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# A pilot study on health risk assessment based on body loadings of PCBs of lactating mothers at Taizhou, China, the world's major site for recycling transformers☆

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ABSTRACT

Our early study reported an extraordinarily high Estimated Daily Intake (EDI) of PCBs of lactating mothers from Taizhou, Zhejiang Province, China (based on a food consumption survey and food basket analysis). The EDI well exceeded the intake limit stipulated by FAO/WHO 70 pg TEQ/kg body weight (bw)/month. The present pilot study provided further information on PCBs body burden in lactating mothers of Taizhou. The total PCBs detected in human milk, placenta and hair samples of these lactating mothers were 363 ng/g lipid, 224 ng/g lipid, and 386 ng/g dry wt. Respectively, three times higher than those samples collected from the reference site (Lin'an). Compared with the previous reported values in the 3rd WHO coordinated study, Taizhou topped the list of 32 countries/regions with regards to WHO-PCB-TEQ values of milk samples, which could be attributed to the relatively higher level of PCB-126 derived from electronic waste. In addition, the corresponding EDI of PCBs of Taizhou mothers (12.9 pg WHO-PCB-TEQ/kg bw/day) and infants (438 pg WHO-PCB-TEQ/kg) were derived from individual congener levels in human milk. The results were also higher than the tolerable daily intakes recommended by WHO (1–4 pg WHO-TEQ/kg bw/day) by 3 and 110 times, for mothers and infants, respectively. A more intensive epidemiological study on the potential health effects of e-waste recycling activities affecting both workers and residents seems to be of top priority, based on findings of this pilot study.

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## 1. Introduction

The rapid increase of electrical and electronic equipment waste (e-waste) has raised global concern (Chi et al., 2011; Peeranart et al., 2013). Due to deficit of well-equipped facilities and appropriate environmental regulations, and high labor costs, large amounts of discarded appliances have been exported from developed countries (mainly North America and Europe) to developing countries (Robinson, 2009). These e-waste receiving countries, usually have lower environmental standards and labour cost, which renders e-waste recycling more profitable (UNEP, 2005). China has been one of these recipient countries, receiving about 70% of global e-waste illegally imported according to the State Environmental Protection Administration of China (Xinhua Net, 2007).

Taizhou region and Guiyu town located in Zhejiang Province and Guangdong Province, respectively, are two notorious e-waste processing sites. In general, the uncontrolled recycling processes of mechanical shredding, acid leaching (outdoor) and baking (indoor) of printed circuit boards, and open combustion of plastics and wires

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(Wong et al., 2007). These e-waste recycling centers have become intensive point source, releasing a wide range of toxic chemicals. In Guiyu town, high emissions of polybrominated diphenyls ethers (PBDEs), polycyclic aromatic hydrocarbons (PAHs), polychlorinated dibenzo-p-dioxins/furans (PCDD/DFs), and heavy metals (e.g. Cr, Cd, Cu and Pb) during uncontrolled recycling activities have brought along pollution problems and threatened the well beings of the local residents and wildlife (Leung et al., 2007; Man and Wong, 2013). In particular, Taizhou region is a major site devoted for recycling transformers and coolants, which have led to the release of polychlorinated biphenyls (PCBs), causing serious environmental and health problems (Xing et al., 2010, 2011).

Due to their long-lasting and lipophilicity characteristics, PCBs are recalcitrant to degradation and persist in animal and environmental reservoirs. Humans can expose to PCBs via inhalation, ingestion, and skin contact (WHO/EURO, 2000; Fitzgerald et al., 2004; Freels et al., 2007). Various health problems, such as dermal toxicity, immunotoxicity, endocrine effects, reproductive effects, and carcinogenicity could be associated with the exposure to PCBs (Knerr and Schrenk, 2006; Longnecker, 2015). Stored PCBs in human tissues could be transferred to the next generation, e.g. to the fetus, by means of transplacental transport (DeKoning and Karmaus, 2000), or to infants through breast-feeding (Chao et al., 2007). Govarts et al. (2012) showed that low-level exposure to PCBs could suppress fetal growth. The lower birth weight was associated with the increase of PCB-153 in cord serum (150 g per 1 μg/L) (Govarts et al., 2012).

