



## Short Communication

## Changes in human hair levels of organic contaminants reflecting China's regulations on electronic waste recycling



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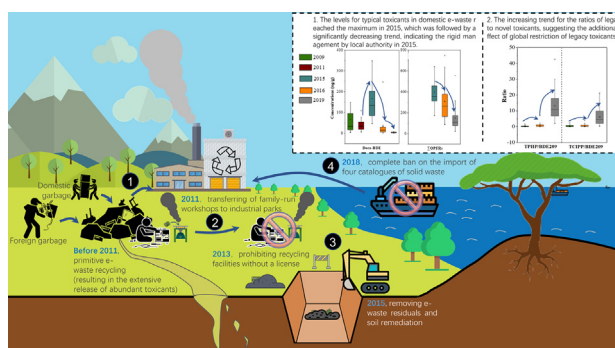
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## HIGHLIGHTS

- Hair levels of PCBs, PBDEs, and OPFRs over ten years were reported from a former e-waste area.
- $\Sigma_9$ PCB,  $\Sigma_3$ Penta-BDE, and  $\Sigma_3$ Octa-BDE levels in hair decreased significant from 2011 to 2019.
- A significant decrease was observed for Deca-BDE and  $\Sigma_8$ OPFR values from 2015 to 2019.
- A significant increase was found for the ratios of TPHP/Deca-BDE and TCIPP/Deca-BDE.
- The temporal changes in levels of chemicals indicates the efficacy of regulations.

## GRAPHICAL ABSTRACT



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## ABSTRACT

To assess the impacts of regulations and laws enhancing the management of e-waste in China, hair samples of local residents and dismantling workers in a former e-waste area in 2016 and 2019, five and eight years after the implementation of legislation and regulations in this area since 2011, respectively. The temporal changes in levels of polychlorinated biphenyls (PCBs), polybrominated diphenyl ethers (PBDEs), and organophosphorus flame retardants (OPFRs) in the hair samples were investigated. Besides, the levels of these organic contaminants in hair samples collected from the same area in 2009, 2011, and 2015 reported in previous studies were used as comparison. The highest median levels of  $\Sigma_9$ PCBs (719 ng/g),  $\Sigma_3$ Penta-BDEs (16.1 ng/g), and  $\Sigma_3$ Octa-BDEs (8.46 ng/g) in hair were found in 2011, with a significant decrease trend was observed from 2011 to 2019 ( $p < 0.05$ ). As for Deca-BDE, the levels reached the maximum in 2015 (133 ng/g), following by a significant decrease to 2016 (7.46 ng/g) and 2019 (2.61 ng/g) ( $p < 0.05$ ). The median levels of  $\Sigma_8$ OPFRs, also decreased significantly ( $p < 0.05$ ) from 2015 (357 ng/g) to 2016 (264 ng/g) and 2019 (112 ng/g). Moreover, a significantly increasing trend was observed for the ratios of triphenyl phosphate (TPHP) and tris(2-

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chloropropyl) phosphate (TCIPP), two predominant OPFRs, to Deca-BDE from 2015 to 2019 ( $p < 0.01$ ), suggesting a shift of “legacy” to “emerging” contaminants released from e-waste recycling in this area. The temporal changes in hair levels of typical organic contaminants in residents and dismantling workers indicated the effectiveness of the regulations on informal e-waste recycling activities and solid waste in China.

