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Polychlorinated biphenyls and chlorinated paraffins in home-produced eggs from an e-waste polluted area in South China: Occurrence and human dietary exposure

Yanhong Zeng[a], Chenchen Huang[a,b], Xiaojun Luo[a,*], Yine Liu[a,b], Zihe Ren[a,b], Bixian Mai[a]

[a] State Key Laboratory of Organic Geochemistry and Guangdong Key Laboratory of Environmental Protection and Resources Utilization, Guangzhou Institute of Geochemistry, Chinese Academy of Sciences, Guangzhou 510640, China
[b] University of Chinese Academy of Sciences, Beijing 100049, China

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ABSTRACT

The levels of polychlorinated biphenyls (PCBs) and short/median-chain chlorinated paraffins (S/MCCPs) in 68 home-produced eggs collected in 2013 and 2016 from an electronic-waste (e-waste) site in South China were measured and the human dietary exposure to these two classes of contaminants via egg consumption was calculated. The levels of PCBs, SCCPs, and MCCPs varied from 236 to 8870 ng/g lipid weight (lw), 477 to 111,000 ng/g lw, and 125 to 91,100 ng/g lw, respectively. There are no significant differences in the levels of PCBs, SCCPs, and MCCPs between 2013 and 2016 ($p > 0.05$). The congener profiles of PCBs and MCCPs were similar to each other between 2013 and 2016; however, the homologue profiles of SCCPs were different. The Toxic Equivalent Quantities (TEQs) of ΣDL-PCBs and the levels of ΣICES-6 PCBs strongly exceeded the limits set by EU Regulation 1259/2011 (2.5 pg World Health Organization-TEQ_{2005} g^{-1} lw for DL-ΣPCBs and 40 ng/ g lw for ΣICES-6 PCBs). The estimated daily intakes (EDI) of PCBs, SCCPs, and MCCPs by adults and children ranged between 5.57 and 1100, 11.8 and 11,900, and 3.62 and 11,400 ng/kg bw/d, respectively. PCBs pose serious health risks for local residents, especially for children, due to the high ratios of EDI (68% in 2013 and 70% in 2016 for adults and 100% for children) in excess of the exposure limits.

1. Introduction

Polychlorinated biphenyls (PCBs) are among the most well-known and studied families of persistent organic pollutants (POPs) with different numbers of chlorine atoms (1 − 10) attached to their biphenyls rings. Mixtures of PCB congeners were widely used as coolants in transformer oil, dielectric fluids, and lubricants from the 1930s to the 1970s (Passatore et al., 2014). Although most governments banned PCB production in the late 1970s, PCBs still represent a global problem due to their persistence and bioaccumulative potential in the environment. Given the widespread exposure to PCBs, the potential adverse health effects are relevant. These compounds are associated with neurological, reproductive, endocrine, and cutaneous diseases (Passatore et al., 2014). Two classes of PCBs have been classified by their toxicological properties, dioxin-like PCBs (DL-PCBs) and six indicator PCBs (ICES-6 PCBs). According to the European Commission regulations, the maximum tolerable levels (MLs) for DL-PCBs and ICES-6 PCBs are 2.5 pg WHO-TEQ/g fat and 40 ng/g fat, respectively (EU, 2011). These levels provide a tool to identify the degree of PCB pollution and take measures for the reduction or elimination of PCBs.

Chlorinated paraffins (CPs) are highly complex technical mixtures of alkanes that can be classified into three categories based on the carbon chain length: short-chain CPs (C_{10}–C_{13}, SCCPs), median-chain CPs (C_{14}–C_{17}, MCCPs), and long-chain CPs (C_{18}–C_{30}, LCCPs) (Houde et al., 2008). CPs have been widely used as additives, secondary plasticizers, and flame retardants in industrial applications for several decades (Tomy et al., 1998). Owing to the high demand from the plastics industry and the regulation of PCBs and other flame retardants, the global production of CPs increased sharply, with a cumulative production of > 7 million tons since the 1930s, which is much higher than that of PCBs (the cumulative global production volume of ~1.3 million tons) (van Mourik et al., 2016). As high-production-volume chemicals, CPs have been detected in various environmental matrices in the past decade (Castells et al., 2008; Harada et al., 2011; Hilger et al., 2013; Chen et al., 2014, 2014; Zeng et al., 2017). Among the CPs, SCCPs have attracted the most concern due to their highest toxicity, bioaccumulative potential, and long-range transport potential and had, therefore, been listed as a group of POPs in the Stockholm

* Corresponding author.
E-mail address: luoxiaoj@gig.ac.cn (X. Luo).

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Convention in May 2017 (Stockholm Convention, 2017). However, compared to other halogenated organics, the knowledge of the fate and levels of CPs is still limited.

Electronic waste (e-waste) is an important source of PCBs and CPs because of the use of primitive e-waste treatment techniques (Zhang et al., 2012; van Mourik et al., 2016). Rudimentary e-waste recycling activities can result in severe PCB and CP contamination and their hydrophobicity and lipophilicity lead to their bioaccumulation in fatty tissues of animals (Luo et al., 2015; Squadrone et al., 2015; Shen et al., 2017; Sun et al., 2017). Consequently, these pollutants enter the food chain. Food is the main route of exposure to POPs, including PCBs and S/MCCPs, for the general population (Xing et al., 2009; Fridén et al., 2011; Song et al., 2011). Labunska et al. (2013, 2014, 2015) showed that dietary intake of PBDEs, PCBs, HBCDs, and "novel" brominated flame retardants in e-waste-impacted foods was an order of magnitude higher than the intake of those chemicals from the control sites. However, data on human dietary exposure to PCBs and CPs associated with e-waste recycling are still limited, or in the case of M/LCCPs, are almost non-existent.

