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Regional Distribution and Human Health Effects of Persistent Organic Pollutants (POPs) in Zhejiang Province

Nanxiang Wu, Ye Yang, Yang Song, Ming Gao and Heru Huang

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Abstract

Zhejiang (ZJ) is a developed province located in the southeast coast of China. In recent years, growing concern has been aroused over the persistent organic pollutants (POPs) pollution associated with electronic and electric waste (e-waste) in this province. This chapter has provided numerous and integrated information concerning POPs pollution level and human health effects in ZJ. The residue levels of major POPs, including DDT, PCDD/Fs, PCBs and PCP/PCP-Na, in the environmental media, local food and human body were relatively higher in polluted areas of intensive e-waste dismantling industry compared with control areas. POPs pollution levels and cancer incidence in both polluted areas and control areas were comparable with the national data. In vitro test and population survey provided evidence that PCBs exposure altered the expression of genes involved in nervous system- and immune system-related diseases, and the CCL22 gene could serve as an effective biomarker for PCBs exposure. Additionally, e-waste management in ZJ province was discussed. Taken together, these data suggest that POPs pollution in ZJ may be correlated to local e-waste recycling activities. In the future, more efforts should be devoted to improve the techniques for e-waste recycling and establish a sound e-waste management framework.

Keywords: e-waste, POPs pollution, health effects, POPs, PCDD/Fs, PCBs

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1. Introduction

Electronic and electric waste (e-waste), referring to obsolete or end-of-life electronic devices such as printers, computers, transformers, television sets, and mobile phones, has become a global concern due to the release of toxic contaminants during the disposal and recycling processing. In recent years, with the rapid economic and technologic development, the amount of e-waste is steadily increasing. It has been estimated that more than 50 million tons of ewaste are generated each year worldwide, and in the United States, over 500 million comput‐ ers become obsolete between 1997 and 2007 [1, 2]. According to statistics, 50–80% of the ewaste from developed countries is legally or illegally exported to developing countries in Asia, 90% of which is transported to China [3]. A recent study reported that 75% of the e-waste from the United States has been transported to southern regions in China, such as Guangdong and Zhejiang (ZJ) provinces [4]. Nevertheless, in the developing countries, rude and uncontrolled e-waste disposal leads to release of considerable amounts of hazardous contaminants into the environment, creating an emerging environmental problem.

Pollutants released during e-waste recycling processing include various heavy metals and persistent organic pollutants (POPs). In recent decades, POPs pollution and the relevant environmental effects associated with e-waste disposal and recycling activities have received growing public attention. These pollutants enter the environment through atmospheric precipitation or surface runoff. Documented studies reported high residue levels of poly‐ chlorinated biphenyls (PCBs), polychlorinated dibenzo-p-dioxins and dibenzofurans (PCDD/ Fs), polycyclic aromatic hydrocarbons (PAHs), and polybrominated diphenyl ethers (PBDEs) were detected in the soil and water column around e-waste dismantling and recycling sites [5, 6]. POPs in the environment enter biological system through food chain, posing great risk to the health of wildlife and human. Knowledge of sources and prevalence of POPs in environ‐ ment and biota are essential to reduce POPs discharge and to diminish environmental burden and human health risk. Recently, a number of researches have been devoted to chemical analysis of POPs in the environment media and biota, and assessment of their toxicity and the mechanistic basis. However, systematic research concerning the sources, prevalence, and health effects of POPs on a large regional scale is limited.

ZJ province, located in the southeast coast of China (**Figure 1**), is well developed in agriculture and industry. In the littoral zone of this province, a cluster of small towns and villages have become intensive e-waste disassembly and recycling centers. Recent studies provided evidence of serious POPs pollution in these coastal areas of this province, voicing public concern over the environmental health effects of e-waste recycling activities [7–9]. However, to the best of our knowledge, there was no systematic investigation about the regional distributions and human health effects of POPs on the whole province scale, 105.5 thousand km² . Moreover, the correlation between the prevalence of major POPs and the incidences of cancers in ZJ province remains unknown.

In the present study, systematic analysis was carried out to identify the burden of major POPs, including DDT, PCBs, PCDD/Fs, PCP/PCP-Na in the environment media, local foods, and human body in ZJ province. As well, the potential link between POPs prevalence and cancer incidence was determined using epidemiological surveys and laboratorial experiments. Furthermore, e-waste disposal and management in ZJ province were discussed.

Figure 1. Sampling locations in Zhejiang province.

2. Distribution of POPs in the environment

2.1. Major POPs levels in environmental media (air, soil, etc.)

2.1.1. PCBs levels in atmospheric particulate matter (PM10)

In the period from August to December in 2009, PM10 samples were collected in ZH, LQ, and LY areas. The sampling sites in ZH and LQ were about 1 km downwind of a dismantling area. LY was selected as the control area. The concentrations of total PCB congeners in PM10 were 348 ng/g dw (5.16 pg/m³), 499 ng/g dw (92.4 pg/m³), and 1139 ng/g dw (127 pg/m³) in LY, LQ, and ZH, respectively. The data of ZH and LQ areas were significantly higher than other areas reported in China, but comparable to those of developed countries.

2.1.2. PCDD/Fs and PCBs levels in soil

Based on documented data about the schistosomiasis history of ZJ province, soil samples (500 g) in 10 areas, that is, CS, JX, JH, YJ, TZ, YH, TT, LY, ZH, and DY, were collected and stored at −20°C for analyzing PCDD/Fs. Concomitantly, sediments in ponds or lakes surrounding the sampling sites were also collected. Both soil and sediment samples were dried at room temperature, freeze-dried, and grinded to pass a 200-mesh sieve for further analysis.

Table 1. Total concentrations of 17 kinds of PCDD/Fs and 18 kinds of PCBs (pg/g dw) in soil of ten regions.

