



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



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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-TEQ₂₀₀₅ 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 biphenyl 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 (C₁₀–C₁₃, SCCPs), median-chain CPs (C₁₄–C₁₇, MCCPs), and long-chain CPs (C₁₈–C₃₀, 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

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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 $-20\text{ }^{\circ}\text{C}$ 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 (gravimetric 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 isoctane 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 ($^{13}\text{C}_{10}$ -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 $^{13}\text{C}_{10}$ -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 \times 0.25 mm i.d. \times 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 \times 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 $^{\circ}\text{C}$, the transfer line temperature was 280 $^{\circ}\text{C}$, and the ion source temperature was set to 200 $^{\circ}\text{C}$. The oven temperature programs was held at 80 $^{\circ}\text{C}$ (held for 3 min) firstly, then increased to 160 $^{\circ}\text{C}$ at 25 $^{\circ}\text{C}/\text{min}$ (held for 6 min), finally increased at 20 $^{\circ}\text{C}/\text{min}$ to 280 $^{\circ}\text{C}$ (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 $^{\circ}\text{C}$ 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

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