Environmental Pollution 250 (2019) 662-668

Contents lists available at ScienceDirect

Environmental Pollution

journal homepage: www.elsevier.com/locate/envpol

High intake of persistent organic pollutants generated by a municipal waste incinerator by breastfed infants *



^a Zhejiang Provincial Center for Disease Control and Prevention, 3399 Bin Sheng Road, Binjiang District, Hangzhou, 310051, China ^b Eye Center of the 2nd Affiliated Hospital, School of Medicine, Zhejiang University, Zhejiang Provincial Key Lab of Ophthalmology, Jiefang Road 88#, Hangzhou, 310009, China

ARTICLE INFO

Article history: Received 14 January 2019 Received in revised form 9 April 2019 Accepted 14 April 2019 Available online 20 April 2019

Keywords: PCDD/Fs PCBs Breast milk Incinerator

ABSTRACT

To monitor the body burden of persistent organic pollutants in mothers and infants living in proximity to a 10-year-old municipal waste incinerator (MWI), levels of polychlorinated dibenzo-p-dioxins and dibenzofurans (PCDD/Fs) and polychlorinated biphenyls (PCBs) in the breast milk of mothers were evaluated, and the estimated daily intake (EDI) level of these pollutants in infants was assessed. In total, 14 mothers from the exposure area and 18 mothers from a control area were recruited for this study. In addition, the levels of 17 PCDD/F congeners and 18 PCB congeners in the breast milk of the mothers were estimated, and risk assessment for their infants by calculating their EDI levels was performed. The mean levels of Σ PCDD/Fs and TEQ- Σ PCDD/Fs in the breast milk of the exposure group were significantly greater than those of the control group (3.36 vs. 1.47 pg/g wet weight; and 0.24 vs. 0.12 pg TEQ/g wet weight, respectively; p < 0.05). With respect to PCBs, the mean levels of the total PCBs (Σ PCBs) and non-dioxin like-PCBs (*SNDL-PCBs*) in the exposure group were significantly greater than those in the control group (269 vs. 141 pg/g wet weight; 230 vs. 115 pg/g wet weight, respectively; p < 0.05). The mean levels of <code>\SigmaPCDD/Fs</code> and <code>TEQ-SPCDD/Fs</code> were positively correlated with the inhabit time of the exposure group (r = 0.636, r = 0.629, p < 0.05, respectively). The mean EDI level of the exposure group in infants was significantly greater than that of the control group (22.0 vs. 13.0 pg TEQ/kg bw day; p < 0.05). In conclusion, mothers and their breast-fed infants living in proximity to the 10-year-old MWI located in the Zhejiang Province of China exhibited a significantly higher body burden of PCDD/Fs and PCBs compared to those living in the control area.

© 2019 Elsevier Ltd. All rights reserved.

1. Introduction

For several decades, incineration has been demonstrated to be an efficient technology for waste disposal. Compared to typical waste treatments such as landfill, incineration exhibits clear advantages, including energy recovery and volume reduction, as well as the elimination of pathogenic agents (Nadal et al., 2008; Zubero et al., 2011). Hence, municipal waste incinerators (MWI) have been widely applied worldwide. According to the *Development Report on Treatment Industry of Urban Domestic Refuse* published in 2017 by the China Environmental Protection Industry (2017), the number of MWIs rose to 220 in Chinese urban areas by 2015, corresponding to twice the number of MWIs in 2010. The Zhejiang Province accounts for the largest number of MWIs. With the rapid increase of MWIs, there is increased concerns for the population living near the MWIs. A report (Industry, 2017) regarding the online data monitoring of MWI emissions revealed that these emissions exceed the approved standard by thousands of times in the first quarter of 2017 in the Zhejiang and Fujian Provinces. Inorganic and organic substances emitted by these MWIs may cause severe health issues in local residents.

Polychlorinated dibenzo-p-dioxins and dibenzofurans (PCDD/ Fs) are the most notable emissions released from waste incinerators due to incomplete combustion (Zubero et al., 2011; Zubero et al., 2017). PCDD/Fs were listed in the 2001 Stockholm Convention on Persistent Organic Pollutants (POPs) as one of the "dirty dozen"





 $^{\,^{\}star}\,$ This paper has been recommended for acceptance by Charles Wong.

^{*} Corresponding author.

E-mail address: zhjchen@cdc.zj.cn (Z. Chen).

¹ P.X. and L.W. contributed equally to this work.

contaminants due to their well-known toxic properties, persistence, and bioaccumulation capacity (Nadal et al., 2008). Polychlorinated biphenyls (PCBs), which are often used in plasticizers. transformers, and lubricants, also can be released during combustion as by-products (Schuhmacher et al., 2013). In 2012, the International Agency for Research on Cancer (IARC) classified two PCDD/ F congeners (i.e., 2,3,4,7,8-PeCDF and 2,3,7,8-TCDD, respectively) and one PCB congener (PCB-126) as carcinogenic to humans, and in 2016, they added PCBs and dioxin-like PCBs (DL-PCBs) as carcinogenic (Agents Classified by the IARC Monographs, 2016). PCDD/Fs and PCBs exhibit stability and lipophilicity, permitting their aggregation in fatty materials such as adipose tissue and breast milk in humans via the food chain (Ulaszewska et al., 2011).

