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Levels and patterns of DDTs in maternal colostrum from an island population and exposure of neonates

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ABSTRACT

Dichlorodiphenyltrichloroethane (DDT) was heavily used in the past in many regions of the world. The occurrence of DDTs in island populations may be elevated if the island is adjacent to major DDT consumption estuaries, such as the Yangtze River Delta. In this study, colostrum samples were collected from maternal-neonate pairs (n = 106) from the Shengsi Island, located directly downstream from the Yangtze River outlet. DDT isomers and enantiomer compositions were analyzed by gas chromatography equipped with mass spectrometer (GC/MS) and GC/MS-MS. The average levels of *p,p'*-DDE, *o,p'*-DDD, *p,p'*-DDD, *o,p'*-DDT, *p,p'*-DDT and total DDTs were 1.32, 0.03, 0.09, 0.08, 0.48, and 1.93 μg g^{-1} lipid weight, respectively. Maternal age and pregnancy body mass index (BMI) were positively associated with levels of DDTs ($p < 0.05$). High (DDE+DDD)/DDT and *p,p'*-DDE/*p,p'*-DDT ratios suggested that current DDT residues originated primarily from historical use of DDT products, but new sources may also contribute partially to some high *o,p'*-DDT/*p,p'*-DDT ratios. Enantiomeric enrichment was found for the (−)-enantiomer of *o,p'*-DDD and the (+)-enantiomer of *o,p'*-DDT, suggesting stereoselective attenuation. Based on breast milk consumption, the average daily intake of DDTs by neonates was 8.33 ± 7.34 μg kg^{-1}bw per day, which exceeded the WHO's tolerable daily intake guideline of 0.01 mg kg^{-1} bw per day by 25%, implying some neonates in the Yangtze River region are potentially at high risk from exposure to DDTs.

1. Introduction

Dichlorodiphenyltrichloroethane (DDT) was widely used in agricultural production and vectors control in the second half of last century. Although DDT was banned in many countries under the Stockholm Convention, it is still listed as a ubiquitous environmental contaminant by the United Nations Environment Programme (UNEP, 2004) because of its uncontrolled use in certain regions and long environmental persistence. Moreover, DDT and its derivatives (DDTs) can undergo biomagnification through successive trophic levels in food chains, posing human health risks (Turusov et al., 2002; Farhang et al., 2005; Ntowa et al., 2008).

China was the second largest producer and consumer of DDT in the world. From 1946 to 1983, the total usage of DDTs in China was 4×10^5tons, accounting for 20% of the global production (Zhou et al., 2014). The Yangtze River Delta, within the watershed of the largest river in China and most important region for agricultural and industrial development (Yang et al., 2005), has been reported for its widespread DDT contamination (Müller et al., 2008). For example, Zhejiang and Jiangsu provinces of the delta region had an estimated total consumption of technical DDT of 6×10^4tons (Zhou et al., 2014), and an analysis of 58 sites in Zhejiang Province showed that the levels of DDTs in soil were from 4.0 to 530 ng g^{-1}(dry weight, dw), suggesting a potential human health risk in this region (Zhang et al., 2012). Studies further concluded that seafood could be the primary source of DDT intake for Chinese residents, accounting for 75% of the total exposure (Nakata et al., 2002).

Shengsi Island, where this study was conducted, is geographically part of Zhoushan Archipelago of Zhejiang Province. It is located at the mouth of the Yangtze River, facing the East China Sea and in the middle of the largest fishing ground (Zhoushan islands) in China (Fig. 1). Seafood is deeply rooted in the diets of this island's residents, which constitutes a critical pathway to accumulate DDTs in human tissues. Hence, there exists a potential for elevated exposure and risk to the island population whose main diet is often seafood (Nakata et al., 2002). A regional survey showed the

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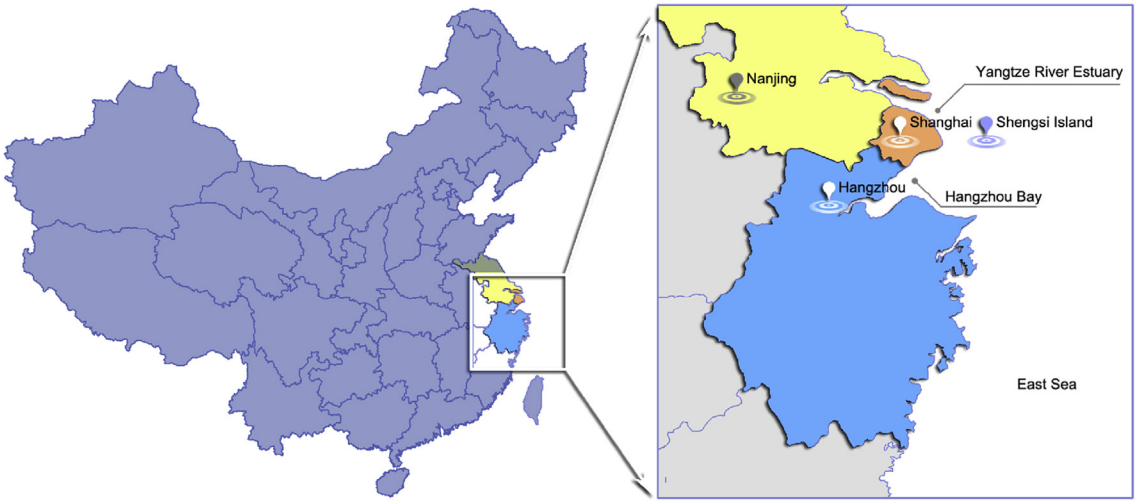