Human milk has been used as a bioindicator in a number of real-time biomonitoring programs (e.g., Malisch and van Leeuwen, 2003; Minh et al., 2004; Turrio-Baldassarri et al., 2008; Ulaszewska et al., 2011a,b). On the other hand, placenta is an important tissue for evaluating pollutant burdens and potential adverse health effects exerted on both the mother and fetus, as a “dual” biomarker” (Chao et al., 2007). Nutrients and toxicants are transferred through placenta, which are closely connected with fetal health, and therefore placenta samples present excellent opportunities for epidemiological studies. It has been generally assumed that the transmission of persistent toxic substances (POPs) via human milk far exceeded the contribution from transmission through placenta (Matsuura et al., 2001), since the placenta might help to reduce the potential exposure of fetus to toxicants preferential screening (Iyengar and Rapp, 2001). There is an urgent need for more studies on placental transfer, in order to provide more information about their association with body burdens of PCBs.

Unlike the analyses of human milk and placenta, which are limited to lactating women, hair samples can be analyzed for any groups of populations and may be used as indicators for long-time exposure, which record accumulation of toxicants over a period of time. The use of hair for this purpose provides a number of benefits, including readily available specimens, stability, non-intrusive sampling, and ease of sample collection, transportation and storage (Covaci et al., 2002; Nakao et al., 2005).

In developed countries, PCBs have shown a declining trend in human milk in recent decades, due to the legislative regulation on the production and application of PCBs since 1974 (Konishi et al., 2001; Bates et al., 2002; Malisch and van Leeuwen, 2003). Nevertheless, there are limited studies available comparing the concentrations of PCBs in more than one matrix of the human body. There is also insufficient information research study focusing on body burdens and associated human health effects of PCBs in China (Wong et al., 2002; Hedley et al., 2006; Sun et al., 2006), especially with regards to body loadings of PCBs in workers engaged in e-waste processing activities (Zhao et al., 2006, 2007; Bi et al., 2007).

Based on the contents of PCBs in various food types collected at Taizhou region, our earlier study estimated the dietary intake of 37 PCBs of lactating mothers was 92.79 ng/kg body weight (bw)/day (Xing et al., 2011). The study involved both local residents and e-waste workers and the estimated dietary intake was derived from their daily consumption of freshwater fish (28%), meat (17%), shellfish (95), vegetables (85%), egg (4%) and marine fish (2%). The total daily intake of 9.78 pg WHO-PCB-TEQ kg bw/day far exceeded the FAO/WHO Tolerable Daily Intake (around 2.33 pg TEQ kg bw/day) (Xing et al., 2011).

The present study aims at (1) investigating PCBs loadings in human milk, placenta and hair of lactating women living or working in the Taizhou e-waste processing site, in comparison with a reference site (Lin'an, also in Zhejiang Province); (2) determining the inter-relationships between the levels of PCBs among human milk, placenta and hair; (3) identifying factors associated with PCB levels in lactating women at the study site; and (4) conducting a health risk assessment based on the PCBs levels in human body and daily intake of PCBs for these lactating women and infants.

## 2. Materials and methods

### 2.1. Sampling sites

The sampling sites have been described in Xing et al. (2011). In Zhejiang Province, Taizhou (28°40′N, 122°21′E) has been a large-scale e-waste recycling site in China, in particular, for recycling transformers and capacitors since late 1980s/early 1990s. In the southern part of Taizhou, Luqiao City was the site for executing e-waste recycling activities (Fig. 1), contributed by 40,000 people working in the e-waste recycling sector (Luqiao Government, China, 2006). In the northwest of Zhejiang Province, Lin'an (30°14′N, 119°43′E), about 245 km away from Taizhou, without electronic waste recycling activities, agriculture is the major industry, has been chosen as a reference site for comparison (Lin'an Government, china, 2006) (Fig. 1).

### 2.2. Sample collection

Three different types of human sample (human milk, placenta, and hair) were collected from 25 women who gave birth between August and December 2005, in both Taizhou and in Lin'an, respectively with the assistance of a local hospitals and a health center, respectively (Luqiao Hospital of Traditional Chinese Medicine, Lin'an Hospital of Traditional Chinese Medicine, and Luqiao Center for Disease Control and Prevention). Milk samples were collected within 1 week postpartum (in the hospital) by manual expression to a solvent-rinsed Pyrex bottle (100 ml) with a Teflon lined cap. Whole placentas were collected by the local gynecologists. Stainless steel scissors were used to collect the hair (~3 g) of breastfeeding mothers, located near the scalp and from the nape of the neck. Samples of placenta and hair were separately packed with aluminum foil and placed into acetone-washed reagent bottles. All human samples were kept at −20 °C until analysis. Consent forms were completed by donors before the collection of specimens. Approval for the study was obtained from the Ethics Committee of Zhejiang Provincial Center for Disease Control and Prevention.