Breast milk is a primary source of intake for infants. Therefore, compared to adults, infants exhibit higher levels of these persistent organic pollutants on a body-weight basis (Lu et al., 2015). A recent study concluded that the levels of 2,3,7,8-TCDDs and TEQ-PCDD/Fs in breast milk are good indicators of their levels in the cord blood, indicative of transfer from a mother to a fetus before birth. The monitoring of breast milk provides an opportunity to estimate the postnatal exposure of infants to contaminants such as PCDD/Fs and PCBs. In addition, sampling breast milk is a convenient, noninvasive route to evaluate the body burden of contaminants in adults and infants.

An MWI located in the Zhejiang Province of China has been in operation for more than 10 years. Previously, our group has revealed that the serum levels of Σ PCDD/Fs and TEQ- Σ PCDD/Fs in local children living within 3 km of the MWI are significantly greater than those living in a control area (3.10 vs. 0.85 pg/g wet weight; 0.36 vs. 0.08 pg WHO-TEQ/g wet weight, respectively). Moreover, the levels of Σ PCDD/Fs in eggs and soil from the exposure area were statistically greater than those from the control area (31.08 vs. 4.02 pg/g dry weight; 1026.00 vs. 291.78 pg/g dry weight, respectively). This study is a continuation of our previous study with focus on the dioxin exposure of the susceptible population, including parturients and their infants. In this study, the levels of PCDD/Fs and PCBs in the breast milk of mothers living in proximity to the 10-year-old MWI is reported, and the exposure of infants via the calculation of the estimated daily intake (EDI) of PCDD/Fs and PCBs by breastfeeding is assessed.

2. Materials and methods

2.1. Study population

The exposure area of our study, which was in the central Zhejiang Province of China, was an agricultural town nearly 1100 km², with an MWI operating for 10 years. The total evaporation of the MWI was 135–165 tons per hour, and the total power was 24 MW. The feedstock of the MWI was mostly unsorted household waste. and the waste capacity was 800-1000 tons per day. Notably, no other industries were emitting PCDD/Fs or PCBs in the exposure area. The control area was also an agricultural town located in western Zhejiang, which was nearly 2200 km², and it was located about 187 km away from the exposure area. The populations, lifestyles, and cultural backgrounds of these two locations were nearly identical. Both these populations were located far away from industrial areas. In total, 14 mothers from the exposure area and 18 mothers from the control area were recruited by local township public health centers. When mothers returned to the township public health centers for physical examination, an informed consent was obtained. Mothers met our standards for recruitment in this study. All of the recruited subjects were physically healthy and non-occupationally exposed to PCDD/Fs and PCBs; in addition, they had resided there for at least 4 years, with a time span ranging from 4 to 42 years. In addition, all of the subjects in the exposure area were residing within 3 km of the MWI. Table 1 summarizes the characteristics of the two groups.

For each subject, 50 mL of breast milk was collected from September to October 2013. All of the samples were collected using unified breast pumps in the morning. These samples were delivered in sterile glass containers and stored at -20 °C until analysis. The baseline data for age, gender, occupation, smoking and drinking habits, and other characteristics, such as the time they had been residing in the area, were collected via a questionnaire. Appropriate informed consent was obtained for all subjects. This study was approved by the Ethics and Human Subject Committee of Zhejiang Provincial Center for Disease Control and Prevention.

2.2. PCDD/F and PCB determination

The PCDD/F and PCB congeners were determined in accordance with the U.S. Environmental Protection Agency (EPA) 1614 and 1668A methods (EPA, 1994, 2003). The experimental procedure was identical to our previous study (Shen et al., 2012; Xu et al., 2014). Briefly, breast milk was weighed and freeze-dried for 36 h, followed by crushing it into a powder using an electric disintegrator (IKA, Guangzhou, China). Then, 10 g of milk powder was spiked with 1 ng of ¹³C₁₂-labeled surrogate internal standards (Wellington Laboratories, Ontario, Canada) and extracted using a mixture of n-hexane and dichloromethane (1:1, v/v) in a Soxhlet apparatus for approximately 12 h. Then, the extracts were purified and fractionated by using sequential multilayer silica gel and acid alumina chromatography columns, followed by the separation of PCDD/Fs and PCBs using an automated pressured extraction acid alumina chromatography column system (Power-Prep™, Fluid Management Systems, Inc., Waltham, MA, USA), All of the evaporated fractions were reconstituted with 30–50 uL of nonane for analysis. Injection standards were added to each PCB and PCDD/F fraction before instrumental analysis. Finally, quantification was performed by the isotope dilution method using an Agilent 6890 gas chromatography system (Agilent Technologies, Palo Alto, CA, USA) coupled to an Autospec Ultima mass spectrometer (Waters Micromass, Manchester, UK). The method detection limit was 0.07 pg/g wet weight for PCDD/Fs and 0.05 pg/g wet weight for PCBs.