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## 1. Introduction

Electrical and electronic waste (e-waste) is one of the fastest growing waste streams in the world in terms of volume and its environmental impacts. The global generation of e-waste grew by 9.2 million metric tons (Mt) since 2014 and is projected to grow to 74.7 Mt by 2030 (Forti et al., 2020). During the last decades, a large amount of e-waste generated in developed countries has been exported legally or illegally to developing countries such as China, India, and some countries in Southeast Asia and Africa. China has become the largest producer and consumer of electrical and electronic equipment in recent years, as well as the country that is most affected by e-waste contamination due to the massive e-waste imports and widespread informal recycling (Perkins et al., 2014; Zeng et al., 2016). Approximately 70% of e-waste generated worldwide has been processed in China (Robinson, 2009). The primitive e-waste recycling activities, such as combusting or roasting circuit boards, peeling and melting plastic, and extracting metals with strong acids, have resulted in the extensive release of abundant toxicants, including heavy metals, polychlorinated biphenyls (PCBs), polybrominated diphenyl ethers (PBDEs), and organophosphorus flame retardants (OPFRs), into the environments, and raised public health concerns (Chen et al., 2011; Huang et al., 2018; Huo et al., 2007). PBDE and PCB exposure have adverse effects on neurodevelopment and fertility by disrupting thyroid hormone (Fromme et al., 2014; Safe, 2003). Some OPFRs are suspected to be mutagenic, carcinogenic, neurotoxic or embryotoxic (van der Veen and de Boer, 2012; Wei et al., 2015).

To address the e-waste problem in China, national and local laws and regulations have been enacted to enhance the management (collection, storage, transport, and disposal) of e-waste (Ni and Zeng, 2009; Puckett et al., 2002; Zeng et al., 2016). China tightened the ban on importation of solid waste, especially e-waste that contains toxic substances, in July 2017, and declared complete ban since 1 January 2018. Longtang is a small town of Qingyuan City, Guangdong Province, and is among the most intensive e-waste recycling sites since 1990s, mainly due to its convenient location and low labor costs. In 2010, the annual production volume of recycled copper (Cu) in Longtang reached 0.68 Mt, with approximately 3 Mt of e-waste dismantled in this area, which were mostly imported from abroad. In this area, family-run e-waste disposal activities were generally conducted in the backyards, sometimes even the living rooms, of the workers' houses, raising continuously high exposure to toxic substances for the local residents and workers (Huang et al., 2018; Poma et al., 2019). A series of regulations have been imposed by the local authority to alleviate e-waste contamination and the associate adverse effects, including: taking steps to restrict family-run recycling workshops and transfer them to industrial parks since 2011 (The People's Government of Qingyuan, 2011), rectifying and cleaning-up scattered small recycling facilities that do not conform to land use planning, prohibiting recycling facilities without a license from April 2013 (The People's Government of Qingyuan, 2013), promoting the dismantling technology, and investing in infrastructure to help recycling e-waste properly. In particular, the extensively removing e-waste residuals and soil remediation from July 2015 are expected to play a key role in preventing the deterioration of e-waste recycling (The People's Government of Qingyuan, 2015).

In recent years, hair has been increasingly used as an alternative non-invasive indicator to assess human exposure to organic contaminants in comparison to other human specimens, for the advantages of its low cost and convenient transportation and storage (Qiao et al.,

2019). In our previous studies, relatively high levels of human exposure to PBDEs, PCBs, and OPFRs for local residents and workers from Longtang town were reported in 2009 (Zheng et al., 2011; Zheng et al., 2013), 2011 (Zheng et al., 2014; Zheng et al., 2016), and 2015 (Qiao et al., 2019). The management in Longtang town was also crucial for reducing exposure of local residents to toxic contaminants released from e-waste recycling. Therefore, in the present study, hair samples of local e-waste dismantling workers/residents were collected in 2016 and 2019 in Longtang town to further investigate the temporal changes in levels of human exposure to these organic chemicals, and to assess the effectiveness of the implementation of legislation and regulations governing e-waste in this area.

## 2. Materials and methods

### 2.1. Chemicals

Seven PBDE congeners, 9 PCB congeners, and 8 OPFRs were selected as target analytes in this study. Detailed information of the analytes is provided in Table S1 of Supporting Information (SI).

