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Dual body burdens of polychlorinated biphenyls and polybrominated diphenyl ethers among local residents in an e-waste recycling region in Southeast China

Xing-Ru Zhao [a,b], Zhan-Fen Qin [a,*], Zhong-Zhi Yang [a], Qian Zhao [a], Ya-Xian Zhao [a], Xiao-Fei Qin [a], Yong-Chuan Zhang [c], Xian-Li Ruan [c], Yin-Feng Zhang [a], Xiao-Bai Xu [a]

[a] State Key Laboratory of Environmental Chemistry and Ecotoxicology, Research Center for Eco-Environmental Sciences, Chinese Academy of Sciences, P.O. Box 2871, Beijing 100085, China
[b] Institute of Water Environment Research, Chinese Research Academy of Environmental Sciences, Beijing 100012, China
[c] Taizhou Hospital, Taizhou City, Zhejiang Province 317000, China

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ABSTRACT

E-waste recycling resulted in serious pollution of polychlorinated biphenyls (PCBs) and polybrominated diphenyl ethers (PBDEs) in Taizhou of Zhejiang Province, China. The aims of this study were to assess dual body burdens of the two pollutants and potential health risk for local residents. Blood samples were collected from two e-waste recycling sites, Luqiao (where PCBs-containing e-wastes were recycled) and Wenling (where PBDEs-containing e-wastes were recycled). The mean ∑PCBs (CB-105, 118, 153, 183, and 180) and ∑PBDEs (BDE-28, 47, 99, 100, 153, 154, 180, and 209) were 204.20 and 117.58 ng g^{-1} lipid in the blood from Luqiao, respectively, while they were 83.80 and 357.44 ng g^{-1} lipid from Wenling, respectively. The PCBs levels among Luqiao residents were comparable to the values reported for US populations, while the PBDEs levels among two study populations were higher than the values from US populations. This is the first report to present dual body burdens of PCBs and PBDEs at so high levels. Based on previous epidemiologic data, it is suggested that dual burdens of PCBs and PBDEs at so high levels might pose health risk for local residents. In addition, no correlation between PCBs or PBDEs concentrations and the ages of the volunteers was observed in the two populations, which was explained by similar exposure time. No correlation of PBDEs with PCBs concentrations suggested different pathways of human exposures to PCBs and PBDEs. Our findings have raised concern about human health risk of dual exposure to PCBs and PBDEs resulting from e-waste recycling.

1. Introduction

In recent years, waste electrical and electronic equipment (WEEE or e-waste) has become a major environmental concern, particularly in developing countries (UNEP, 2005; Leung and Wong, 2007). It has been estimated that 50–80% of the global e-wastes is illegally imported to Asia, 90% of which is sent to China for recycling (Wong et al., 2007). Because the techniques used in recycling operations are primitive in some regions in China, various hazardous chemicals are released into the environment, including persistent organic pollutants (POPs), especially polychlorinated biphenyls (PCBs) and polybrominated diphenyl ethers (PBDEs). PCBs were widely used in transformers and capacitors as dielectric fluids, heat exchangers or hydraulic fluids before the 1970s. Commercial production of PCBs, identified as one of the twelve priority POPs in the Stockholm Convention of 2000, has been banned since the 1970s and 1980s due to their bioaccumulation and a wide range of toxicities for animals and humans. PBDEs as flame retardants are used in various electronic equipment, such as computers, printers, copying machines, television sets, mobile phones (Darnerud, 2003; Wang et al., 2007). Due to bioaccumulation and potential toxicities, the production of penta- and octaBDE was banned in the European Union in 2004 and in several states in the United States (e.g. California, Maine, and Hawaii) in 2006. Very high levels of PCBs or PBDEs were found in various environmental matrices and biotic samples in some e-waste recycling regions in China (Zhao et al., 2006; Wu et al., 2008; Luo et al., 2009). The PCBs and PBDEs burdens in human body and potential health risk in e-waste recycling regions in China are of concern, with increasing data on adverse health effects of PBDEs (Main et al., 2007; Turyk et al., 2008; Dallaire et al., 2009; Meeker et al., 2009).

Guiyu town in Guangdong Province and Taizhou region in Zhejiang Province are two of largest e-waste recycling regions in China. A research group reported high PBDEs levels and low PCBs levels in the blood of e-waste recycling workers from Guiyu, where PBDEs-containing e-wastes were recycled (Bi et al., 2007; Qu et al., 2007). The e-waste recycling activities in Taizhou region are

* Corresponding author. Tel.: +86 10 6291 9177; fax: +86 10 6292 3563.
E-mail address: qinzhanfen@rcees.ac.cn (Z.-F. Qin).