Longtang is one of the largest e-waste recycling centers in Qingyuan, Guangdong Province (Zhang et al., 2012) and high levels of hazardous chemicals were detected in the local environmental matrix (Chen et al., 2011; Zhang et al., 2013; Chen et al., 2014, 2014; Xiao et al., 2014). In order to mitigate e-waste contamination, laws and regulations have been imposed by the Qingyuan municipal government since 2010 (South Reviews., 2018). The consequences of the implementation of the legislation and the current local environmental contamination status are extremely important to estimate the environmental risks of e-waste. Free-range eggs are considered a suitable indicator of ambient POP levels owing to their high fat content and the intensive contact of hens with the environment (Van Overmeire et al., 2009; Windal et al., 2009). Therefore, the aim of this study was to investigate the levels of PCBs and CPs (only SCCPs and MCCPs) in free-range eggs from Longtang and the human dietary exposure to PCBs and S/MCCPs through the consumption of these eggs. The presence of MCCPs in eggs is of interest because of their increasing annual production in China and potential biomagnification in the food chain (Herzke et al., 2013; van Mourik et al., 2016).

2. Materials and methods

2.1. Sampling

The study area (N:23.60°, E:113.03°) was located in a village in Longtang Town, Qingyuan County, Guangdong Province (Fig. S1). This area was once a typical e-waste recycling site with dozens of homes, and homes in there have relatively intensive distribution (Fig. S1). Before the regulations, the e-waste were usually dismantled in the family backyard. However, since 2010, laws and regulations have been passed to ban the e-waste disposal in Qingyuan, and almost the family-run e-waste workshops were transferred to the local industry park. Therefore, in the present study, in order to assess the implementation of the regulations, we focused to study the levels of the dominant pollutants (PCBs and CPs) and their level changes in local free-range eggs about three and six years after the laws enacted by Qingyuan government since 2010. During the two sampling periods (2013 and 2016), seven families were chosen as the target families for egg collection, which located in different parts of the study area. About five fresh eggs were collected from each target families, while at the second sampling period, eggs were only collected from six families, as no hens was raised in one target family. All the eggs were laid by free-range hens, and these hens were generally feed by local produced grain and hunted for food around the farmhouse. Finally, a total of 68 home-produced eggs were collected in 2013 ($n = 38$) and 2016 ($n = 30$). After sampling, the home-produced eggs were washed with deionized water and transported to our laboratory. The egg content was transferred to clean glass jars and stored at −20 °C until further analysis.

2.2. Sample preparation and analysis

Samples were extracted and cleaned up using previously published methods with minor modifications. The purification of PCBs were similar to those reported in Gao et al. (2009) and the clean-up methods of CPs were similar to those reported in Chen et al. (2011) and Sun et al. (2017). Briefly, approximately 2 g (dry weight) of homogenized samples were extracted using the Soxhlet method with 200 mL hexane/dichloromethane (1:1, v/v) for 48 h, after being spiked with surrogate standards for PCBs (PCB 30, PCB 65, and PCB204) and an internal standard for CPs (ε-HCH). An aliquot of the extract was used to determine the lipid content (gravimetrical method) and the rest of the extract was used for the analysis of PCBs and CPs. The concentrated H_2SO_4 (AR) and the multilayer florisil/silica gel column (i.d. = 1.0 cm), which consists of florisil, neutral silica, and sulfuric acid silica, were used to remove the lipids and other substances from the samples. The column was eluted with 55 mL hexane collected for PCBs and 50 mL dichloromethane collected for CPs in sequence. Then, two parts of the eluent were concentrated to near dryness under a gentle stream of nitrogen and reconstituted in 300 μL isooctane for PCBs and CPs. Prior to the instrumental analysis, the first part was spiked with known amounts of internal standards (PCB 24, PCB 82, PCB198) for PCBs detection and the second part was spiked with known amounts of the surrogate standards ($^{13}C_{10}$-trans-chlordane) for CPs detection. All of the standard substances (PCB 30, PCB 65, PCB 204, PCB 24, PCB 82, PCB 198, ε-HCH and $^{13}C_{10}$-trans-chlordane) were purchased from AccuStandard, America.

A total of 159 individual PCB congeners were analyzed in this study and the details of the congeners are provided in the supplementary information (SI). The quantitative analysis of PCBs was performed using an Agilent 7890A gas chromatograph (GC) with an Agilent 5975C mass spectrometer (MS) in electron impact (EI) ionization mode. For the separation of individual PCB congeners, a DB-5 MS column (60 m × 0.25 mm i.d. × 0.25 μm film thickness) was used. Details of the instrumental parameters are provided elsewhere (Hu et al., 2008).