### 2.3. Questionnaires

In order to study the dietary habit and personal information of the participating mothers, questionnaires derived from the second round of WHO's PCDDs, PCDFs, and PCBs exposure study with slight modifications (WHO, 1996), were completed by these donating mothers. WHO's Survey was translated from English to Simplified Chinese with some questions modified to suit the life style of Chinese living in the sampling sites. For example, for the kind of fish

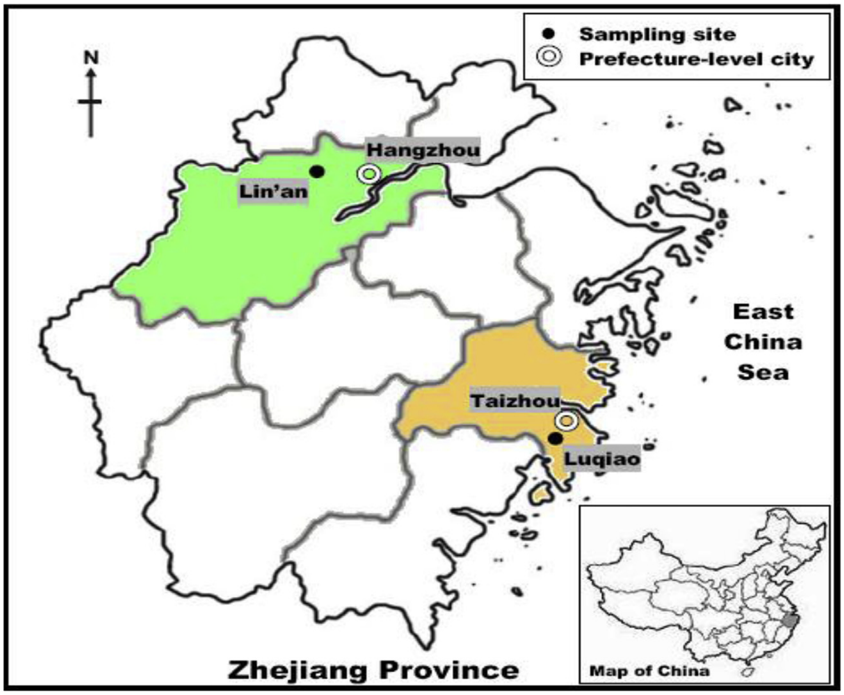


**Fig. 1.** Locations of sampling sites (Taizhou & Lin'an) (Chan et al., 2007; Man et al., 2014).

and fish products that respondents would eat, the local species such as big head carp, and crucian carp are asked, instead of "Tuna fish". In addition, a few questions were added about the occupational history in e-waste recycling sites. The information obtained included data of lifestyle, sociodemographic variables, duration of exposure through e-waste recycling activities, locations of residences, and status of breastfeeding and food intake patterns (WHO, 1996; Yang et al., 2002b).

### 2.4. Sample analyses

Chemical analyses for human samples (milk, placenta and hair) from Taizhou and Lin'a were conducted by the State Key Laboratory for Freshwater Ecology and Biotechnology, Institute of Hydrobiology (Chinese Academy of Sciences) in Wuhan, Hubei province. Samples of human milk were extracted and then cleaned up, according to Standard Methods 608 (USEPA, 1988) and 3640 A (USEPA, 1994). Placenta samples were pretreated and freeze-dried. Hair samples were washed for 5 min with a commercial detergent (1%) and then by distilled water (Covaci et al., 2002; Nakao et al., 2005), and oven-dried at 70 °C. Placenta and hair samples spiked with surrogate standards (PCB30 and PCB209) were extracted using acetone and n-hexane based on Standard Method 354 °C (USEPA, 1996a). The lipid contents of human milk and placenta were obtained from sample extracts, by evaporation. The samples were further cleaned up, based no Standard Methods 3630C (USEPA, 1996b), and 3620B (USEPA, 1996c), accordingly. A Gas chromatography (HP6890) equipped with a $^{63}Ni$ electron capture detector and a capillary DB-5 column (5% phenyl/95% methyl silicone, 60 m, 0.25 mm i.d. 0.25 μm film thickness, J&W Scientific, Folsom, CA, USA) was used to analyze PCBs. The temperature programme and operation conditions of GC analysis has been published in Zhao et al., 2007 (please refer to supplementary data) Twenty-three PCB congeners (PCB 28, 52, 77, 81, 99, 101, 105, 114, 118, 123, 126, 128, 138, 153, 156, 157, 167, 169, 170, 180, 183, 187, 189), which included 12 dioxin-like (DL) PCBs and 6 indicator PCBs were quantitatively analyzed. The sum of toxicity equivalents (TEQs) of the 12 DL-PCBs (WHO-PCB-TEQ) were obtained according to WHO-TEF-1998 (Van den Berg et al., 1998) (Table S1).