2.3. Estimated daily intake

The daily intake of PCDD/Fs and PCBs by infants via

n two areas.

Table 1	
Demographic information of subjects	i

Variable	Exposure	Control	р
Number	14	18	-
Age (years) ^a	27 ± 6	28 ± 5	0.54
Height (cm) ^a	156.4 ± 3.3	157.2 ± 4.6	0.58
Weight 1 (kg) ^{a e}	49.8 ± 5.9	52.4 ± 8.3	0.34
Weight 2 (kg) ^{a e}	65.5 ± 9.2	66.8 ± 6.8	0.64
Inhabit time (years) ^b	8 ± 10	19 ± 7	0.00^{**}
Smoking (Yes/No) ^c	0/14	1/17	-
Passive smoking (Yes/No) ^c	5/9	8/10	0.62
Drinking (Yes/No) ^c	0/14	2/16	-
Distance (km) ^d	2.7 ± 1.9	-	-
Infants' age (months)	8 ± 3	8 ± 3	0.94

*p < 0.01.

t-test b

Non-parametric test.

Chi-square test. d

Distance away from the MWI.

^e Weight 1 means weight before parturition; Weight 2 means weight after parturition.

breastfeeding was estimated according to previous studies (Chan et al., 2007; Deng et al., 2012) with some modifications. The EDI was calculated by the following formula:

EDI = concentration* daily milk consumption/infant weight

EDI: estimated daily intake (pg TEQ/kg bw day)

Concentration: levels of PCDD/Fs and PCBs in the breast milk samples (pg TEQ/g wet weight). As the lipid levels were not determined, the lipid content was not included in this formula, but the EDI level was not affected after unit conversion.

Daily breast milk consumption: 750 mL/day (780 g/day) for 0–6 months, 600 mL/day (624 g/day) for 7–12 months, and 500 mL/day (520 g/day) for 13–24 months, according to domestic publications (Society, 2016; Sun and Yang, 2013).

Infant weight: mean values of body weight in different age groups from the *Reference standard for growth and development of children under 7 years old in China*, published by the National Health Commission of the People's Republic of China (to be observed in Supplementary Material, Tables S1 and S2).

2.4. Statistical analyses

The t-test, chi-square test, and non-parametric test were employed to compare the demographic data of the subjects between the two groups. With respect to the data obtained for PCDD/ Fs and PCBs, the typical method to deal with values below the detection limit is to substitute a fraction of the detection limit. especially one-half fraction. However, these fabricated values were reported to render poor statistical estimates, as well as obscure patterns and trends in the data (Helsel, 2006). Hence, according to the Helsel (2006), the Regression on Order Statistics (ROS) model by script, referred to as Nondetects And Data Analysis (NADA), is used in the R statistics package to predict the values under the detection limit in this study. Then, the normality test was employed to assess the distribution of the PCDD/F and PCB levels. If the data fitted the normal distribution, two independent sample *t*-tests were performed to compare the mean levels of the two groups. Otherwise, the non-parametric test was performed. Meanwhile, the Pearson correlation was utilized to evaluate the levels of PCDD/Fs and PCBs associated with subject characteristics, including the time they had resided in an area (inhabit time) and the distance of their residence from the MWI (inhabit distance). For PCDD/F and PCBs data, the R statistics package version 3.5.3 was used. For other statistical analyses, the Statistical Package for Social Science version 16.0 was used. A p value < 0.05 was statistically significant for all the tests.

3. Results and discussion

3.1. Subject characteristics

In this study, 14 mothers and 18 mothers were recruited from the exposure and control areas, respectively. Table 1 summarizes the demographic results of the subjects. Significant differences in the age, height, and body weight before or after parturition, or in passive smoking status between the two groups (p > 0.05), were not observed. However, the inhabit times of these two groups differed (p > 0.05). With respect to smoking and drinking habits, the number of subjects was limited to make a comparison. Within the exposure area, the mean inhabit distance of the 14 subjects residing from the MWI was 2.7 km. Furthermore, the age of the infants did not exhibit significant differences between the two groups (p > 0.05).

