Title

Characterization of PCBs and BFRs in human breast milk from the coastal area of the highly industrialized city of Surabaya, Indonesia

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Aim

- To evaluate legacy and novel BFRs contamination in the milk of mothers living in the coastal area of the highly industrialized city of Surabaya, Indonesia.
- To assess the potential health risks to humans through breastfeeding of infants.

Analytical Procedures

Study location and sampling site. Surabaya City is the capital of East Java Province. It is the second-largest city in Indonesia and is characterized by high population growth (Hakim, 2009). Contamination by heavy metals in the human milk and blood of children has been reported in this city, particularly in the coastal area of Kenjeran, as well as in riverine and coastal sediments (Arifin, 2011). Additionally, our previous studies have

reported contamination by legacy POPs (PCBs, PBDEs, and HBCDs) in the environment and biota collected from Surabaya City (Ilyas et al., 2010; Ilyas et al., 2011a; Ilyas et al., 2011b; Ilyas et al., 2013).

Sample collection and questionnaire. During this study, a total of 67 human milk samples were collected from mothers living in the Benowo dumpsite (n=30) and in the Kenjeran coastal area, Surabaya City (n=37), from August to September 2008 and stored in the Environmental Specimen Bank (es-Bank) of the Center for Marine Environmental Studies (CMES), Ehime University, Japan. However, because of the insufficient number of samples for the analysis of organic compounds, this study only analyzed human milk samples from the coastal area (n=28). A questionnaire survey was also conducted to collect related information from all mothers, such as age, body weight, height, dietary fish, parity, lactation period, and occupation, following the procedure described previously by Sudaryanto et al. (2008), Tue et al. (2010), and Thomsen et al. (2010a) with minor modifications.

Chemical analysis. Forty-two PBDE congeners from mono- to deca-BDE, BTBPE, DBDPE, and 3 HBCD isomers were analyzed in this study. The concentrations of PBDE congeners and HBCD isomers were summed to determine the total concentration, which was expressed as nanograms per gram of wet weight (ng g^{-1} ww) and nanograms per gram of lipid weight (ng g^{-1} lw). Unless otherwise specified, identification, quantification, and instrumental analysis of PBDEs were performed following the methods previously described by Sudaryanto et al. (2008), Malarvannan et al. (2009) and Tue et al. (2010). The median recovery values were in the range of 68–115% for PBDEs, BTBPE, and DBDPE and 90% for HBCDs. A blank sample was also analyzed in every batch to monitor the potential contamination and interference of the samples. Repeatability obtained from three replicate analyses was maintained at less than 20% of the variance. Concentrations below the detection limit (DL) were assigned zero values.

Statistical analysis. Statistical analyses were performed using Microsoft Excel. Differences between contaminant levels were determined using the Mann-Whitney U-

test, whereas correlations between congener levels and contaminants were evaluated using Pearson's product-moment correlations. Significance was set at a probability (p) value < 0.05. The levels of contaminants and hazard quotients were calculated and plotted using Box and Whisker.

Results and discussion

Participant characteristics. The participants in this study were breastfeeding mothers living in the coastal areas of Kenjeran, Surabaya. Factors predicted to influence contaminant levels were identified as follows: age (22–43 years), weight (36 – 60 kg), lactation period (0.5 - 30 months); parity (1–4 children), city inhabitant (1–41 years), fish diet (a little to very much), and occupation period as a housewife (1-25 years).