### 2.5. QA/QC

For checking any interference or contamination, a blank sample was included and analyzed in each batch of 16 samples. Standard reference materials of SRM 1945 (organics in Whale Blubber) and 2978 (Mussel Tissue (Organic Contaminants-Raritan Bay, NJ)) obtained from National Institute of Standards and Technology, USA were analyzed for PCB congeners, and the recovery percentages ranged from 65 to 120%.

### 2.6. Body loading estimation

Estimations of body burdens (toxicity equivalents (TEQs) ng) and Estimated Daily Intake (EDI) (pg/kg bw/day) of DL-PCBs for mothers and infants were based on USEPA (2000) and Yang et al. (2002a) and the equations are listed in the supplementary data. All the assumptions for the calculation derived from different papers were listed in the supplementary data.

The changes of concentrations of human milk during breastfeeding and the body weight of an infant during the first year can be predicted using procedures proposed by Yang et al. (2002a). Based on the first-order kinetics described in Rogan et al. (1991) (decline in the chemical contaminant of 20% over a 6 month period), the concentrations in breast milk, after initial breast-feeding, was corrected for every subsequent three months. Infant weight variations with time were assumed to be 5 kg at 0–3 months, 7 kg at 4–6 months.

### 2.7. Data analyses

SPSS 16.0 software program was used for statistical analyses. PCBs detected in samples were characterized by descriptive statistics. Mann-Whitney *U* test or analysis of variance (ANOVA) was used to assess the differences among groups. Pearson correlation coefficients were used for determining the relationships among different parameters and the association of PCB levels among the three types of human tissues. In addition, associations between the potential dietary exposures and total PCB concentrations in human samples were tested by multiple linear regression analysis. The significance level was set at $p < 0.05$.

## 3. Results and discussion

### 3.1. Characteristics of the sample population

Table S2 lists personal characteristics of the studied populations. There was no manifest difference for physiological characteristics (e.g. age, body weight and body mass index (BMI) among the participants at the two study sites. Most of the mothers at Lin'a were primipara (80%), compared with only 48% at Taizhou. All the infants were singletons and none were born with any congenital disorders or abnormalities. There was no significant difference for children's body weight and male/female ratio between two sites. Although a low birth ratio of boys to girls have been reported in the Arctic (ratio of girls to boys up to 2), possibly due to PCBs pollution (Times Online, 2007), the percentage of newly born male infants (64%) was slightly higher at Taizhou than Lin'an (50%), which might be due to the limited size of the sample population.

### 3.2. Body burden of PCBs

The concentrations of total PCBs, DL-PCB, and WHO-PCB-TEQ for human specimens are listed in Table 1. The human milk samples from Taizhou contained high levels of PCBs (363 ng/g lipid), and thus contributed to an elevated WHO-PCB-TEQ value (143 pg WHO-PCB-TEQ/g lipid). These values were significantly higher than those obtained at Lin'an (116 ng/g lipid for total PCBs, 70.6 pg WHO-PCB-TEQ/g lipid). More importantly, these human milk PCB levels (WHO-PCB-TEQ) in Taizhou and Lin'an exceeded the recommended tolerable level for raw milk and dairy products (3 pg WHO-PCDD/F-TEQ/g lipid) stipulated by the European Union by around 48 and 25 times, respectively (Commission Regulation (EC), 2006).