Fig. 1. Map of Yangtze River Delta showing the geological location of Shengsi Island.

occurrence of hexachlorocyclohexane (HCH), DDTs and chlordane in 11 mollusk species from coastal areas along the Yangtze River Delta, with DDTs exhibiting the highest levels ranging from 6.22 to 398.19 ng g^{-1} (wet weight, ww) (Zhou et al., 2014). The analysis of 13 types of seafood from four counties around Zhoushan Island found levels of organochlorine pesticides (OCPs)from 259 to 3460 ng g^{-1}(lipid weight, lw), with DDTs accounting for 32.1–89.0% of the total OCPs (Wang et al., 2014). Because of its unique geographical location, inhabitants of Shengsi Island are likely subject to serious long-term exposure to DDTs in China, warranting a close scrutiny (Kunisue et al., 2004).

Pregnant women and their neonates are the most sensitive population groups to chemical exposure. Breast milk, collected non-invasively, is an ideal matrix for evaluating long-term cumulative exposure of mothers (Fürst et al., 1994; Wong et al., 2005; Devanathan et al., 2009). In particular, colostrum secreted during the very early days after neonatal birth, could reflect the first critical exposure period for breast milk feeding. Colostrum is unique because of its lower fat and higher growth factors, secretory IgA, lactoferrin, oligosaccharides, anti-inflammatory cytokines, antioxidants, soluble CD14 and other protective components than the mature milk (Meier et al., 2010). As the first and often primary dietary source for neonates, chemicals from the mothers may be more easily transferred through colostrum to neonates, resulting in exposure of breast-fed infants to DDTs (Mishra and Sharma, 2011). However, the transfer of DDTs through colostrum to neonates has not been well studied in the east coastal region of China. Moreover, earlier surveys on DDTs in breast milk largely confined on the total DDT level by accounting for only *p,p'*-DDE (dichlorodiphenyldichloroethylene) and *p,p'*-DDT. A less understood aspect is the chiral signature of *o,p'*-DDD (dichlorodiphenyldichloroethane) and *o,p'*-DDT that may be used as a powerful tool to infer sources of contamination (Yang et al., 2005).

To help address these issues, we examined contamination profiles of DDTs in colostrum of mothers inhabiting in the Shengsi Island downstream from a major DDT use region, investigated maternal characteristics influencing the exposure patterns, identified the source of contamination, with a particular emphasis on the enantiomeric enrichment of *o,p'*-DDD and *o,p'*-DDT, and estimated the exposure risk of DDTs to neonates via breastfeeding. Results from this study contributed to a better understanding of exposure of DDTs among island inhabitants, and provided guidance on establishing safety thresholds in breastfeeding in this and similar contaminated areas (Niu et al., 2013; Gao et al., 2015).

2. Materials and methods

2.1. Sampling site and sample collection

Shengsi Island (Fig. 1) is in a marginal area of the East China Sea. It faces Yangtze Estuary and Hangzhou Bay, shares a marine border with the Pacific Ocean to the east, Putuoshan to the South, Shanghai to the west, and the Yellow Sea to the north. The island has land coverage of 88 km^2, and a population of close to 80,000 as of the end of 2013.

A total of 120 pregnant women who gave births between July 2011 and May 2012 living on the Shengsi Island were recruited to participate in the study. Screening criteria were puerperal mothers without any maternal history of serious illness, obvious clinical symptoms, or drug addictions, and neonates without congenital diseases. Seven individuals were excluded after the initial screening. After excluding the undersized samples (n = 4) and faulty samples (n = 3), a total of 106 maternal-neonatal pairs were included in this investigation. The colostrum (about 50 mL) was collected manually during the first five days after neonatal birth. The colostrum samples were stored at −20 °C before analysis.

All the donors were asked to finish a questionnaire which included questions on maternal age, pre-pregnancy body mass index (BMI, defined as the body mass divided by the square of the body height, and is universally expressed in the unit of kg m^{-2}), pregnancy weight gain, reproductive history (parity), education level, birth gender and weight. All the information obtained is concluded in Table S3 of SI. Informal consent was obtained from each donor. The study protocol was approved by the ethics committee of the participating hospitals.

2.2. Sample preparation and extraction

Extraction and cleanup of DDT residues from the colostrum was carried out according to the U.S. EPA method 608 with some modifications (Kumar et al., 2006). All the colostrum samples were thawed and homogenized prior to extraction. A 2-mLsubsample was spiked with the surrogate standards2, 4, 5, 6-tetrachloro-*m*-xylene (TCMX) and decachlorobiphenyl (PCB 209) (J&K Chemical, Beijing, China) in 30-mLglass centrifugation tubes (Chen et al., 2008). Each sample was then added with 0.5 mL formic acid,