A total of 50 soil samples were collected (a mixture sample of five individual samples in each area), and the residue levels of 17 kinds of PCDD/Fs were analyzed (**Table 1**). The detected PCDD/Fs concentrations were in the range of 98–5099 pg/g dw, with the mean to be 1205 pg/g dw. The highest concentration of PCDD/Fs was in CS, which was about 2- to 50-fold higher than those of other areas. OCDD was the predominant congener, accounting for 66.9– 95.8% of the total concentration, which suggested the identical source of PCDD/Fs in all sampling areas. Relatively lower levels (4.5 pg/g dw) were found in the soil in Beijing [10].

According to the results in our study, PCDD/Fs contamination in CS and JD district of JH was worse than that in other areas, which may arise from historic contamination. More specifically, CS and JH were the two areas of high schistosomiasis incidence in the past. Sodium penta‐ chlorophenate (PCP-Na) has been widely used in these two areas for controlling oncomelania in the last decades, unintentionally resulting in the formation of the main by-product of PCDD/ Fs.

Additionally, levels of 18 kinds of PCBs in the soil samples were determined (**Table 1**). PCBs concentrations ranged from 1100 to 72,156 pg/g dw, with the mean of 14,553 pg/g dw. LQ showed the highest PCBs concentration, which was about 2- to 70-fold of the concentrations in other areas. Comparatively, PCBs levels in these areas were lower than those in heavily polluted regions reported by Chu et al. (430–788 ng/g) [11], but higher than those detected in the soil of Beijing (0.39–13 ng/g, mean of 3.1 ng/g) [12], Qingdao (3.06–14.88 ng/g, mean of 8.04 ng/g) [13], and Yangtze River Delta (mean of 1636.8 ng/kg in rice field, 919.2 ng/kg in vegetable field, and 553.5 ng/kg in historical vegetable field) [14].

The data revealed PCBs contamination was most severe in YJ and LQ. This may be explained by the fact that LQ was an area of intensive e-waste dismantling plants, and rough management, open burning, and random discharge of industrial waste resulted in heavy environmental pollution of PCBs.

2.1.3. PCP/PCP-Na and DDT levels in soil

As presented in **Table 2**, PCP/PCP-Na contamination in the five sampling areas was relatively mild at concentrations of 0.4–1.9 ng/g dw, which were far lower than the standard value of the former Soviet Union (0.5 μg/g). The residue levels in different areas were in an ascending order as YJ, JH, YH, CS, and LQ. Higher level detected in LQ was speculated to be implicated with the local e-waste dismantling industry. Additionally, with the exception of AX, PCP/PCP-Na concentrations in other seven sites in YH area were in the same order of magnitude (**Table 3**).

Note: These data were determined by the authors. n, number of soil samples.

Table 2. Concentrations of PCP/PCP-Na and DDT (ng/g dw) in soil of five regions.

Table 3. Concentrations of PCP/PCP-Na and DDT (ng/g dw) in soil of several YH areas in HZ.

Note: These data were determined by the authors. n, number of soil samples.

As for DDT, notable difference was observed in the residue levels in these areas, with concen‐ trations of different areas in an ascending order of JH, YH, YJ, CS, and LQ (**Table 2**). LQ was found to have the highest level of p, p' –DDT. o, p' –DDT was not detected in YH district in HZ except DT (**Table 3**).

2.2. Major POPs levels in food and fish

It has been well recognized that more than 90% of human exposure to POP is attributed to food consumption. Estimation of POPs levels in food is the most important for risk assessment of POPs to human health. During 2009, residues of major POPs were monitored in late rice, wild crucian, and eggs in ten areas in this province (**Table 4**). PCBs concentrations were also determined in seafood, breast milk, and dairy products [15, 16].

Table 4. PCBs concentrations (pg/g dw) and TEQ (pg/kg) in late rice, egg, crucian in different areas (n = 5).

2.2.1. PCBs levels

Due to the low-fat content in rice, PCB concentrations in late rice were shown to be at a low level, ranging from 31 to 807 pg/g dw. Highest concentration was detected in LQ, implying severe environmental pollution in this area. Undoubtedly, eggs and wild crucian in these areas, containing high-fat content, were found to have more PCB accumulation.

The LQ district of TZ, one of the large-scale e-waste dismantling areas in southern China with a 20-year history for dismantling, has been heavily polluted by PCBs. Random discharge of untreated transformer oil containing PCB mixtures, as well as open burning of plastic pipe, might be the important reasons for PCBs pollution in the soil. In recent years, due to improved dismantling technique and integrated management of dismantling industry, the polluted land has been partially restored and the soil ecosystem has been improved. However, due to persistence property, PCBs can highly bioaccumulate in various organisms through food chain. The concentration of total PCBs detected in wild crucian in LQ reached 700 ng/g dw, far above other areas. Moreover, diet survey was conducted to identify PCBs exposure of population via food intake in these areas. The results showed that consumption of fish caused an average exposure of 60.4 pg/WHO-TEQ/kg per person per day, far exceeding the WHO standard value 4 pg/WHO-TEQ/kg, implying that the wild crucian in this area was not fit for consumption. PCBs residue levels in different areas were found in the order of LQ > HY > WL in rice, LQ > YH > LH > WL in eggs, and LQ > HY > WL > YH in crucian.

2.2.2. PCP/PCP-Na levels

Residue levels of PCP/PCP-Na in wild crucian of different areas ranged from 0.49 to 0.75 ng/g, in the order of JX > YH > LQ (**Table 5**). No significant difference of PCP/PCP-Na concentrations was observed among the six sampling sites in YH district in HZ (**Table 6**).

Note: These data were determined by the authors. n, number of soil samples.

Table 5. Concentrations of PCP/PCP-Na and DDT (ng/g dw) in crucian in some regions.

Note: These data were determined by the authors. n, number of soil samples.

Table 6. Concentrations of PCP/PCP-Na and DDT (ng/g dw) in crucian in several YH areas in HZ.