### 2.2. Sample collection and analysis

Longtang town, once known as “the renewable copper city of China”, in where e-waste recycling activities have been conducted for more than 30 years, is located in Qingyuan city, Guangdong province. Three villages (Baihe village: N 23°36', E 113°04'; Banchong village: N 23°32', E 113°03'; Yangwu village: N 23°34', E 113°02') that involved in e-waste recycling activities for many years were selected for the sampling campaigns. A total of 42 and 37 e-waste dismantling workers/residents were recruited in 2016 and 2019, respectively. The workers/residents should have inhabited in the e-waste area for more than five years, and have not had hair care, such as hair coloring or perming, within two years. The study was approved by the Ethics Committee in the School of Life Sciences, Sun Yat-sen University. Written informed consent was obtained from all the participants, and they were clearly informed about the scope and nature of the study. Sample collection, preparation, and instrumental analysis were conducted according to the methodologies reported in our previous study (Tang et al., 2021), and given in details in the SI.

Additionally, the data for hair contaminants in 2009 (Zheng et al., 2011; Zheng et al., 2013), 2011 (Zheng et al., 2014; Zheng et al., 2016), and 2015 (Qiao et al., 2019) from the same area were collected as well.

### 2.3. Quality assurance and quality control

Procedural blanks and spiked matrices (hair) were set for experimental quality control. Two procedural blank samples were included in each batch of 22 samples. Only trace amounts were detected in the blanks, and the average levels of target chemicals in blanks were subtracted from those detected in the hair samples, respectively. The mean recoveries for the target chemicals in the spiked hair matrices ( $n = 6$ ) were in the range of 72–113% with relative standard deviation (RSD) below 16%.

The limits of quantification (LOQs) were calculated as the mean value of target analytes detected in procedure blanks plus three times of those of the standard deviation for individual chemical that was

detected in the procedural blank samples. For the undetected chemicals in procedural blanks, LOQs were estimated as a signal-to-noise ratio of 10 ( $S/N = 10$ ). The LOQs were 0.01–6.89 ng/g for PBDEs, 0.05–9.76 ng/g for PCBs, and 0.02–7.74 ng/g for OPFRs, respectively. Recoveries of all ISS ranged between 87 and 108%, with an RSD < 18%.

#### 2.4. Statistical analysis

Statistical analysis was performed for target chemicals with detection frequency (DF) higher than 60% using SPSS 22.0 (SPSS Inc., Chicago, IL, USA). Concentrations below the LOQs were assigned as LOQ\*DF during statistical analysis. One-way analysis of variance (ANOVA) was conducted to determine the possible differences among chemical concentrations in hair samples collected in different years. Statistical significance was set at  $p < 0.05$ .

### 3. Results and discussion

The levels of target analytes in hair samples are summarized in Table 1. The DFs for PBDE congeners were 5–92% and 0–86% in 2016 and 2019, respectively, and were 81–95% and 0–69% for PCB congeners in 2016 and 2019, respectively; these values were much lower than those for PBDEs (84–100%) and PCBs (97–100%) in hair samples collected in 2009, 2011, and 2015. However, the DFs for OPFRs in 2019 (46–97%) and 2016 (67–100%) were comparable to those in 2015 (61–100%) (Table 1).

Fig. 1a–e shows the changes in levels of PBDEs, PCBs and OPFRs in human hair collected in 2009 (Zheng et al., 2011; Zheng et al., 2013), 2011 (Zheng et al., 2014; Zheng et al., 2016), 2015 (Qiao et al., 2019), 2016, and 2019 from residents and e-waste dismantling workers in Longtang town. The median  $\Sigma_9$ PCB levels in hair were 142, 719, 178, 125, 1.41 ng/g in 2009, 2011, 2015, 2016, and 2019, respectively. The levels of  $\Sigma_9$ PCBs increased from 2009 to 2011, which was probably due to the continuous dismantling of obsolete transformers and capacitors that containing PCBs in Longtang town during this period. Then, a significant decrease was observed for the  $\Sigma_9$ PCB levels from