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mainly executed in Luqiao and Wenling. The recycling of e-wastes in Luqiao began since the late 1970s, where a large number of PCBs-containing transformers and capacitors were treated. The recycling of e-wastes in Wenling began since the late 1990s, where various PBDEs-containing e-wastes were disassembled and shattered. Some reports have demonstrated that PCBs pollution is serious in Luqiao (Zhao et al., 2006; Shen et al., 2009), resulting in high PCBs burdens among local residents (Ling et al., 2008; Shen et al., 2008; Wen et al., 2009). Recently, high PBDEs levels were found in soils (25 479 ng g^{-1} dry weight), sediments (up to 3661 ng g^{-1} dry weight), and biotic samples from Wenling (Liang et al., 2008; Yang et al., 2008). Correspondingly, high levels of PBDEs were also found in human hair from Wenling and near residents (Zhao et al., 2008). Because Luqiao is 25 km away from Wenling, it is suggested that the dual pollution of PCBs and PBDEs will occur in human body among these populations due to the diffusion. In a recent study, Zhao et al. (2009a) reported PBDEs and PCBs in tissue samples of surgical patients who were diagnosed at hospitals in Wenling. Overall, available information on dual exposure to PCBs and PBDEs among Taizhou populations, especially Wenling population, is limited. The aim of this study is to detect PCBs and PBDEs concentrations in the blood to assess dual body burdens of PCBs and PBDEs and potential health risk for local residents in Luqiao and Wenling.

2. Material and methods

2.1. Study region characteristics and sample collection

Taizhou (28°01′–29°20′N, 120°11′–121°56′E) is situated at the east of Zhejiang Province, Southeast China. The region has a coastline of 745 km, accounting for 28% of that of Zhejiang Province. Luqiao and Wenling are situated in the southeast part of Taizhou. In Luqiao, numerous villages have received scrap metals, obsolete electric capacitors, electric generators, and cable wires since the late 1970s. In Wenling, there are approximately 30 villages heavily involved in recycling e-wastes containing PBDEs with a 10 year-history. These e-wastes are disassembled and shattered into powder to select the usable materials, and discarded e-waste powder is stacked around some villages.

Volunteers were recruited from residents in a typic e-waste recycling site of Luqiao and of Wenling in November 2006. Ten mililiter of whole blood from each volunteer was drawn from a cubital vein by medical professionals, and transferred into a hexane-rinsed brown glass battle. The blood samples were immediately frozen and transported to the laboratory and stored at −20 °C until analysis. Volunteers' information on age, sex, occupational exposure, and lifestyle characteristics were obtained by a questionnaire survey.

2.2. Chemicals

Standard solutions for PCBs analysis, including $^{13}C_{12}$-labeled internal standard (EPA 68A–LCS), injection standard (EPA 68A–IS) and calibration curve standard (EPA 68A–CVS), were obtained from Wellington Laboratories (Gulph, Canada). Standard solutions for PBDEs analysis, including $^{13}C_{12}$-labeled internal standard (EO-5277) and injection standards (EO-5275) were obtained from Cambridge Isotope Laboratories, Inc. (MA, USA). Methylene dichloride, *n*-hexane, and nonane at pesticide grade were purchased from Tedia (Fairfield, OH, USA). Anhydrous sodium sulfate, sulfuric acid, sodium hydroxide, and anhydrous ethanol were analytical grade (Beijing Chemical Factory, Beijing, China).

2.3. Sample extraction and cleanup

PCBs and PBDEs were analyzed following US EPA 1668A and US EPA 1614. Briefly, the samples were spiked with 1 ng of a $^{13}C_{12}$-PCBs mixture (EPA 68A–LCS mixture) and a $^{13}C_{12}$-PBDEs mixture (EO-5277), and mixed with anhydrous sodium sulfate and allowed to equilibrate for 12–24 h. The samples were extracted using *n*-hexane:acetone 50:50 (v/v) in a sonication extractor; the extracts were purified and fractionated with multi-layer silica gel columns. Then, the PCB fractions were further purified with basic alumina columns, and the PBDE fractions were purified with Florisil columns. All fractions were concentrated for injection into analysis. $^{13}C_{12}$-labeled injection internal standards EPA 68A-IS and EO-5275 were added to each PCBs fraction and PBDEs fraction, respectively. Then, an aliquot of the extract was injected immediately into GC. Quantification was performed using an isotope dilution method.

2.4. Instrumental analysis

PCBs and PBDEs were analyzed by high-resolution GC/MS using an Agilent 6890 gas chromatograph coupled to an Autospec Ultima mass spectrometer (Waters Micromass, Manchester, UK) operating in EI mode at 35 eV at a trap current of 600 μA. The MS analysis and quality control are conducted following EPA methods 1668A. The GC was equipped with a CTC PAL autosampler. A 1-μL aliquot of the sample was injected in splitless mode (splitless time of 1 min for PCBs) onto a DB-5 MS fused silica capillary column (60 m × 250 μm i.d., film thickness 0.25 μm) with helium as carrier gas at a constant flow rate of 1.0 mL min^{-1}. The oven temperature program for PCBs was as follows: start at 80 °C, held for 2 min, increased to 150 °C at 15 °C min^{-1}, increased to 270 °C at 2.5 °C min^{-1} and held for 3 min, and finally increased to 320 °C at 15 °C min^{-1} and held for 1 min. For PBDEs, A 1-μL aliquot of the sample was injected in pulsed splitless mode (345 kPa, splitless time of 1 min for PBDEs) onto a DB-5 MS fused silica capillary column (15 m × 250 μm i.d., film thickness 0.25 μm) with helium as carrier gas at a constant flow rate of 1.5 mL min^{-1}. The program started at 80 °C, held for 1 min, increased to 200 °C at 10 °C min^{-1}, increased to 300 °C at 20 °C min^{-1} and held for 15 min. The injector and interface temperatures were 275 °C and 320 °C, respectively.