3.2. Levels of PCDD/Fs and PCBs in breast milk

The levels of 17 PCDD/F congeners in the breast milk samples were determined. A majority of the PCDD/F congeners in the exposure group were significantly greater than those in the control group (p < 0.05). With regard to the mass concentrations, the mean levels of Σ PCDD/Fs and TEQ- Σ PCDD/Fs in the exposure group were significantly greater than those in the control group (3.36 vs. 1.47 pg/g wet weight; 0.24 vs. 0.12 pg TEQ/g wet weight, respectively; p < 0.05). In addition, the major congeners in the exposure group were OCDD, 2,3,4,7,8-PeCDF, and 1,2,3,4,6,7,8-HpCDD, while those in the control group were OCDD; 1,2,3,4,6,7,8-HpCDF, and 1,2,3,6,7,8-HxCDF (Fig. 1). In addition to the PCDD/Fs, the 18 PCB congeners were determined, and the levels of 4 of these congeners in the exposure group were significantly greater than those in the control group (p < 0.05). With respect to dioxin like-PCBs (DL-PCBs), the mean levels of ΣDL -PCBs and TEQ- ΣDL -PCBs did not exhibit statistical differences (38.8 vs. 26.0 pg/g wet weight; 0.05 vs. 0.03 pg TEQ/g wet weight, respectively; p > 0.05). The major DL-PCB congeners were PCB-118, PCB-105, and PCB-156 in the exposure and control groups (Fig. 2). However, the mean levels of the total PCBs (SPCBs) and non-dioxin like-PCBs (SNDL-PCBs) in the exposure group were significantly higher than those in the control group (269 vs. 141 pg/g wet weight, 230 vs. 115 pg/g wet weight, respectively; p < 0.05). The mean levels of $\Sigma(PCDD/Fs + DL-PCBs)$ and TEQ- Σ (PCDD/Fs + DL-PCBs) in the exposure group were greater than those in the control group, but only TEQ- Σ (PCDD/Fs + DL-PCBs) exhibited statistical significance (42.1 vs. 27.5 pg/g wet weight, p > 0.05; 0.28 vs. 0.16 pg/g wet weight, p < 0.05; respectively). The correlations between the levels of PCDD/Fs and PCBs in the breast milk and subject characteristics, including the inhabit time and inhabit distance from the MWI, were analyzed. The mean levels of Σ PCDD/Fs and TEQ- Σ PCDD/Fs positively correlated with the inhabit time in the exposure group (r = 0.636, r = 0.629, p < 0.05, respectively), but not in the control group (p > 0.05). Significant correlations were not observed between the SDL-PCB or TEQ- Σ DL-PCB level and the inhabit time (p > 0.05). Furthermore, the correlations between the contaminant levels and inhabit distance were not significant in the exposure group (p > 0.05).

To monitor the body burden of persistent organic pollutants in mothers and infants living in proximity to a 10-year-old MWI in the Zhejiang Province of China, the levels of PCDD/F and PCB in the breast milk of mothers were evaluated, and the EDI level of their infants was assessed. To comprehensively compare these results with previous studies, the levels of PCDD/Fs and PCBs in mothers living near MWI in different countries were investigated (Table 2). Owing to a design deficiency in this study, the lipid levels of the breast milk were not determined; hence, the TEQ levels of PCDD/Fs and PCBs are expressed by pg TEQ/g wet weight. To compare these results with others, the results were multiplied by 25 to transform the TEQ levels into pg TEQ/g lipid based on our previous experience that the level of lipids in breast milk was approximately 4%. Thus, the estimated level of PCDD/Fs in the exposure group was 6.0 pg TEQ/g lipid; this value is considerably less than those reported in most of the previous studies conducted in Spain (Schuhmacher et al., 2004; Schuhmacher et al., 1999; Schuhmacher et al., 2009), Portugal (Reis et al., 2007), Germany (Deml et al., 1996), and Japan (Tajimi et al., 2005), but it is greater than that in one study conducted in Spain (Schuhmacher et al., 2013) and another study in Italy (De Felip et al., 2008). Furthermore, our results were compared with a national survey of PCDD/Fs and PCBs in human milk in China: The obtained result was greater than the level indicated by the survey, which was 3.12 pg TEQ/g lipid (Li et al., 2009). Different from two earlier studies (De Felip et al., 2008; Reis et al., 2007), mothers living near the MWI exhibit a significantly higher body

100% –				
90% -				
80% -				
70% - 60% -				
50% -				
40% -				
30% -			_	
20% -			_	_
10% -				_
0% -	Exposure	Control	Exposure	Control
	Concer	ntration	T	ËQ
□OCDF	0.09	0.09	0.00003	0.00003
□ 1,2,3,4,7,8,9-HpCDF	0.08	0.06	0.0008	0.0006
■ 1,2,3,4,6,7,8-HpCDF	0.14	0.13	0.001	0.001
■2,3,4,6,7,8-HxCDF	0.10	0.08	0.01	0.01
■1,2,3,7,8,9-HxCDF	0.03	0.08	0.003	0.008
■1,2,3,6,7,8-HxCDF	0.22	0.11	0.02	0.01
■1,2,3,4,7,8-HxCDF	0.21	0.10	0.02	0.01
■2,3,4,7,8-PeCDF	0.24	0.07	0.07	0.02
■1,2,3,7,8-PeCDF	8-PeCDF 0.00		0.00	0.003
■2,3,7,8-TeCDF	0.07	0.10	0.007	0.01
■OCDD	1.70	0.41	0.0005	0.0001
■1,2,3,4,6,7,8-HpCDD	0.23	0.10	0.002	0.001
■1,2,3,7,8,9-HxCDD	0.08	0.05	0.008	0.005
■1,2,3,6,7,8-HxCDD	5,7,8-HxCDD 0.09		0.009	0.006
■1,2,3,4,7,8-HxCDD	0.05	0.00	0.005	0.00
■1,2,3,7,8-PeCDD	0.08	0.04	0.08	0.04
■2,3,7,8-TeCDD	0.00	0.00	0.00	0.00

^a 0.00 means not detectable or predictable.