2.2.3. DDT levels

The total DDT concentrations in crucian of three areas were in an ascending order as YH, LQ, and JX. Among all DDT congeners, o, p′ –DDT had the lowest residue level and was not detected in fish sampled in JX. Obvious difference of DDT concentrations was found in fish collected in the six sampling sites. In GZ, only p, p' –DDE was detected. O, p' –DDT was not detected in all sites except DH (**Table 6**). DDT levels in fish were shown in the order as DH > WH > YH > LZ > DT > GZ.

2.3. Total toxic equivalents (TEQs) of PCBs and PCDD/Fs

As shown in **Figure 2**, TEQs in various foods of LQ were higher compared with YH, and crucian was shown to have the highest TEQ (10.87 pg/g ww). TEQs detected in other food were 3.77 pg/g ww in duck meat, 2.80 pg/g ww in egg, 2.43 pg/g ww in chicken meat, 0.08 pg/g ww in rice, and 0.22 pg/g ww in vegetable. In YH area, total TEQ was shown to be highest in duck meat (0.74 pg/g ww), and TEQs in other food were 0.69 pg/g ww in egg, 0.55 pg/g ww in crucian, 0.44 pg/g ww in chicken meat, 0.002 pg/g ww in vegetable, and 0.0002 pg/g ww in rice. The results revealed that PCBs TEQ in animal-originated food in LQ and YH shared the same order as crucian > egg > chicken and duck meat. Regarding PCDD/Fs, total TEQ was different in the two areas, with the order of duck > crucian > chicken > rice > vegetable in LQ and duck > egg > chicken > crucian > rice > vegetable in YH.

The data indicated that, apart from eggs in LQ and crucian in LQ and YH, the other kinds of food made great contribution to the total TEQ value.

3. Human body load of major POPs

Health effects of environmental contaminants on humans and wildlife are usually assessed through external exposure test, and the exposure levels of human population to toxicants are generally estimated by using equations with parameters for exposure routes (oral, dermal, or inhalation), on the basis of the analytical data of toxicant concentrations in ambient environmental media (air, water, or food). Estimates based on external exposure and multiple hypotheses often have big error, since just the approximate doses received by organisms are predicted. The predicted exposure doses generally deviate from the absolute internal exposure level because there are many undefined factors. Assessing toxicity of environmental pollutants by external exposure cannot provide insightful information for environmental conservation, human health protection, and formulation of law and regulation. Study of health effects by monitoring the internal exposure has become a significant and effective mean for risk assessment of environmental pollutants.

Biological monitoring is an effective mean for identifying internal exposure levels by using advanced analytical techniques to measure the concentrations of parent chemicals and metabolite in the whole body or tissues. Due to the advantages of speediness and exactness, biomonitoring has become the important mean for measuring internal exposure doses in biological system. Biomonitoring data provide scientific basis for establishing environment sanitary criterion and medical diagnosis standard and assessing the effectiveness of public health measures.

3.1. Body burden of major POPs in special population

Residue levels and fingerprint of PCDD/Fs and PCBs were determined in fat, breast milk, and blood of general population in this province. The average total concentrations in fat, breast milk, and children's blood samples were 108, 55.0, and 208 pg/g lipid for 17 kinds of PCDD/Fs; 32.8, 8.0, and 9.8 ng/g lipid for all 12 kinds of DL-PCBs; and 154, 15.8, and 28.3 ng/g lipid for all indicator PCBs. The TEQs in these samples were 9.22, 3.09, 11.7 pg/g lipid for PCDD/Fs, and 16.2, 3.56, 11.9 pg/g lipid for PCBs. Similar pattern of PCDD/Fs and PCBs fingerprint was obtained in several kinds of food (fish and eggs) and in human body, implying that food consumption was the main route for human exposure to these POPs. PCDD/Fs concentrations in this study are notably different from those detected in the body fat of westerners, which may be implicated with different eating habits between easterners and westerners.

Body load of PCBs and PCDD/Fs was investigated in occupational population and specific population. Analytical data indicated 90% detection rate of these pollutants in the cerumen of occupational population in dismantling areas and 50% detection rate in non-occupational population. No PCBs but low levels of DL-PCBs were detected in the control subjects. Significant difference of PCBs levels was observed between population in dismantling areas and control group. Data of correlational analysis revealed a positive correlation between PCBs levels in cerumen and service length of workers.

3.2. Concentrations and TEQ of PCDD/Fs and PCBs in body fat

A total of 24 body fat samples were collected, numbered and stored at −20°C for chemical analysis.

3.2.1. PCDD/Fs levels

Concentrations and TEQ of 17 kinds of PCDD/Fs and 18 kinds of PCBs in body fat are shown. PCDD/Fs concentrations were in the range of 33.9–504 pg/g lipid, with mean of 108 pg/g lipid. These data are comparable to those reported in Spain (109 pg/g lipid) [17], higher than Turkey (73.3 pg/g lipid) [18], but lower than Japan (171 pg/g lipid) [19]. Kiviranta et al. [20] reported PCDD/Fs concentrations in human body fat ranged from 171 to 1180 pg/g lipid. As for PCDD/ Fs, OCDD was found to be the predominant congener, accounting for 68% of the total concentrations. Other main congeners included 2,3,4,7,8-PeCDF, 1,2,3,6,7,8-HxCDD, 1,2,3,4,6,7,8-HpCDD, and 1,2,3,4,7,8-HxCDF, respectively constituted 6.61, 4.14, 3.62, and 3.23%. The proportional composition of PCDD/Fs is similar to that reported in other countries.

TEQ in fat was calculated using the revised WHO TEQ factor (WHO-PCDD/F TEF 98, 05) [21, 22]. In this study, the average TEQ for WHO-PCDD/F TEF 98 was 9.22 pg/g lipid (1.64–20.3 pg/g lipid), comparable to the data in Turkey (9.2 pg/g lipid) [18], Japan (11.9 pg/g lipid) [19], Korea (12.8 pg/g lipid) [23], and India (14.4 pg/g lipid) [24], but significantly lower than European countries (17.8–48 pg/g lipid) [20]. Numerous studies provide evidence that food consumption is the main route for PCDD/Fs exposure. Due to different dietary habits, consumption of animal-originated food by easterners is far less than westerners, which may be one of the most important reasons for the different PCDD/Fs residues in body fat [25].