SCCPs and MCCPs were analyzed using an Agilent 6890 N gas chromatograph (GC) coupled with an Agilent 5975C mass spectrometer (MS), operated in electron capture negative ionization (ECNI) and selective ion-monitoring (SIM) modes. A DB-5HT capillary column (15 m × 0.25 mm i.d., 0.10 μm film thickness; J&W Scientific, CA) was used to separate the structural isomers of CPs. The injector temperature was set to 250 °C, the transfer line temperature was 280 °C, and the ion source temperature was set to 200 °C. The oven temperature programs was held at 80 °C (held for 3 min) firstly, then increased to 160 °C at 25 °C/min (held for 6 min), finally increased at 20 °C/min to 280 °C (held for 15 min), as described by Chen et al. (2011). To enhance the instrument sensitivity, all monitored ions of SCCPs and MCCPs were divided into four groups: C_{10} and C_{15}, C_{11} and C_{16}, C_{12} and C_{17}, and C_{13} and C_{14}. Therefore, four individual injections were needed to analyze all the selected CP congeners for each sample. For the identification of congener groups, their retention time range, signal shape, and isotope ratio were analyzed synthetically. As for the quantification procedures for SCCP (C_{10}–C_{13} with Cl_5–Cl_{10}) and MCCP (C_{14}–C_{17} with Cl_5–Cl_{10}), it was based on CP carbon, chain length and chlorination degree, as described in Reth et al. (2005).

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

All experimental glassware was heated at 450 °C and washed with acetone, dichloromethane, and hexane prior to use to avoid sample cross-contamination. During the analysis, a procedural blank was processed for each batch of 12 samples to monitor background interference and contamination. Trace amounts of a few PCBs (PCB 117, PCB 128, PCB170, and PCB175, range: 1.83–6.60 ng/g lipid weight (lw)) were

detected in the procedural blanks ($n = 6$) and subtracted from the samples. The recoveries of CPs in three spike samples ranged from 83.9% to 96.1%, and the relative standard deviation were < 5% for all targets. The percent recoveries (AV ± SD) of the surrogate standards ($n = 68$) were 98 ± 7, 101 ± 7, 106 ± 7, and 108 ± 18 for PCB 30, PCB 65, PCB 204, and $^{13}C_{10}$-trans-chlordane, respectively. No surrogate corrections were made to final concentrations. Instrumental QC was performed by regular injection of solvent blanks and standard solutions each day. The method detection limit (MDL) was defined as the mean value of target compounds detected in procedure blanks plus three times the standard deviations. For the undetectable compounds in blanks, the MDL was estimated as a signal-to-noise ratio of 10. Based on an average lipid level of 0.5 g in eggs and a constant volume of 300 μL, the MDLs for CPs and PCBs were in the range of 30–45 ng/g lw and 0.1–4.0 ng/g lw, respectively, except for PCB 117/128/170/175 (ranged from 2.1 to 8.0 ng/g lw).

2.4. Statistical analysis

The statistical analysis was performed using SPSS20.0. The non-parametric Kolmogorov-Smirnov Z test was used to test the normality of the distribution. The concentration data were log-transformed when data did not follow a normal distribution. To determine the difference in the concentrations (ng/g, lw) of PCBs, SCCPs, and MCCPs in home-produced eggs between 2013 and 2016, the independent-samples *t*-test was used. The paired-samples t-test was used to determine the difference in the composition profiles of CPs in the same sampling year because of the interactions between the contributions of different CP carbon chain lengths. The level of significance was set at $p < 0.05$.

3. Results and discussion

3.1. Levels of PCBs and CPs in the chicken eggs

The concentrations of ΣPCBs, SCCPs, and MCCPs, in free-range eggs collected in 2013 and 2016 are presented in Table 1 and Table S1 (SI). The statistics for PCBs and CPs in both periods are also shown in Table 1. The detection frequencies and the concentrations of ICES-6 PCBs (PCB28, PCB52, PCB101, PCB138, PCB153, and PCB180) and DL-PCB congeners (PCB77, PCB81, PCB105, PCB114, PCB118, PCB123, PCB126, PCB156, PCB157, PCB 167, PCB169, and PCB189) are presented in Table S2 (SI).

3.1.1. Levels of PCBs

The concentrations of ΣPCBs (sum of 159 individual PCB congeners) varied from 254 to 4580 ng/g lw with a median of 1130 ng/g lw in 2013 and from 236 to 8870 with a median of 1360 ng/g in 2016. The concentrations of PCBs were higher in 2016 than in 2013 but the differences were not significant ($p = 0.075$, Table 1). Concentrations of PCBs in the present study were much higher than those in food samples (< 1 ng/g wet weight (ww)) from a Norwegian cohort (Xu et al., 2017) and those in chicken eggs (range: 23.1–35.9 ng/g lw) from supermarkets in Madrid, Spain (García-Bermejo et al., 2017), while they were comparable to those (1180 ng/g lw) in chicken eggs from another e-waste recycling site in eastern China (Labunska et al., 2015), indicating a distinct influence of e-waste recycling on the levels of PCBs in local biota samples.

The levels of ICES-6 PCBs (the sum of six indicator PCB congeners) ranged from 68 to 1380 ng/g lw in 2013 and from 61 to 1730 ng/g lw in 2016. The concentrations of ICES-6 PCBs in this study exceeded the maximum level (ML) of PCBs in free-range eggs (40 ng/g lw) set by the European Commission (EU, 2011). The median values of ICES-6 PCBs in eggs (310 ng/g lw for eggs in 2013 and 398 ng/g lw for eggs in 2016) were almost ten times the ML. The ML for DL-PCBs was 2.5 pg WHO-$TEQ_{2005} g^{-1}$ lw (EU, 2011; Labunska et al., 2015). The levels of DL-PCBs were comparable to those of ICES-6 PCBs (43–1270 ng/g lw in 2013 and 57–2220 ng/g lw in 2016). Moreover, the median values of DL-PCBs in eggs in 2013 and 2016 were 7.7 and 11.2 pg WHO-$TEQ_{2005} g^{-1}$ lw, which were three to five times the ML. Up to 80% of egg samples had a DL-PCB TEQ exceeding the EU ML.

