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A national survey of polychlorinated dioxins, furans (PCDD/Fs) and dioxin-like polychlorinated biphenyls (dl-PCBs) in human milk in China

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ABSTRACT

The study examined the levels of dioxin-like compounds in 24 pooled samples that were comprised of 1237 individual human milk samples from 12 provinces of China. The samples were taken in different regions to evaluate the body burden of these contaminants and assess their health risk to breast-feeding infants of China. The pooled samples were analyzed for 17 polychlorinated dibenzo-p-dioxins and dibenzofurans (PCDD/Fs) and 12 dioxin-like polychlorinated biphenyls (dl-PCBs) congeners. The population of the provinces involved in this study accounts for approximately 50% of the total Chinese population. The range of upper-bound total-TEQ in samples was from 2.59 pg TEQ g^{-1} lipid to 9.92 pg TEQ g^{-1} lipid with a mean of 5.42 pg TEQ g^{-1} lipid and a median of 5.11 pg TEQ g^{-1} lipid. PCDD/Fs-TEQ and total-TEQ in human milk from rural areas were lower than those from urban areas. The positive correlations were found between total-TEQ level in human milk and the consumption of aquatic food and meat respectively. The mean estimated daily intake of PCDD/Fs and dl-PCBs by infants was 28.0 pg TEQ kg^{-1} bw per day with a range from 14.2 pg TEQ kg^{-1} bw per day to 48.6 pg TEQ kg^{-1} bw per day. In the study, both the TEQ body burden of the sample population and estimated daily TEQ intake of breast-feeding infants were lower than those of developed countries. Continuous surveillance on PCDD/Fs and dl-PCBs levels in human milk is needed to correctly evaluate both the environmental impact and human health risk in China.

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1. Introduction

Polychlorinated dibenzo-p-dioxins and dibenzofurans (PCDD/Fs) and dioxin-like polychlorinated biphenyls (dl-PCBs) are a group of structurally related chemicals that are ubiquitous, persistent and lipophilic pollutants. These compounds can be accumulated in animal and human tissue causing toxic responses. Due to the potential for adverse health and reproductive outcomes, scientists, public health officials, and environmental regulators are concerned about their levels.

Human milk is a convenient specimen for monitoring of PCDD/Fs and PCBs because relatively large volumes can be collected non-invasively. The concentrations of these compounds in human milk can reflect the levels and trends of contamination in the local environment. Human milk has been used for monitoring human body burdens of PCDD/Fs, PCBs for decades in some countries such as, German (Raab et al., 2007). The Stockholm Convention will rely on human milk monitoring to aid in the assessment of the effectiveness of the Treaty in reducing emissions of POPs into environment. Since 1987, the World Health Organization (WHO) has coordinated three rounds of exposure studies on Dioxin using human milk (Hedley et al., 2006). However, in China, only Hong Kong participated in the third round of the WHO study initiated in 2000 (Hedley et al., 2006). The studies on PCDD/Fs and PCBs in human milk from mainland China have been relatively limited. Studies already undertaken have a bias to areas with heavy contamination such as Taizhou, an electronic waste (e-waste) recycling site in China (Chan et al., 2007). To be nationally representative, women of various socioeconomic backgrounds and geographic location should be included (Landrigan et al., 2002).

In the present study, the current levels of PCDD/Fs and dl-PCBs in human milk from 12 provinces of China was examined in order to evaluate the body burden in various geographic regions and assess the health risk of these contaminants to breast-feeding infant. In our previous study, the dietary intake of PCDD/Fs by adults from China was estimated by total dietary study (TDS) in 2000 (Li et al., 2007). For evaluation of the effect of various dietary habits between regions, the survey was performed in the same regions, as the previous TDS.

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2. Materials and methods

2.1. Donor selection and sample collection

The approach for donor selection and sampling human milk was based on the 'Guidelines for Developing a National Protocol' of the Fourth WHO-Coordinated Survey of Human milk for Persistent Organic Pollutants in Cooperation with UNEP (WHO, 2007). Some modifications were made for the special condition within China, for example, the maximum age of mother was 35 in this study instead of 30 in the Guidelines. Mothers donating milk were informed of the nature and purpose of the study and signed the consent forms.

As shown in Fig. 1, the samples were collected from 12 provinces including Heilongjiang, Liaoning, Hebei, Henan, Shanxi, Ningxia, Jiangxi, Fujian, Shanghai, Hubei, Sichuan and Guangxi. The population of the provinces involved in this study accounts for about 50% of the total population of China.

In each province, one urban site and two rural sites were selected for sampling. In each urban area, 50 donors were selected, and in each rural area, 30 donors were selected. A written questionnaire was completed to record the content of the face-to-face interview for each mother. The information included date of birth, place of birth, residence record, dietary habits (vegetarian/non-vegetarian), consumption of animal origin food including aquatic food, meat, egg and milk, occupation before pregnancy, and the indoor using of DDT at home.

