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Body Loadings and Health Risk Assessment of Polychlorinated Dibenzo-*p*-dioxins and Dibenzofurans at an Intensive Electronic Waste Recycling Site in China

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This study is one of the very few investigating the dioxin body burden of a group of child-bearing-aged women at an electronic waste (e-waste) recycling site (Taizhou, Zhejiang Province) (24 ± 2.83 years of age, 40% were primiparae) and a reference site (Lin'an city, Zhejiang Province, about 245 km away from Taizhou) (24 ± 2.35 years of age, 100% were primiparae) in China. Five sets of samples (each set consisted of human milk, placenta, and hair) were collected from each site. Body burdens of people from the e-waste processing site (human milk, 21.02 ± 13.81 pg WHO-TEQ_{1998}/g fat (World Health Organization toxic equivalency 1998); placenta, 31.15 ± 15.67 pg WHO-TEQ_{1998}/g fat; hair, 33.82 ± 17.74 pg WHO-TEQ_{1998}/g dry wt) showed significantly higher levels of polychlorinated dibenzo-*p*-dioxins and polychlorinated dibenzofurans (PCDD/Fs) than those from the reference site (human milk, 9.35 ± 7.39 pg WHO-TEQ_{1998}/g fat; placenta, 11.91 ± 7.05 pg WHO-TEQ_{1998}/g fat; hair, 5.59 ± 4.36 pg WHO-TEQ_{1998}/g dry wt) and were comparatively higher than other studies. The difference between the two sites was due to e-waste recycling operations, for example, open burning, which led to high background levels. Moreover, mothers from the e-waste recycling site consumed more foods of animal origin. The estimated daily intake of PCDD/Fs within 6 months by breast-fed infants from the e-waste processing site was 2 times higher than that from the reference site. Both values exceeded the WHO tolerable daily intake for adults by at least 25 and 11 times, respectively. Our results implicated that e-waste recycling operations cause prominent PCDD/F levels in the environment and in humans. The elevated body burden may have health implications for the next generation.

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Introduction

Electronic waste, "e-waste" or "waste electrical and electronic equipment" (WEEE) defined by the European Community Directive 2002/96/EC, is a waste type consisting of any broken or unwanted electrical or electronic appliance. Obsolete e-waste has become a serious problem. The average lifespan of computers in developed countries has dropped from 6 years in 1997 to 2 years in 2005 (*1*). According to the State Environmental Protection Administration of China, 70% of worldwide e-waste has been sent to China (*2*). In the western United States, up to 80% of the e-waste collected for recycling is exported to Asia, of which 90% is sent to China due to lower environmental standards and cheaper labor (*3*). Moreover, in China alone, 4 million PCs are discarded annually (*4*).

There are several e-waste processing sites in China, of which Taizhou region in Zhejiang Province and Guiyu town in Guangdong Province are the most intensive (*2*). The "recycling" is done by rudimentary methods, which include burning wire piles to recover metals, melting circuit boards over coal grills to release valuable chips, cooking computer casings to remove combustible plastics and isolate metals, and extracting metals in acid baths (*5*, *6*). Our previous studies conducted in Guiyu showed that persistent organic pollutants (POPs) and heavy metals caused serious contamination in air, soil, sediment, and freshwater (*5*−*11*). Dioxins (PCDD/Fs), i.e., polychlorinated dibenzo-*p*-dioxins (PCDDs) and polychlorinated dibenzofurans (PCDFs), are of particular concern as burning of solid waste releases large amounts of these toxic compounds (*12*). Open burning and acid leaching were found to release the largest amount of dioxins of all e-waste operation processes (*8*), and open dumping sites are potential sources of dioxins due to uncontrolled burning (*13*). Therefore, uncontrolled burning and disposal of waste could impose serious threats to the health of workers and local residents. The World Health Organization (WHO) has coordinated exposure studies on dioxins in human milk since 1987 (*14*−*16*). Human milk samples were collected from several regions in China to determine levels of dioxins (*17*−*21*). However, information concerning human body burdens of dioxins at e-waste recycling sites is limited. Thus, the major objective of this study is to carry out a health risk assessment to investigate the body burdens of dioxins of local residents at Taizhou region.