^a 0.00 means not detectable or predictable.

burden of PCDD/Fs than those living in a non-exposed area. The discrepancies between the as-obtained results and others were most likely due to regional differences, subject diversity, and various technical differences. Another important reason was the sampling year: Most countries exhibited a strong downward trend for these POPs. Notably, the data demonstrated that the mean level of PCDD/Fs positively correlates with the time spent living in the exposure area, indicative of the cumulativity of PCDD/Fs in local residents. In this study, OCDD accounted for the majority of PCDD/ Fs, which was in agreement with most of the previous studies (De Felip et al., 2008; Deml et al., 1996; Schuhmacher et al., 2004; Schuhmacher et al., 1999; Schuhmacher et al., 2013; Tajimi et al., 2005).

Regarding the PCBs, the estimated level of DL-PCBs was 1.25 pg TEQ/g lipid in the exposed mothers in this study; this value is less than those in previously reported studies in Spain (Schuhmacher et al., 2009, 2013), Italy (De Felip et al., 2008), and in Japan (Tajimi et al., 2005). Compared to the national survey (Li et al., 2009), the mean level of DL-PCBs in this study was nearly the same as that reported previously (1.46 pg TEQ/g lipid). Furthermore, PCB-118 and PCB-156 accounted for the majority of PCBs in our study as well as in previous studies (Table 2).

In 2011, another national survey (Zhang et al., 2016), which was conducted in 16 provinces of China, reported that the mean levels of Σ PCDD/Fs and TEQ- Σ PCDD/Fs in breast milk are 133.3 pg/g lipid and 4.9 pg TEQ/g lipid, respectively; these corresponding values increase to ~32.6% and 99.9% during 2007-2011. However, the DL-

PCB concentrations did not exhibit any statistical difference between the two surveys. As that study calculated the TEQ levels by TEF 1998, which differed from our study that was calculated by TEF 2005, this study was not compared with this national survey, but instead it was compared with the first one (Li et al., 2009). The rapid increase of PCDD/Fs in breast milk revealed that mothers and their infants in China are subject to considerable health risks. Furthermore, previous studies reported that the general population is considered rather than the population living near the MWI. In future, more attention should be focused on the PCDD/F pollution, among residents living near MWIs.

3.3. Estimation of the daily intake of infants

In this study, the infant EDI levels in the exposure group ranged from 11.0 to 45.7 pg TEQ/kg bw day and from 8.35 to 19.4 pg TEQ/kg bw day in the control group (Table S3). The mean level of EDI in the exposure group was considerably greater than that in the control group (22.0 vs. 13.0 pg TEQ/kg bw day; *p* < 0.05).

The EDI level was 22.0 pg TEQ/kg bw day in our study, which was considerably greater than the tolerable daily intake (TDI) of 4 pg TEQ/kg bw day as proposed by the World Health Organization (WHO) (van Leeuwen et al., 2000); this value is also greater than the TDI proposed by the Scientific Committee on Food of the European Commission, which is 2 pg TEQ/kg bw day (EC, 2001). As the studies considering mothers living near MWIs seldom report the associated EDI levels, this study has to be compared with those

Fig. 1. PCDD/Fs congener profiles in breast milk from two areas (%)^a.

100% -				
90% -				
80% -				
70% -				
60% -				
50% -				
40% -				
30% -				
20% -				
10% -				
0% -				
070	Exposure	Control	Exposure	Control
	Concer	ntration	T	EQ
■ PCB 189	0.53	0.40	0.00001	0.00001
■ PCB 167	3.24	0.89	0.00009	0.00003
■ PCB 157	1.28	1.12	0.00004	0.00003
■PCB 156	5.29	3.60	0.0006	0.0001
■PCB 123	0.32	0.13	0.000001	0.000004
■PCB118	19.5	13.7	0.008	0.0004
■PCB 114	1.30	0.84	0.00003	0.00003
■PCB 105	5.74	4.18	0.0007	0.0001
■PCB 169	0.24	0.16	0.001	0.005
■PCB 126	0.38	0.23	0.01	0.02
■PCB 81	0.12	0.11	0.000004	0.00003
■ PCB 77	0.82	0.60	0.00005	0.00006

Fig. 2. PCBs congener profiles in breast milk from two areas (%).

Table 2	
Mean levels of PCDD/Fs and PCBs in the breast milk of residents living near incinerator	's ^{a #} .