It is evident that the total PCB levels in the three types of studied human specimens (milk, hair and placenta) collected from Taizhou were significantly higher far exceeded ($p < 0.05$) (7.6, 5.0 and 3.1 times, respectively) than the values recorded from the reference site (Lin'an). This could be attributed to the frequent e-waste recycling activities in Taizhou. However, the rather high WHO-PCB-TEQ values detected in placenta and hair samples of the reference site (Lin'an) were unexpected. These values were comparable and even higher than those levels obtained from Taizhou. This was due to the relatively higher distribution of PCB 169, and its relatively high TEF value (0.01) (Srogi, 2008). Hair is an outstanding indicator to intrinsic exposure of human body to POPs originated from internal and external sources (Zhang et al., 2007). Therefore, the contribution of PCB-169 in higher PCB-TEQs levels of the hair samples collected from the reference site (Lin'an) was probably due to the hair surface exposure to exogenous sources such as atmospheric deposition (Schramm, 2008). Kunisue and Tanabe (2016) stated three possible processes: the formation of coplanar PCBs, including the emission from commercial PCBs mixtures; release from incineration; and products from solar photolysis of higher chlorinated PCBs which may usually contribute to lesser extent when compared with other sources. It had been identified that PCB-169 was mainly released through heating processes but not via volatilization in the atmosphere in Japan (Ogura et al., 2004). It is suspected that a substantial amount of PCB-169 could have been emitted from the combustion of crop residues during our sampling period (September–November 2005) at Lin'an (Zhang et al., 2008), where open burning of crop residues (notably rice straw) after harvest is commonly practiced in rural China. There is no e-waste recycling industry in Lin'an. In fact, open burning of crop residues, a common practice worldwide, especially in Asia, releases polychlorinated dibenzo-dioxins and dibenzofurans (PCDD/PCDF) and

**Table 1**
The average concentrations of PCB congeners in human specimens from the e-waste recycling site Taizhou (TZ) and a reference site Lin'an (LA).

| Congener | Hair(ng/g dry wt) | | | | Placenta(ng/g lipid) | | | | Milk(ng/g lipid) | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | TZ (n = 25) | | LA (n = 25) | | TZ (n = 25) | | LA (n = 25) | | TZ (n = 25) | | LA (n = 25) | |
| | Mean | SD | Mean | SD | Mean | SD | Mean | SD | Mean | SD | Mean | SD |
| PCB28 | 39.4 | 61.0 | 2.44 | 2.37 | 16.9 | 13.5 | 1.03 | 1.94 | 12.7 | 16.5 | 8.21 | 8.64 |
| PCB52 | 65.0 | 98.0 | 0.0900 | 0.300 | 1.92 | 2.32 | 0.720 | 1.88 | 5.40 | 9.98 | 28.8 | 50.0 |
| PCB101 | 70.0 | 92.1 | 0.920 | 3.04 | 9.97 | 11.3 | 2.95 | 7.53 | 11.0 | 9.85 | 12.7 | 22.9 |
| PCB138 | 0.200 | 1.00 | 0.100 | 0.470 | 20.5 | 20.7 | 7.49 | 19.3 | 74.4 | 109 | 3.38 | 3.89 |
| PCB153 | 0.00 | 0.00 | 0.00 | 0.00 | 0.00 | 0.00 | 0.00 | 0.00 | 48.6 | 69.4 | 0.0700 | 0.330 |
| PCB180 | 8.74 | 13.2 | 1.18 | 4.21 | 7.82 | 8.72 | 3.64 | 9.26 | 18.1 | 23.5 | 1.19 | 1.99 |
| ∑Indicator PCBs | 183 | 250 | 4.72 | 8.48 | 57.1 | 38.8 | 15.8 | 38.5 | 170 | 216 | 54.4 | 84.5 |
| PCB77 | 33.8 | 25.4 | 20.7 | 13.6 | 54.8 | 15.8 | 7.60 | 12.1 | 1.60 | 1.93 | 17.3 | 8.70 |
| PCB81 | 16.3 | 8.35 | 15.0 | 13.0 | 45.0 | 13.4 | 3.48 | 7.22 | 0.860 | 1.02 | 13.5 | 5.68 |
| PCB126 | 0.00 | 0.00 | 0.00 | 0.00 | 0.00 | 0.00 | 0.500 | 0.960 | 1.12 | 1.00 | 0.0200 | 0.100 |
| PCB169 | 2.93 | 2.84 | 7.06 | 5.65 | 10.0 | 10.6 | 6.92 | 4.97 | 1.21 | 0.920 | 6.30 | 4.75 |
| ∑non-ortho PCBs | 53.0 | 32.0 | 42.8 | 29.5 | 110 | 32.4 | 18.5 | 21.4 | 4.79 | 3.85 | 37.2 | 19.1 |
| PCB105 | 0.340 | 1.69 | 0.00 | 0.00 | 1.08 | 4.20 | 0.00 | 0.00 | 21.7 | 38.42 | 1.20 | 3.710 |
| PCB114 | 0.00 | 0.00 | 0.0100 | 0.0500 | 0.110 | 0.570 | 0.290 | 0.690 | 3.30 | 4.40 | 0.160 | 0.580 |
| PCB118 | 82.4 | 112 | 0.400 | 1.23 | 21.2 | 23.0 | 1.28 | 3.21 | 80.0 | 115 | 10.0 | 16.9 |
| PCB123 | 3.26 | 4.29 | 0.110 | 0.460 | 2.93 | 10.9 | 0.0900 | 0.430 | 1.44 | 2.14 | 0.10 | 0.410 |
| PCB156 | 0.100 | 0.390 | 0.0300 | 0.080 | 4.93 | 5.17 | 0.940 | 2.20 | 11.8 | 18.5 | 0.850 | 0.700 |
| PCB157 | 5.02 | 8.90 | 0.00 | 0.00 | 0.0600 | 0.280 | 1.06 | 4.85 | 2.58 | 3.82 | 0.300 | 1.46 |
| PCB167 | 7.61 | 12.7 | 0.0900 | 0.430 | 0.430 | 1.88 | 0.0700 | 0.350 | 4.15 | 5.71 | 0.00 | 0.00 |
| PCB189 | 0.0700 | 0.320 | 0.0100 | 0.0700 | 5.10 | 10.3 | 1.14 | 1.59 | 1.27 | 1.05 | 2.00 | 4.97 |
| ∑mono-ortho PCBs | 98.8 | 137 | 0.660 | 1.41 | 35.88 | 29.5 | 4.87 | 7.99 | 126 | 185 | 14.6 | 39.6 |
| ∑dl-PCBs | 152 | 162 | 43.4 | 29.6 | 146 | 51.7 | 23.4 | 27.9 | 131 | 186 | 518 | 40.3 |
| $\sum_{23}$ PCBs | 386 | 465 | 51.0 | 37.2 | 224 | 97.0 | 44.6 | 77.0 | 363 | 470 | 116 | 108 |
| ∑WHO-PCB-TEQ (pg/g lipid) | [a]45.5 | [a]33.1 | [a]74.2 | [a]58.3 | 116 | 108 | 122 | 113 | 143 | 126 | 70.6 | 47.9 |