Experimental Section

Sampling Site Description. The e-waste recycling site is Taizhou region (TZ) which is located in Zhejiang Province, eastern China (Figure 1). Taizhou is situated at the central coastal section of Zhejiang (28 °N latitude and 122 °E longitude) and has a coastline of 745 km, accounting for 28% of that of Zhejiang Province (*22*). E-waste recycling activities are mainly executed in a city called Luqiao situated in the southern part of Taizhou, with a total area of 274 km^2 and a population of 400 000.

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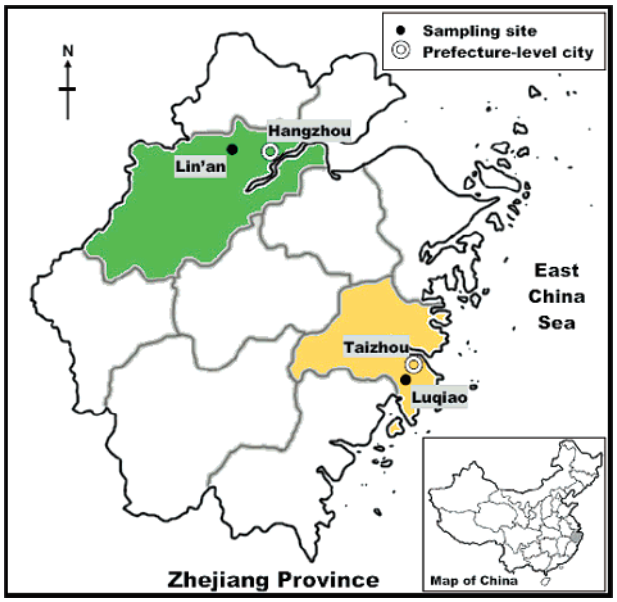

FIGURE 1. Sampling locations at Zhejiang Province, China.

The e-waste recycling activities in Taizhou started in the late 1970s and dealt mostly with domestically generated e-waste (*23*). In the early 1990s, Taizhou began to process imported e-waste. Currently, the imported e-waste contributes 90% of the total and originates mainly from Japan, the United States, Western European countries, and Russia. In recent years, Taizhou has become one of the main receivers of e-waste in China. It mainly receives scrap metals, obsolete electric capacitors, household appliances, electric generators, and cable wires. The annual volume of dismantled e-waste has exceeded 2.2 million metric tons, and the sales volume has reached 4 billion Chinese Yuans (approximate U.S. $5.16 hundred million). Forty thousand people are working in the e-waste recycling sector. Most of the recycling operations involve open air burning, acid leaching, and physical dismantling by hammer, chisel, screw driver, and bare hand.

The reference site is Lin'an city (HZ) (30 °N latitude and 118 °E longitude), Hangzhou prefecture, Zhejiang Province (*24*). It is about 245 km away from Taizhou. Lin'an is situated inland, at the northern part of Hangzhou, and it is a city with a land area of 3127 km^2 and a population of 520 000.

Study Population. Human milk, placenta, and hair samples were collected at the two study sites from 10 women who gave birth between August and December 2005. These 10 women were selected randomly under the conditions that they had been living in the study site for at least 2 years and they were willing to donate sufficient amounts of hair and milk for the chemical analysis. A set of specimens was collected from each individual. The donors were accessed through Luqiao Hospital of Traditional Chinese Medicine, Lin'an Hospital of Traditional Chinese Medicine, and the local Centers for Disease Control and Prevention. Before the collection of specimens, the donors completed an informed consent. A survey on their personal characteristics was conducted (see Table S1, Supporting Information).