Country	Year	Group	Sample size	PCDD/ Fs	DL- PCBs	PCDD/F major congeners	DL-PCB major congeners	References
Spain	1998	Exposure	15	11.82 ^b		OCDD; 1,2,3,4,6,7,8-HpCDD; 1,2,3,6,7,8-HxCDD	1	Schuhmacher et al. (1999)
	2002	Exposure	15	11.9	1	OCDD; 1,2,3,6,7,8-HxCDD; OCDF	1	Schuhmacher et al. (2004)
	2007	Exposure	15	7.6	4.8	1	/	Schuhmacher et al. (2009)
	2012	Exposure	20	4.79	2.48	OCDD; 1,2,3,6,7,8-HxCDD; 2,3,4,7,8-PeCDF	Ĩ	Schuhmacher et al. (2013)
Italy	2008 2009	Exposure	21	3.78	4.87	OCDD; 2,3,4,7,8-PeCDF; 1,2,3,4,6,7,8-HpCDF	PCB118; PCB170; PCB156	(De Felip et al., 2008) ^N
		Control 1	22	4.67	5.27	OCDD; 2,3,4,7,8-PeCDF; 1,2,3,6,7,8-HxCDD	PCB170; PCB118; PCB156	
		Control 2	16	4.70	6.28	OCDD; 2,3,4,7,8-PeCDF; 1,2,3,6,7,8-HxCDD	PCB118; PCB170; PCB156	
Portugal	1999 2003	Exposure	73	9.5	/	1,2,3,7,8-PeCDD; 2,3,4,7,8-PeCDF; 1,2,3,6,7,8- HxCDD	1	(Reis et al., 2007) ^N
		Control	108	9.1	1	1,2,3,7,8-PeCDD; 2,3,4,7,8-PeCDF; 2,3,7,8-TeCDD	1	
Germany	1993	Exposure	7	12.4 ^b	,	OCDD; 1,2.3,4,6,7,8-HpCDD; 1,2,3,6,7,8-HxCDD	, I	Deml et al. (1996)
Japan	1999 	Exposure	240	14.9	10.6	OCDD; 1,2,3,6,7,8-HxCDD; 2,3,4,7,8-PeCDF	PCB118; PCB156; PCB105	Tajimi et al. (2005)
China	2013	Exposure	14	0.24 ^c	0.05 ^c	OCDD; 2,3,4,7,8-PeCDF; 1,2,3,4,6,7,8-HpCDD	PCB118; PCB105; PCB156	This study ^P
		Control	18	0.12 ^c	0.03 ^c	OCDD; 1,2,3,4,6,7,8-HpCDF; 1,2,3,6,7,8-HxCDF	PCB118; PCB105; PCB156	

#/means not available.

^a pg WHO-TEQ/g lipid. ^b pg I-TEQ/g lipid. ^c pg WHO-TEQ/g wet weight. ^NNegative: No significant differences were observed between subjects living near the incinerators and those living far away.

^PPositive: Significant differences were observed between subjects living near the incinerators and those living far away.

conducted on general population. The national survey conducted in 2007 reported a higher mean level of EDI (23.70 pg TEQ/kg bw day) compared to that observed here (17.0 pg TEQ/kg bw day), ranging from 11.8 to 40.7 pg TEQ/kg bw day (Li et al., 2009). Twelve provinces of China were enlisted in the national survey, but the provinces did not include the Zhejiang Province; hence, the background levels of PCDD/Fs and PCBs in Zhejiang have not yet been clarified. Typically, the EDI level in this study was considerably less than those in most of the previous studies, e.g., 60.3-80.4 pg TEQ/ kg bw day in Greece (Costopoulou et al., 2013), 48.2 pg TEQ/kg bw

day in Shenzhen of China (Deng et al., 2012), 31.8 pg TEQ/kg bw day for newborns in Taiwan (Chao et al., 2005), and 60 pg TEQ/kg bw day in Korea (Yang et al., 2002). On the one hand, different background levels of PCDD/Fs and PCBs accounted for the discrepancies between these studies. On the other hand, the parameters applied herein were adapted to the domestic population, which was different from other studies; this may have also caused disparities between the results. However, compared to the control area in our own province, the EDI level in the exposure area was significantly increased, indicating that the proximity to the MWI increases not only the body burden of PCDD/Fs and PCBs in mothers but also health risks of their infants.

As has been mentioned in the introduction, the environmental samples of eggs and soil collected from the same exposure area as this study revealed significantly higher levels of PCDD/Fs than those from the control area. Furthermore, the levels of TEQ- Σ PCDD/ Fs in the eggs, soil, and particulate matter less than $2.5 \,\mu m \,(PM_{2.5})$ from the exposure area were significantly higher (4.54 vs. 0.45 pg WHO-TEQ/g dry weight; 2.21 vs. 0.47 pg WHO-TEQ/g dry weight; 0.06 vs. 0.02 pg TEQ/m³, p < 0.05, respectively; data from another study that are not published). The elevation of contaminants in these environmental factors revealed that the dietary intake and respiratory exposure can be potential exposure pathways for contaminants emitted by MWIs and affecting local populations. Notably, in China, mothers who traditionally to eat more fatty food, such as trotters, produce more breast milk. Due to the lipophilicity of PCDD/Fs and PCBs, the mothers might consume these contaminants more often than the general population. Undoubtedly, infants fed on breast milk exhibit an increased intake of these contaminants.