3.2.2. PCBs levels

Both DL-PCBs and indicator PCBs were detected in all fat samples. The average total concentration of PCDD/Fs was 32.8 ng/g lipid (4.11–125 ng/g lipid), comparable to those reported in Japan (29.8 ng/g lipid) [19], higher than the levels in Turkey male fat (14.0 ng/g lipid) [18], but lower than the levels in women from Spain (56.0 ng/g lipid) [26] and those detected in south China (237 ng/g lipid) [27]. PCB118 was found to be the predominant congener of DL-PCBs, followed by PCB156 and PCB105. All the three PCBs made up 77.6% of the total DL-PCBs concentrations. In other studies, consistent results were obtained, showing PCB118, PCB156, and PCB105 are the main congeners of all DL-PCBs [18, 19, 26].

As for indicator PCBs, previous studies indicated PCB153 was the predominant congener, which had been detected in all environmental media. A recent study in Europe reported PCB153 concentration in human body fat was 232 ng/g lipid, and all indicator PCBs levels were 389–855 ng/g lipid with a mean of 606 ng/g lipid [20]. In our study, indicator PCBs concentra‐ tions were 8.75–745 ng/g lipid, with mean of 154 ng/g lipid. Consistent with other studies, PCB153 was the predominant congener, but its concentration (52.5 ng/g lipid) was far lower than that detected in Europe.

As regards the PCBs TEQ, PCB126 (83.8 pg/g lipid) made a major contribution (90%) to the total TEQ. The detected PCBs TEQ was in the range of 1.4–61.6 pg/g lipid with mean of 16.2 pg/g lipid, which is higher than that of other countries in Asia, but lower than that of developed countries in Europe (**Figure 3**).

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Figure 3. Comparison of PCDD/Fs- and PCBs-TEQ in human body fat from various countries [17–19, 23, 24, 28–30].

3.3. Concentrations and TEQ of PCDD/Fs and PCBs in breast milk

Breast milk contains fatty acid, protein, endogenous hormone, and antibody, which are essential for infant growth and development. WHO recommends exclusive breastfeeding for infants in the first 6 months of life. However, there is a lot of evidence indicating that many POPs, such as PCDD/Fs and PCBs, are transferred from mother to infant via breast milk which is rich in fat. Detection of PCDD/Fs and PCBs levels in breast milk not only reflects the exposure risk of local population to these pollutants, but also indicates the health effects on infant by breastfeeding. The organization of Stockholm Convention has evaluated the impact of implementing emission reductions of POPs, based on the monitoring data of POPs in breast milk worldwide. WHO has successively initiated three programs for monitoring breast milk, whereas, in China, apart from Hong Kong which participated in the third program initiated by WHO, quite limited efforts have been made to monitor POPs in breast milk [31]. POPs levels in breast milk have been reported in e-waste dismantling areas, while systematic study is scarce. In this study, a total of 74 breast milk samples collected in areas of no e-waste dismantling industry have been monitored to identify the PCDD/Fs and PCBs levels in general population [32]. The breast milk samples (25–100 ml each) were numbered and stored at −20°C, then freeze-dried, grinded, and sealed for further pretreatment and analysis.

3.3.1. PCBs levels

The detection limit for PCBs analysis was 0.05 pg/g lipid, and the recovery rate of isotope internal standards was 58–89%. Analysis of the blank control and the standard reference material (WMF-01) conformed to the requirement for quality control. All six kinds of indicator PCBs and 12 kinds of DL-PCBs have been detected in all breast milk samples. The mean of total concentration and total TEQ for PCB congeners were $23,881 \pm 9718$ pg/g lipid (13,643– 45,205 pg/g lipid) and 3.56 \pm 1.06 pg/g lipid (2.92–6.31 pg/g lipid), respectively. The top five congeners include PCB138, PCB153, PCB118, PCB180, and PCB105, respectively, accounting for 37.58, 19.07, 11.79, 7.70, and 7.00% of the total concentration. The proportional composition of PCB congeners is shown in **Figure 4**.

A previous study determined PCBs levels in breast milk of general population in 12 regions of China [31], showing PCBs levels in industrially developed areas were significantly higher than those in underdeveloped areas. The data of our country are relatively lower than those of developed countries (4.9–57.2 pg/g lipid) [31]. The worldwide PCBs levels in breast milk have regional difference. For instance, the data in East Asia (China, Korea, and Japan) are very similar, but much lower than those in US and European countries (10–100 pg/g lipid). Different dietary habit may partially explain this difference. It is well known that 90% of POPs including PCBs in human body is obtained via food consumption, especially animal-originated products which are mostly favored by westerners. Our recent studies also found PCBs fingerprints detected in food and body tissues were accordant [21, 34], providing evidence of the major role of food consumption in human PCBs exposure.

In addition, PCBs levels in breast milk were analyzed in different age groups. However, due to narrow age range (21–30), no positive relevance was observed between PCBs concentrations and age (R2 = 0.220). Further investigation will be conducted in population of wide age range.

3.3.2. PCDD/Fs levels

Levels of PCDD/Fs in breast milk from urban and rural residents were measured to determine whether regional environment had effects on body load of POPs. Generally, our results showed lower PCDD/Fs levels in breast milk in our country compared with developed countries, consistent with the nationwide data [31]. It should be noticed that the data of urban groups $(71.4 \pm 40.8 \text{ pg}$ /lipid, n = 23) were significantly higher than those of rural groups $(38.6 \pm 38.1 \text{ m})$ pg/lipid, n = 51). Consistently, previous studies revealed that the data of developing countries were lower than those of developed countries [31, 35–38]. Numerous POPs, such as PCDD/Fs and PCBs, are mostly the by-products of industrial activities, and they enter the ecosystem mainly via atmospheric precipitation and surface runoff, transfer and bioaccumulate via the food chain, and eventually accumulate in human body. Although a limited number of samples have been analyzed in our study, the data absolutely indicate a positive correlation between the body load of POPs and the local industrialized levels.