Data Collection. Socio-demographic data and food consumption habits of the study population were obtained from face-to-face interviews and semiquantitative food intake questionnaires, respectively. During the interview, the donors answered questions modified from the second round of WHO's PCDDs, PCDFs, and PCBs exposure study (*14*). The intakes of different types of food were assessed (see Table S2, Supporting Information). Weekly intake (g/week) for each food item was computed for each individual. Dietary change over the course of pregnancy was ignored. The response rate was 100%.

Sample Collection. Human milk (~100 mL) was collected from each donor when the infant was 4−5 days old. The sample was manually expressed and collected in hexane-rinsed reagent bottles with Teflon-lined caps. Placenta was collected on the day of delivery and individually placed into cleaned glass containers. Hair (~3 g) was collected on the first day after the delivery and was sampled from near the scalp and from the nape of the neck using stainless steel scissors. It was then put into a sealable polyethylene bag. All the specimens were frozen immediately after collection and stored at −20 °C until chemical analysis.

Laboratory Analysis. The chemical analyses were conducted by The State Key Laboratory for Freshwater Ecology and Biotechnology, Institute of Hydrobiology, Chinese Academy of Sciences, China. Human milk and placenta samples were freeze-dried and then homogenized. Hair was washed with commercial detergent (1%) for 5 min, followed by distilled water, to remove on-surface impurities, e.g., hair care products, which may interfere with the chemical analysis (*25*). It was then oven-dried at 70 °C. Each sample was spiked with surrogate EDF-8999 and then Soxhlet-extracted based on U.S. EPA Method 3540C (*26*). The lipid contents of human milk and placenta samples were gravimetrically determined from an aliquot of the extract. A cleanup process was then carried out by using three columns filled with silica gel (*27*), activated basic alumina (*28*), and florisil (*29*), respectively. Internal standard (EDF-5999, $^{13}C_{12}$ 1,2,3,4-TCDD, and $^{13}C_{12}$ 1,2,3,7,8,9-HxCDD) (10 μL) was added to the concentrated sample for high-resolution gas chromatography/high-resolution mass spectrometry (HRGC/HRMS) analysis (*30*). The analyses were performed on an Agilent 5890II gas chromatograph (Agilent Technologies, U.S.A.) coupled to a Finnigan MAT 95s mass spectrometer (Thermo Electron, U.S.A.) equipped with a CTC A200S autosampler (CTC, Switzerland) at a resolution of 10 000. An RTX-DIOXIN2 capillary column (60 m × 0.25 mm i.d. × 0.25 μm f.t., Restek Co.) was used for the compound separation. The PCDD/Fs were identified by mass spectrometry in the EI and multiple ion detection modes.

Expression of PCDD/F Concentration. Levels of PCDD/Fs were indicated as mass concentrations and toxic equivalency (TEQ) of WHO 1998 (*31*). The body burdens were expressed as picograms of dioxin TEQs per gram of fat in human milk and placenta. They were also expressed as picograms of dioxin TEQs per gram of dry weight when comparison was made among three types of samples. The undetected values were treated as zero.

Data Analysis. The Statistical Package for Social Sciences (SPSS of Windows, version 11.0; SPSS Inc., Chicago, IL) was used for the quantitative data analysis. The differences among groups were assessed by the Student's t test or analysis of variance (ANOVA). Pearson correlation coefficients were used for determining the relationship among different parameters. The significant level was $p < 0.05$ and two-tailed. Data are presented as means ± SD.

Results and Discussion

Factors Affecting Dioxin Levels: Demographic Characteristics and Food Intake. Various factors were reported to be associated with the POP concentrations in human milk such as parity, age of mother, food intake preferences, and period of breast-feeding (*32*). In this study, the total TEQs increased with the length of residence in TZ ($r = 0.993$ and $p = 0.01$ for human milk, $r = 0.950$ and $p = 0.013$ for placenta) and the number of spontaneous abortions of the donor ($r = 0.929$ and $p = 0.023$ for hair). Increased spontaneous abortion was observed in dioxin-contaminated sites at Seveso and Chapaevsk, Russia (*33*). However, Takekuma et al. (*34*) did

