DOCTORAL THESIS

Human exposure and health risk assessment of polychlorinated biphenyls at two major electronic-waste recycling sites in China

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ABSTRACT

This study aimed to characterize PCB concentrations and distributions in different environmental media (air, dust and sediment), food samples and human specimens from two major electronic-waste recycling sites (Guiyu and Taizhou) in China; investigate human exposure to PCBs via dietary intake, inhalation, dust ingestion and dermal absorption; and study the bioaccessibility of PCBs in different food items.

Total diet studies, covering 10 major food groups (freshwater fish, marine fish, shellfish, meat, egg, viscera, vegetables, cereal, fruit and drinking water), were conducted at the two e-waste recycling sites compared with a reference site (Lin'an). The exposure to PCBs through dietary intake in Taizhou was estimated at 92.79 ng/kg/day, with contribution by freshwater fish (28%), rice (28%), meat (17%), shellfish (9%), vegetables (8%), egg (4%) and marine fish (2%). With regards to WHO-PCB-TEQ values, freshwater fish (40%), shellfish (24%), duck liver (18%) and rice (12%) were the main dietary sources with total daily intake (9.78 pg WHO-PCB-TEQ/kg/d) exceeding the FAO/WHO Tolerable Daily Intake (70 pg TEQ/kg body wt/month). At Guiyu, PCBs were only detected in seven species of freshwater fish from Lianjiang River, with an average level of 17.27 ng/g. The dietary intakes of PCBs by Guiyu and Lin'an mothers were 7.31 and 5.36 ng/kg/d, respectively.

Air samples containing gaseous and particulate phases were collected from e-waste workplaces and residential areas of the three study sites. The total concentration of PCBs at the open burning site (OBS) at Guiyu (472 ng/m^3) was the highest in the ambient air worldwide, followed by the residential area (RA) of Guiyu (19.5 ng/m^3), transformer recycling workshops (17.6 ng/m^3) at Taizhou, the residential area at Taizhou (3.37 ng/m^3), and the lowest at the residential area of Lin'an (0.46 ng/m^3). There was no significant difference between indoor and outdoor air at Taizhou. The

mean exposure rates for total PCBs via gaseous phase and particulate phase were estimated to be 1461 ng/d and 438 ng/d, respectively, for the e-waste workers working at OBS, which were significantly higher (p<0.01) than the corresponding mean exposure rates for the local residents at RA in Guiyu (388 ng/d for total air) and Taizhou (67 ng/d for total air). This indicated the importance of inhalation exposure for the e-waste workers engaged in OBS at Guiyu.

The mean concentration of total PCBs in dust was 310.8 ng/g dry wt at printed circuit board recycling workshops, which was significantly higher (p<0.01) than that at RA (44.57 ng/g dry wt) at Guiyu and Gurao Town (3.41 ng/g dry wt). In Taizhou, the highest levels of PCBs were found in the transformer recycling workshops with an average of 2824 ng/g dry wt, also significantly higher than that at RA (572.0 ng/g dry wt) and Lin'an (42.39 ng/g dry wt). WHO-PCB-TEQs levels in the workshops of Taizhou reached 2216 pg/g dry wt, due to the high abundance of PCB 126 (21.47 ng/g dry wt), which contributed 97% of WHO-PCB-TEQs. In general, dust PCB levels at Taizhou were higher than that at Guiyu, but higher levels of PCBs in the combusted residues of e-waste at Guiyu could be expected as extremely high levels of PCBs were shown in the air at OBS. Transformer recycling workers exposed to PCBs via dust ingestion and dermal absorption were estimated to be 7.75×10^{-4} and 3.60×10^{-4} pg WHO-PCB-TEQ/kg/d.

The mean concentrations of total PCBs in human milk, placenta and hair samples collected from Taizhou were 363 ng/g lipid, 224 ng/g lipid, and 386 ng/g dry wt, respectively, 7.6, 5.0 and 3.1 times, respectively, higher than those for the reference site (Lin'an). The mean PCB levels in human milk collected from Guiyu (0.68 ng/g lipid) was the lowest among the three study sites. When comparing with results obtained in the 3rd WHO-coordinated study, Taizhou topped the list of 32 countries/regions with

regards to WHO-PCB-TEQ values, due to the relatively higher contribution of PCB-126.

The average daily intake (ADI) levels of dioxin-like PCBs via food consumption, inhalation exposure, dust ingestion and dermal absorption were higher than the calculated values based on human milk for the e-waste workers and residents at both e-waste recycling sites, assuming 100% absorption rate. The average bioaccessibilities of PCBs in bighead carp, oriental weatherfish, spinach and Chinese white cabbage were 2, 3, 25 and 27%, respectively. Taking into account the bioaccessibility values, the ADI values would be significantly lower.

Dietary exposure (99.6% of the total exposure) was the dominant exposure route to PCBs intake for Taizhou residents. On the contrary, e-waste workers at Guiyu were exposed to PCBs mostly via inhalation (99.7%). Total PCBs ADI levels based on estimation of multiple route exposure were 39.9 and 97.4 ng/kg/day leading to carcinogenic risks of 8/100000 and 19/100000 for e-waste workers at Guiyu and Taizhou, respectively.

The PCBs homologue patterns in different matrices at the two e-waste recycling sites were reviewed, and it was revealed that from the source to the top of the food chain, high molecular weight PCBs, which are more toxic, tended to accumulate in the human body along the food chain for both e-waste recycling sites.

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