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Habitat- and species-dependent accumulation of organohalogen pollutants in home-produced eggs from an electronic waste recycling site in South China: Levels, profiles, and human dietary exposure☆

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ABSTRACT

Organohalogen pollutants (OHPs) including chlorinated paraffins (CPs), polybrominated diphenyl ethers (PBDEs) and other halogenated flame retardants (OHFRs) (dechlorane plus (DP), decabromodiphenyl ethane (DBDPE), 1,2-bis(2,4,6-tribromophenoxy) ethane (BTBPE), hexabromobenzene (HBB), hexabromocyclododecanes (HBCDs) and tetrabromobisphenol A (TBBPA)) originating from an e-waste recycling area in Guiyu, southern China were investigated in chicken and goose eggs. As expected, OHP concentrations were higher in chicken eggs collected from the location (site 1) approaching the e-waste recycling center than from the location (site 2) far from the e-waste recycling center. Also, much higher OHP levels were observed in goose eggs foraging in residential area (site 2) than that in agricultural area (site 1), suggesting a clear habitat dependent OHP bioaccumulation pattern both concerning distance from e-waste activities and type of foraging habitat. Goose eggs exhibited higher short chain chlorinated paraffins (SCCPs) concentrations but lower PBDE and OHFR levels than chicken eggs. The proportion of high brominated PBDEs (hepta-to deca-BDEs) was lower in goose eggs than that in chicken eggs and showed a clear decrease from site 1 to site 2. DP isomeric composition f_{anti} values (the ratio of the *anti*-DP to the sum of the *anti*- and *syn*-DP) in goose eggs were significantly lower than those in chicken eggs ($p < 0.001$). These differences are likely a reflection of factors such as the species-specific differences in habitat preference and the differing environmental behaviors of the pollutants owing to their inherent properties (such as solubility and vapor pressure). Our findings suggested a high dietary intake of OHPs *via* home-produced eggs. For BDE99 there is a potential health concern with respect to the current dietary exposure *via* eggs.

1. Introduction

Organohalogen pollutants (OHPs) are generally brominated or chlorinated chemicals that are widely found in e-wastes (Robinson, 2009). Polybrominated diphenyl ethers (PBDEs) are typical brominated pollutants, and have been banned or restricted in some regions of the world (Betts, 2008). Other brominated pollutants, e.g., decabromodiphenyl ethane (DBDPE), tetrabromobisphenol A (TBBPA), hexabromocyclododecanes (HBCDs), hexabromobenzene (HBB), and 1,2-bis(2,4,6-tribromophenoxy) ethane (BTBPE), were used in plastics, textiles, electronic circuitry and some of them used as alternatives for the legacy OHPs, such as PBDEs (Covaci et al., 2011). Chlorinated paraffins (CPs) and dechlorane plus (DP) belong to the class of chlorinated pollutants and are extensively used in many types of polymers (Boer et al., 2010; Xian et al., 2011). Recently, toxicological research indicated that CPs are carcinogenic (Boer et al., 2010), and DP may induce apoptosis in the liver of juvenile Chinese sturgeon (Liang et al., 2014). These studies provide further concerns on the potential threat of CPs and DP to human health and ecosystems. Available data on these halogenated flame retardants (HFRs) indicated that these chemicals might also be persistent, bioaccumulative, and toxic (Covaci et al., 2011). So far, numerous studies have reported high concentrations of both legacy OHPs and newer-generation chemicals in various environmental

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media and biological samples at e-waste recycling sites (Ben et al., 2013; Zhang et al., 2011).

Guiyu, located in Guangdong Province, on the southern coast of China, is one of the largest e-waste recycling centers in the country. Nearly 80% of the families residing here have members involved in e-waste operations (Li et al., 2008). Primitive e-waste processes like manual separation, open burning, and strong acid digestion were adopted by the locals, which have caused severe OHP pollution in Guiyu (Gao et al., 2011; Labunska et al., 2013a). It is expected that most of these contaminants, once in the environment, can accumulate in living organisms (Covaci et al., 2011). The species-specific nature of bioaccumulation of OHPs has been reported in various field studies (Chen et al., 2010; Sun et al., 2014), with bioavailability and biotransformation being suggested as the likely reasons for this. The differing living and feeding habits of animals are also highlighted as important factors in determining the OHP profiles. However, little is known about the OHP compositional differences of biotas occupying different habitats. Moreover, dietary intake is a significant pathway of human exposure to OHPs, largely attributed to consumption of dairy products with a relatively high fat content. Thus, the possible adverse effects of OHPs from these hot spots (e-waste recycling sites) on local animals and residents have become particularly concerning.