TABLE 1. Concentration (pg/g and pg WHO-TEQ/g) of PCDD/Fs in Human Specimens from the E-Waste Recycling Site (TZ) and the Reference Site (HZ)[a,b,c]

		e-waste recycling site (TZ)			reference site (HZ)		
congener	WHO-TEF	human milk pg/g fat	placenta pg/g fat	hair pg/g dry wt	human milk pg/g fat	placenta pg/g fat	hair pg/g dry wt
PCDD congeners							
2,3,7,8-TCDD	1	3.59	4.88	2.31	3.11	4.74	4.48
1,2,3,7,8-PeCDD	1	4.65	10.56	7.45	1.47	2.46	0.326
1,2,3,4,7,8-HxCDD	0.1	2.11	2.43	2.42	1.21	1.66	0.106
1,2,3,6,7,8-HxCDD	0.1	4.76	1.10	4.08	2.80	0.717	0.106
1,2,3,7,8,9-HxCDD	0.1	2.30	0.606	2.10	0.948	0.539	0.065
1,2,3,4,6,7,8-HpCDD	0.01	10.2	7.06	12.4	5.08	5.85	1.31
OCDD	0.0001	80.9	49.8	22.6	41.0	52.7	6.80
∑PCDD-TEQ		9.27	15.93	10.74	5.13	7.55	4.84
PCDF congeners							
2,3,7,8-TCDF	0.1	9.54	5.95	46.6	1.35	1.60	1.16
1,2,3,7,8-PeCDF	0.05	6.86	8.70	23.7	0.404	0.725	0.142
2,3,4,7,8-PeCDF	0.5	15.4	31.4	24.6	6.06	6.51	0.705
1,2,3,4,7,8-HxCDF	0.1	10.4	14.53	14.8	3.81	3.74	1.01
1,2,3,6,7,8-HxCDF	0.1	9.23	6.76	15.2	3.08	2.77	0.744
2,3,4,6,7,8-HxCDF	0.1	6.12	2.73	14.0	2.66	1.55	0.740
1,2,3,7,8,9-HxCDF	0.1	0.323	0.372	2.71	0.134	0.436	0.0652
1,2,3,4,6,7,8-HpCDF	0.01	13.0	4.49	24.9	6.29	4.84	2.05
1,2,3,4,7,8,9-HpCDF	0.01	1.23	1.01	2.01	0.134	0.967	0.0652
OCDF	0.0001	1.45	2.03	6.62	0.417	7.18	1.87
∑PCDF-TEQ		11.7	19.22	23.1	4.22	4.36	0.752
∑PCDD/F-TEQ		21.0	35.1[c]	33.8[c]	9.35	11.9	5.59

[a] Abbreviations: HpCDD, hetachlorodibenzodioxin; HpCDF, heptachlorodibenzofuaran; HxCDF, hexachlorodibenzofuaran; HxCDD, hexahetachlorodibenzodioxin; OCDD, octachlorodibenzodioxin; OCDF, octachlorodibenzofuaran; TCDD, tetrahetachlorodibenzodioxin; TCDF, tetrachlorodibenzofuaran. [b] Detection limit (pg/g): milk, TCDD/F = 0.10−0.13, PeCDD/F = 0.10−0.17, HxCDD/F and HpCDD/F = 0.07−0.13, OCDD/F = 0.13−0.23; placenta, TCDD/F = 0.06−0.08, PeCDD/F = 0.06−0.10, HxCDD/F and HpCDD/F = 0.04−0.08, OCDD/F = 0.08−0.14; hair, TCDD/F = 0.15−0.20, PeCDD/F = 0.15−0.25, HxCDD/F and HpCDD/F = 0.10−0.20, OCDD/F = 0.20−0.35. [c] Significant difference between TZ and HZ samples ($p < 0.05$).

not find any significant correlation between residence years and TEQ values of human milk. Dioxin levels in human specimens were found to be positively correlated with the mother's age (*17*, *34*, *35*). Nevertheless, in the present study, the levels did not significantly vary with age ($p < 0.05$). This result was consistent with other studies related to POPs (*19*, *36*). Narrow age range, recent exposure of the contaminants (*37*), and limited sample size are possible reasons for the result. Parity did not show any correlation with body burden in this study ($p < 0.05$). Others studies also revealed such result (*36*, *38*). In contrast, Beck et al. (*35*) and Iida et al. (*39*) reported dioxin levels in human milk from primiparae were higher than multiparae. TEQ values in human milk from the dumping site of India showed a negative correlation with the number of deliveries (*38*).