[a] Pg/g dry wt. For hair.

dioxin like PCB (Black et al., 2012), has exerted potential impacts on air quality, atmosphere and climate (Oanh et al., 2011, 2015).

Furthermore, the reasons for high levels of PCBs in Lin'a were not very clear. Other possible reasons could be: (a) Lin'a is also located in an area with relatively high PCB contamination in China (Xing et al., 2005); (b) Lin'a may have other polluted sources other than e-waste recycling operations, such as chemical plants or power plants in the vicinity (UNEP, 2002); and (c) the participating mothers in Lin'a may not be local residents, and came from other polluted areas, e.g. Taizhou; The PCBs pollution in Lin'a should be investigated further.

Another similar study conducted at Taizhou (Wen et al., 2008) indicated that the average PCB levels in hair of male e-waste workers was 1600 ng/g dry weight (wt.), 4 times higher than the present results, while WHO-PCB-TEQ levels in hair were half of the present results. PCB-126 (with a TEF value of 0.1) was the dominant constituent in dioxin-like PCBs in the male workers, followed by PCB-118, 156, and 105. By contrast, PCB-169, 118, 77, 157 were the top 4 contributors to WHO-PCB-TEQs in the present study. Differences in sampling period, gender and occupations of the engaged populations might be the reasons for the observed variations between the two studies. Nevertheless, both studies showed that the workers/residents were exposed to high levels of PCBs in Taizhou, the world's major site for recycling transformers.

### 3.3. Comparison of PCB profiles in different human samples of Taizhou

Penta- and hexa-chlorobiphenyls contributed to the highest proportion (almost 80%) in milk samples, while different patterns were found in hair and placenta samples (Fig. 2). Low chlorinated PCBs (i.e. tri-, tetra- and penta-chlorobiphenyls) occupied about 90% of the total PCBs in hair samples, seemed to link with the patterns of commercial PCBs (e.g. Aroclor 1242, Aroclor 1254, PCB3 and PCB5) used in transformers or capacitors (ATSDR, 2000; Breivik et al., 2002; Xing et al., 2005).