Contaminant levels. Concentrations of PBDEs (0.089 – 13 ng g^{-1} lw), DBDPE (<DL – 3.5 ng g^{-1} lw), HBCDs (<DL – 5.7 ng g^{-1} lw), and BTBPE (<DL – 0.03 ng g^{-1} lw). Higher levels of PBDEs than HBCDs, DBDPE, and BTBPE indicated high PBDEs consumption in Surabaya City. In addition, the detection of NBFRs in human milk, particularly DBDPE, indicates the heavy and widely used by industries. The mean concentration of PBDEs (1.6 ng g-1 lw) was in the ranges of the concentrations reported in several cities (Jakarta, Bogor, Purwakarta, and Lampung) in Indonesia (0.49 – 13 ng g^{-1} lw) (Sudaryanto et al., 2008a), and lower than reported from Malate, the Philippines (7.8 ng g^{-1} lw) (Malarvannan et al. 2009), Venice, Italy (240 ng g^{-1} lw) (Ingelido et al. 2007), and from Belgium (BDE-47 (1.7 ng g^{-1} lw); BDE-153 (0.4 ng g^{-1} lw)) (Pirard et.al., 2003). The median concentration of HBCDs (0.023 ng g^{-1} lw) was lower than reported in Norway (0.86 ng g^{-1} lw) (Polder et al., 2008). The mean and median concentrations of DBDPE were 0.48 and 0.091 ng g^{-1} lw, respectively, while concentrations of BTBPE were almost undetectable in the human milk samples. The Mann-Whitney U test indicated no significant difference between legacy and novel BFRs ($p > 0.05$).

Congener patterns. In all samples, PBDEs were dominated by congeners BDE-47 and BDE-209, whereas HBCDs were dominated by the α-isomer. However, other congeners selected from di- to deca-BDEs were also detected. The variation among samples was strongly distressed by BDE-209, BDE-47, and BDE-15. Variations were also

strongly influenced by BDE197+ 204, BDE-183, BDE-153, BDE-99, BDE-100, and BDE-201, indicating that PBDEs accumulation varied among individual mothers. Darnerud et al. (2001) have reported that the major route of human exposure to PBDEs was through food. On the other hand, direct exposure to PBDEs could also occur through the daily usage of commercial products containing BFRs and/or inhalation of indoor house dust, mainly PBDE lower congeners such as BDE-47 and BDE-99. It has been reported that higher molecule PBDEs, such hepta-BDE to deca-BDE, were less efficient in transferring from blood to breast milk compared to lower molecule PBDEs, as well as low bioaccumulation potential (Darnerud et al., 2001). The occurrence of some higher molecule PBDEs, such as BDE-209, BDE-201, and BDE-183, in the present study could be due to the large amount of Deca-BDE product mixture usage instead of the products of debromination of this congener. The high retention of deca-BDE in body fat has also been reported (Darnerud et al., 2001). Further studies are needed to reveal the exposure pathways to legacy BFRs, such as PBDEs and HBCDs, in the coastal area of Surabaya. Similar congener profiles of PBDEs and HBCDs in human milk have been reported in several countries, including Vietnamese e-waste recycling sites (Tue et al., 2010), the Philippines (Malarvannan et al., 2009), and Norway (Thomsen et al., 2010b). It has also been found in other locations in Indonesia (Sudaryanto et al. 2008a).

Factors controlling levels. The PBDEs, HBCDs, DBDPE, and PBDE congeners were correlated using Pearson product-moment correlations at a significance level of $p < 0.05$. BDE-183, BDE-153, BDE-99, BDE-47, BDE-28, and BDE-15 showed significant correlations with human milk debromination. High exposure to the Deca-BDE product mixture may explain the significant correlations between the congeners BDE-209, BDE-207, BDE-201, and BDE-197. The significant correlations between PBDEs, congener BDE-209, and DBDPE suggest that Surabaya City has used novel BFRs, such as DBDPE, as flame retardants alongside Deca-BDE product mixtures. The study found that HBCDs (primarily α-HBCD) and PBDEs had different accumulation dynamics and exposure pathways; therefore, their levels did not correlate. Our previous study found a good correlation between HBCDs (predominantly -HBCD isomer) and PBDEs in

sediments and soils from this location (Ilyas et al., 2011; Ilyas et al., 2013). Thus, the accumulation of α-HBCD isomers in the biota and human milk is more likely.

Future study

Further analysis will be conducted on the potential human exposure to PCBs, BFRs, and OCPs through seafood consumption. Finally, we estimated the health risks of seafood exposure by using the target hazard quotient (THQ) and carcinogenic risk (CR).

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