2.5. Quality control

Prior to analyze the human bloods, the pig blood samples were performed for the initial precision and recovery. Recoveries of spiked blood samples with the EPA 68A–LCS mixture and EO-5277 mixture were in the range of 65–110%, RSD <20%. Analytical quality control was applied during analysis. The continuous monitoring of laboratory contamination was conducted based on the determination of a blank sample throughout the whole analytical procedure, including extraction, cleanup, and quantification. Simultaneously, the ongoing precision was performed, and the recovery ranged 50–130%, RSD <30%, which satisfied the requirement of EPA method 1668A and method 1614.

2.6. Statistical analysis

All the statistical analyses were performed using SPSS software version 13.0 (SPSS, Chicago, USA). The Wilcoxon rank-sum test was used to test the difference of PCBs and PBDEs concentrations between two sexes or two study populations. Correlations between PCBs or PBDEs and ages and between PCBs and PBDEs were tested using the Spearman's coefficient correlation. A p value of <0.05 was considered to indicate statistical significance.

3. Results and discussion

3.1. Levels of PCBs and PBDEs

PCB-105, 118, 153, 183, and 180 were main PCB congeners in all of these samples. Table 1 shows the concentrations of these PCBs congeners and ∑PCBs as well as main PBDE congeners (BDE-28, 47, 99, 100, 153, 154, 180, and 209) and ∑PBDEs in the samples from both Luqiao residents ($n = 27$) and Wenling residents ($n = 23$). Because no significant difference in ∑PCBs and ∑PBDEs was observed between the males and the females in each study population, the data of the males and the females from each study population were combined to analyze.

The mean ∑PCBs in the blood samples from Luqiao was 204.20 ± 234.88 ng g^{-1} lipid (range 28.70–1044.26 ng g^{-1} lipid), which was significantly higher than the mean from Wenling (83.80 ± 52.08 ng g^{-1} lipid, range 26.88–218.80 ng g^{-1} lipid) (Table 1). Correspondingly, the mean level of CB-153, as one of dominant congeners, was significantly higher among Luqiao residents (55.34 ng g^{-1} lipid) than among Wenling residents (19.00 ng g^{-1} lipid). High body burdens of PCBs among Luqiao residents were related to the recycling of PCBs-containing e-wastes for three decades. In Wenling, no significant PCBs pollution source was found. Considering transport potential of PCBs (Pier et al., 2003; Wang et al., 2009), we suggest that PCBs pollution in Wenling might be mainly derived from the PCBs diffusion from Luqiao because the sampling site in Wenling is approximately 25 km away from the e-waste recycling site in Luqiao. The PCBs level in this study was consistent with the PCBs value (with a mean CB-153 concentration of 41.5 ng g^{-1} lipid) on Luqiao residents reported by Ling et al. (2008). To our knowledge, the PCBs levels among Luqiao residents seemed to be the highest relative to other populations in China, including Guiyu population (with a mean ∑PCBs of 69 ng g^{-1} lipid) (Bi et al., 2007). Compared with Europe general populations, the PCBs levels among Luqiao residents were also higher, as reported in previous studies (Table 2). For example, Guvenius et al. (2003) reported the median of ∑PCBs (105, 118, 153, 183, and 180) in maternal blood plasma of individuals from Stockholm was 134 ng g^{-1} lipid, while Thomas et al. (2006) reported the median ∑PCB (105, 118, 153, 183, and 180) in human blood serum samples from the general population in the UK was 118 ng g^{-1} lipid. Even, the PCBs levels among Luqiao residents were comparable to some values reported for US populations (Sjödin et al., 2004; Morland et al., 2005), who are thought to have high PCBs burdens, although it was lower than those from famous PCBs-pollution areas, such as Yusho in Japan (Todaka et al., 2009) and Brescia in Italy (Donato et al., 2008). Although the PCBs levels among Wenling residents were lower than general European and US populations, it was also higher relative to other Chinese populations except for Luqiao residents (Bi et al., 2007).

