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Contamination by PCBs and BFRs in Vietnamese Human Milk Associated with Recycling of E-waste

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Abstract—The present study investigated the human exposure to PCBs, PBDEs and HBCDs in three Vietnamese e-waste recycling sites (EWRS) using breast milk as bioindicator. The general contamination patterns followed the order of PCBs > PBDEs > HBCDs except for some recycling workers with higher levels of PBDEs than PCBs. Compared with the urban site, human exposure to PBDEs in two EWRS was significantly higher, up to two orders of difference for a number of recyclers. These highly PBDE-exposed individuals also had significantly higher levels of HBCDs than the urban population. In EWRS, PBDE and HBCD positively correlated with each other but not with PCBs, suggesting different sources of PCBs and BFRs. Levels of penta- to deca-PCBs were found to be associated with consumption of lipid-rich food whereas levels of the lower PCB congeners and BFRs were related to recycling activities. Comparing the estimated daily intake of PBDEs with their reference doses, potential health risks associated with feeding mother milk were suggested for children of Bui Dau recycler mothers. To our knowledge, these preliminary results are the first to be reported on human exposure to BFRs related to e-waste recycling in Vietnam.

Keywords: breast milk, e-waste, HBCD, PBDE, PCB, risk assessment, Vietnam

INTRODUCTION

E-waste, obsolete electrical and electronic products containing hazardous substances, has become a subject of growing environmental concern in Asian developing countries due to a large volume of illegal import from developed nations (The Basel Action Network, 2002). Asian e-waste recycling sites (EWRS), due to primitive recycling techniques employed and uncontrolled disposal of unwanted materials, are potential sources of a wide range of toxic substances including brominated flame retardants (BFRs), polychlorinated biphenyls (PCBs), dioxins and heavy metals as reported in China (Wong *et al*., 2007; Liu *et al*., 2008). However, except for China, the contamination status of EWRS in most

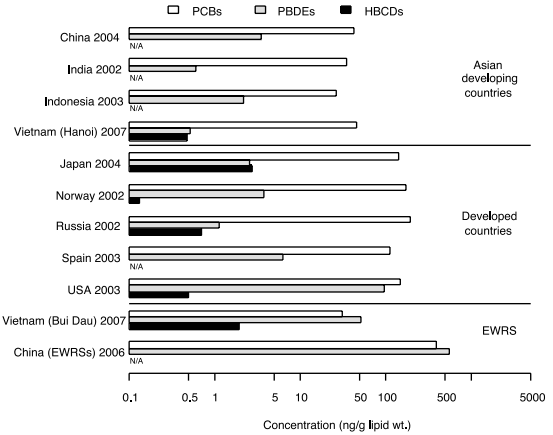

Fig. 1. Comparison of residue levels of PCBs and BFRs in human milk from Vietnam with other countries: China (Kunisue *et al*., 2004; Bi *et al*., 2006), India (Subramanian *et al*., 2007), Indonesia (Sudaryanto *et al*., 2008), Japan (Kunisue *et al*., 2006; Eslami *et al*., 2006; Kakimoto *et al*., 2008), Norway (Polder *et al*., 2008b), Russia (Polder *et al*., 2008a), Spain (Gómara *et al*., 2007), USA (She *et al*., 2007), Chinese EWRS (serum, Bi *et al*., 2007).

Asian countries has largely been unknown. The exposure pathways of toxic substances from e-waste to human was also not clearly understood. This report aimed to investigate the human exposure to PCBs, PBDEs and HBCDs related to e-waste recycling in Vietnam using breast milk as bioindicator in view of levels, distribution and possible influencing sociodemographic factors such as age, lactation time, dietary habit and occupational exposure to e-waste, etc.

MATERIALS AND METHODS

Sample collection

Human milk samples were collected from four locations in the Red River delta region, northern Vietnam: a typical urban area, Hanoi (HN), for reference and from three recycling villages in the provinces of Hai Phong (Trang Minh - TM, e-waste recycling) and Hung Yen (Bui Dau - BD, e-waste recycling and Dong Mai - DM, battery recycling). The samples were kept in ice during transport and ultimately stored at –20°C until analysis. Informed consents and exposure questionnaires were obtained from all donors.