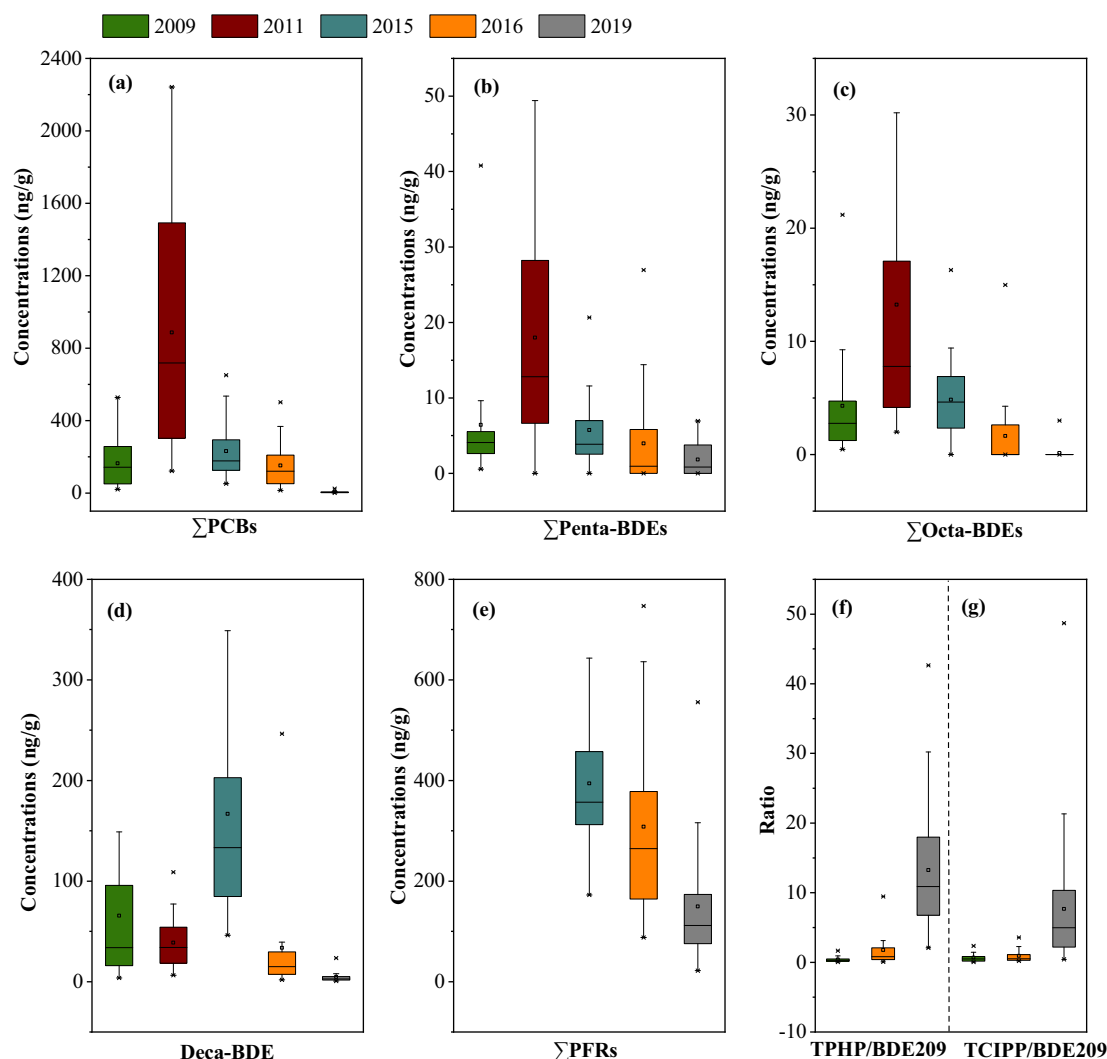
2011 to 2019 ( $p < 0.05$ , Fig. 1a). The observed decline fit well with the e-waste disposal regulations in this region since 2011, suggesting the efficacy of regulatory policies on reducing human exposure to PCBs. PCBs were used primarily as electrical insulating fluids in capacitors and transformers, hydraulic fluid, and lubricating fluid, and have been banned since 1970s (Erickson and Kaley, 2011). During our field trips to Longtang in 2011, the authors learned from owners of the family-run workshops that obsolete transformers and capacitors for recycling had no longer been available in this region since 2011 due to the regulations.