Eight PBDE congeners (28, 47, 99, 100, 153, 154, 183, and 209) were detected in all blood samples from Luqiao. Unfortunately, BDE-209 in 11 of 24 samples from Wenling was not quantified due to BDE-209 pollution during extraction. ∑PBDEs in 27 blood samples from Luqiao ranged from 39.88 to 395.86 ng g^{-1} lipid, with an mean of 117.58 and a median of 80.94 ng g^{-1} lipid (Table 1). ∑PBDEs in 13 blood samples from Wenling ranged from 132.46 to 714.16 ng g^{-1} lipid, with an mean of 357.44 and a median of 314.88 ng g^{-1} lipid (Table 1). There was significant difference in the ∑PBDEs level between two populations, accurately, the mean ∑PBDEs was three times higher from Wenling than Luqiao. The result was consistent with serious PBDEs pollution in Wenling resulted from recycling wasted computers, televisions, etc., which contain a great deal of PBDEs. In Luqiao, local e-waste recycling possibly contributed less to PBDEs pollution because recycled e-wastes mainly included transformers, capacitors, electric generators, and other hardware, which hardly contain PBDEs. In a previous study, we characterized the PBDEs diffusion from Wenling to the surrounding regions, including Luqiao (Zhao et al., 2009b). Therefore, we concluded that the PBDEs pollution in Luqiao might be partly derived from the PBDEs diffusion from Wenling. In addition, plastic manufacture as a local source might also contribute to the PBDEs pollution in Luqiao.

The PBDEs burdens among Wenling residents were lower than among occupational workers (with a mean PBDEs of 600 ng g^{-1} lipid) from Guiyu, as reported by Bi et al. (2007). In the literature, North Americans have higher body burdens of PBDEs than Europeans, with Tetra- and Penta-BDE as dominant congeners, while PBDEs burdens in populations in Asia are comparable to those in Europe (Sjödin et al., 2003; Hites, 2004; Wang et al., 2007). Hites (2004) concluded that the median concentration of PBDEs in blood samples from the US population was about 35 ng g^{-1} lipid. In this study, the median concentrations of ∑PBDEs in the blood samples from Wenling and Luqiao were much higher than the value from the US

Table 1
Concentrations of PCB and PBDE (ng g^{-1} lipid) in blood samples from the two study populations in Taizhou.

	Luqiao ($N = 27$, male 16, female 11)			Wenling ($N = 23$, male 7, female 16)		
	Mean ± SD	Median	Range	Mean ± SD	Median	Range
Age	39.2 ± 9.1	35	27–55	49.5 ± 11.9	49	25–72
CB-105	21.04 ± 33.22	10.32	2.20–147.90	8.16 ± 6.64	5.84	1.48–30.44
CB-118	50.58 ± 60.33	26.6	6.10–258.02	31.14 ± 24.72	24.97	4.76–76.04
CB-138	60.30 ± 67.26	36.78	7.66–309.02	21.58 ± 13.28	17.40	6.17–54.00
CB-153	55.34 ± 58.64	32.28	6.78–258.90	19.00 ± 11.20	15.72	7.46–52.94
CB-180	15.04 ± 17.62	6.76	1.78–69.44	3.90 ± 1.44	3.80	1.64–6.76
∑PCB	204.20 ± 234.88	118.38	28.7–1044.26	83.8 ± 52.08	75.26	26.88–218.80
CV of ∑PCB	1.15			0.62		
BDE-28	2.30 ± 2.86	1.28	0.62–14.90	31.28 ± 28.58	21.10	1.00–127.66
BDE-47	5.76 ± 9.44	3.64	1.22–51.18	44.72 ± 47.26	25.30	2.88–222.48
BDE-99	1.96 ± 3.50	1.06	0.44–17.84	7.90 ± 5.84	5.74	1.14–23.04
BDE-100	0.88 ± 0.94	0.66	0.08–4.98	4.12 ± 4.06	3.12	0.64–21.24
BDE-153	14.60 ± 14.60	10.46	2.38–68.54	26.12 ± 21.66	21.90	8.40–119.40
BDE-154	0.96 ± 1.12	0.78	0.14–6.12	2.30 ± 2.44	1.70	0.54–12.60
BDE-183	3.92 ± 2.12	3.44	1.00–11.62	7.26 ± 4.22	6.72	2.60–22.76
BDE-209	87.20 ± 79.64	64.54	17.16–378.29	210.48 ± 136.8[a]	171.60[a]	84.52–555.76[a]
∑PBDE	117.58 ± 81.56	80.94	39.88–395.86	357.44 ± 177.8[a]	314.88[a]	132.46–714.16[a]
CV of ∑PBDE	0.69			0.50[a]		

[a] Data from 12 samples because no data were obtained from other 11 samples due to BDE-209 pollution.

Table 2
Mean and median (indicated in parentheses) concentrations of PCB and PBDE (ng g^{-1} lipid) in blood samples from Taizhou in China compared with those from other countries/regions in the literature.