García-Bermejo et al. (2017) reported 1.762–2.953 ng/g lw of ICES-6 PCBs (median 2.355 ng/g lw) and 0.26–1.80 pg WHO-$TEQ_{2005} g^{-1}$ lw of DL-PCBs (median 1.2 pg WHO-$TEQ_{2005} g^{-1}$ lw) in chicken eggs from supermarkets in Madrid, Spain. Significantly higher levels of ICES-6 PCBs and DL-PCBs-TEQ in chicken eggs collected from e-waste or industry-influenced areas were reported by other studies. Labunska et al. (2015) measured the levels of ICES-6 PCBs and DL-PCBs-TEQ in chicken eggs from e-waste recycling sites (Taizhou) as 388 ng/g lw and 6.41 pg/g lw, respectively. Squadrone et al. (2015) detected 117–218 ng/g lw of ICES-6 PCBs and 30–103 pg WHO-TEQ_{2005} g^{-1} lw of DL-PCBs in free-range eggs at a sampling site close to a secondary aluminum smelter in northern Italy. Shen et al. (2017) reported relatively high levels of ICES-6 PCBs (range: 3.00–55.40 ng/g lw) and DL-PCBs-TEQ (0.3–32.0 pg/g lw) in chicken eggs collected from the vicinity of e-waste disassembling sites. These values were comparable to the results of this study.

Table 1
Concentrations (ng/g, lipid weight) of ΣPCBs, ΣICES-6 PCBs, SCCPs, MCCPs, and ΣDL-PCB TEQs (pg WHO-$TEQ_{2005} g^{-1}$ lw) in home-produced eggs.

Sampling year		2013					2016					
	ML[a]	Mean	SE[b]	Median	Range	HR[c] (%)	Mean	SE	Median	Range	HR (%)	*p*-Value[d]
ΣPCBs		1470	218	1130	254–4580		2240	360	1360	236–8870		0.075
ΣICES-6 PCBs[e]	40[g]	431	66.1	310	68-1380	100%	783	90	398	61–1730	100%	0.125
ΣDL-PCBs		360	61.7	250	43–1270		511	85.7	368	57–2220		0.146
ΣDL-PCB TEQs[f]	2.5[h]	10.9	1.86	7.7	1.29–38.3	81.6%	15.6	2.59	11.2	1.76–66.8	86.7%	0.145
SCCPs		2680	736	926	477–26,200		10,100	4700	1490	611–111,000		0.133
MCCPs		6100	2520	1030	125–90,700		6830	3220	999	297–91,100		0.590

[a] Maximum level.
[b] Standard error.
[c] High risk, defined as the ratios of PCB levels that exceeded the maximum levels.
[d] *p*-Value of t-test between 2013 and 2016.
[e] ΣICES-6 PCBs expressed in the sum of PCB28, PCB52, PCB101, PCB138, PCB153, and PCB180.
[f] ΣDL-PCBs-TEQ expressed in the sum of PCB77, PCB81, PCB105, PCB114, PCB118, PCB123, PCB126, PCB156, PCB157, PCB 167, PCB169, and PCB189 (pg WHO-TEQ_{2005}/g, lipid weight) (Van den Berg et al., 2006).
[g] Maximum level for sum of six non-dioxin-like PCBs set by the European Commission (EU, 2011).
[h] Maximum levels for the sum of twelve dioxin-like PCBs derived by subtraction of value for the sum of dioxins (WHO-PCDD/F-TEQ_{2005}) from the sum of dioxins and DL-PCBs (WHO-PCDD/F, PCB-TEQ_{2005}) (EU, 2011).

3.1.2. Levels of CPs

SCCPs and MCCPs were detected in all samples. The total concentrations of SCCPs in eggs ranged from 477 to 111,000 ng/g lw, with median values of 926 ng/g lw in 2013 and 1490 ng/g lw in 2016. MCCP concentrations were between 125 and 91,100 ng/g lw, with median values of 1030 ng/g lw in 2013 and 999 ng/g lw in 2016. The median concentrations of SCCPs and MCCPs in egg samples were comparable with the levels of PCBs (1130 and 1360 ng/g lw in 2013 and 2016, respectively) in this study. Similar to those of PCBs, there are no significant differences in SCCP and MCCP levels between 2013 and 2016 ($p = 0.133$ for SCCPs and $p = 0.590$ for MCCPs, Table 1), which means that these types of compounds remain at relatively high levels in environmental and biological systems for a long period of time.

To date, very limited data on CPs are available in chicken eggs. To our knowledge, there is only one study that reported the quantifiable levels of SCCPs in gull eggs from the Ebro Delta in Spain (Morales et al., 2012). SCCPs were detected at 4.54 ± 0.04 ng/g ww in *L. michahellis* and 6.36 ± 0.02 ng/g ww in *L. audouinii*, which were almost two orders of magnitude lower than those (294 ± 80.6 ng/g ww in 2013 and 1230 ± 562 ng/g ww in 2016, Table S1) reported in this study. The concentrations of SCCPs in the present study were also much higher than those in composite food samples from non-e-waste sites, such as Beijing (8.5–28 ng/g ww in 2009), Japan (< 1.1 ng/g ww in 2009), and Seoul (< 50–56 pg/g ww in 2007) (Harada et al., 2011). However, compared to the SCCP levels (mean values: 4900–12,000 ng/g lw) in terrestrial resident birds collected between November 2011 and May 2012 from the same sampling sites, mean levels of SCCPs in this study (926 and 1490 ng/g lw) were much lower (Luo et al., 2015). This result implies that wild birds have more chance to accumulate SCCPs than domestic fowls. This conclusion should be drawn cautiously because of the different tissues (muscle and eggs) measured in two studies. Since there is no available data of the MCCP levels in chicken eggs, even in terrestrial bird species in China, we were unable to compare these samples.