Three commercial PBDE mixtures, i.e. Penta-, Octa-, and Deca-BDEs, have been produced and used. The median levels in hair in the five sampling years were 4.42, 16.1, 4.36, 2.58, and 0.78 ng/g for  $\Sigma_3$ Penta-BDEs, were 2.76, 8.46, 4.63 ng/g, n.d. (not detected), and n.d. for  $\Sigma_3$ Octa-BDEs, and were 33.8, 34.0, 133, 7.46, and 2.61 ng/g for Deca-BDE (BDE 209), respectively. Similar to those for PCBs, the highest levels for  $\Sigma_3$ Penta-BDEs and  $\Sigma_3$ Octa-BDEs were found in 2011; as for Deca-BDE, no significant difference was observed between 2009 and 2011, and the levels reached the maximum in 2015, which was followed by a significantly decreasing trend from 2015 to 2019 (Fig. 1b–d). PBDEs were additive flame retardants used in various products such as electrical appliances, plastics, polyurethane foam, furniture, and textiles. Penta- and Octa-BDE mixtures were extensively used in North America and Europe, while Deca-BDE was used in the largest quantity in China (Liu et al., 2016). The delay for the peak hair levels of Deca-BDE implied a shift from importation to domestic generation of e-waste being dismantled in this area from 2011 to 2015. Additionally, the historical residues of e-waste dismantling in 2011, resulting in the high levels of PBDEs in the surrounding environments including soils, air and suspended particles, could also contribute to the high hair levels in 2015. However, the robust influence by the regulation implemented in 2015 in this area probably the main reason for the declining of Deca-BDE. The regulation implemented in 2015 was unprecedentedly strict. According to the local authorities, there were a total of 2358 e-waste recycling workshops in Longtang town by the end of 2014; however, 556 workshops were moved into the e-waste recycling industrial parks, and the rest were

**Table 1**  
Summary of organic contaminant concentrations in hair from Longtang Town, South China over a 10-year long biomonitoring (ng/g).

