for exposure to PCDD/Fs via inhalation and dust ingestion

As we know, apart from the daily intake, inhalation and dust ingestion may make important contributions to the total exposure to POPs. The average daily intake was calculated using the model released by US EPA (See Supplementary materials). As shown in Fig. 3, because of the reduction in the PCDD/F levels in exposed air, the inhalation exposure doses were relatively lower than ever before (Li et al., 2007). However, when dust ingestion exposure was added, the total doses were higher than the average intake doses for Chinese people (adult: 0.72 pg TEQ/(kg•day), child: 1.08 pg TEQ/(kg•day)). Our interviews with local residents and authorities showed that most of the food currently consumed by local residents was from other towns and provinces, which indicated that there was little place-specific consumption in the recycling regions and that the average intake doses of PCDD/Fs by Chinese people could be used for total daily intake estimation and comparison in this study. For ordinary residents, it is estimated that food intake accounts for approximately 90% of the tolerable daily intake (TDI) (Robinson, 2009). It seems that this estimate was not suitable for the scenarios of these recycling related residents. Residents in Guiyu recycling areas were at the highest risk of PCDD/Fs exposure, ranging from 1.06 to 3.00 pg TEQ/(kg•day) and 9.50 to 33.4 pg TEQ/(kg•day) for adults and children, respectively. Because of the differences in the exposure factor between children and adults, the daily intake doses of children were approximately 10 times those of adults. Although the exposure doses of adults were under the recommended TDI of PCDD/Fs established by the WHO (1–4 pg TEQ/

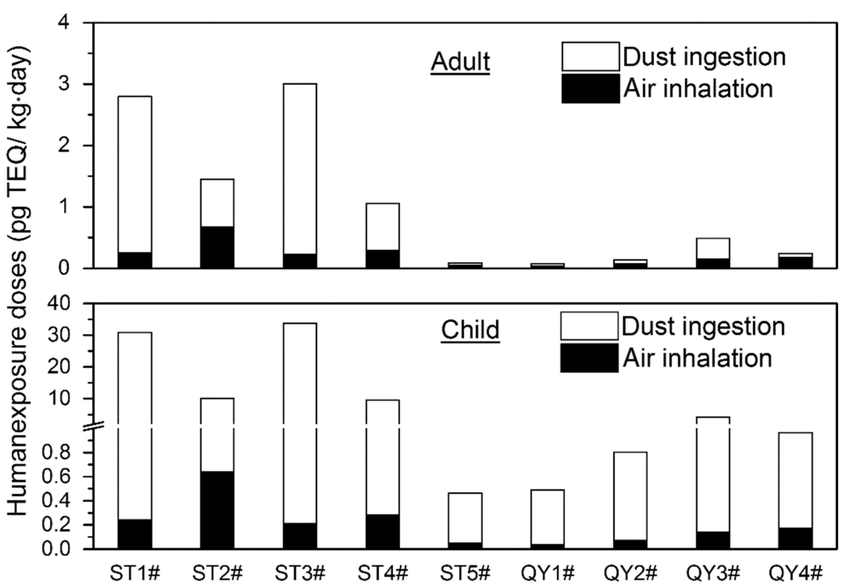

Fig. 3 – Estimated exposure doses of adult and children in the studied areas.

(kg•day), children may take in more than 2 times this value. Therefore, children in Guiyu may be more susceptible to the poor air quality and surrounding environment and suffer an elevated prevalence of respiratory symptoms in recent years (Zeng et al., 2016).

3. Conclusions

In this study, the PCDD/Fs contamination status of two typical e-waste recycling regions after stricter regulation was studied and compared. The atmospheric PCDD/Fs and particle concentrations in both regions were significantly reduced compared with any previous levels, which could be the result of stricter control measures and application of an environmental technique. Different current PCDD/F emission sources and pollution statuses were found between Guiyu and Qingyuan. In future years, the authorities can focus on regulating the industrial processes of the circuit board baking and plastics recycling in Guiyu while supervising the flue gas treatment facilities in centralized industry parks in Qingyuan to address the PCDD/F problems. Although the existing measures showed a significant reduction in the release of PCDD/Fs, the pollution status is still considered severe in Guiyu town, and further industrial reforms and science investigation are urgently needed.

Acknowledgments

This work was supported by the National Natural Science Foundation of China (No. 91543202) and the Ministry of Environmental Protection of the People's Republic of China (Nos. 201409022, 201409080 and 201509063) and South China Institute of Environmental Science (No. PM-zx021-201510-015).

Appendix A. Supplementary data

Supplementary data to this article can be found online at http://dx.doi.org/10.1016/j.jes.2017.07.009.

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