Both chickens and geese are poultry, but their life habits are different. Chickens are essentially terrestrial fowls while geese are a kind of water fowl and often have open grazing access to both land and water bodies. Since eggs are a high-fat component of the daily human diet and are considered to be a good indicator of ambient persistent organic contaminant levels (Windal et al., 2009), chicken and goose eggs are a good option for investigating the bioaccumulation characteristics of OHPs in biotas in different habitats. Therefore, in the present study, home-produced chicken and goose eggs were collected from families living in the e-waste recycling areas in Guiyu. The primary aim of this study was to investigate the residual levels and compositional patterns of OHPs in chicken and goose eggs affected by e-waste recycling, in order to determine the possible differences in species living in different habitats around the e-waste disposal center and to estimate the daily dietary intake of OHPs *via* consumption of these eggs by the local residents.

2. Materials and methods

2.1. Sampling

A total of 40 samples, including 17 chicken eggs and 23 goose eggs, were collected from two sites (site 1, N 23°19′38″ E 116°21′39″; and site 2, N 23°19′4″ E 116°21′23″) in an e-waste recycling region in Guiyu in December 2013 (Fig. S1). Site 1 was located in an industrial park at a distance of about 2 km from the e-waste recycling workshops. Chickens at site 1 foraged around the residential areas, while geese at site 1 spent the daytime in local paddy field or watercourses and rested at a farmhouse during the night. Site 2 is located in at a distance of about 2.5 km from the e-waste recycling workshops. Both chickens and geese at site 2 foraged exclusively in residential areas (Fig. S2). Eggs freshly laid by the free-range poultry were collected from site 1 (8 chicken eggs and 9 goose eggs) and site 2 (12 chicken eggs and 11 goose eggs). The eggs were cleaned with deionized water and the egg contents were transferred to clean glass jars, which were then stored at −20 °C until chemical analysis.

2.2. Sample preparation

All the samples were freeze-dried and homogenized into powder, weighed (1.0 g) and spiked with the surrogate standards ($^{13}C_{10}$-*trans*-chlordane, PCB30, PCB65, PCB204, BDE77, BDE181, BDE205, $^{13}C_{12}$-BDE-209, $^{13}C_{12}$-labeled α-, β-, γ-HBCD and ^{13}C-labeled TBBPA) prior to extraction. The lipid content was determined gravimetrically on an aliquot of the extract (one tenth of the extract), while the rest of the extract was divided into two equal parts for further purification. One part was used for the determination of CPs, PBDEs, DBDPE, DP, BTBPE, and HBB. The other part was used for the determination of TBBPA and HBCDs. Detailed sample preparation procedures are described in the Support Information (SI).

2.3. Instrumental analysis

For the CP (C_{10}–C_{17} with Cl_5–Cl_{10}) analysis, an Agilent 6890 gas chromatograph (GC) connected to an Agilent 5975C mass spectrometer (MS) operated in electron capture negative ionization (ECNI) mode was equipped with a DB-5HT (15 m × 0.25 mm i.d., 0.10 μm film thickness) capillary column. The injector temperature was set to 250 °C, the transfer line temperature was 280 °C, and the ion source temperature was 200 °C. The oven temperature program is detailed in the SI. All monitored ions of short chain chlorinated paraffins (SCCPs) and medium chain chlorinated paraffins (MCCPs) were divided into four groups: 1) C_{10} and C_{11}, 2) C_{12} and C_{13}, 3) C_{14} and C_{15}, and 4) C_{16} and C_{17} to improve the instrument sensitivity. Therefore, for each sample, four individual injections were needed in order to analyze all the selected CP congeners. Congener groups were identified by the retention time range, the signal shape, and the correct isotope ratio. The quantification of SCCP (C_{10}–C_{13} with Cl_5–Cl_{10}) and MCCP (C_{14}–C_{17} with Cl_5–Cl_{10}) was conducted according to the CP carbon chain length and the degree of chlorination as described previously (Reth et al., 2005).

PBDEs, DBDPE, DP, BTBPE, and HBB were analyzed using a GC/MS (Agilent 6890N/5975C MSD; Agilent Technology, CA) in ENCI mode. Di-through hepta-BDEs, DP, and HBB were separated with a DB-XLB (30 m × 0.25 mm i.d., 0.25 μm film thickness) capillary column. For octa-through deca-BDEs, BTBPE, and DBDPE, a DB-5HT (15 m × 0.25 mm i.d., 0.10 μm film thickness) capillary column was used. The oven temperature programs are detailed in the SI.

For the analysis of TBBPA and HBCDs, an Agilent 1200 series liquid chromatography (LC) system coupled to an Agilent 6410 electrospray triple quadrupole mass spectrometer equipped with a XDB-C_{18} column (50 mm × 4.6 mm i.d., 1.8 μm, Agilent, CA) was used. Details of the analytical methodology used for separation and quantification of TBBPA and HBCDs have been published previously (Feng et al., 2012).