3.2. Profiles of contaminants in home-produced eggs

3.2.1. PCBs

Congeners PCB28(31), 47(48,75), 66, 74, 99, 105, 118, 138, 153, and 180(193) are dominant in egg samples (Fig. 1). Fig. 2 shows the DL-PCB and ICES-6 PCB profiles of the analyzed egg samples. Among DL-PCBs (Fig. 2a), the most abundant compound was PCB118 (63.4% in 2013 and 60.4% in 2016) followed by PCB105 (24.9% in 2013 and 25.5% in 2016) and PCB156 (5.5% in 2013 and 4.9% in 2016), already identified by other studies as dominant congeners (Squadrone et al., 2015; García-Bermejo et al., 2017). Major contributors to ICES-6 PCBs were PCB138 (32.5% in 2013 and 33.4% in 2016) and PCB 153 (32.5% in 2013 and 32.1% in 2016) (Squadrone et al., 2015, García-Bermejo et al., 2017) (Fig. 2b). However, for ICES-6 PCBs, several differences were found between the profiles in this study and other studies. PCB 52 (19%) and 101 (14%) were dominant in chicken eggs from supermarkets in Spain (García-Bermejo et al., 2017), but they were not relevant (PCB 52: 0.51% in 2013 and 1.8% in 2016, and PCB101: 1.4% in 2013 and 2.4% in 2016) in our samples. These different profiles observed between our egg samples (collected from e-waste influenced sites) and eggs from non-e-waste sites indicate the influence of multiple types of local sources on e-waste recycling sites. These sources are including PCB-containing transformer oil and/or dielectric fluids and/or lubricants, as well as unintentional formation of PCBs from combustion processes (Labunska et al., 2015). Moreover, no significant differences in PCB congener profiles were found between the eggs collected in 2013 and 2016 ($p > 0.05$), indicating that the PCB sources in the area were relatively stable (Figs. 1 and 2). The new input of PCBs to the environment has gradually disappeared since the PCB production was banned in the 1970s.

3.2.2. S/MCCPs

Homologue profiles of SCCPs and MCCPs in eggs collected in 2013 and 2016 are shown in Fig. 3. The homologue group profiles of SCCPs in eggs in 2013 were different from those in 2016. In egg samples collected in 2013, an almost equal abundance of SCCP homologue groups (C_{10}: 0.23, C_{11}:0.26, C_{12}:0.25, and C_{13}:0.26) was observed, which is consistent with the results of the previous reports on some terrestrial bird species (the great tit and oriental magpie-robin) collected from the same areas (Luo et al., 2015). Moreover, this is also similar to earlier results, which showed an almost equal abundance of SCCP homologue groups observed in three fish species (*turbot*, *Navodon septentrionalis*, and *capelin*) from Liaodong Bay, North China, and C_{10} and C_{11} accounted for 53.9% of the total SCCPs (Huang et al., 2017). In egg samples collected in 2016, a different distribution of homologue groups was observed from those collected in 2013. Homologues with 10 and 11 carbon atoms (69.0%) were dominant and significantly more abundant than C_{12} and C_{13} homologues ($p < 0.001$). C_{10} and C_{11} homologue groups have been also found to dominant in finless porpoise and in organisms (zooplankton, invertebrates, and fishes) (Ma et al., 2014; Zeng et al., 2015). However, a different homologue profiles of SCCPs were also reported in the earlier study, which showed a very low abundance (< 10%) of C_{10} homologue profiles in catfish, crucian carp, and mud carp, while C_{11} homologues were the most abundant group in both catfish (36.6%) and snakehead (31.3%) (Sun et al., 2017).

Since the regulations for e-waste import and disposal were imposed by the Qingyuan municipal government in 2010, the e-waste industry in Longtang has been suffering. Moreover, according to our investigation, in 2013, there were still some family-run e-waste workshops in Longtang, but in 2016, there were almost no workshops left in our sampling sites, indicating that SCCP sources for local biota in 2013 are likely influenced by both domestic and overseas e-waste items (SCCP commercial mixtures dominant by C_{12}), while sources for SCCPs in eggs collected in 2016 are dominated by non-point sources in the study area. The release of SCCPs from polyvinyl chlorinated products containing Chinese commercial mixtures (dominant by C_{10} and C_{11}) is expected to continue as these products are used extensively and remain unregulated in China, although the contribution from historical SCCP residues in the surrounding area from former e-waste recycling activities cannot be ruled out. Moreover, different SCCP congener group patterns were found in environmental matrices between e-waste-impacted and non-e-waste-impacted regions (Chen et al., 2011). Therefore, different SCCP congener group patterns in the environment during the two periods are expected and that is the reason for different homologue group profiles in egg samples in 2013 and 2016.