The difference in homologue pattern observed in different human tissues was possibly due to their differences in lipid contents, as well as molecular size. The highest lipid content was found in milk, followed by placenta, and the lowest in hair samples. As a consequence, due to their lipophilic nature, the concentrations of higher molecular weight PCBs tended to accumulate in human milk (Schecter et al., 1998). In addition to hexa- and hepta-chlorobiphenyls, penta-chlorobiphenyls also accounted for around 40% of the total PCBs in milk (Fig. 2). PCB-126 was a representative penta-chlorobiphenyl congener with a high TEF value (0.1) (Srogi, 2008). A significant correlation between total WHO-PCB-TEQs and PCB-126 was noted for human milk ($r = 0.985$), indicating PCB-126 is a reliable marker for assessing the toxicity of dioxin-like PCBs exposure in Taizhou. Furthermore, Mori et al. (2014) indicated that lower molecular weight PCBs were more readily passed from mother to fetus via placental transfer. Hence, PCB congeners with lower molecular weight (i.e. tri-, tetra- and penta-chlorobiphenyls) dominated (more than 70%) in the profile of placenta (Fig. 2).

### 3.4. Factors affecting PCB levels in human milk

It is commonly observed that factors such as parity, maternal age, dietary type, body mass index, residence period, breast-feeding time and number of previously breast-fed children are closely connected with the body burden of PCBs (Subramanian et al., 2007; Devanathana et al., 2009; Tsang et al., 2011). Nevertheless, the present results indicated that there was no significant correlation between PCB levels in human milk and most of the aforementioned factors. Only maternal age was found to have prominent correlations with the PCB contents in milk ($p > 0.05$, $p = 0.056$). In fact, our previous study also reported similar observation. Significant correlation could not be identified between the loading of PCBs in human milk and dietary intake of different food items types (e.g. freshwater fish, shellfish and meat), due to the fact that multiple food sources of PCBs in Taizhou, including freshwater and marine fish, rice, meat, shellfish, egg and vegetables (Xing et al., 2010). The age interval (24–37 years), unknown recent exposure of PCBs, limited sample size of this study, and complicated exposure routes were possible reasons for the limited association obtained between PCB concentrations in breast milk and other investigated factors including parity, maternal age, dietary type, body mass index, residence period, breast-feeding time and number of previously breast-fed children.

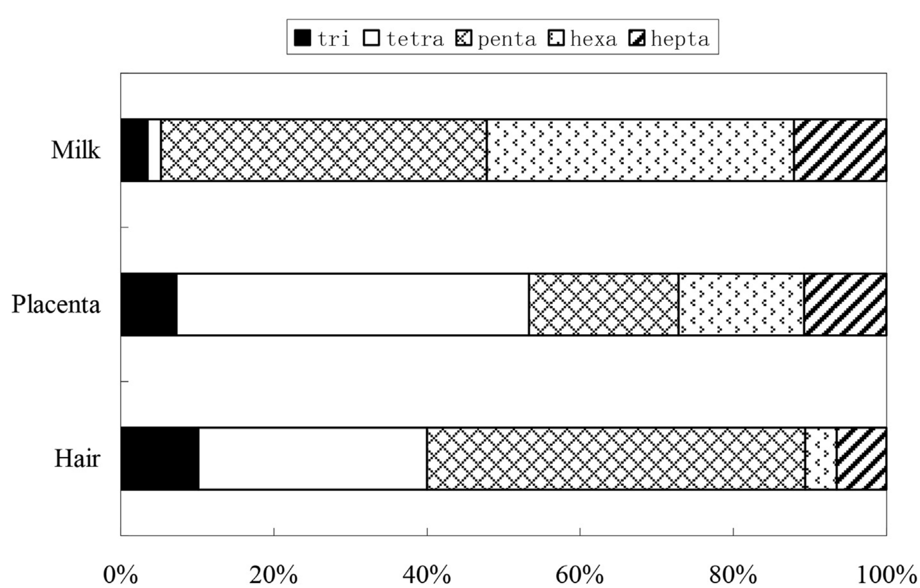


**Fig. 2.** PCB congeners profile in different human tissues collected from Taizhou.

### 3.5. Body loading estimation

The EDI of Taizhou mothers and infants were 12.9 and 438 pg WHO-PCB-TEQ/kg bw/day which were 3.2 and 110 times, respectively higher than the tolerable daily intakes suggested by the WHO (1–4 pg WHO/TEQ/kg bw/day) (IPCS, 1998) (Table 2). As placental transfer of PCBs had not been taken into account for the EDI calculation, this estimation of PCB intake by infants in Taizhou could be underestimated (Tsukimori et al., 2013). Acute exposure to such a high level of PCBs would deteriorate the thyroid hormone system and immunological functions of newborn babies (Stine and Brown, 2006). The persistent toxicant might even be accumulated in the infant body, interfering with its normal growth and development (Wong et al., 2012).