The results of the present study showed that there was a statistically significant increase in TZ milk dioxin levels with the intakes of crab ($r = 0.888$ and $p = 0.044$) and egg ($r = 0.921$, $p = 0.026$). Moreover, the TEQ values for TZ placenta also significantly correlated with the intake of crab ($r = 0.924$, $p = 0.025$), whereas TZ milk fat proportion showed a significant correlation with the intake of shellfish ($r = 0.886$, $p = 0.045$). There were statistical correlations between HZ body loadings and the intake of foods of animal origin: bivalve ($r = 0.954$, $p = 0.012$ for human milk, $r = 0.920$, $p = 0.027$ for placenta), pork ($r = 0.904$, $p = 0.035$ for human milk, $r = 0.953$, $p = 0.012$ for placenta), beef/lamb ($r = 0.917$, $p = 0.028$ for human milk), chicken ($r = 0.913$, $p = 0.028$ for human milk), and animal stomach ($r = 0.981$ and $p = 0.019$ for hair). However, there was no significant correlation between total TEQs and the personal characteristics. These indicated that consumption of foods of animal origin was the main dietary exposure to dioxins. According to the results of dioxin surveys conducted in Taiwan and Japan, fish and shellfish contained the highest TEQ levels among foods of animal origin (*40*, *41*). Previous investigations showed that this group was the principal dietary source of PCDD/F intake accounting for 50−80%, while the secondary source was meat and eggs (*40*−*42*).

Comparison: Dioxin Body Burden of Mothers in the E-Waste Recycling Site and the Reference Site. The PCDD/F concentrations in human milk, placenta, and hair collected from the two sites are shown in Table 1. The total TEQ value of TZ human milk (21.02 ± 13.81, 11.59−44.60 pg WHO-TEQ/g fat) was about 2 times higher than that of HZ human milk (9.35 ± 7.39, 2.04−21.35 pg WHO-TEQ/g fat), although the difference was not significant ($p > 0.05$). However, TZ placenta (31.15 ± 15.67, 18.15−43.20 pg WHO-TEQ/g fat) and TZ hair (33.82 ± 17.74, 14.57−62.43 pg WHO-TEQ/g dry wt) showed statistically higher concentrations than the samples from HZ (placenta, 11.91 ± 7.05, 4.89−13.39 pg WHO-TEQ/g fat; hair, 5.59 ± 4.36, 2.41−13.38 pg WHO-TEQ/g dry wt) ($p < 0.05$). Background pollution level, dietary habit, and personal characteristics of the sample donors are the most important factors affecting the body burden. Although previous studies showed that less than 2% of human dioxin intake is from direct inhalation (*43*), TZ people are exposed more to the toxic chemicals via inhalation, in addition to dermal contact and consumption of local foods due to relatively high background contamination level. Human exposure to dioxins begins with atmospheric emissions (*35*), of which incineration releases the largest quantity of dioxins (*12*). Open burning of e-waste releases extremely high levels of dioxins (203−1100 pg WHO-TEQ/g dry wt in soil) (*8*). Moreover, significant amounts of dioxin-related POPs, e.g., PAHs (2065 μg/kg dry wt in soil, 124 ± 44.1 μg/m^3 in total suspended particles of air samples) and PBDEs (33 000−97 400 ng/g dry wt in combusted residue of plastic chips and cables) were found at the areas near e-waste recycling workshops (*5*−*7*). Hence, the background contamination in TZ should be more serious than that in HZ. This can be further confirmed by the dioxin hair analysis since dioxin levels in hair reflect those in the atmosphere (*44*−*47*). Our results showed that the level of dioxins in TZ hair (33.82 ±