3.4. Concentrations and TEQ of PCDD/Fs and PCBs in human blood

Occurrence of POPs in the environment may originate from multiple sources. For instance, PCDD/Fs can be formed during natural events such as volcanic eruption, or be created by industrial process such as exhaust emission by steelmaking industry and waste incinerating factory or sewage discharges by paper mill. Statistics show that over one million tons of ewaste are generated annually, 70% of which are introduced in China for dismantling and recycling heavy metals such as copper and gold. LQ and GY are the top two biggest e-waste dismantling areas in China. In LQ, over 60 thousand of people work on e-waste dismantling industry, generating 1.4 billion dollar a year. However, this pillar industry brings about potential hazard to the environment accompanied with enormous economic benefits. Com‐ pared with the adults, children have the least chance of occupational exposure to POPs. POPs concentrations in children's blood can exactly reflect the health effects of environmental pollutants. Therefore, our study for the first time analyzed POPs concentrations in children's blood in these areas. Briefly, blood samples of children were collected by the local Centers for Disease Control in LY, LQ, and TT. LQ was considered as the heavily polluted area because of the intensive e-waste dismantling industry, and TT and LY are selected as mildly polluted area and control area, respectively [39].

3.4.1. PCBs levels

The average concentration of PCB mixtures in children's blood sampled in LQ was 40.6 ± 7.01 ng/g lipid, higher than that in LY (20.7 \pm 6.90 ng/g lipid) and TT (20.7 \pm 8.09 ng/g lipid). This result indicated more PCBs intake of children in LQ through various routs such as food consumption, implying serious pollution in LQ. Improved dismantling techniques, rigorous

regulatory process, and scientific guideline in the dismantling industry of LQ are required to alleviate the environmental effects.

Our results are consistent with the data reported previously [40]. DL-PCBs concentrations in LQ, LY, and TT were 16.0 ± 3.32 , 7.32 ± 3.53 , and 6.68 ± 3.05 ng/g lipid, respectively, far lower than the levels in cord blood reported by Zhao et al. [40] (348 ng/g lipid) but higher than the levels in pregnant women's blood in Japan (5.9–34.3 ng/g lipid) [41, 42]. Due to lipophilic property, PCBs tend to accumulate in tissues of high-fat levels, such as lipid and breast milk. It has been reported that worldwide PCBs levels, including indicator and DL-PCBs, were 30– 1800 ng/g lipid in breast milk [43] and 389–4242 ng/g lipid in human fat tissues [20]. The predominant congeners in children's blood included PCB118, PCB105, PCB153, PCB138, and PCB28, which was consistent with the results previously detected in lipid tissues.

3.4.2. PCDD/Fs levels

Different from the results of PCBs, PCDD/Fs levels in children's blood of the heavily polluted area LQ (206 \pm 157 pg/g lipid) were shown to be higher than the moderately polluted area TT $(160 \pm 102 \text{ pg/g lipid})$, but lower than the control area LY $(282 \pm 261 \text{ pg/g lipid})$. Integrated analysis of the data for the three areas was performed, and the average PCDD/Fs level in children's blood was 208 ± 172 pg/g lipid (54.4–784 pg/g lipid) (**Figure 5**). Our previous study also revealed that PCDD/Fs levels in food (primarily fish and egg) in LY were about 3- to 5 fold of those in LQ. In 1960s, PCB-Na was abundantly produced and extensively applied to control schistosomiasis in LY. But during the production of PCP-Na, a large amount of PCDD/ Fs was generated. PCDD/Fs are persistent and can bioaccumulate through food chain, eventually enter human body [44]. These results in our studies indicate there still exists great health risk to the environment and population due to historically widespread application of PCP-Na in LY.

Figure 5. Comparison of PCBs concentrations (ng/g lipid) in children's blood between polluted areas (LQ) and control areas (LY and TT) [39].

Comparatively, in Korean, PCDD/Fs levels in the blood were reported to be 12.3 pg/g lipid, 10-fold less than the results of our study [23]. However, our results were comparable to the data reported in pregnant women's blood in Japan (196 pg/g lipid) [42]. Additionally, the data of LQ were comparable to those reported previously. Because of the least chance of occupa‐ tional exposure for children, the data detected in children's blood more likely reflect human health effects of pollutants.

4. PCDD/Fs and PCBs pollution characteristics and correlation analysis

As stated above, POPs are persistent and lipophilic, can migrate globally via atmospheric precipitation and water flow, and bioaccumulate through food chain. In Western countries, due to a long history of production and application of PCB/PBDE, POPs levels in food are commonly higher than developing countries. It is generally believed that food consumption is the main route for human exposure to POPs. Other exposure routes include air breath, skin contact, and mother-to-child transmission.

Due to persistent property of POPs, it is speculated that POPs pollution profile in different environmental media is stable. Study of POPs in various environment media is of great significance for fingerprint analysis and understanding of sources and transport of pollutants.

In our study, systematic analysis was conducted on POPs residues in environmental media (source water, soil, sediment, air), food (eggs, rice, freshwater fish, vegetable, livestock), and body tissues (breast milk, blood, and fat). Pollution characteristic of PCDD/Fs and PCBs in different environmental media was further analyzed.

PCB congener compositions in different media are very similar. All 6 kinds of indicator PCBs and 12 kinds of DL-PCBs were detected in breast milk. The predominant congener PCB138 makes up 32.86% of total concentration. The abundance of other major PCB congeners was in a descending order as PCB153 (26.85%), PCB118 (14.43%), PCB28 (8.61%), PCB180 (5.89%), and PCB105 (5.44%). These major PCB congeners account for 94.08% of total concentration. In body fat, the abundance of major congeners was in the order as PCB153 > PCB 138 > PCB 180 > PCB 118 > PCB 28 > PCB 105, totally consisting 94.34%. In human blood, PCB28 was most abundant. Notably, the proportion of PCB180 in fat was 20.24%, higher than that in breast milk (5.89%) and blood (4.01%), which can be explained by the fact that high-chlorinated congeners more easily bioaccumulate in high-fat tissues [45].