Targeted analytes	2009 (n = 31)			2011 (n = 35)			2015 (n = 31)			2016 (n = 42)			2019 (n = 37)		
	DF (%)	Median	Range	DF (%)	Median	Range	DF (%)	Median	Range	DF (%)	Median	Range	DF (%)	Median	Range
BDE 47	100	1.90	0.37–24.2	88	5.79	nd–26.6	84	1.45	nd–9.03	74	1.55	nd–23.0	54	0.78	nd–6.94
BDE 99	100	2.18	0.20–51.3	88	7.32	nd–36.1	97	2.54	nd–11.6	83	2.18	nd–30.0	0	–	nd
BDE 100	100	0.31	0.07–6.59	97	1.26	nd–42.3	100	0.63	0.03–2.27	36	–	nd–4.61	3	–	nd–3.66
BDE 153	100	1.00	0.17–8.76	97	1.94	nd–29.0	97	1.34	nd–6.56	62	0.43	nd–4.30	0	–	nd
BDE 154	100	0.26	0.05–4.31	100	0.99	0.20–44.8	97	0.54	nd–1.74	5	–	nd–6.51	0	–	nd
BDE 183	100	1.65	0.18–12.6	100	4.48	0.72–10.6	97	2.56	nd–7.99	38	–	nd–7.27	6	–	nd–2.99
BDE 209	100	33.8	3.71–513	100	34.0	6.46–109	100	133	46.2–426	92	10.1	nd–208	86	2.61	nd–23.5
$\Sigma$ PBDEs		45.5	4.82–522		70.0	19.2–179		147	49.8–449		17.5	nd–221		3.55	nd–26.45
PCB 28	100	31.3	3.59–124	100	207	32.4–479	100	52.9	14.0–296	88	21.5	nd–97.7	9	–	nd–6.75
PCB 52	100	14.9	1.64–61.6	100	76.2	8.83–288	100	30.4	6.80–125	86	23.0	nd–73.1	35	–	nd–7.86
PCB 95	100	12.9	1.25–59.0	100	83.6	13.1–291	100	17.6	7.63–50.8	83	12.0	nd–67.7	13	–	nd–2.33
PCB 101	100	13.8	1.75–59.0	100	103	17.0–374	97	17.1	n.d–110	86	18.3	nd–130	13	–	nd–3.34
PCB 105	100	8.22	1.80–49.2	100	55.5	6.14–199	100	11.3	1.63–78.0	86	14.2	nd–60.3	0	–	nd
PCB 118	100	16.9	2.79–83.7	100	96.1	12.8–383	100	21.0	3.35–150	95	11.7	nd–79.0	16	–	nd–2.83
PCB 138	100	13.2	1.99–34.7	100	36.0	8.65–219	100	14.1	2.58–82.6	88	7.31	nd–53.4	31	–	nd–0.74
PCB 153	100	11.0	0.14–48.4	100	34.2	5.59–167	100	10.6	1.77–65.2	90	8.37	nd–61.4	69	0.80	nd–3.58
PCB 180	100	4.39	0.71–11.2	100	7.67	1.88–31.5	100	3.95	0.38–15.2	81	2.33	nd–34.5	50	0.20	nd–2.34
$\Sigma$ PCBs		142	19.4–527		719	122–2240		178	51.3–650		125	nd–500		1.41	nd–24.1
TPHP							100	34.2	11.9–286	93	80.9	nd–437	86	28.9	nd–86.0
TBOEP							97	80.6	nd–341	90	6.81	nd–20.5	70	1.82	nd–24.7
TEHP							100	55.4	26.7–224	100	32.8	10.1–78.9	92	10.1	nd–57.5
EHDPP							100	15.6	6.44–96.7	98	53.9	nd–186	89	30.6	nd–165
TCEP							61	1.88	nd–51.6	71	5.61	nd–68.7	95	8.76	nd–45.2
TCIPP							100	67.2	12.6–747	100	12.7	0.50–193	97	13.1	nd–82.0
TDCIPP							100	31.4	7.48–166	67	205	nd–151	46	–	nd–127
TCrP							100	23.0	9.69–260	98	11.4	nd–134	92	1.30	nd–77.5
$\Sigma$ OPFRs								357	173–916		264	87.9–747		112	21.9–556

DF (%), detection frequency;  
nd: not detected.



**Fig. 1.** Regulations on e-waste recycling and the changes in levels of organic contaminants in hair from Longtang town, South China over a 10-year long biomonitoring. (PCBs including 9 congeners, i.e., PCB28, PCB52, PCB95, PCB101, PCB105, PCB118, PCB138, PCB153, and PCB180; Penta-BDEs including 3 congeners, i.e. BDE47, BDE99 and BDE100; Octa-BDEs including 3 congeners, i.e. BDE153, BDE154, and BDE183; Deca-BDE i.e. BDE209; and PFRs including 8 chemicals, i.e., triphenyl phosphate (TPHP), tris(2-butoxyethyl) phosphate (TBOEP), tris(2-ethylhexyl) phosphate (TEHP), 2-ethylhexyl diphenyl phosphate (EHDHP), tris(2-chloropropyl) phosphate (TCEP), tris(2-chloropropyl) phosphate (TCIPP), tris(1,3-dichloro-2-propyl) phosphate (TDCIPP), and tricresyl phosphate (TCrP)).

closed by the end of 2015. Meanwhile, infrastructure for environmental protection was promoted to clean up e-waste remains in 2015 (The People's Government of Qingyuan, 2015).