Country	CB-153	∑PCBs	BDE-28	BDE-47	BDE-99	BDE-100	BDE-153	BDE-154	BDE-183	BDE-209	∑PBDEs	References
US	(4.4)	ND	ND	(11)	(2.9)	(1.8)	(1.5)	(0.3)	<LOD	ND	(21)	Bradman et al. (2007)
US	60.5	ND	ND	13.3	3.2	2.7	3.2	0.6	0.5	ND	ND	Morland et al. (2005)
US	(35)	ND	ND	(34)	(11)	(5.9)	(7.3)	(1.0)	ND	ND	(61)	Sjödin et al. (2004)
Sweden	(56)	(176)	(0.07)	(0.83)	(0.19)	(0.17)	(0.56)	(0.04)	(0.06)	ND	(2)	Guvenius et al. (2003)
UK	(41)	(170)		(0.8)	<LOD	(0.8)	(1.7)	(0.6)	(0.3)		(5.6)	Thomas et al. (2006)
Spain	ND	ND	<LOD	(2.6)	(2.3)	(1.2)	(0.8)	<LOD	(0.1)	(1.1)	(9.7)	Gómara et al. (2007)
Japan	ND	45.7 (38)	ND	ND	ND	ND	ND	ND	ND	ND	3.3 (3.0)	Inoue et al. (2006)
Japan	ND	184.6 (138.8)	ND	ND	ND	ND	ND	ND	ND	ND	8.5 (1.3)	Koizumi et al. (2005)
Japan	ND	ND	0.2 (0.1)	0.9 (0.7)	0.2 (0.2)	0.3 (0.2)	0.7 (0.6)	0.2 (0.1)	0.3 (0.2)	9.2 (6.9)	13 (9.5)	Takasuga et al. (2004)
Korea	78.6	ND	0.3	5.7	2.7	1.4	4.7	0.3	2.0	ND	17.2	Kim et al. (2005)
China	ND	69(52)	ND	ND	ND	ND	ND	ND	ND	340 (310)	580 (600)	Bi et al. (2007)
China	(8.3)	ND	(2.6)	(6.7)	(1.4)	(1.0)	(9.0)	(0.8)	(4.7)	(83.5)	ND	Qu et al. (2007)
China	55.3 (32.3)	204.2 (118.4)	2.3 (1.3)	5.8 (3.6)	2.0 (1.1)	0.9 (0.7)	14.6 (10.5)	1.0 (0.8)	3.9 (3.4)	87.2 (64.5)	117.6 (80.9)	This study
China	19.0 (15.7)	83.8 (75.3)	31.3 (21.1)	44.7 (25.3)	7.9 (5.7)	4.1 (3.1)	26.1 (21.9)	2.3 (1.7)	7.3 (6.7)	210.5 (171.6)	357.4 (314.9)	This study

ND means no data.

population. As reviewed in Table 2, the PBDEs level in this study was also higher than those reported for human blood from Europe and Japan (Guvenius et al., 2003; Inoue et al., 2006; Thomas et al., 2006; Gómara et al., 2007). To our knowledge, this is the first report to present dual body burdens of PCBs and PBDEs at so high levels.

3.2. Potential health risk of high burdens of PCBs and PBDEs for residents

Numerous studies have demonstrated that PCBs have wide adverse effects in experimental animals, including developmental neurotoxicity, thyroid disrupting activities, etc. Although some epidemiologic have not supported that PCBs have adverse effects on human health, the significant association of PCBs burdens to some human health signs found in some surveys has highlighted potential health risk of PCBs for humans. Rogan and Gladen (1992) reported that prenatal PCB exposure was associated with poorer performance on the Psychomotor Index from Bayley Scales of Infant Development in children from general population in North Carolina. A study from the Netherlands indicated that prenatal exposure to "background" PCB concentrations were associated with poorer cognitive functioning in preschool children (Patandin et al., 1999). Walkowiak et al. (2001) reported that PCB exposure was associated with deficits in mental and motor scores in Germany children up to seven-months of age. In consistent with neurodevelopmental deficits, some epidemiologic surveys also demonstrated that PCBs might be responsible for abnormalities of thyroid functions (Turyk et al., 2007; Bloom et al., 2009; Langer et al., 2009). In these reports, PCBs burdens associated with adverse health effects were not unusually high. For example, the PCBs level associated with T4 decrease reported by Turyk et al. (2007) was similar to those found in the general US population. Prenatal exposure to PCBs associated with poorer cognitive functioning in Netherlandish preschool children was only "background" level (Patandin et al., 1999). In the present study, the PCBs level found in Luqiao residents exceed or close to the levels associated with adverse health effects. Therefore, potential health risk of PCBs for local population should be paid much attention. Similar to PCBs, PBDEs have been also shown to have developmental neurotoxicity and endocrine disrupting activities in animal experiments. Recently, there is increasing evidence that PBDEs have corresponding adverse effects on human health. For example, based a study combined children's development with pollutant burdens in their mothers, Roze et al. (2009) concluded that transplacental transfer of PBDEs were associated with the neurodevelopment of children at school age. Turyk et al. (2008) found that increased thyroglobulin antibodies and increased T4 in adult males were associated with PBDE exposure. Dallaire et al. (2009) also reported a positive association between PBDEs burden and T3 level, although they suggested that effects of PBDEs on thyroid homeostasis require further investigation despite. Also, Meeker et al. (2009) suggested that PBDE concentrations in general house dust were related to hormone levels in men despite the sample size in this study was small. PBDEs levels in breast milk showed an association with congenital cryptorchidism in newborn boys (Main et al., 2007). All the above findings were resulted from surveys on general populations with general exposure level of PBDEs. In the present study, PBDEs burdens were higher in two study populations, especially in Wenling population, than in general populations. Thus, we suggest that high PBDEs burden might pose high health risk on Taizhou populations. In particular, the health risk of dual burdens of PCBs and PBDEs is noteworthy. Previous studies showed that PBDE and PCB can interact to enhance neurotoxic effects on experimental animals (Eriksson et al., 2006; He et al., 2009). Therefore, a further epidemiologic study is required imperatively to evaluate the association of dual burdens of PCBs and PBDEs with some human health signs in Taizhou populations.