Regarding MCCPs, the C_{14} homologue was dominant with an average proportion of 0.39 in 2013 and 0.41 in 2016, and C_{15} was the second most abundant group (0.24 in 2013 and 0.23 in 2016). The MCCP homologue profiles in two sampling periods were similar, different from the SCCP profiles. Moreover, the congener distribution of MCCPs in the present study was also consistent with that in fish from Liaodong Bay, North China (Huang et al., 2017), and in biota from the European Arctic (Reth et al., 2006). The homologue patterns of MCCPs appear identical across the globe; e.g., in southern China (Wang et al., 2013), India, Pakistan (Chaemfa et al., 2014), Canada, and the Baltic Sea (Muir, 2010). Thus, similar MCCP group profiles in biota samples are likely caused by the identical MCCP group patterns in the environment.

4. Estimation of daily intakes

The estimated human daily intakes (EDI) of PCBs, DL-PCBs, SCCPs, and MCCPs by adults and children via the consumption of chicken eggs in Longtang are shown in Table 2. The EDI of DL-PCBs was calculated by multiplying the egg consumption rate (CR, g/d) by TEQ concentrations (C_{TEQ}, pg TEQ/g ww) in eggs and dividing it by body weight (BW, kg) (Eq. (A)),

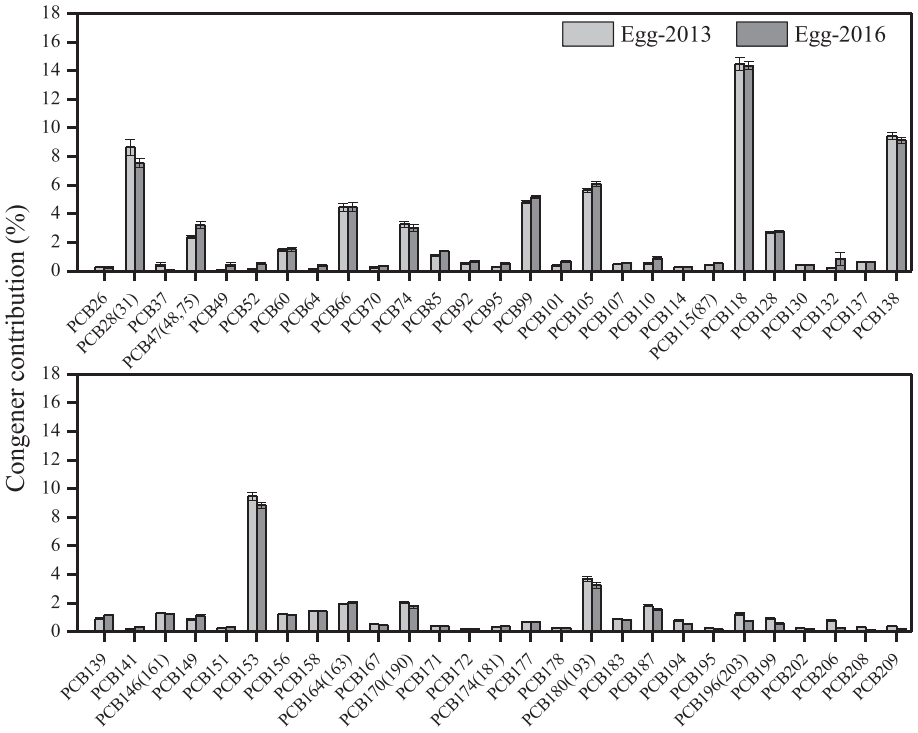

Fig. 1. Average profiles of PCBs (the congener percent > 0.2%) observed in home-produced eggs (2013 and 2016). Values are mean ± standard errors.

$$EDI = \frac{C_{TEQ} \times CR}{BW} \quad (A)$$

while the EDI for other compounds was calculated by multiplying the egg consumption rate (CR, g/d) and concentrations (C_i, pg/g ww) in eggs and dividing it by body weight (BW, kg) (Eq. (B)).

$$EDI = \frac{C_i \times CR}{BW} \quad (B)$$

The average egg consumption was 14.0 g/d/person for total population in rural areas in Guangdong province according to the report by Ma et al. (2005). The body weight for adults and children employed in our calculations were 63 kg and 14.65 kg, respectively (Zhou et al., 2012, MOHOC (Ministry of Health of China), 2012).

The Agency for Toxic Substances & Disease Registry established the Minimal Risk Levels (MRL) for ΣPCBs as 20 ng/kg bw/d (Agency for Toxic Substances, 2000). The World Health Organization (WHO) established the tolerable daily intake (TDI) for DL-PCBs as a range of 1–4 pg TEQ/kg bw/d (van Leeuwen et al., 2000) in 2000. The TDI for SCCPs is 100 μg/kg bw/d according to International Programme on Chemical Safety (IPCS, 1996) and 6 μg/kg bw/d for MCCPs according to Environment Canada (2008). The risk assessment was performed by comparing EDI with these values (Table 2).