PCBs residues in seafood, eggs, and freshwater fish which are universally consumed by local people have been analyzed to determine the sources of PCBs in human body. The major six kinds of PCB congeners make up 79.67, 88.09, and 80.71% of total concentration, respectively, in seafood, eggs, and fish. The fingerprints of the major PCB congeners in food and human body were very similar, suggesting food consumption was the main route for human PCBs exposure. In both food and human tissue samples, OCDD was the most abundant congener, and the levels of other PCDD/Fs congeners were relatively low.

Additionally, our study showed that the fingerprints of PCBs and PCDD/Fs in body fat, breast milk, and blood in general population were very similar to those in main food samples such as fish and eggs, implying the main contribution of food consumption to PCDD/Fs exposure

of general population. We speculate that there is a possibility to control the hazard of PCDD/ Fs to human health by adjusting the diet structure.

5. Body burden in general population and special population

Analysis of PCBs and PCDD/Fs in peripheral blood was conducted in healthy population in LQ, LY, TT, YH, and ZH. LY showed the highest TEQ value, followed by LQ, TT, YH, and ZH. The results of PCBs and PCDD/Fs fingerprints were consistent with those stated above.

Additionally, PCBs concentrations in breast milk, umbilical cord blood, and mothers' and children's blood in polluted sites were higher than the reference sites. The TEQ values of PCBs and PCDD/Fs in breast milk were also higher in polluted sites.

In the dismantling areas, cerumen from occupational population or non-occupational popu‐ lation was collected for analysis of PCBs and DL-PCBs (n = 30). Cerumen of farmers in TY town about 10 km far from the polluted area was considered as control group ($n = 30$). The detection rate was 90% in occupational population and 50% in non-occupational population in the dismantling area. No PCBs have been detected in the control groups. The difference of PCBs and DL-PCBs between the three groups was significant. Correlation analysis revealed a positive relevance between PCBs levels and seniority of dismantling workers.

6. Correlation between major POPs pollution and cancer

6.1. Statistics of cancer incidence and financial loss

Prevalence of diseases, as well as cancer-related financial loss and mortality, has been estimated in TZ and YH. In both regions, the total number of patients and total medical expense in both polluted area and control area increased over time during 2004–2010, whereas the proportion of cancer patients and cancer-related financial loss remained constant during these years. Statistical data indicated that cancer incidence during 2004–2010 was 2.6% in polluted area and 3.4% in control area. The growing number of cancer patients and increasing financial loss were hypothesized to arise from the increasingly improved social security system and self-health-care consciousness. The cost for cancer therapy in the control area during 2004– 2010 totaled 8 million RMB Yuan with per capita cost of 9.2 thousand RMB Yuan, while in the polluted area, these data were shown to be higher with total cost of 17.96 million RMB Yuan and per capita cost of 12 thousand RMB Yuan.

In addition, retrospective epidemiological study was conducted to determine the morbidity and mortality of cancer and other kinds of disease in this province by stratified sampling. The statistical data indicated no significant difference between the polluted area and control area (data not shown).

6.2. Biomarker responses by exposure to major POPs

In this section, in vitro cell culture experiments, coupled with population survey, were conducted to screen the sensitive biomarkers following PCBs exposure by testing gene expression.

6.2.1. In vitro experiment

In vitro, effects of PCB153, the predominant congener in the environment, and biota in this province, on gene transcription profile in human B lymphoblasts, were investigated using gene chip technique (Human-12T Beadchip, Illumina) [46]. The data indicated PCB153 exposure caused notable change in the transcription level of 161, 191, and 1006 genes, respectively, at concentrations of 25, 100, and 200 μmol/L. Among these genes, 15 genes' expression was altered by PCBs at all exposure concentrations, specifically, upregulation in 4 and downregu‐ lation in 11 genes. These results were further validated by real-time PCR assay, and we found CCDC92 and TMEM175 were upregulated while CCL22, STK38L, and GZMK were downre‐ gulated following varying exposure periods.

It has been reported that CCDC92 and TMEM195 influence the function of B lymphocyte and T lymphocyte, respectively. CCL22 regulates immune system. Altered CCL22 expression was reported to be potentially associated with cancers. GZMK plays critical role in clearing virus and tumor cells. Altered transcription of STK38L impacted cell cycle and encouraged apoptosis. Therefore, these in vitro tests implied PCB153 potentially disrupted the transcription level of genes relevant to immune system and cancer.

6.2.2. Population survey

The expression of five genes which had altered transcription in the in vitro test, including CCDC92, TMEM175, CCL22, STK38L, and GZMK, was determined in population survey. Furthermore, the transcription levels were compared between the polluted area and the control area [47].

Peripheral blood (2 ml) from population in dismantling areas and in pollution-free areas was collected $(n = 60)$. The subjects were grouped by age. Total RNA in peripheral blood from men and women aged 30–40 and 50–60 years was isolated, with three subjects in each group. Quality-inspected RNA samples from three subjects in the same group were mixed for further analysis of gene chips using Human-12T Illumina Beadchip.