3.3. Correlations between PCBs or PBDEs and ages

Many studies have reported that PCBs concentrations increase with the ages of the individuals (Johnson-Restrepo et al., 2005;

Kim et al., 2005; Harden et al., 2007). However, no correlation between PCBs concentrations and the ages of the volunteers were found in the two study populations (Fig. 1). The range of ages in the study population was 27–55 years in Luqiao, while it was 25–72 years in Wenling. Because PCBs pollution in Taizhou was mainly derived from the recycling of PCBs-containing e-wastes since the late 1970s, these volunteers had similar life-time exposure independent of the individual's age, which can explain the lack of the correlation of PCBs concentrations with ages in this study. Similar to PCBs, PBDEs concentrations were also found to not correlate with ages in the two study populations. The observation was consistent with some of previous reports (Sjödin et al., 1999, 2000; Johnson-Restrepo et al., 2005; Kunisue et al., 2007). Generally, no correlation of PBDEs burdens with the ages was explained by similar PBDEs-exposure time. The world production and use of PBDEs have begun since 1970s, even much later in some regions, whereas the subjects involved in PBDEs burdens were generally older than 30 years in these studies (Johnson-Restrepo et al., 2005; Kunisue et al., 2007). In this study, our result on no age-difference in PBDEs burden seemed to might be also explained by similar PBDEs-exposure time (approximately 10 years), which began from the late 1990s in Taizhou. However, several latest studies showed age-difference in PBDEs burden (Sjödin et al., 2008; Toms et al., 2008, 2009a; Haraguchi et al., 2009). Toms et al. (2008, 2009a) reported that infants had higher PBDEs burdens than adults, and children at the age of 2–5 years had much higher PBDEs burdens than infants. Sjödin et al. (2008) reported that the younger and older burdened higher level of PBDEs than adults. In these studies, different age means not only different exposure time but also different exposure background and pathway as well as different eliminating process resulting from the difference in lifestyle and metabolism. In another word, the age-difference observed in the latest studies might suggest that PBDEs burden depend heavily on exposure background and metabolism rather than exposure time (Sjödin et al., 2008). In this study, regrettably, we cannot examine the association between PBDEs burdens and age stages, like the study conducted by Sjödin et al. due to the limit of small sample size.

3.4. Correlation between PCBs and PBDEs

In this study, no significant correlation was found between PBDEs and PCBs concentrations in blood samples from the two study populations (Fig. 2). In previous studies on human adipose tissues of Belgium and the US, no significant correlation was observed between PBDEs and PCBs. Recently, Bi et al. (2007) also reported that PBDE concentrations did not correlate with PCBs in serum samples from e-waste recycling workers in Southern China. These studies suggested that there might be different pathways of human exposures to PBDEs and PCBs (Johnson-Restrepo et al., 2005; Naert et al., 2006). Dietary exposure is believed to be the most important pathway of human exposure to PCBs. Diet is also believed to be one of important pathway of human exposure to PBDEs (Fraser et al., 2009). Recently, inhalation is also suggested to be a possible exposure pathway because high levels of PBDEs have found in air and dust (Jones-Otazo et al., 2005; Frederiksen et al., 2009; Toms et al., 2009b). In this study, coefficients of variation (CV) of PCBs and PBDEs among Luqiao residents were 1.15 and 0.69, respectively, while they among Wenling residents were 0.62 and 0.50, respectively. The result that less variability of PBDEs occurred among individuals relative to PCBs from two study populations, despite small sample size, also seemed to suggest that there is homologous source(s) for PBDEs exposure besides diet. Maybe, inhalation as a homologous source for PBDEs exposure made an important contribution to PBDEs burden among Taizhou populations. By contrast with our results, Kunisue et al. (2007) reported that significantly positive correlations between PBDEs and PCBs in Japanese adipose tissues and explained that there might be similar exposure pathways of the two pollutants for Japanese citizens, probably via fish intake. We believe that diet and inhalation or other factors may all contribute to exposure to PBDEs, and the relative importance of each of these exposure pathways will be heavily dependent on the local pollution background and human lifestyle. When local diets contain high levels of PBDEs, dietary exposure will become a dominant pathway of PBDEs rather than other factors (such as inhalation). Thus, a significant correlation between PBDEs and PCBs concentrations will be found in human tissues, whereas, PBDE will not correlate with PCBs.