2.4. Quality assurance and quality control (QA/QC)

In order to avoid any interference and contamination, all glassware was rinsed with solvents (acetone, dichloromethane and hexane) and heated at 450 °C prior to use. Spiked samples were analyzed to validate the sample preparation method. During the sample analysis, each batch of 11 samples included one procedural blank. Trace amount of a few PBDEs were detected in the procedure blanks, but the levels were less than 1% of the analyzed concentration in the samples. The average recoveries of the spiked surrogate standards ranged from 63 to 98%, 72–117%, 50–108% and 66–107% for CPs ($^{13}C_{10}$-*trans*-chlordane), PBDEs (BDE77, BDE181, BDE205 and $^{13}C_{12}$-BDE-209), TBBPA (^{13}C- TBBPA) and HBCDs (^{13}C-α-, β-, γ-HBCD), respectively. Instrumental QC was performed by regular injection of solvent blanks and standard solutions. The method detection limit (MDLs) was defined as a signal to noise (S/N) ratio of 10. The MDLs were in the range of 0.2–1.8 ng/g lipid weight (lw) for CPs, 0.2–0.4 ng/g lw for PBDEs and OHFRs, and 0.7–1.7 ng/g lw for HBCDs and TBBPA.

3. Results and discussion

3.1. Inter-site and inter-species differences in OHP levels

Descriptive statistics for the levels of CPs (only SCCPs detected), PBDEs, and OHFR (including DBDPE, TBBPA, HBCDs, DP, BTBPE, and HBB) in home-produced eggs from the e-waste recycling area (Guiyu) are summarized in Table 1.

As presented in Table 1, SCCPs (C_{10}–C_{13}), PBDEs, DP, TBBPA, HBCDs, DBDPE, BTBPE, and HBB were found in all analyzed chicken eggs, with their concentrations ranging from 2300 to 6800, 620 to 46,000, 46 to 2200, 1.6 to 260, 56 to 7600, 4.5 to 190, 0.4 to 78, 2.0 to 6.7 ng/g lw, respectively. Generally, the ranges of OHP concentrations reported here for chicken eggs were one to two orders of magnitude higher than those observed in chicken eggs from the other sites (Covaci et al., 2009; Hu et al., 2011; Labunska et al., 2014, 2015); however, they were comparable to those observed in chicken eggs collected from Qingyuan (an e-waste recycling region in South China) (Zheng et al., 2012) (Table S1). The OHP (except for SCCPs, TBBPA and HBB) levels in chicken eggs from site 2 were significantly lower than those from site 1 (One-way ANOVA: $p < 0.05$), an observation which was *a priori* expected, due to the greater distance between the farm and the e-waste recycling center (Fig. S1). Compared to SCCPs, TBBPA and HBB, the other OHPs (except for HBCDs) are less volatile due to their lower vapor pressure (Vapor pressure of subcooled liquid (P_L/Pa): $10^{-7} - 10^{-2.29}$ for SCCPs, TBBPA and HBB, $10^{-14.5} - 10^{-7.03}$ for DP, BTBPE, DBDPE, and nona-to deca-BDE (the dominant PBDE congeners in chicken eggs)) (Drouillard et al., 1998; Kuramochi et al., 2008; Wang et al., 2008), which indicated a superior atmospheric migration ability of SCCPs, TBBPA, and HBB than that of the other OHPs. In the present study, no obvious decrease of the concentrations of SCCPs, TBBPA, and HBB were seen, while the clear decrease of the concentrations of the other OHPs from site 1 to site 2 may be due to the high migration abilities of SCCPs, TBBPA, and HBB compared to the other OHPs. Therefore, the differing compositional patterns of SCCPs, TBBPA, and HBB in chicken eggs from the two sites indicated that the environmental distribution pattern of these pollutants was potentially influenced by their physicochemical properties.