This cross-sectional study exhibited several limitations. First, the sample size was limited, possibly leading to sampling bias. Second, the lipid levels of breast milk samples were not examined due to a design deficiency; hence, the contaminant concentrations are only calculated by wet weight. Third, a questionnaire about the daily diets of the subjects was not included, limiting the assessment of exposure to the contaminants. Finally, the information on the infants was not adequate for further analysis. The second round of investigation will start in 2019, still focusing on the body burden of POPs in residents living in proximity to the MWI, but all of the limitations mentioned will be improved.

4. Conclusion

In this study, the levels of PCDD/Fs and PCBs in the breast milk of mothers living in proximity to a 10-year-old MWI located in the Zhejiang Province of China was evaluated, and the EDI level of their infants was assessed. In conclusion, mothers and their breast-fed infants living in proximity to the MWI exhibited significantly higher body burden of PCDD/Fs and PCBs.

Conflicts of interest

The authors declare they have no existing or potential competing financial interests.

Acknowledgments

This study is supported by the Foundation of the Ministry of Health and Science and Technology (WKJ-ZJ-1606), the National Natural Science Foundation of China of China (81502786, 81472986, 81670833), National Health Commission of China (WKJ-ZJ-1606), the Science and Technology Department of Zhejiang Province (2017C37018), Program of Zhejiang Medical technology (2018KY337, 2015KYA055, 2012KYA047), Zhejiang Province Key Research and Development Program (2019C03091), and Fundamental Research Funds of the Central Universities. We are grateful to the study population and to all people who contributed to this work.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envpol.2019.04.069.

References

- IARC, 2006. Preamble to the IARC monographs on the evaluation of carcinogenic risks to humans.
- Chan, J.K., Xing, G.H., Xu, Y., Liang, Y., Chen, L.X., Wu, S.C., Wong, C.K., Leung, C.K., Wong, M.H., 2007. Body loadings and health risk assessment of polychlorinated dibenzo-p-dioxins and dibenzofurans at an intensive electronic waste recycling site in China. Environ. Sci. Technol. 41, 7668–7674.
- Chao, H.R., Wang, S.L., Su, P.H., Yu, H.Y., Yu, S.T., Papke, O., 2005. Levels of polychlorinated dibenzo-p-dioxins and dibenzofurans in primipara breast milk from Taiwan: estimation of dioxins and furans intake for breastfed infants. J. Hazard Mater. 121, 1–10.
- Costopoulou, D., Vassiliadou, I., Leondiadis, L., 2013. Infant dietary exposure to dioxins and dioxin-like compounds in Greece. Food Chem. Toxicol. 59, 316–324.
- De Felip, E., Abballe, A., Casalino, F., di Domenico, A., Domenici, P., Iacovella, N., Ingelido, A.M., Pretolani, E., Spagnesi, M., 2008. Serum levels of PCDDs, PCDFs and PCBs in non-occupationally exposed population groups living near two incineration plants in Tuscany, Italy. Chemosphere 72, 25–33.
- Deml, E., Mangelsdorf, I., Greim, H., 1996. Chlorinated dibenzodioxins and dibenzofurans (PCDD/F) in blood and human milk of non occupationally exposed persons living in the vicinity of a municipal waste incinerator. Chemosphere 33, 1941–1950.
- Deng, B., Zhang, J., Zhang, L., Jiang, Y., Zhou, J., Fang, D., Zhang, H., Huang, H., 2012. Levels and profiles of PCDD/Fs, PCBs in mothers' milk in Shenzhen of China: estimation of breast-fed infants' intakes. Environ. Int. 42, 47–52.
- EC, 2001. Opinion of the Scientific Committee on Food on the Risk Assessment of Dioxins and Dioxin-like PCBs in Food.
- EPA, U.S., 1994. Method 1613 Tetra- through Octa-Chlorinated Dioxins and Furans by Isotope Dilution HRGC/HRMS.
- EPA, U.S., 2003. Method 1668, Revision A Chlorinated Biphenyl Congeners in Water, Soil, Sediment, Biosolids, and Tissue by HRGC/HRMS. EPA-821-R-07-004.
- Helsel, D.R., 2006. Fabricating data: how substituting values for nondetects can ruin results, and what can be done about it. Chemosphere 65, 2434–2439.
- Industry, C.A.o.E.P., 2017. Development Report on Treatment Industry of Urban Domestic Refuse in 2017. China Environmental Protection Industry, pp. 9–15.
- Li, J., Zhang, L., Wu, Y., Liu, Y., Zhou, P., Wen, S., Liu, J., Zhao, Y., Li, X., 2009. A national survey of polychlorinated dioxins, furans (PCDD/Fs) and dioxin-like polychlorinated biphenyls (dl-PCBs) in human milk in China. Chemosphere 75, 1236–1242.
- Lu, D., Lin, Y., Feng, C., Wang, D., She, J., Shen, H., Wang, G., Zhou, Z., 2015. Levels of polychlorinated dibenzo-p-dioxins/furans (PCDD/Fs) and dioxin-like polychlorinated biphenyls (DL-PCBs) in breast milk in Shanghai, China: a temporal upward trend. Chemosphere 137, 14–24.
- Nadal, M., Perello, G., Schuhmacher, M., Cid, J., Domingo, J.L., 2008. Concentrations of PCDD/PCDFs in plasma of subjects living in the vicinity of a hazardous waste incinerator: follow-up and modeling validation. Chemosphere 73, 901–906.
- Reis, M.F., Sampaio, C., Aguiar, P., Mauricio Melim, J., Pereira Miguel, J., Papke, O., 2007. Biomonitoring of PCDD/Fs in populations living near Portuguese solid waste incinerators: levels in human milk. Chemosphere 67, S231–S237.
- Schuhmacher, M., Domingo, J.L., Kiviranta, H., Vartiainen, T., 2004. Monitoring dioxins and furans in a population living near a hazardous waste incinerator: levels in breast milk. Chemosphere 57, 43–49.
- Schuhmacher, M., Domingo, J.L., Llobet, J.M., Kiviranta, H., Vartiainen, T., 1999. PCDD/F concentrations in milk of nonoccupationally exposed women living in southern Catalonia, Spain. Chemosphere 38, 995–1004.
- Schuhmacher, M., Kiviranta, H., Ruokojarvi, P., Nadal, M., Domingo, J.L., 2009. Concentrations of PCDD/Fs, PCBs and PBDEs in breast milk of women from Catalonia, Spain: a follow-up study. Environ. Int. 35, 607–613.
- Schuhmacher, M., Kiviranta, H., Ruokojarvi, P., Nadal, M., Domingo, J.L., 2013. Levels of PCDD/Fs, PCBs and PBDEs in breast milk of women living in the vicinity of a hazardous waste incinerator: assessment of the temporal trend. Chemosphere 93, 1533–1540.
- Shen, H., Ding, G., Wu, Y., Pan, G., Zhou, X., Han, J., Li, J., Wen, S., 2012. Polychlorinated dibenzo-p-dioxins/furans (PCDD/Fs), polychlorinated biphenyls (PCBs), and polybrominated diphenyl ethers (PBDEs) in breast milk from Zhejiang, China. Environ. Int. 42, 84–90.
- Society, C.N., 2016. Guidelines for feeding infants aged 7-24 months. J. Clin. Pediatr. 34, 381–387.
- Sun, Z.q., Yang, Z.y., 2013. Evaluation of breast milk intake in exclusively breast-fed infants aged 0-6 months. Acta Nutr. Sin. 35, 134–136.
- Tajimi, M., Uehara, R., Watanabe, M., Oki, I., Ojima, T., Nakamura, Y., 2005.