Chemical analysis

Approximately 40 g of sample was freeze-dried then Soxhlet-extracted with 300 ml diethyl ether and 100 ml hexane for 7 h. The extract was then treated and the analytes (PCBs, PBDEs and HBCDs) were quantified according to the methods described elsewhere (Isobe *et al*., 2007). Procedural blanks were

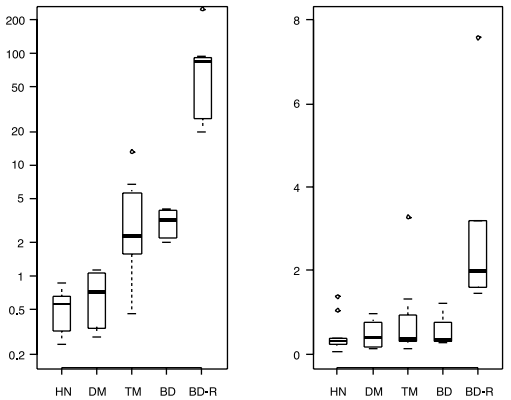

Fig. 2. Comparison of BFR concentration in breast milk from the populations of Hanoi (HN), Dong Mai (DM), Trang Minh (TM), Bui Dau non-recyclers (BD) and Bui Dau recyclers (BD-R) in 2007.

analyzed simultaneously with samples to check for interferences and contamination. Concentrations were expressed on a lipid weight basis unless otherwise specified.

Statistics

The Wilcoxon rank sum test was used for significant difference in concentrations of contaminants between groups. Principal component analysis (PCA) was used to explore the relationship among chemicals as well as between chemicals and sociodemographic parameters obtained from the questionnaires. Only compounds detected in at least 70% of the samples were included in this analysis. All calculations were performed using the statistical software package R version 2.8.0 at 95% confidence interval.

RESULTS AND DISCUSSION

PCBs

Total PCB levels in human milk were not significantly different in the four locations, with median of respectively 46, 50, 33 and 28 ng/g for HN, DM, TM and BD. This indicates that exposure to PCBs is rather uniform in the Vietnamese population. PCB levels in Vietnamese human milk are among the highest in Asian developing countries but lower than in developed nations (Fig. 1). The contamination levels in Vietnamese EWRS, in term of total PCBs, are also lower than in Luqiao, the largest Chinese disassembly site of electrical waste (Zhao *et al*., 2007).

In term of individual PCB congeners, EWRS tend to have higher concentrations of *tri* and *tetra* congeners than in HN: mean total CB-28, -44, -49,

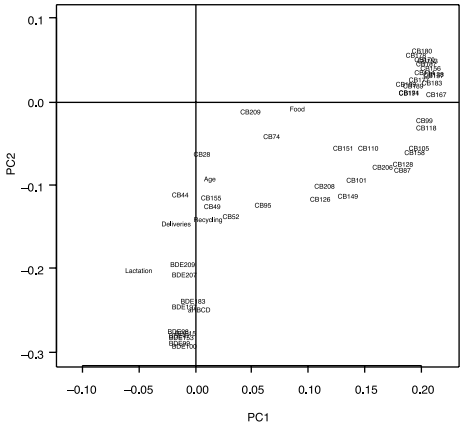

Fig. 3. Principal component analysis on chemical concentrations and sociodemographic parameters.

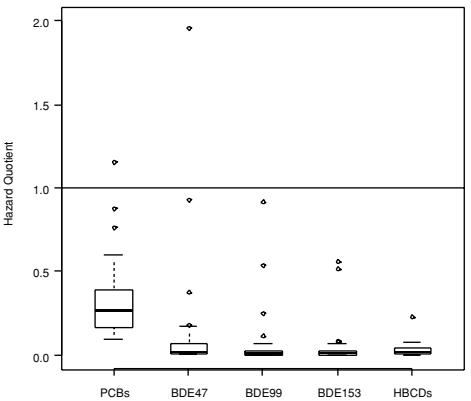

Fig. 4. Hazard quotients of PCBs and BFRs in mother milk for consumption by infants.