SCCPs, PBDEs, DP, TBBPA, HBCDs, DBDPE, BTBPE and HBB were frequently detected (>50% detection frequency, except for BTBPE (20% detection frequency)) in the goose eggs, with concentration ranges of nd–150,000, 230–7500, 14–450, 3.5–890, nd–110, nd–11, nd–14, and 1.8–17 ng/g lw, respectively (Table 1). Comparison with previous studies reveals that the OHP concentrations obtained in our study were at the high end of values previously reported for OHPs in seabird eggs from Spain, Norway, and the Great Lakes basin (Gauthier et al., 2007; Huber et al., 2015; Muñoz-Arnanz et al., 2011). In some cases, concentrations of OHPs in goose eggs in the present study exceed those found in duck eggs from Taizhou, Zhejiang, another major e-waste recycling site in China (Labunska et al., 2013b, 2015) (Table S1). Unexpectedly, the OHP levels (except for SCCPs and TBBPA) for goose eggs from site 2 were generally higher than those from site 1, a phenomenon differing from that observed for chicken eggs, as discussed above. Geese from site 1 spent the daytime in local paddy fields or watercourses and returned to the residential areas only during the night, while geese in site 2 foraged exclusively in the residential areas. Studies have shown that higher concentrations of PBDEs are distributed in the residential areas of Guiyu (Zhang et al., 2014). This may explain why PBDE levels in goose eggs from site 2 are higher than those from site 1. Although no data available for the other OHPs in residential and agricultural soils from e-waste polluted region in Guiyu, it is believed that higher concentrations of the other OHPs are distributed in the residential areas, as most recycling workshop are located in the residential areas of Guiyu. Moreover, the OHP distribution characteristics in the goose eggs from the two sites suggested that the habitat of animals is an important factor in determining the OHP profiles.

Inter-species differences in OHP contributions for eggs were observed. As described in Fig. 1, SCCP was the predominant compounds in goose eggs (averages of 97.8% and 64.5% for site 1 and site 2, respectively, % of total OHP concentrations). However, PBDEs seems to a more dominant compounds in chicken eggs, as the contributions of PBDEs in chicken eggs (averages of 70.1% and 42.6% for site 1 and site 2, respectively) were much higher than those in goose eggs (averages of 2.0% and 29.9% for site 1 and site 2, respectively) from the same site. Further, the contributions of OHFR in chicken eggs (averages of 17.2% and 8.7% for site 1 and site 2, respectively) were higher than those (averages of 0.2% and 5.6% for site 1 and site 2, respectively) in goose eggs too. Additionally, larger standard deviations were obtained for goose eggs than those for chicken eggs (Table 1). These differences are likely to be a reflection of the species-specific differences in habitat preference (geese spend a substantial portion of time in local watercourses whereas chickens are exclusively terrestrial). Moreover, large standard deviations is a consequence of very high levels in few eggs.

Table 1
Average and median values, and concentrations ranges of OHPs (in ng/g lw) in home-produced eggs from an e-waste recycling site, South China.

		Chicken egg		Goose egg	
		Site 1	Site 2	Site 1	Site 2
n[a]		8	9	12	11
Lipid (%)	Average	10.3	10.1	14.4	14.2
	Median	10.6	10.0	14.2	13.9
	Range	7.9–12.2	6.9–13.6	12.7–17.1	10.5–17.4
SCCPs	Average	3400	4000	60,000	4600
	Median	3100	3400	nd[b]	1700
	Range	2300–5500	2600–6800	nd–150,000	nd–11,000
PBDEs[c]	Average	14,000	3500	1200	2100
	Median	7700	3100	400	1700
	Range	1400–46,000	620–5500	230–5900	470–7500
DP[d]	Average	450	100	31	320
	Median	120	75	21	78
	Range	30–2200	46–165	14–123	30–450
TBBPA	Average	46	420	85	27
	Median	20	27	6	18
	Range	1.6–180	1.6–260	3.5–890	3.5–110
HBCDs	Average	1900	190	2.1	44
	Median	1200	170	1.3	2.0
	Range	90–7600	56–300	nd–5.8	nd–110
DBDPE	Average	78	9.5	2.3	3.8
	Median	33	8.4	1.9	3.4
	Range	10–190	4.5–13	nd–11	nd–8.6
BTBPE	Average	24	1.1	0.5	1.7
	Median	15	0.6	nd	nd
	Range	1.1–78	0.4–2.1	nd–0.5	nd–14
HBB	Average	3.6	3.7	2.9	3.9
	Median	3.4	3.3	2.1	2.5
	Range	2.3–6.7	2.0–6.8	1.8–7.9	1.8–17

[a] Number of samples.
[b] Not detected.
[c] Sum of BDE 28, 47, 66, 85, 100, 99, 138, 154, 153, 183, 197, 203, 196, 208, 207, 206 and 209.
[d] Sum of *anti*- and *syn*-DP.