The results showed that CCL22 expression declined in PCBs exposure subjects compared with the control group, consistent with the data of in vitro test [48]. GZMK and MTDH expression was upregulated, but the expression of CCDC92, STK38L, and TMEM175 had no change in PCBs exposure groups. The CCL22 gene, located on the q arm of chromosome 16, is a member of the cys-cys (CC) chemokine family, encoding proteins critical for chemotactic activity of monocyte, dendritic cell, NK cell, and T cell. CCL22 primarily functions to regulate immune system by influencing biological process of T lymphocyte, such as transfer of T lymphocyte to the inflammatory sites. Previous studies demonstrated that several kinds of inflammatory diseases, such as atopic dermatitis, rheumatoid arthritis, psoriatic arthritis, and osteoarthritis, occurred with dysregulated expression of CCL22. Moreover, aberrant CCL22 expression was reported to be related with hepatitis C virus infection, acute leukemia, lung cancer, gastric cancer, abdominal aortic aneurysm, and esophageal squamous cancer. PCBs exposure may be correlated with atopic dermatitis, rheumatoid arthritis, osteoarthritis, chronic hepatitis, lung cancer, gastric cancer, and belly aneurysm. In the present study, downregulated CCL22 gene expression was found in PCB153-exposed human B lymphoblasts and in the peripheral blood of PCBs exposure population, suggesting PCBs might disturb the function of immune system by inhibiting CCL22 and eventually cause inflammatory diseases. Nevertheless, further studies are required for investigation of the underlying mechanisms.

The transcription of 68 genes showed significant difference between PCBs exposure popula‐ tion and control population, including 37 upregulated genes and 31 downregulated genes. These genes were found to distribute on all except chromosomes 6, 10, 16, 20, x, and y. They are primarily involved in ribosomal peptide synthesis, pathogenic bacterial infection, cytoskeleton actin regulation, insulin signal pathway, Jak-STAT signal pathway, and endocytosis. Sexually, there were 21 genes in men showing different expression between PCBs-exposed group and control group, including 10 upregulated genes and 11 downregulated genes. These genes mainly function to regulate the signal pathway of ribosome, cytoskeleton actin, and phagocytosis. In women, 316 genes had significant change in expression level in PCBs-exposed population, including 181 upregulated and 135 downregulated genes. These genes primarily regulate ribosome, metabolism, oxidative phosphorylation, Alzheimer's disease, Parkinson's disease, cytoskeleton actin, Huntington chorea, cancer, chemokine signal transduction, smallcell lung cancer, infection pathogens, Jak-STAT signal pathway, and endocytosis. Our study demonstrated that there was sexual difference in response to PCBs exposure, and women were more susceptible. The expression of genes associated with nervous system in women was altered, which was not observed in men.

In general, the results of the present study indicated PCBs exposure caused altered expression of genes involved in nervous-system- and immune-system-related diseases and cancers, despite inconsistent results of in vitro cell culture test and population survey. Additionally, our study showed women were more sensitive to PCBs exposure, and CCL22 might serve as a powerful and effective biomarker of PCBs exposure.

6.3. Animal experiments: rat, mouse, and zebrafish

6.3.1. Toxicity of circuit board powder to male mice

In this section, circuit board powder was used to elucidate the toxic effects of pollutants in ewaste, including heavy metals and various POPs, on organs of male mice, especially the reproductive system [49]. Male ICR mice were fed either normal chow diet or mixed diet containing circuit board powder. The weight and food intake were recorded periodically. After 90-day exposure, the animals were sacrificed and the organs, including brain, heart, liver, spleen, lung, kidney, and testis, were weighed and histopathologically examined. Organ coefficients were calculated, and PBDEs levels in the liver and brain were determined. Sperm motility and the relevant kinetic parameters were measured, and the expression of Connexin43 protein in testis was tested by immunofluorescence. The acute oral LD50 of male mice after 24-h exposure to circuit board powder was higher than 10,000 mg/kg. After a 90-day sub‐ chronic exposure, the organ coefficients of liver (4.63 \pm 0.39), kidney (1.72 \pm 0.29), brain (1.02 \pm 0.13), and lung (0.51 \pm 0.04) in exposed groups were significantly higher than those in the control group (liver: 3.99 ± 0.42 , kidney: 1.38 ± 0.16 , brain: 0.85 ± 0.15 , lung: 0.46 ± 0.06). Pathological damage occurred in the liver and kidney of exposed animals. PBDEs levels in liver (175.54 ng/g ww) and brain (29.60 ng/g ww) of exposed animals were enormously elevated compared with those in control group (liver: 2.16 ng/g ww, brain: 0.12 ng/g ww). However, no significant difference was observed between the tested groups and control group in terms of organ coefficient, pathological section and Connexin43 expression in testis, and motility and kinetic parameters of sperm. In general, oral exposure to circuit board powder caused pathological changes in the liver and kidney of adult male mice, but had no toxic effects on the reproductive system.

6.3.2. Chronic oral toxicity of circuit board powder to rat

Toxicity of circuit board powder to rats was investigated by subchronic oral exposure experi‐ ments [50]. Briefly, SD rats were fed either control diet or mixed diet containing circuit board powder at doses of 10, 20, and 50 g/kg. A chronic exposure experiment of 90 days was conducted, followed by a 45-day recovery test. After 90-day exposure period, organ coeffi‐ cients in each group were calculated and blood biochemical indexes were measured. Addi‐ tionally, contents of thyroxine T3, T4, and testosterone (T) were determined after 45- and 90 day exposure and 45-day recovery exposure. The results indicated that there was no statistical difference in body weight between the exposed and control groups. The organ coefficients in exposed female rats were significantly higher than those in control animals. After 45- and 90 day exposure, the contents of T3, T4, and T were significantly increased in all groups exposed to the circuit board powder when compared to the control. No difference of T3, T4, and T contents was observed between higher dose groups and control group following 45-day recovery exposure. The findings suggested that exposure to circuit board powder caused notable liver damage and significant increase of plasma T3, T4, and T levels in rats.

6.3.3. Toxicity of circuit board powder leachate to zebrafish

Chemical analysis was conducted to determine the concentrations of heavy metals in circuit board powder and leachate [51]. As expected, a large amount of heavy metals, including nickel, cadmium, iron, copper, manganese, and lead, was detected, with the top three of copper, lead, and iron. However, in the circuit board powder leachate, no lead was detected, and the concentrations of the other five metals were extremely low, suggesting less possibility of water pollution due to circuit board powder stack. Furthermore, toxicity data indicated that circuit board powder had no toxicity to adult zebrafish following 28-day exposure at the tested concentrations.