The sampling was carried out from August to November in 2007. Mothers provided the samples at the local contact places where collection was supervised. At least 50 mL of milk was collected from each mother. The samples were collected directly to the pre-washed collecting jars and were stored at −20 °C until analysis.

It was initially targeted to collect 1320 samples. Finally, 1237 individual samples were collected. The ages of mothers ranged from 18 to 35 with a mean of 25.6. According to the questionnaires, none of the mothers smoked, and none used DDT in their home.

2.2. Sample pooling and analysis

The individual samples were grouped according to regions. For each province, the individual samples from the urban area were pooled into one pooled sample and individual samples from the two rural areas were pooled into one pooled sample. As a result, 24 pooled samples were created. The information of each pooled sample is listed in Table 1.

A 100 mL pooled sample was freeze-dried before being blended with anhydrous sodium sulfate. After spiking with $^{13}C_{12}$-labeled internal standard, samples were Soxhlet-extracted with a mixture of *n*-hexane and dichloromethane (1:1, 250 mL) for 24 h. Gravimetric lipid determination was performed after solvent evaporation. Bulk lipid removal was achieved by shaking the extract with acid-modified silica gel and a further clean-up was completed using a Power Prep instrument (Fluid Management Systems, Waltham, MA, USA) with multiple silica columns, alumina columns and carbon columns. Two fractions containing PCDD/Fs and dl-PCBs congeners were then collected. After concentrating the sample to approximately 10 ul, the $^{13}C_{12}$-labeled injection standards for PCDD/Fs and dl-PCBs were added to the final extracts, respectively.

The identification and quantification was performed by a GC-HRMS (MAT95XP ThermoFinnigan, Germany) with a DB-5MS cap-

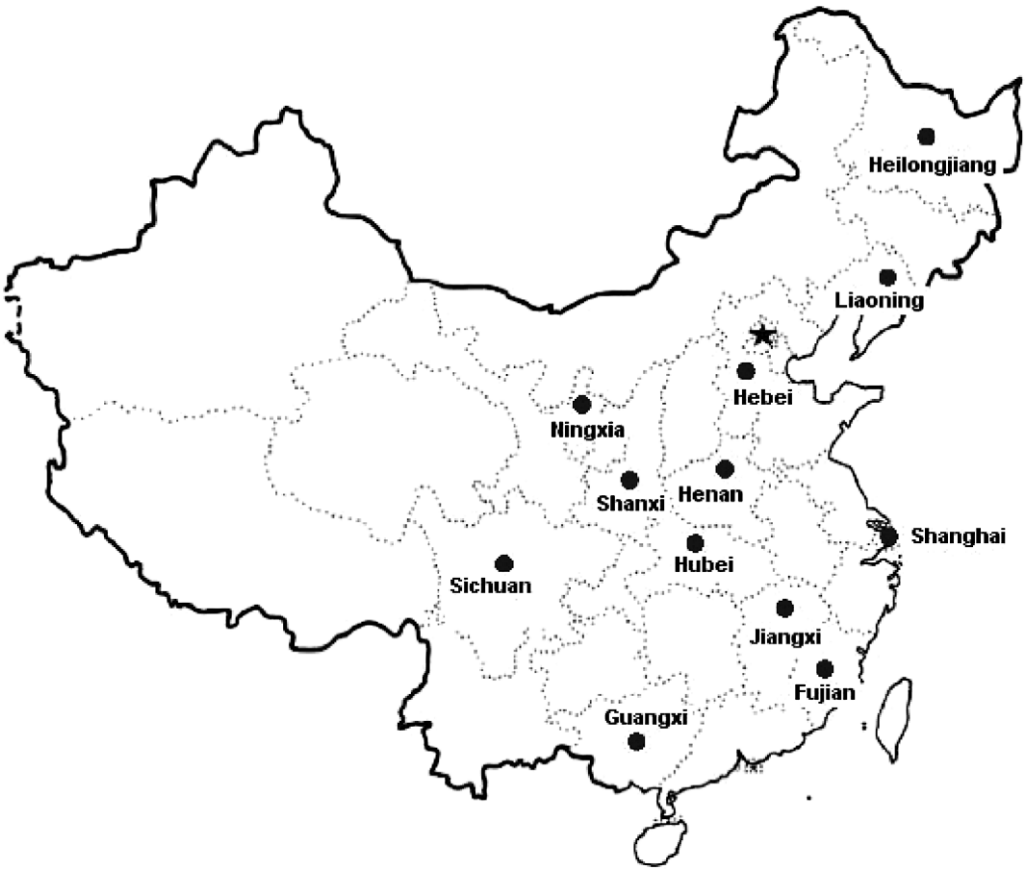

Fig. 1. The provinces for sampling.

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