17.74 pg WHO-TEQ/g dry wt) was significantly higher than that in HZ samples (5.59 ± 4.43 pg WHO-TEQ/g dry wt) ($p < 0.05$). Moreover, Nakao et al. (*45*) pointed out that hair collected from municipal solid waste (MSW) incineration workers contained total TEQ levels (4.37 pg WHO-TEQ/g) that were 2.5 times higher than those from the general population (1.74 pg WHO-TEQ/g). This implied that the higher background dioxin level at TZ compared with HZ was due to the e-waste recycling operation, particularly burning of wastes. Thus, the body loadings were significantly greater for TZ mothers than for HZ mothers.

Such difference could also be explained by personal characteristics and dietary intake. Of all the personal characteristics, parity and the number of breast-fed children showed significant differences between the two sites ($p < 0.05$) (see Table S1, Supporting Information). Forty percent of TZ mothers were primiparae, while all HZ mothers were primiparae. These women were selected according to the criteria mentioned in the Study Population section. Previous studies showed that increased parity or increased period of breast-feeding was directly related to a lighter body burden of PCDD/Fs (*35*, *38*, *39*). However, this phenomenon was not observed in this study. Thus, these two parameters did not appear to affect the body burden. On the other hand, according to the results of the food consumption survey, TZ mothers generally consumed more foods of animal origin, whereby the consumption of pork and animal stomach was significantly higher ($p < 0.05$) (see Table S2, Supporting Information). Foods of animal origin are considered to be the major dietary sources of dioxins since POPs are fat-seeking and accumulate in the fat of animals (*48*). As mentioned before, statistical analysis showed that higher intakes of this food class lead to greater body loadings in TZ mothers ($p < 0.05$).

Congener Concentrations and Homologue Profiles. The most toxic congener, 2,3,7,8-TCDD, was present in all samples from both sites. On average, 2,3,7,8-TCDD, 2,3,4,7,8-PeCDF, and 1,2,3,7,8-PeCDD contributed 7−80%, 6−45%, and 6−30%, respectively, to the total TEQs. These three congeners constituted up to 65−92%. The homologue profiles indicate the environmental fate and sources of dioxins (*49*). The samples of each TZ or HZ donor showed similar profiles to those of the representative profiles (obtained by using the average concentrations). All samples from both sites, except TZ hair, showed similar homologue profiles where PCDDs (TCDD, 18−64%; OCDD, 20−40%) contributed largely to the total concentration (see Figures S1a−S1d and S1f, Supporting Information). In contrast, TZ hair showed a distinct homologue profile which was dominated by tetra through hexachlorinated furans (73%) (see Figure S1e, Supporting Information). The reasons for the contradiction among TZ samples are unknown. The detailed fates of dioxins after entering human bodies via different exposures are not clearly recognized. It is suggested that the profile for hair might indicate more recent exposure of dioxins since hair directly contacts with the exposure media (atmosphere), whereas those for milk and placenta might reflect the historical exposures. However, this speculation may only be applied in highly contaminated areas since such difference was not observed in HZ samples. The profile for TZ hair closely resembled that for hair of workers at an emission source, such as an MSW incinerator (*45*). Both profiles were markedly different from those for the reference group or for the general population. Nakao et al. (*44*) also concluded that although the hair samples had been washed for chemical analysis, dioxins produced in the combustion process in the MSW incinerator was one of the exposures leading to dioxins in workers' hair as similar isomer patterns were also found in precipitation ash. Therefore, high levels of dioxins in TZ hair were caused by atmospheric exposure in addition to body accumulation. This gave further support that the e-waste recycling activity accounted for the fresh input of dioxins, leading to greater body burdens in TZ mothers.