3.5. Congener patterns of PCBs and PBDEs

PCB-105, 118, 153, 183, and 180 were dominant PCB congeners in all samples from two study populations. There was no sex-difference in the PCB congener pattern in blood samples from each study population. The PCB congener pattern from Luqiao residents did not differ from that from Wenling residents. As the above mentioned, the results showed that PCBs pollution source was same in two study region, i.e. PCBs pollution in Wenling might be mainly derived from the PCBs diffusion from Luqiao.

BDE-209 was the dominant congener in Luqiao residents, which accorded with previous reports on Chinese populations (Bi et al., 2007; Qu et al., 2007; Jin et al., 2009). However, we found signifi-

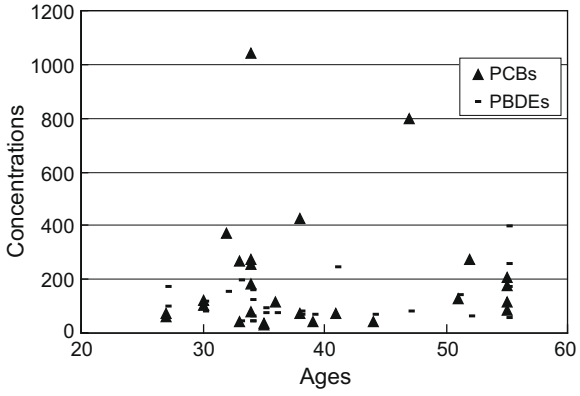

Fig. 1. Scatterplot of PCBs and PBDEs concentrations (ng g^{-1} lipid) in blood samples by ages of Luqiao residents ($n = 27$). There was no significantly linear correlation between PCBs ($r = 0.32$, $p = 0.87$) or PBDEs ($r = 0.35$, $p = 0.08$) and ages.

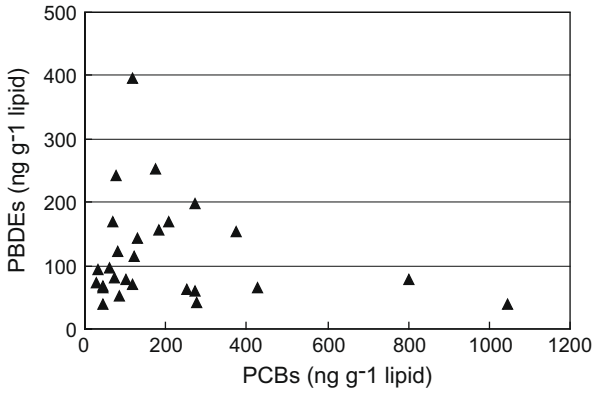

Fig. 2. Scatterplot of PBDEs concentrations by PCBs concentrations in the blood from Luqiao residents ($n = 27$). There was no significant correlation between PCBs and PBDEs ($r = -0.18$, $p = 0.37$).

cant difference in the percentage contribution of BDE-209 to ∑PBDEs between two sexes despite small sample size (Fig. 3). The percentage contribution of BDE-209 to ∑PBDEs in female blood samples was 0.83 ± 0.10% (range 74–96%), while it was 0.59 ± 0.19% (range 27–93%) in male blood samples. Correspondingly, the percentage of low brominated PBDEs (47 and 99) in the blood from the females was significantly lower than that from the males. In accord with Luqiao residents, BDE-209 was also the dominant congener among Wenling residents. Due to the lack of BDE-209 data in 11 of 23 samples from Wenling, the sex-difference in congener pattern cannot be compared. In previous studies, the sex-difference in PBDEs congener pattern has been never reported (Gómara et al., 2007). The ratio of BDE-99 to BDE-47 is believed to reflect a normal metabolized congener pattern (Sjödin et al., 2005; Bradman et al., 2007). We compared the ratio of BDE-99 to BDE-47 in the males with that in the females among Luqiao residents (data not shown), and found no significant difference, suggesting that the sex-difference in PBDEs congener pattern might not result from the difference in the metabolism between the males and the females. We have no ready explanation for this finding other than guess that the sex-difference PBDEs congener pattern might be related to different exposure due to sex-differences in lifestyle and occupation. Unfortunately, we have obtained quantitative information although we have known that almost all volunteers are involved in recycling e-wastes to different degree in Luqiao.

