DOCTORAL THESIS

Dietary exposure, human body loadings, and health risk assessment of persistent organic pollutants at two major electronic waste recycling sites in China

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Dietary Exposure, Human Body Loadings, and Health Risk Assessment of Persistent Organic Pollutants at Two Major Electronic Waste Recycling Sites in China

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ABSTRACT

Total diet studies, covering 9 major food groups (freshwater fish, marine fish, shellfish, meat, poultry, egg, viscera, vegetables, and cereal), were conducted in two major e-waste recycling sites (Guiyu and Taizhou) which are located at coastal areas and a reference site (Lin'an) which is located inland to investigate the human dietary exposure to PBDEs and DDTs. The dietary intake of PBDEs by Guiyu women was 931 ± 772 ng/kg bw/day of which the intake of BDE-47 was 584 ng/kg bw/day exceeding the US EPA's reference dose (100 ng/kg/day). The intakes of PBDEs by Taizhou and Lin'an mothers were 44.7 ± 26.3 and 1.94 ± 0.86 ng/kg bw/day, respectively. Seafood, particularly freshwater fish, was the main dietary source of PBDEs in Guiyu and Taizhou, accounted for 88-98 %, while pork (41 %) was the main contributor in Lin'an. The levels of PBDEs in river fish from Guiyu (76.1-11353 ng/g wet wt) were the highest throughout the world. The exposure to DDTs through dietary intake in Taizhou was estimated at 52.1 ± 49.5 ng/kg bw/day, dominated by viscera (34 %) and seafood (38 %) (total 72 %). The value for Guiyu was 31.5 ± 34.8 ng/kg bw/day, largely contributed by vegetables (29 %), seafood (24 %), and poultry (34 %) (total 87 %), while that for Lin'an was estimated at 13.0 ± 6.51 ng/kg bw/day, dominated by pork (41 %), poultry (22 %) and vegetables (13 %) (total 76 %). The estimated daily intakes of DDTs of the study sites were far below the JMPR Provisional Tolerable Daily Intake (10,000 ng/kg bw/day).

Levels of PCDD/Fs in wild freshwater fish from Guiyu (4.64 ± 2.69 pg WHO-TEQ/g wet wt) were up to 12 and 107 times higher than Taizhou group (0.90 ± 0.38 pg WHO-TEQ/g wet wt) and Lin'an market fish (0.08 ± 0.01 pg WHO-TEQ/g wet wt) respectively and were at the high end of the worldwide range. The exposure to

PCDD/Fs via fish consumption by Guiyu women (1.95 ± 1.25 pg WHO-TEQ/kg bw/day) was at least 5 times higher than that by Taizhou (0.37 ± 0.36 pg WHO-TEQ/kg bw/day) and Lin'an women (0.03 ± 0.03 pg WHO-TEQ/kg bw/day). The maximum intakes of PCDD/Fs in Guiyu (4.31 pg WHO-TEQ/kg bw/day) exceeded the higher end of the WHO Tolerable Daily Intake (1-4 pg WHO-TEQ/kg bw/day). The results of dioxin-like activities of fish samples determined by H4IIE-luc cell bioassay demonstrated that H4IIE-luc cell bioassay was a very sensitive, cost-effective screening tool for assessing the overall dioxin-like toxicity in the samples.

Milk, placenta and hair were collected to determine the body burdens of PCDD/Fs, PBDEs and DDTs of lactating women at the study sites. Concentrations of PCDD/Fs in human milk (21.0 ± 13.8 pg WHO-TEQ/g fat), placenta (31.2 ± 15.7 pg WHO-TEQ/g fat) and hair (33.8 ± 17.7 pg WHO-TEQ/g dry wt) from Taizhou were significantly higher than those from Lin'an (milk: 9.35 ± 7.39 pg WHO-TEQ/g fat; placenta: 11.9 ± 7.05 pg WHO-TEQ/g fat; and hair: 5.59 ± 4.36 pg WHO-TEQ/g dry wt) and were comparatively higher than other studies. The daily intakes of PCDD/Fs by Taizhou and Lin'an infants via breast-feeding were estimated at 102.98 ± 67.65 and 45.83 ± 36.22 pg-TEQ/kg body wt/day, respectively. Both exceeded the WHO Tolerable Daily Intake by at least 25 and 11 times.

The $\sum_7$PBDE levels in milk samples from Guiyu (94.1 ± 86.4 ng/g fat) and Taizhou (70.7 ± 114 ng/g fat) were significantly higher than those from Lin'an (1.43 ± 0.81 ng/g fat) and topped the list of the nationwide and worldwide figures. The total PBDE levels of placenta (19.5 ± 30.0 ng/g fat) and hair (110 ± 210 ng/g dry wt) from Taizhou exceeded those from Lin'an (placenta: 1.02 ± 0.36; hair 3.57 ± 2.03 ng/g dry wt) by at least 19 times. Levels of DDTs in the body of donors from Guiyu (milk: 305 ± 109 ng/g fat) and Taizhou (milk: 360 ± 319 ng/g fat; placenta: 122 ± 109 ng/g fat; hair: 79.9 ± 215 ng/g dry wt) were also significantly higher than those from Lin'an (milk: 190 ±

131 ng/g fat; placenta: 49.2 ± 30.2 ng/g fat; hair: 10.8 ± 7.09 ng/g dry wt). The estimated daily intakes of PBDEs by Guiyu, Taizhou and Lin'an infants were 461 ± 423, 346 ± 559, and 7.01 ± 3.95 ng/kg bw/day, respectively; and those of DDTs were 1.69 ± 1.86, 1.48 ± 0.79, and 0.95 ± 0.73 μg/kg body wt/day, respectively. Therefore, at all study sites, breast-fed infants were faced with greater dietary exposure to PCDD/Fs, PBDEs and DDTs, except the case of PBDE in Guiyu, than their mothers.

The present study revealed that food and human specimens collected from the e-waste recycling sites were more contaminated with PCDD/Fs and PBDEs than the reference site because of the uncontrolled e-waste recycling operations which led to high background levels. Such differences were also observed in the case of DDTs, due to greater application of DDT at the coastal areas. The elevated levels of PCDD/Fs and PBDEs in food and human body observed in Guiyu and Taizhou may impose health implications for the next generation.

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