3.2. Inter-species differences in OHP composition profiles

The SCCP profiles in egg samples from the investigated sites seemed to resemble each other, irrespective of SCCP levels. Both C_{12}-CPs and C_{13}-CPs are predominant in all samples. The average proportions of summed C_{12}-CPs and C_{13}-CPs were 67.1 ± 1.4% (site 1) and 79.7 ± 6.4% (site 2) in the chicken eggs, and 66.6 ± 2.8% (site

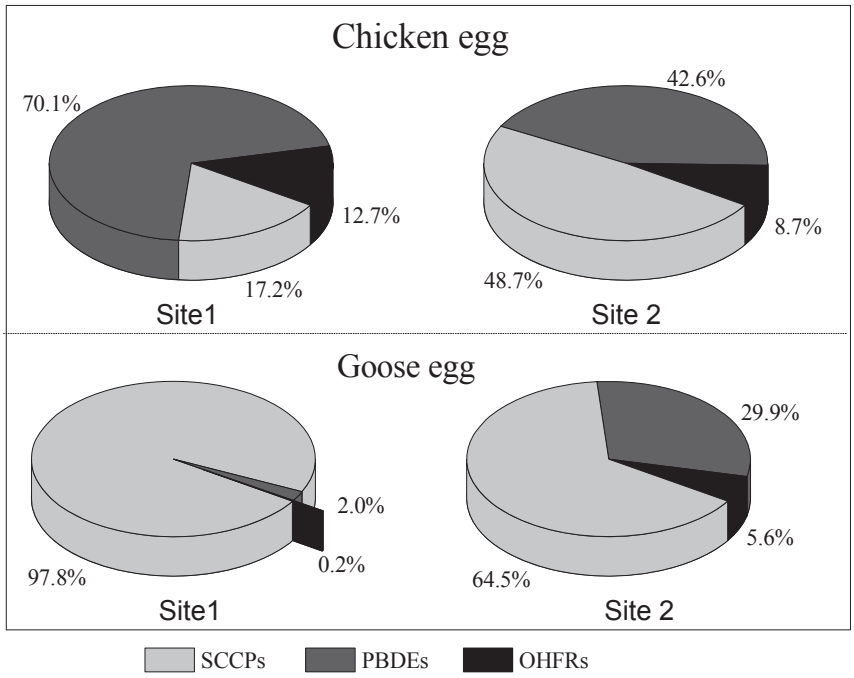

Fig. 1. Relative contributions of CPs, PBDEs, and OHFRs (including DP, TBBPA, HBCDs, DBDPE, BTBPE, and HBB) to total OHPs in home-produced eggs from an e-waste recycling site, South China.

1) and 70.1 ± 3.5% (site 2) in the goose eggs, which could be related to the fact that congeners with more carbon atoms are more prone to bioaccumulation due to their higher K_{OW}. The GC-NCI-MS chromatogram of SCCPs in eggs of the present study was similar to that of a standard solution (Fig. S3), and this profile of SCCPs were also found in gull eggs from the Ebro delta Natural Park (Morales et al., 2012). Since no SCCP profile data are available on the poultry eggs, no comparison could be drawn in the current study.

As for PBDEs, although BDE209 was the most abundant PBDE congener, accounting for 50.4% (site 1) and 66.2% (site 2) in the chicken eggs, and 44.3% (site 1) and 38.0% (site 2) in the goose eggs, the proportion of high brominated PBDEs (hepta-deca-BDEs) was lower in goose eggs than that in chicken eggs and showed a clear decrease from site 1 to site 2 (Fig. S4). Therefore, principal component analysis (PCA) was conducted to compare the PBDE congener profiles between the two species as well as the two sampling sites (site 2 is located further away from the e-waste recycling workshop than site 1). As shown in Fig. 2a, the principal component 1 (PC 1) explained 54% of the total variance and was characterized by high loading of high brominated PBDE congeners (hexa-to deca-BDE). Principal component 2 (PC 2) accounted for 30% of the total variance and was highly associated with lower brominated PBDE congeners (tri-to penta-BDE).

Overall, the factor loadings in the current study indicated clear inter-site and inter-species differences in the PBDE profiles. On the one hand, egg samples from site 1 and site 2 could be discriminated by factor 2 (Fig. 2b). All samples from site 1 had negative factor 2 scores, which resulted from the relatively high content of high brominated congeners (hexa-to deca-BDEs), while samples from site 2 generally had positive factor 2 scores, corresponding to a high proportion of low brominated congeners (tri-to penta-BDEs). Studies have suggested that heavier BDEs are readily deposited in the region of their emission whereas lighter PBDEs are transported over longer distances (Jiang et al., 2012). This may explain why egg samples from site 2 were characterized by lower brominated congeners and samples from site 1 by higher brominated congeners. On the other hand, although the distribution patterns of chicken and goose eggs from the two sites were asymmetric, chicken and goose eggs from the same site could be differentiated by factor 1. This obvious partition in PBDE profiles between chicken and goose eggs indicated different pollution patterns between the aquatic and terrestrial samples, likely due to the species-specific differences in habitat preferences.