-52 and -70 were 11 ng/g in DM ($p = 0.15$), 3.8 ng/g in TM ($p < 0.05$) and 4.4 ng/g in BD ($p = 0.4$) vs. 2.3 ng/g in HN. Concentrations of *penta* and higher chlorinated PCBs, on the other hand, were higher in HN than in EWRS ($p < 0.05$): 42 ng/g vs. 25 ng/g in DM, 28 ng/g in TM and 23 ng/g in BD. This difference in accumulation of PCB congeners may be related to exposure pathways which will be discussed in a later section.

BFRs

PBDE concentrations in human milk from EWRS, with the exception of the battery recycling site (DM), were significantly higher than in the urban site (Fig. 2). In TM, the PBDE levels in recyclers and non-recyclers were similar with a

median higher than in HN by a factor of 4. In BD, non-recyclers had comparable PBDE levels with TM, whereas the recycler group had a much greater median level, higher than the urban level by a factor of 150, indicating high exposure to BFRs from e-waste. These recycling workers were also the only group with higher HBCD levels than the urban populations (factor 6). In a comparison of BFR levels in human milk from different countries (Fig. 1), the Vietnamese urban had ones of the lowest levels in the world but had higher HBCD levels than Norway (Polder *et al*., 2008b). TM was similar to other Asian developing countries whereas BD was in comparable range with the world's highest levels of PBDEs (US, She *et al*., 2007) and HBCDs (Japan, Kakimoto *et al*., 2008) in non-occupationally exposed populations. However BD levels were still an order lower than serum levels of Chinese e-waste dismantling workers from Guiyu (Bi *et al*., 2007).

The BDE congener pattern in HN milk seems to indicate a secondary exposure through diet, with the dominant being BDE-47, followed by BDE-153; and BDE-209 at non-detectable levels. For EWRS, primary exposure to Octa and DecaBDE technical mixtures from e-waste was likely to occur as BDE-209 was detected in most of the samples with varying proportions up to 50% of total PBDEs. Other highly brominated congeners also featured more prominently in EWRS, especially in BD where BDE-197 and BDE-207 were comparable to BDE-47.

Influencing factors

PCA results (Fig. 3) suggest that the exposure sources of *tri-*, *tetra*-CBs and BFRs were different than higher chlorinated PCBs: higher PCBs were aligned with consumption rate of lipid-rich food (Food variable) whereas lower PCBs and BFRs aligned with the time involved in e-waste recycling (Recycling variable). As such, the urban population tend to accumulate more higher PCBs due to a richer diet whereas the e-waste recycling population, most notably in BD tend to accumulate more BFRs.

Infant health risk

The health risks for breastfed infants associated with PCBs and BFRs in mother milk were assessed using hazard quotients (HQs), ratios between the estimated daily intakes (DIs) of chemicals with corresponding reference doses (RfDs). The DIs were calculated based on an assumed consumption rate of 700 g milk/day by a 5-kg infant (Oostdam *et al*., 1999). The RfDs used were: 1 μg/kg/day for PCBs (Oostdam *et al*., 1999), 0.1, 0.1, 0.2 μg/kg/day for BDE-47, -99, -153 (EPA, 2008a, b, c) and 0.2 μg/kg/day for HBCDs (European Chemicals Bureau, 2007). As shown in Fig. 4, the majority of HQ values are below 1, indicating low risk from the target contaminants. The highest HQs for PBDEs, with several values close to or over 1, belong to BD recycler mothers indicating potential health risk for their children. Higher HQs of PCBs are possibly due to a longer usage history of PCBs in Vietnam and not related to e-waste recycling.

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