### 3.6. Global comparison with PCB levels in human tissues

The PCB concentrations in human milk in China and the Asian countries derived from different studies are listed in Table S3. The total PCBs and WHO-PCB-TEQ in human milk in Taizhou (363 ng/g lipid and 143 pg/g lipid) and Lin'an (116 ng/g lipid and 70.6 pg/g lipid) of the present study were comparable to Pingqiao (208 ng/g lipid and 8.07 pg/g lipid), which was another e-waste recycling site located in Zhejiang Province (Zhao et al., 2007). The current findings outweighed the values reported from other Asian countries or cities, such as Hong Kong (N.A. and 3.79 pg/g lipid) (Wong et al., 2013), Guangzhou (33.0 ng/g lipid and N.A.) (Wong et al., 2002), Dalian (42.0 ng/g lipid and 3.00 pg/g lipid) (Kunisue et al., 2004), Chennai, India (34.0 ng/g lipid and N.A.) (Subramanian et al., 2007), Jakarta, Indonesia (33.0 ng/g lipid and N.A.) (Sudaryanto et al., 2005) and Bien Hoa, Vietnam (N.A. and 1.59 pg/g lipid) (Nghi et al., 2015). Compared with the results obtained in the 3rd WHO-coordinated study, the content of indicator PCB in the human milk samples obtained from Taizhou ranked tenth in the list, which was much higher than those collected from other regions in China, including Lin'a and Guiyu (Fig. S2A). Taizhou even ranked number one in the list regarding WHO-PCB-TEQ values (Fig. S2B), as a result of relatively higher contribution of TEF value by PCB 126 (0.1) (Srogi, 2008).

The PCB levels in human placenta and hair are summarized in Tables S4 and S5. On a global scale, placenta and hair samples collected from Taizhou and Lin'an illustrated extremely high total PCB levels and WHO-PCB-TEQ levels. This is particularly true for Taizhou, with total PCB levels in human tissues (placenta: 224 ng/g lipid; hair: 386 ng/g dry wt.) which were higher than the values reported in other regions, such as Italy (98.7 ng/g lipid) and Japan (0.870 ng/g lipid) for placenta (Bergonzi et al., 2009; Tsukimori et al., 2013); Philippine (dumpsite: 31.0 ng/g dry wt.; non-dumpsite: 26.0 ng/g dry wt.) and Iran (Ahvaz: 17.0 ng/g dry wt.; Noushahr: 8.00 ng/g dry wt.; Countryside of Noushahr: 5.50 ng/g dry wt.) for hair (Malarvannan et al., 2013; Behrooz et al., 2012).

**Table 2**
Body loading estimation of PCBs levels for mothers and infants in e-waste recycling site.

| Body loading estimation | PCB-TEQs (TZ) | PCB-TEQs (LA) |
|---|---|---|
| Body burden (ng TEQ) | 2366 | 108 |
| $EDI_{mother}$ (pg/kg bw/day) | 12.9 | 6.38 |
| $EDI_{infant}$ (pg/kg bw/day) | 438 | 216 |
| [a]pg WHO-TEQ/kg bw/day | 1.00–4.00 | 1.00–4.00 |

Note: EDI = Estimated Daily Intake; WHO = World Health Organization; TEQ = toxicity equivalents; TZ = Taizhou; and LA = Lin'an.
[a] Tolerable daily intakes recommended by the WHO (IPCS, 1998).

## 4. Conclusion

The human milk samples collected from Taizhou and Lin'a showed extraordinarily high PCBs levels (363 and 116 ng/g lipid, respectively). In terms of TEQ levels in human milk, Taizhou topped the list, comparing with the findings obtained from countries or cities in the 3rd round WHO-coordinated study. In respect of placenta and hair samples, the two studied sites included in the present study both contained PCB levels at the high end worldwide. It seems obvious that the PCBs levels in human tissues of the lactating mothers of the present study are extremely high, when compared with data generated from other studies conducted in different countries.

The EDI of PCBs by Taizhou mothers and infants derived from their loadings in human milk were 12.9 pg WHO-TEQ/kg bw/day and 438 pg WHO-TEQ/kg bw/day, respectively. These extremely high body burdens reflected the severe potential health threats imposed on the residents, as well as on their next generation in Taizhou. Nevertheless, this is only a pilot study and human exposure to PCBs from e-waste processing activities would involve multiple environmental compartments through various pathways. Therefore, further investigation on the potential health impacts caused by e-waste recycling operations through epidemiological studies is needed in order to generate a more comprehensive and accurate assessment.

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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.2017.04.069.

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