The *anti*-DP fraction (f_{anti}), defined as the ratio of the *anti*-DP concentration to the sum of the *anti*- and *syn*-DP concentrations, was calculated to evaluate the differences in DP congener profiles. The f_{anti} values in chicken eggs from site 1 and site 2 were 0.74 ± 0.06 and 0.76 ± 0.02, respectively (Fig. 3). The values were comparable to the ratios reported in chicken eggs from other e-waste sites (average values ranged from 0.63 to 0.75) (Zheng et al., 2012) and ratios in terrestrial bird eggs from Spain (which ranged between 0.62 and 0.75) (Guerra et al., 2011). The average f_{anti} value in goose eggs from site 1 and site 2 was 0.64 ± 0.04, which was similar to that calculated for white stork (*Ciconia ciconia*) (an aquatic bird) eggs from the Madrid (0.64 ± 0.07) and Doñana National Park (0.66 ± 0.12) (Muñoz-Arnanz et al., 2011), and herring gull (*Larus argentatus*) (an aquatic bird) eggs from the Laurentian Great Lakes (0.69 ± 0.08) (Gauthier and Letcher, 2009). However, f_{anti} values in goose eggs were significantly lower than those in chicken eggs in the current study ($p < 0.001$), which indicates a higher proportion of *anti*-DP in chicken eggs than in goose eggs. These differences could be due to the differing behavior of DP isomers, which is influenced by their intrinsic properties (such as solubility). Though no clear data for the physical properties of *anti*-DP and *syn*-DP has been reported until now, according to their retention times on the capillary columns, *anti*-DP was supposed to be more lipophilic and less water-soluble than *syn*-DP. Therefore, f_{anti} isomer ratios in goose eggs were expected to be lower than those in chicken eggs. A one-way ANOVA revealed that no significant inter-site differences were found in f_{anti} values ($p > 0.05$). To investigate the differences between the two locations in more detail, more samples are needed.

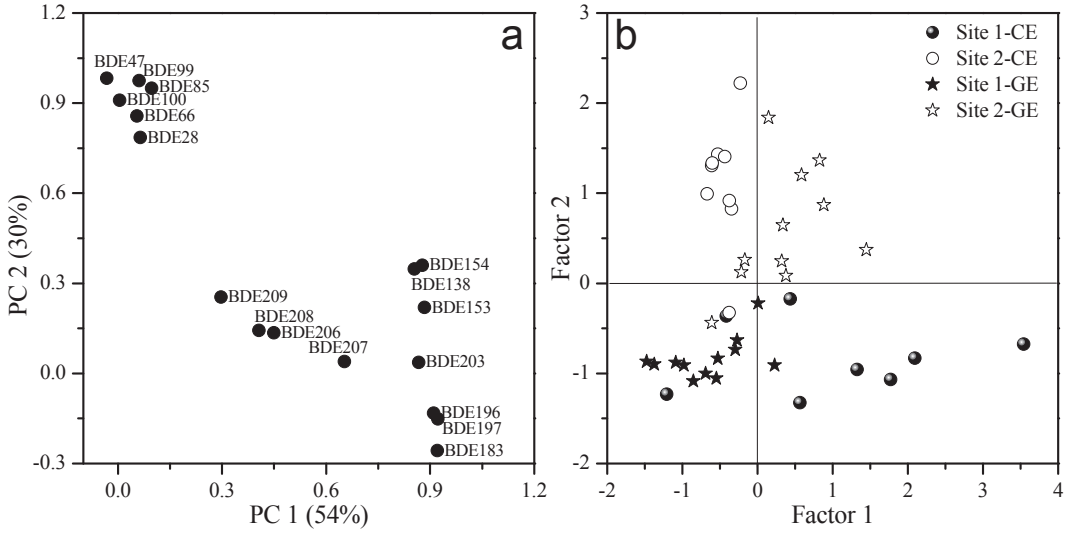

Fig. 2. Principal component analysis results based on the log 10 transformed concentrations of PBDE congeners (PC1, 54% variance; PC2, 30% variance) in home-produced eggs (CE: chicken egg, GE: goose egg) from two sampling sites (Site 2 is located further away from the e-waste recycling workshop than site 1) situated near an e-waste recycling center, South China. The figure legends represent the factor loadings (a) and factor scores (b).

In the current study, α-HBCD was the predominant diastereoisomer among the HBCDs, contributing almost 100% to ΣHBCD concentrations for eggs, and was detected in 100% and 78% of the chicken and goose eggs, respectively. The β- and γ-HBCD were only detectable (>50% detection frequency) in chicken eggs, with average contributions of 0.15% and 0.09%, respectively. The predominance of α-HBCD in terrestrial and aquatic bird eggs was also reported in home-produced chicken eggs from another e-waste site in South China (Zheng et al., 2012), in chicken eggs from the Hubei province of China (Hu et al., 2011), in Swedish peregrine falcon (*Falco peregrinus*) eggs (Janák et al., 2008), and herring gull eggs from the Laurentian Great Lakes (Gauthier et al., 2007). Moreover, the chiral signatures of α-HBCD, expressed as (+)-α-HBCD divided by Σα-HBCD (EFs), were calculated for chicken eggs, but no EF values were obtained for goose eggs due to their limited levels. All chicken eggs showed enrichment of (−)-HBCD, with calculated EFs of 0.43 ± 0.10 and 0.34 ± 0.13 for site 1 and site 2, respectively. The preferential enrichment of the (−)-HBCD was also found in chicken eggs from another e-waste site in South China (Zheng et al., 2012) and in herring gull eggs from Germany (Esslinger et al., 2011).