Although BDE-209 was the dominant congener among two study populations, there was obvious difference in the PBDEs congener pattern of female blood samples between Luqiao and Wenling. In Luqiao females, BDE-153 was the next most abundant congener, followed by BDE-47. In Wenling females, BDE-47 was the next most abundant congener, followed by BDE-28 and BDE-153 (Fig. 4). We calculated the ratio of the sum of high brominated PBDEs (BDE-153, 154, 183 and 209) to low brominated PBDEs (BDE-28 and 47). The ratios ranged from 7.56 to 97.93, with a mean of 28.24 among Luqiao residents, while it ranged from 0.87 to 8.70, with a mean of 2.13 among Wenling residents. The result showed that high brominated congeners contributed relatively larger to ∑PBDEs in Luqiao females than in Wenling females. It might be explained by different pollution source, i.e. PBDEs pollution in Wenling mainly be resulted from e-waste recycling, while there might be other sources in Luqiao except from PBDEs diffusion from Wenling.

High brominated PBDE congeners had been believed to be less accumulative in biota due to large molecular size and high octanol–water partition coefficient (Boon et al., 2002; Hites, 2004). However, in consistent with recent reports, our study has strongly demonstrated that high brominated PBDE congeners as well as low brominated congeners are bioavailable in humans. Due to the short half life of BDE-209, high level of BDE-209 in blood samples implied that local populations were continuously exposed to BDE-

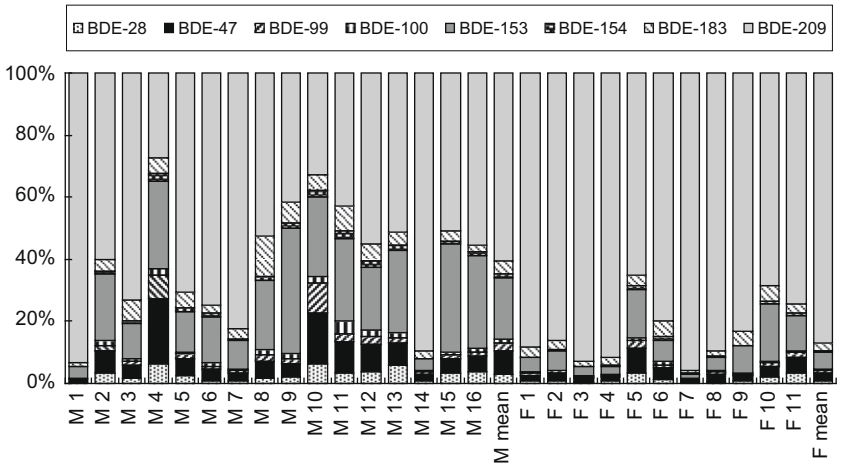

Fig. 3. Percentage contributions of PBDE congeners to the sums of PBDEs concentrations in the blood from male residents (M1–16) compared with females (F1–11) in Luqiao.

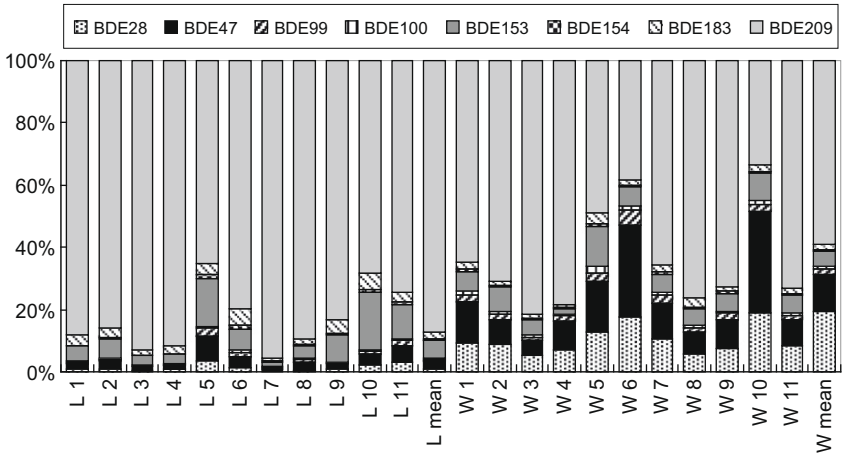

Fig. 4. Percentage contributions of PBDE congeners to the sums of PBDEs concentrations in the blood from Luqiao female residents (L1–L11) compared with those from Wenling female residents (W1–W11).

209 (Thuresson et al., 2006). In previous studies with no measures of BDE-209, PBDEs burden in humans maybe be underestimated (Bi et al., 2006). In addition, based on the fact of great geographic variance in PBDE congener pattern, we believe that PBDE-congener patterns in humans might heavily depend on exposure background rather than other factors.

In conclusion, we found that Taizhou populations simultaneously burdened high levels of PCBs and PBDEs due to mixed pollution resulting from the recycling of PCBs- and PBDEs-containing e-wastes. Based on previous epidemiologic data, it is suggested that dual burdens of PCBs and PBDEs at so high levels in local populations might pose potential health risk. A further study with large sample size is required to evaluate potential health effects of high dual burdens of PCBs and PBDEs for Taizhou populations.

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Appendix A. Supplementary material

Supplementary data associated with this article can be found, in the online version, at doi:10.1016/j.chemosphere.2009.12.013.

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