4.1. PCB intake

The range of exposure of adults to total PCBs was 5.57–255 ng/kg bw/d and of children was 23.9–1100 ng/kg bw/d (Table 2). The percentage of EDI in adults that exceeded the MRL (20 ng/kg bw/d) was 68% in 2013 and 70% in 2016, while in children, 100% of EDI (range: 23.9–1100 ng/kg bw/d) exceeded the MRL, indicating the serious risk for local residents, especially for children. We also estimated the daily intake of total DL-PCBs based on their TEQ. The adult intake

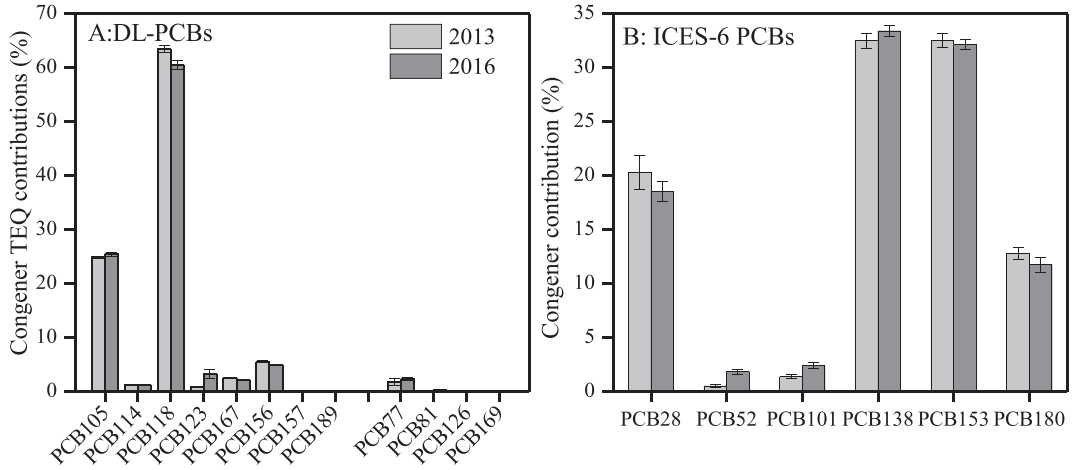

Fig. 2. Congener TEQ contributions of individual DL-PCBs to total DL-PCB levels (A) and congener contributions of individual ICES-6 PCBs to total ICES-6 PCB levels (B) in home-produced eggs. Values are mean ± standard errors.

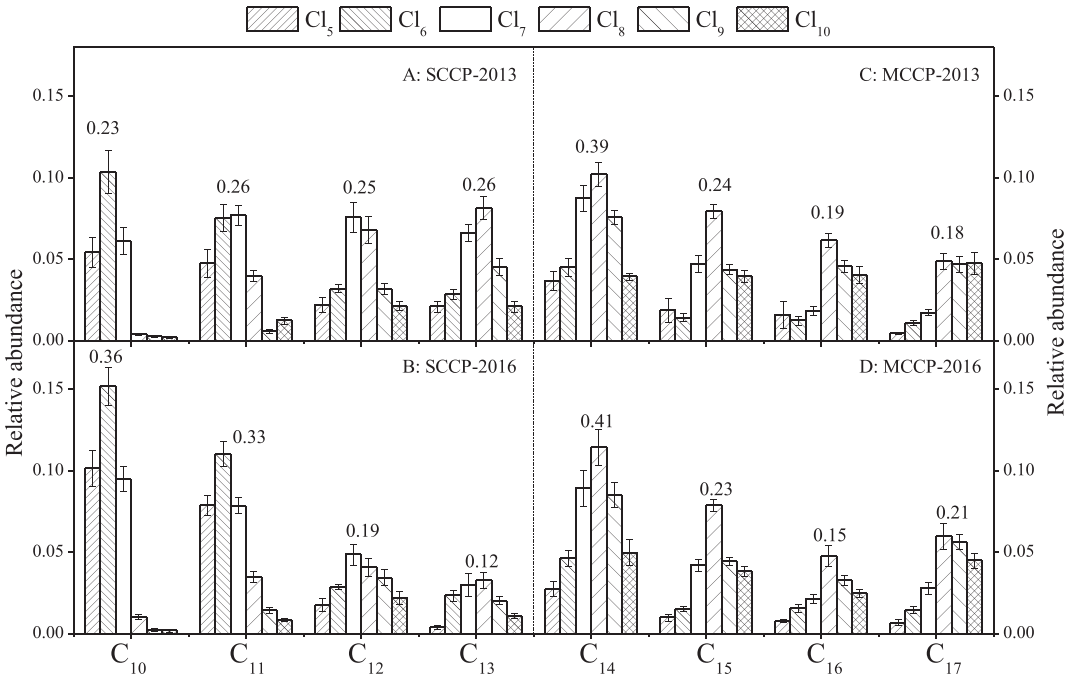

Fig. 3. Congener group patterns of SCCPs and MCCPs in home-produced eggs. Values are mean ± standard errors.

(0.03–1.92 pg TEQ/kg bw/d) was within the TDI (1–4 pg TEQ/kg bw/d) for DL-PCBs and > 90% of the DL-PCB estimates for children did not exceed the TDI (mean values of 1.20 and 1.86 pg TEQ/kg bw/d in 2013 and 2016, respectively) (Table 2). Our estimates for DL-PCBs were comparable to those reported in e-waste polluted areas. Labunska et al. (2015) reported that EDI of DL-PCBs by adults and children from e-waste recycling sites in eastern China were 0.35 and 1.51 pg TEQ/kg bw/d, respectively. Shen et al. (2017) reported that the EDI of DL-PCBs via consumption of chicken eggs from e-waste disassembling sites was 25.8 pg TEQ(d)$^{-1}$ (corresponding to 0.4 and 1.76 pg TEQ/kg bw/d for adults and children, respectively). However, our estimates for DL-PCBs were much higher than the estimates for hen eggs from markets or locations far from the polluted areas. Shen et al. (2017) reported that the daily intake of DL-PCBs via consumption of hen eggs was 6.4 pg TEQ(d)$^{-1}$ (corresponding to 0.1 and 0.4 pg TEQ/kg bw/d for adults and children, respectively). Squadrone et al. (2015) reported that estimates of DL-PCBs in eggs in sampling zone 3 (far from the secondary aluminum smelters) were 0.1 and 0.2 pg TEQ/kg bw/d for adults and children (9-year-old). Moreover, the highest EDI of DL-PCBs by children was 8.26 pg TEQ/kg bw/d, which was twice the WHO TDI upper limit for all 12 DL-PCBs combined. Exposure to high concentrations of PCBs and dioxin-like POPs can pose adverse health effects, including increases in birth defects and decreased child height. Therefore, dietary exposure to PCBs via home-produced chicken eggs in polluted areas should be limited to reduce the risk to human health, especially to children's health.