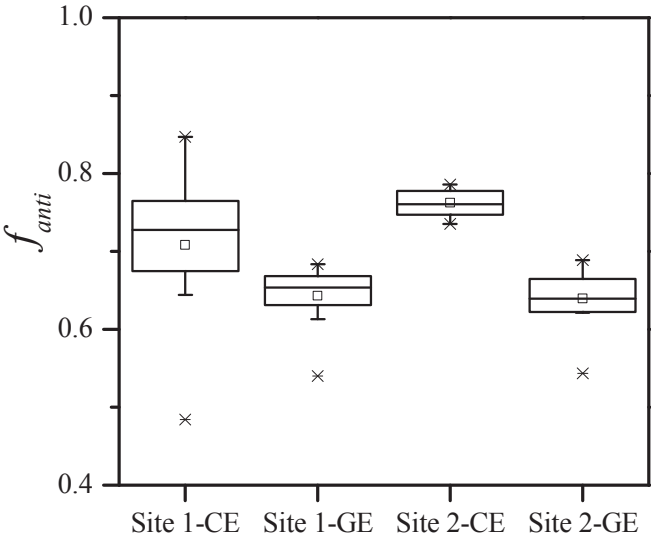

Fig. 3. Boxplot of the f_{anti} values (the ratio of the *anti*-DP concentration to the sum of the *anti*- and *syn*-DP concentrations) in home-produced eggs (CE: chicken egg, GE: goose egg) from an e-waste recycling site, South China.

3.3. Estimation of daily OHP intake of local residents

In the present study, we estimated the dietary OHP intake of local residents *via* egg consumption (including chicken and goose eggs). Daily OHP intake was calculated by multiplying average food consumption rates by the OHP levels (ng/g wet wt.) detected in the egg samples. Food consumption rate assumed for chicken egg considered in our study was drawn from the published study, that reports an average daily consumption rate for eggs (type unspecified) of 22.9 g/day in Guiyu, China (Chan et al., 2013). However, official figures on daily consumption of goose eggs in China, and in the Guiyu area in particular, are not available. Information from goose farm owners during our sampling program indicated that an average of one goose egg per week for a person. On the basis of the average goose egg weight in our study (160.6 g, n = 23), the average goose egg exposure values is 22.9 g/day. It will underestimate potential health consequences for people who consume both chicken eggs and goose egg in the study areas when we only report the EDI values of OHPs *via* chicken eggs. However, it will overestimate potential health consequences for people who consume only chicken eggs or goose eggs when we report the total EDI values of OHPs *via* goose eggs and chicken eggs. In this case, we calculated EDI of OHPs from chicken egg and goose egg consumption separately and EDI values for OHPs *via* chicken egg and goose egg consumption were used to compare with the reference values, respectively.

The estimated daily intake (EDI) values of SCCPs, PBDEs, and OHFRs *via* chicken eggs from the e-waste recycling sites were in the ranges 5900–18,700, 1900–102,000, and 540–19,600 ng/day, respectively. The EDI values of SCCPs, PBDEs and OHFRs *via* goose eggs were in the ranges nd-450000, 900–29,000, and 80–6300 ng/day, respectively. Most previous studies on the EDI values of OHPs *via* eggs have been primarily focused on PBDEs, with no information for SCCP and sparse information for OHFRs so far. The average EDI value of PBDEs *via* chicken eggs in our study (mean of

Table 2
Overview of estimated intake and margin of exposure for PBDEs *via* consumption of home-produced eggs from an e-waste recycling site in Guiyu, South China.