Use of Human Specimens as Biological Indicators for Dioxin Contamination. Different types of human specimens showed advantages and disadvantages for assessing exposure. Dioxins in hair indicated body accumulation and atmospheric deposition onto the hair surface (*25*). Hair can be applied to all population groups. Its sampling is very convenient and noninvasive (*50*). Relatively high lipid content (3.5−4%) shows stability for the chemical analysis (*51*, *52*). However, it is difficult to distinguish between exogenous and endogenous contamination (*53*). Concentrations may be varied if hair from different positions of the head is collected (*25*). Moreover, different concentrations are found along the length of hair (*54*). Placenta provides a good indication for prenatal exposure (*21*). The variations among different donors can be reduced due to the same sampling time, usually on the day of labor. Moreover, placenta is ideal for biomonitoring as it is large enough to allow assay of several pollutants during the same sampling. Nevertheless, it is restricted to females and not easily available. Tissues need to be obtained by more invasive procedures (*53*). Human milk reflects maternal body burden and the postnatal transfer of dioxin from mothers to infants (*15*). Moreover, collection of human milk is convenient and noninvasive and is easily replicated. The high content of fat (2−5%) makes the extraction method easier and improves the precision of the measurements. However, human milk is restricted to lactating women. Furthermore, all the milk samples should be collected at the same breast-feeding time as dioxin levels vary along the breast-feeding period (*35*).

When comparing the three types of specimens from TZ, hair contained the highest dioxin concentration (33.82 ± 17.74 pg WHO-TEQ/g dry wt; $p = 0.001$), followed by human milk (2.96 ± 1.30 pg WHO-TEQ/g dry wt), and then placenta (2.06 ± 1.30 pg WHO-TEQ/g dry wt). Samples from HZ also showed a similar trend whereby PCDD/F concentrations in hair (5.59 ± 4.43 pg WHO-TEQ/g dry wt) were significantly higher than those in placenta (0.63 ± 0.28 pg WHO-TEQ/g dry wt; $p = 0.032$); however, there was no significant difference with those in human milk (2.20 ± 1.45 pg WHO-TEQ/g dry wt; $p > 0.05$). The TEQ values of the three types of TZ specimens showed significant positive correlations with each other ($r = 0.995$ and $p < 0.001$ for human milk and placenta; $r = 0.900$ and $p = 0.037$ for human milk and hair; $r = 0.892$ and $p = 0.042$ for placenta and hair). For HZ samples, only dioxin concentration in human milk showed a significant positive correlation with concentration in placenta ($r = 0.930$, $p = 0.022$). These results appeared to indicate that at highly contaminated sites, hair may be used as a reliable bioindicator for dioxin body burden. However, further investigations are needed for proving this hypothesis and for finding out the causes leading to different homologue profiles for different types of samples.

Comparison with Worldwide and Domestic PCDD/F Levels. *Human Milk.* All TZ samples and 80% of the HZ samples exceeded the EU's maximum permitted level in milk (3 pg WHO-TEQ/g lipid) (*55*). The concentrations of the former were high, whereas those of the latter were moderate when compared with the 26 countries involved in the WHO Third Exposure Study on Human Milk Dioxins (*15*) (see Figure S2, Supporting Information). For example, the concentrations in TZ human milk would be ranked number 2 after Egypt, whereas HZ showed concentrations similar to Ireland and Norway. Moreover, TZ human milk showed the highest concentrations among the samples collected from other parts of China during 2000−2005 (see Table S3, Supporting Information). HZ samples contained comparable concentrations with those from Dalian and Shenyang (*19*), Taiwan (*17*,

21), Shijiazhuang, and Tangshan (*56*). This further confirmed that the relatively heavy body burden of TZ mothers was due to e-waste recycling activities which are absent from the areas of other studies. Schecter and co-workers (*57*, *58*) investigated the dioxin contamination levels in an area which had been heavily sprayed with sodium pentachlorophenol (Na−PCP) to control schistosomiasis. Na−PCP is a pesticide which is contaminated with higher chlorinated PCDDs. Human milk samples collected from the sprayed areas contained concentrations that were 4-fold lower than those from TZ. However, those collected from the residents living near Ya-Er Lake, which had been heavily polluted by effluent from a chloroalkali manufacturing plant nearby, contained extremely high TEQ values (*59*).