7. E-waste management in ZJ province

7.1. Status of e-waste recycling

Due to large-amount imports from developed countries, coupled with domestic use, the amount of e-waste in China has been steadily increasing in recent decades [52]. China has been becoming one of the biggest centers for e-waste dismantling and recycling in the world [53].

ZJ province is one of the most developed regions in China, where the replacement of appliances is very fast and the amount of obsolete electronic devices is quite large. In December 2003, the National Development and Reform Commission (NDRC) initiated the national pilot program for e-waste management system, and ZJ was the only province selected to implement the pilot project due to the large-amount e-waste from domestic generation and imports from devel‐ oped countries [54]. Since then, a large number of e-waste recycling centers in ZJ province have been established. The e-waste recycling sites in this province are mostly distributed in the southeast coastal areas [55]. The regions of developed e-waste recycling industry include LQ and WL areas in TZ city, and ZH area in NB city. TZ is the biggest e-waste recycling center in ZJ province with a nearly 30-year history for e-waste disassembly and has one of the largest e-waste recycling facilities in the world [56, 57]. According to the statistical data, more than 100 thousand people in TZ worked on the e-waste recycling activities, and the annual e-waste amount reached 2 million [8]. The e-wastes disposed in these areas mainly include electrical machine, transformer, and electric wire and cable [56].

7.2. Management of e-waste recycling industry

Given the rapid increase of e-waste, as well as the potentially concomitant environmental effects and health risk to human and wildlife [58], it is essential to establish sound and environmentally benign management system for e-waste [59]. Legislative Affairs Office of ZJ province subsequently published Pilot Measures for Recovery Processing of Waste Electrical and Electronic Products in ZJ province. This pilot measures apply to natural person, legal entity, and any other organization engaged in production, use, sell, repair, and import of electrical and electronic equipment or e-waste disposal and recycling activities, and aim to reduce the use of hazardous chemical in electronic appliances and the pollution generated during the manufacture, recycling, and disposal of these products. Furthermore, as the biggest e-waste recycling center of this province, TZ has enforced a series of policy measures and regulations for e-waste management. The Economic and Trade Department of ZJ province also made great contribution to establishment of e-waste recycling and disposal system and implementation of the pilot project in many e-waste disposal sectors. Thus, in recent years, a great advance has been obtained in e-waste recycling management in ZJ province.

7.3. Problems in e-waste disposal and recycling

With the growing amount of e-waste, it has been widely recognized the importance of establishing a sound and regulated e-waste management system in ZJ province. Reclamation of precious materials for reuse from e-waste has practical significance for development of circular economy in ZJ province. Despite recent improvement in e-waste recycling industry, there still exist a lot of problems in e-waste disposal and recycling framework in this province. Specifically, the responsibility of the government, producer, assembler, importer, or dealer for the collection and recycling of e-waste is not very explicit. Secondly, although a great many formal e-waste recycling centers with large-scale solid waste incinerators equipped with exhaust treatment device have been established, due to lack of sound recycling network, a large amount of e-waste flows to informal small-scale family workshops for disposal and recycling using crude and primitive methods such as manual disassembly and open burning, which bring about potential detrimental environmental effects. Thirdly, the cooperative management and joint law enforcement of different sectors are not coordinated and effective. Additionally, the reward system and subsidy system in the provincial government require further improvement to ensure the benefits of business owners of formal e-waste recycling companies and encourage their initiatives. Based on these remained problems, in the future, the e-waste recycling industry should be regulated through establishing sound legislation, such as extended producer responsibility (EPR) legislation, and the informal e-waste disposal and recycling processes should be replaced by large-scale facilities.

8. Conclusion

Our study for the first time determined the residue levels of major POPs in the environmental media, local food, and human body in ZJ province. Fingerprints of PCBs and PCDD/Fs were identified in various kinds of food, body fat, human blood, and breast milk. Body burden of PCBs and PCDD/Fs in special populations (children, women, and occupational population) was compared between polluted sites and reference sites. POPs pollution and cancer incidence in polluted areas and control areas were surveyed, and the correlation was analyzed. Furthermore, the results of in vitro and in vivo tests provided evidence that the CCL22 gene could be used as a effective biomarker for PCBs exposure. Acute toxicity, subacute toxicity, and subchronic toxicity of circuit board powder to experimental animals were as well investigated.

Generally, the present study provided integrated information for the assessment of POPs pollution level in ZJ province. The data suggest that the pollution status of major POPs in the environment is undesirable and should be noteworthy. Especially, the pollution levels in east coastal areas, such as LQ and ZH, were shown to be more serious than the middle and west areas, which may result from the long history of e-waste dismantling activities in these areas. However, due to long-range transport of POPs through atmosphere and biomagnification via food chain, there was mild pollution of PCBs and PCDD/Fs in the midwest areas. In the littoral zone, rude and primitive e-waste recycling processing, such as manual dismantling and open incineration, may be one of the most important reasons for POPs pollution in the ambient environment.

In recent years, due to some techniques improvement in e-waste dismantling industry and the widespread application of pollution control measures, the environmental quality in some areas of intensive e-waste recycling industry has been improved to some extent. However, there still exist many obstacles and challenges involved in combating e-waste and improving the environment in ZJ province. In the future, more efforts should be devoted to propaganda and enforcement of pollution control regulations, and should optimize the e-waste recycling management framework, development of advanced techniques for e-waste disposal and recycling, regular monitoring of environmental pollution level, and implementation of comprehensive health surveillance of the human population for cancer control in this province.

Author details

Nanxiang Wu* , Ye Yang, Yang Song, Ming Gao and Heru Huang

*Address all correspondence to: zamewu@mail.hz.zj.cn

Zhejiang Academy of Medical Sciences, Hangzhou, Zhejiang, China

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