The levels of  $\Sigma_8$ OPFRs, which were analyzed solely in hair samples collected in 2015, 2016 and 2019, also decreased significantly ( $p < 0.05$ ), with median values of 357, 264 and 112 ng/g, respectively (Fig. 1e). The rigid management in 2015 are probably the main reason for the reduction in the levels of PBDEs and OPFRs in hair. Additionally, with the restrictions on PBDEs, the production and use of OPFRs as the alternatives are expected to increase yearly (van der Veen and de Boer, 2012; Wei et al., 2015). As thus, the potential risks of human exposure to OPFRs would also increase. Triphenyl phosphate (TPHP) and tris(2-chloropropyl) phosphate (TCIPP) are two predominant OPFR chemicals. In the present study, the ratios of TPHP/Deca-BDE (BDE 209) and TCIPP/Deca-BDE were calculated. The values for TPHP/Deca-BDE were 0.1–1.7 (median: 0.3), 0.1–9.5 (median: 0.8), and 2.1–42.6 (median: 10.9) in hair samples collected in 2015, 2016, and 2019, respectively; and those for TCIPP/Deca-BDE were 0.1–2.4 (median: 0.5), 0.2–4.1 (median: 0.6), and 2.1–48.7 (median: 5.0), respectively. A significantly increasing trend was observed for both the ratios from 2015 to 2019 ( $p < 0.01$ ; Fig. 1f–g). This was probably the additional effect of global restriction of PBDEs by the Stockholm Convention on

Persistent Organic Pollutants in 2009 and 2017, and OPFRs are used as the major alternatives (van der Veen and de Boer, 2012; Wei et al., 2015). This indicates a shift from “legacy” to “emerging” contaminants released from e-waste recycling in this area.

Moreover, the changes in the hair levels of these contaminants over the past decade were also a result of increasingly rigid solid waste import policies implemented by the Chinese government. In 2008, China made an announcement on the issuance of the catalogue of banned and restricted import solid waste. The catalogue was adjusted or new catalogues were issued successively in 2009, 2011, 2015, 2017, and 2018. From February to November 2013, Chinese customs launched the “Green Fence Operation”, an enhanced enforcement campaign to minimize illegal imports of waste. The newly revised “Law on the Prevention and Control of Environmental Pollution by Solid Wastes” in 2020 announced a complete ban of import of solid waste from January 1, 2021. Our field survey also revealed that the domestically generated e-waste has accounted for most of the e-waste dismantled in Longtang town in 2019.

#### 4. Conclusions and implications

In summary, our work on hair over the past decade (2009–2019) indicates significant declining trends in the levels of organic contaminants



such as PCBs, PBDEs, and OPFRs in hair of local residents and workers, which provides evidence for the effectiveness of the regulations on informal e-waste recycling activities and solid waste in China. Despite the significant progress that has made, there is still existence of primitive e-waste recycling facilities across the country and continue high exposure to toxic substances for some cohorts. In addition, with the strict enforcement of solid waste imports, domestic generation of e-waste will become the predominant source for recycling in China. It is very much possible that the additive chemicals in the domestic e-waste differs from those in the oversea waste, for example, due to the type of product, year of manufacture, and standard for additives. Therefore, a shift in the contaminants released from e-waste recycling as well as the associated environmental impacts and health consequences are expected. This warrants continuous efforts to monitor human exposure of e-waste recycling employees. Human hair would be a promising bio-monitoring matrix in future studies.

### CRediT authorship contribution statement

**Bin Tang:** Investigation, Formal analysis, Writing – original draft, Writing – review & editing, Visualization. **She-Jun Chen:** Investigation, Formal analysis, Writing – review & editing. **Jing Zheng:** Conceptualization, Writing – review & editing, Funding acquisition, Project administration. **Shi-Mao Xiong:** Methodology, Validation, Investigation, Writing – review & editing. **Xiao Yan:** Investigation, Writing – review & editing. **Wei-Keng Luo:** Investigation, Visualization, Writing – review & editing. **Bi-Xian Mai:** Funding acquisition, Resources, Writing – review & editing, Supervision. **Yun-Jiang Yu:** Resources, Writing – review & editing, Supervision.

### Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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### Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.scitotenv.2021.150411>.

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