Placenta. Concentrations in TZ samples were 2−3-fold higher than specimens from the general population of central Taiwan (*21*, *60*), the United States (*61*), and Germany (*62*) and were comparable with those from Japan (*63*) (see Table S3, Supporting Information). However, those collected from six mothers with Yu-Cheng disease caused by the consumption of PCB-contaminated rice oil in Taiwan showed markedly higher dioxin levels (*64*) than all other studies.

Hair. There are only a few studies on dioxins in human hair (see Table S3, Supporting Information). The highest TEQ value was found by the present study, followed by Luksemburg et al. (*65*) who collected samples from barber shops at an e-waste recycling site (Guiyu, Guangdong Province) in China. The total TEQs in hair and ash samples reported by this study were relatively high (21.0 and 5858.33 pg WHO-TEQ/g, respectively), suggesting that e-waste processing activities release a significant amount of dioxins. Hair collected from residents living close to Ya-Er Lake contained concentrations that were 25 times lower than TZ hair (*66*). However, human milk collected from Ya-Er Lake revealed higher levels than those from TZ (*59*). Such contradiction may imply that there was fresh input of dioxins to the atmospheric environment in TZ as hair is a good indicator for atmospheric dioxins (*44*, *46*, *47*, *63*).

Health Risk Assessment for Infants. In order to evaluate the risk faced by infants via breast-feeding, the estimated daily intake (EDI) of PCDD/Fs should be determined (see the Supporting Information for the calculation). TZ infants consumed 102.98 ± 67.65 pg TEQ/kg body wt/day which is 2 times higher than that of HZ infants (45.83 ± 36.22 pg TEQ/kg body wt/day). Both values would exceed the WHO tolerable daily intake (TDI) (1−4 pg TEQ/kg body wt/day) (*67*) by at least 25 and 11 times, respectively. Assuming that 90% of the ingested dioxins are absorbed (*68*), the infant's exposure would still be serious. The WHO TDI includes the exposure to dioxin-like PCBs; thus, the EDI for TZ infants might underestimate the risk. Although the TDI has been estimated for humans over their entire lifetime, the acute exposure via high intake exceeding the toxicological limit during breast-feeding is of concern due to the long half-life of the compounds and immature body defense of infants. Studies found that postnatal exposure to dioxins affects thyroid hormone system and immunological functions of infants and children (*69*, *70*). Moreover, it should be noted that infants and children absorb dioxins at a faster rate than adults due to their high growth rate (*71*). In addition, TZ infants are at greater risk due to higher background levels, implying that they would absorb more dioxins via dermal contact, inhalation, and prenatal exposure. However, breast-feeding should be encouraged, given that the infant receives nutritional advantages and protection against infectious diseases (*17*, *18*).

This is the first study investigating the PCDD/F levels in human milk, placenta, and hair collected from mothers at an intense e-waste recycling site and a reference site in China. Although a limited number of samples were analyzed, the results indicated that e-waste recycling operations caused elevated PCDD/F levels in humans and in the environment. The body burdens of dioxins in people from the e-waste processing site were ranked among the highest when compared on an international basis. The elevated body burden may impose health implications for the next generation. This study would be more comprehensive if the sample size is increased. However, the costs of chemical analysis of dioxins should be considered. On the other hand, work should be done on reducing the negative impacts of the e-waste recycling activities to the environment and humans. For example, education supported by the central and local governments should be carried out to teach local residents realize e-waste recycling is a problem and should be addressed. Moreover, further investigations on epidemiological studies of health impacts caused by e-waste recycling operations, dioxin levels in local foods, and kinetics about the POP partitions among different human tissues should be conducted.

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Supporting Information Available

Experimental Section, data collection, Discussion section, health risk assessment for infants, Tables S1−S3, and Figures S1a−S1f and S2. This material is available free of charge via the Internet at http://pubs.acs.org.

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