Levels of dioxins and polybrominated diphenyl ethers and longitudinal changes of dioxins in human milk in northern China

Suju SUN
Associate professor,
Hebei Medical University,
Department of Occupational and Environmental Medicine, College of Public Health

Objectives
Persistent organic pollutants (POPs) are a group of chemicals which enter the environment due to accidental emissions or to human activities such as mining, industrial emissions, agricultural practices, waste disposal, combustion, heating, and traffic. There are widely distributed levels in air, water and other environment. As their high stability and lipophilic properties, they accumulate in the food chain and are toxic to humans and wildlife. The studies on polychlorinated dibenzodioxins and dibenzofrans (PCDD/Fs), dioxin-like polychlorinated biphyls (dl-PCBs) and polybrominated diphenyl ethers (PBDEs) in human milk from Mainland China have been relatively limited. So far, no longitudinal comparison on PCDD/Fs, dl-PCBs have been carried out in China.

The aims of the present study are to investigate the spatial differences of PCDD/Fs, dl-PCBs and PBDEs in northern China and to elucidate longitudinal comparison of PCDD/Fs, dl-PCBs in humans living in Shijiazhuang area. In the same time, we investigated the dietary habits of donors from these areas to assess the dietary risk factor and to identify the associate between food contamination and these chemicals.

Methods
We collected human milk samples from 60 breast-feeding mothers in Tianjin, 48 in Yantai and 50 in Shijiazhuang from November 2006 to April 2007, and from 30 mothers in Shijiazhuang from November 2002 to February 2003, respectively. The three areas are located in northern China. Shijiazhuang is a large inland industrial city with a population of 9.2 million. Tianjin is a large harbor and industrial city, with a population of approximately 9.5 million. Yantai, with its well-
developed fisheries, is a coastal city with a population of approximately 6.5 million people. All of the specimens were collected within 1–8 weeks postpartum. Informed consent was obtained from all of the subjects. For the samples of 2007, we selected randomly 20 samples from each area for detection of PCDD/Fs, dl-PCBs such as non-ortho PCBs (no-PCBs) and mono-ortho PCBs (mo-PCBs) and PBDEs by high-resolution gas chromatography/high-resolution mass spectrometry (HRGC/HRMS). All the human milk samples in Shijiazhuang in 2002 were also determined the PCDD/Fs and dl-PCBs by HRGC/HRMS.

Results

PCDD/Fs and dl-PCBs concentrations were expressed as total toxic equivalency (TEQ) pg g⁻¹ fat (WHO-2005-TEF). The average concentrations of PCDD/Fs and PCDD/Fs plus dl-PCBs were highest in Tianjin. The PCDD/Fs levels were 5.08 TEQ pg g⁻¹ fat in Tianjin, 3.95 TEQ pg g⁻¹ fat in Shijiazhuang and 3.58 TEQ pg g⁻¹ fat in Yantai. The concentrations of PCDD/Fs plus dl-PCBs were 7.54 TEQ pg g⁻¹ fat in Tianjin, 6.24 TEQ pg g⁻¹ fat in Shijiazhuang and 6.69 TEQ pg g⁻¹ fat in Yantai. Mothers from Yantai had the highest total no-PCBs and total dl-PCBs levels. Total no-PCBs in Yantai measured 2.97 TEQ pg g⁻¹ fat, versus 2.01 TEQ pg g⁻¹ fat in Shijiazhuang and 2.28 TEQ pg g⁻¹ fat in Tianjin. Total dl-PCBs levels were 3.11 TEQ pg g⁻¹ fat in Yantai, 2.29 TEQ pg g⁻¹ fat in Shijiazhuang, 2.46 TEQ pg g⁻¹ fat in Tianjin. For total mo-PCBs, the highest concentration was found in Shijiazhuang: 0.28 TEQ pg g⁻¹ fat versus 0.19 TEQ pg g⁻¹ fat and 0.14 TEQ pg g⁻¹ fat in Tianjin and Yantai, respectively.

Comparing with 2002, the levels of PCDD/Fs and dl-PCBs in TEQ and the level of PCDF and mo-PCBs in pg g⁻¹ fat in human milk in Shijiazhuang were higher in 2007.

The highest concentration of PBDEs was found in Yantai compared with that in Shijiazhuang and Tianjin. The average concentrations of PBDEs were 3.71 ng g⁻¹ fat, 3.42 ng g⁻¹ fat and 4.16 ng g⁻¹ fat in human milk from Shijiazhuang, Tianjin and Yantai, respectively.

Discussion

Our results showed that differences in human milk of PCDD/Fs and dl-PCBs in the three areas might be due to differences in industrial level and dietary habits. As a historical base for the chemical industry in China, Tianjin has been the producer of plastic, hexachlorocyclohexane (HCH), lindane, chlorobenzene, and pentachlorophenol for approximately 30-50 years. Compared to developed countries, the level of PCDD/Fs and dl-PCBs in the three areas of northern China are relatively low. But the levels of PCDD/Fs and dl-PCBs in human milk in Shijiazhuang have increased from 2002 to 2007. For the congener profiles, we found comparable congener profiles among human milk from Shijiazhuang, Tianjin, Yantai, Dalian and Shenyang. But the congener profiles in human milk from these areas were different from Hong Kong, Germany and Japan. This
implied that there might be similar main source in these regions in China, however, which were not same to the Hong Kong, Germany and Japan.

The total PBDE levels found in this study were compared with those reported for other countries. The mean levels of total PBDEs in the three regions were lower than that of the United Kingdom and that of USA. The levels of total PBDEs in Shijiazhuang and Tianjin were comparable to those reported in southern China, Japan and Germany, while the concentration in Yantai was found to be comparable to Taiwan.

We found the mild correlation between total PBDE concentration and intake of sea fish and pork, but there was no relationship after adjusts by other food intake, age, BMI and education through a multiple regression analysis. It suggested that sources of exposure to PBDEs in northern China involve factors other than food intake.

The congener profiles of PBDEs in the three areas were relatively similar. High brominated congeners (BDE209, BDE207, BDE197, and BDE153) and low brominated congeners (BDE15 and BDE28) were the predominant congeners in the samples from the three areas.

**Conclusion**

Our results show different distribution of PCDD/Fs, dl-PCBs and PBDEs in the human milk among Tianjin, Yantai and Shijiazhuang of northern China. The levels of PCDD/Fs plus dl-PCBs in the average TEQ in the human milk from Shijiazhuang in 2007 were higher than that of in 2002. The consumption of freshwater fish might be a major contributor to total mo-PCBs, and sea fish consumption was a major contributor to total no-PCBs in human milk in these areas. Rigorous preventive measures and monitoring is needed to ensure a decreasing trend in dioxin emissions and sustainable development in China.