4.2. SCCP and MCCP intake

The total dietary exposure to SCCPs (11.8–11,900 ng/kg bw/d) was within the corresponding TDI (100 μg/kg bw/d) according to IPCS (1996), which was much higher than the exposure to SCCPs in food samples from Japan (81 ng/kg bw/d in 2009), while the intake of SCCPs in food samples from Beijing (390–1000 ng/kg bw/d in 2009) was within the range of this study (Harada et al., 2011) (Table 2). The mean dietary intake of MCCPs in our study was below the TDI, with a

Table 2
Overview of estimated dietary intakes (EDI) for PCBs and CPs via consumption of home-produced eggs.

Sampling year		2013			2016		
	MRL[c]/TDI[d]	Adult	Child	HF[e] (%)	Adult	Child	HF[e] (%)
ΣPCBs[a]	20[g]	37.4 ± 5.46 (5.57–113)[f]	161 ± 23.5 (23.9–486)	68%/100%	62.1 ± 9.91 (6.26–255)	267 ± 42.6 (26.9–1100)	70%/100%
ΣDL-PCBs[b]	1-4[h]	0.28 ± 0.05 (0.03–0.95)	1.20 ± 0.20 (0.12–4.07)	0%/5.3%	0.43 ± 0.07 (0.05–1.92)	1.86 ± 0.31 (0.20–8.26)	0%/3.3%
SCCPs[a]	100000[i]	65.4 ± 17.9 (11.8–645)	281 ± 77.0 (50.5–2770)	0%/0%	274 ± 125 (13.0–2766)	1180 ± 538 (55.8–11,900)	0%/0%
MCCPs[a]	6000[j]	151 ± 61.9 (3.62–2230)	651 ± 266 (15.6–9590)	0%/3%	193 ± 93.2 (6.31–2640)	829 ± 401 (27.1–11,400)	0%/3.3%

[a] ng/kg bw/d.
[b] pg TEQ/kg bw/d.
[c] Minimal risk levels.
[d] Tolerable daily intake.
[e] Health risks (adult/child), defined as the ratio of EDIs exceeded the MRL/TDI values.
[f] Mean ± standard error (range).
[g] The minimal risk levels derived by the Agency for Toxic Substance & Disease Registry (Agency for Toxic Substances, 2000).
[h] The range of TDI for dioxin-like chemicals established by the (van Leeuwen et al., 2000).
[i] The tolerable daily intake (TDI) for SCCPs according to (IPCS, 1996).
[j] The tolerable daily intake (TDI) for MCCPs according to Environment Canada (2008).

range of 151–829 ng/kg bw/d (mean values) for adults and children. However, the maximum EDI for MCCPs (11.4 μg/kg bw/d) in this study was twice the TDI (6 μg/kg bw/d) reported by Environment Canada (2008).

Our estimates for SCCPs or MCCPS were generally below the TDI values, but note that our exposure estimate is only based on the consumption of chicken eggs and local residents in our study area have additional dietary (such as seafood, vegetables, and meat) and non-dietary exposure (such as indoor air inhalation and dust ingestion), which was not considered in this study. According to the standard of food intake in China, the egg consumption only account for 2.5% of the total daily food intake, and as reported by Fridén et al. (2011), the relative contribution of adult exposure to S/MCCPs vie diet is ~85%, therefore, assuming that the CP levels were similar among different type of food, our estimates based wholly on home-produced eggs can only explain 2.1% of exposure intake for local residents. Moreover, the evidence to date suggests that the toxicological effects of SCCPs are generally more obvious than those of MCCPs, and SCCPs have been reported to cause liver toxicities in rats and trout, and "possibly carcinogenic to human" (Nilsen et al., 1980, IARC (International Agency for Research on Cancer), 1990, Cooley et al., 2001). Therefore, our results may be of great concern in China because of the large production and consumption of SCCPs/MCCPs in the country. Consequently, it is necessary to conduct a comprehensive assessment of dietary exposure to CPs by targeting different food types. In addition, concerning the insufficient knowledge related to the ecological toxicity of CPs, more toxicological investigation for CPs are required.

5. Conclusions

We measured high levels of PCBs, SCCPs, and MCCPs in eggs from a former e-waste recycling site. Concentrations of ΣICES-6 PCBs and TEQs of ΣDL-PCBs strongly exceeded current European maximum limits. Consuming these eggs implicates a serious health risk for local residents, especially for children. There were no significant changes in the PCB, SCCP, and MCCP levels between 2013 and 2016, indicating that these compounds remain at relatively high levels in environmental and biological systems for a long period of time. Therefore, it is necessary to raise awareness among the local population and take appropriate measures to reduce the intake of contaminants by home-produced eggs.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envint.2018.04.006.

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