Correlation coefficients between the dioxin levels in mother's milk and the distances to the nearest waste incinerator which was the largest source of dioxins from each mother's place of residence in Tokyo, Japan. Chemosphere 61, 1256–1262.

- Ulaszewska, M.M., Zuccato, E., Capri, E., Iovine, R., Colombo, A., Rotella, G., Generoso, C., Grassi, P., Melis, M., Fanelli, R., 2011. The effect of waste combustion on the occurrence of polychlorinated dibenzo-p-dioxins (PCDDs), polychlorinated dibenzofurans (PCDFs) and polychlorinated biphenyls (PCBs) in breast milk in Italy. Chemosphere 82, 1–8.
- van Leeuwen, F.X., Feeley, M., Schrenk, D., Larsen, J.C., Farland, W., Younes, M., 2000. Dioxins: WHO's tolerable daily intake (TDI) revisited. Chemosphere 40, 1095–1101.
- Xu, P., Lou, X., Ding, G., Shen, H., Wu, L., Chen, Z., Han, J., Han, G., Wang, X., 2014. Association of PCB, PBDE and PCDD/F body burdens with hormone levels for children in an e-waste dismantling area of Zhejiang Province, China. Sci. Total Environ. 499, 55–61.
- Yang, Y.H., Chang, Y.S., Kim, B.H., Shin, D.C., Ikonomou, M.G., 2002. Congener-distribution patterns and risk assessment of polychlorinated biphenyls, dibenzo-pdioxins and dibenzofurans in Korean human milk. Chemosphere 47, 1087–1095.
- Zhang, L., Yin, S., Li, J., Zhao, Y., Wu, Y., 2016. Increase of polychlorinated dibenzo-pdioxins and dibenzofurans and dioxin-like polychlorinated biphenyls in human milk from China in 2007-2011. Int. J. Hyg Environ. Health 219, 843–849.
- Zubero, M.B., Aurrekoetxea, J.J., Ibarluzea, J.M., Rivera, J., Parera, J., Abad, E., Rodriguez, C., Saenz, J.R., 2011. Evolution of PCDD/Fs and dioxin-like PCBs in the general adult population living close to a MSW incinerator. Sci. Total Environ. 410–411, 241–247.
- Zubero, M.B., Eguiraun, E., Aurrekoetxea, J.J., Lertxundi, A., Abad, E., Parera, J., Goni-Irigoyen, F., Ibarluzea, J., 2017. Changes in serum dioxin and PCB levels in residents around a municipal waste incinerator in Bilbao, Spain. Environ. Res. 156, 738–746.