		Estimated daily intake (EDI, ng/kg bw/day)[a]								Margin of exposure (MOE)[b]			
		BDE47		BDE99		BDE209		ΣPBDEs		BDE47		BDE99	
		Adult[c]	Child[d]	Adult[c]	Child[d]	Adult[c]	Child[d]	Adult[c]	Child[d]	Adult	Child	Adult	Child
RfD		100		100		7000		na[e]					
$D_{r,h}$[f]										172		4.2	
Chicken egg	LB[g]	1.8	7.7	4.1	18	23	99	235	55	97	22	1.0	0.2
	UB[h]	12.5	54	13	58	907	3900	6943	1614	14	3.2	0.3	0.1
Goose egg	LB[g]	2.2	9.3	1.7	7.4	248	1067	1192	14	80	19	2.4	0.6
	UB[h]	19.2	82	18	79	245	1053	1988	462	9.0	2.1	0.2	0.1

[a] EDI = (C_{egg} × consumption of 22.9 g/day)/human body weight, C_{egg} is the ΣPBDE or individual BDE congener concentration in eggs, ng/g, wet weight.
[b] MOE = $D_{r,h}$ (ng/kg b.w./day)/EDI(ng/kg b.w./day).
[c] Based on an average adult weight of 63 kg.
[d] Based on an average child weight of 14.65 kg.
[e] na: not applicable.
[f] Reference values promulgated by the EFSA Panel on Contaminants in the Food Chain., Efsa journal 2011,9(5):2156.
[g] LB: Lower bound, based on the lowest PBDE concentration in eggs.
[h] UB: Upper bound, based on the highest PBDE concentration in eggs.

19,000 ng/day) was comparable to that (mean range of 4200–20,000 ng/day) reported in chicken eggs from another e-waste recycling site in southern China (Zheng et al., 2012), but was one to two orders of magnitude higher than the values estimated in foodstuffs widely consumed by the population of Catalonia, Northeast Spain (75.4 ng/day) (Domingo et al., 2008), Belgium (38.5 ng/day) (Gomara et al., 2006), and Sweden (37–64.9 ng/day) (Darnerud et al., 2006). For OHFRs, the EDI values *via* home-produced chicken eggs in the current study (median of 4200 ng/day) lay within the range of the mean values (970–4530 ng/day) of chicken eggs from another e-waste site in southern China. Moreover, the EDI values of PBDEs *via* goose eggs from the e-waste sites in the present study (mean of 5500 ng/day) were slightly higher than those (mean range of 159–5124 ng/day) reported in duck eggs from another e-waste recycling site in eastern China (Labunska et al., 2013b).

Concerning the available toxicity data, only the EDI values of PBDEs in the egg samples were normalized to body weight for further discussion. Normalization to body weight for adults was based on an average body weight of 63 kg (Zhou et al., 2012), and for children on an average body weight of 14.65 kg (China, 2012). By comparison of the calculated human dietary intake associated with the body burden at the chronic human daily dietary intake ($D_{r,h}$) with the EDIs, the margin of exposure (MOE) was calculated, where MOE = $D_{r,h}$ (amount/kg b.w./day)/EDI (amount/kg b.w./day) (EFSA Panel on Contaminants in the Food Chain., 2011). The calculated MOE provides a more appropriate dose metric for a direct comparison of effects in animals and humans as it represents the internal body burden of a toxicant. The CONTAM Panel concluded that a MOE larger than 2.5 might indicate that there is no health concern (EFSA Panel on Contaminants in the Food Chain., 2011). As indicated in Table 2, our estimates of exposure to individual key PBDE congeners (BDE47, BDE99 and BDE209) for adults and children did not exceed the reference dose (RfD) values promulgated by the U.S. Environmental Protection Agency (100 ng/kg bw/day, 100 ng/kg bw/day, 7000 ng/kg bw/day, respectively) (EPA, 2008a, 2008b, 2008c). However, the MOE for BDE47 *via* consumption of goose eggs for child with a high consumption (UB) is 2.1, and the MOEs for BDE99 *via* consumption of eggs for adults and child, are in the range of 0.2–2.4 and 0.1–0.6, respectively (Table 2). These MOEs are smaller than 2.5, and thus indicate a potential health concern. This is of great concern, as PBDEs was reported to have adverse impacts on child neurobehavioral development (Eskenazi et al., 2013). Furthermore, our exposure estimate is only based on consumption of chicken eggs or goose eggs, and local residents of our study area will receive additional dietary and non-dietary exposure. These results highlight the potential for adverse human health impacts arising from exposure to PBDEs at e-waste recycling sites in Guiyu.

In conclusion, the present study has shown that elevated levels of OHPs were found in home-produced eggs from an electronic waste recycling site in southern China. Inter-site and inter-species differences in OHPs levels and profiles, especially for PBDEs, were found in the egg samples, which are likely due to various factors including species-specific differences in habitat preference, accessibility of domestic fowl to contaminated locations, and the proportion of time spent foraging in the contaminated areas. Additionally, our results indicated that SCCPs are more prone to distribution through water than PBDEs due to their relatively higher hydrophilic properties. High EDI values of OHPs *via* home-produced eggs highlight the potential for adverse human health impacts, especially adverse impacts on child neurobehavioral development due to the high PBDE exposure.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.envpol.2016.05.039.

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