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DDT levels in human milk in Hong Kong, 2001–02

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ABSTRACT

High levels of *p,p′*-DDT were detected in Hong Kong breast milk sampled in 1976 and 1985. Monitoring DDT levels in human breast milk in this region is important to identify trends in exposure. As part of the 2002–03 WHO/EURO coordinated exposure study, the concentrations of DDT and its metabolites were determined in 10 pooled milk samples classified by geographic origin and dietary history, comprising milk samples from 238 primiparous mothers giving birth in Hong Kong. Analysis was performed by Gas Chromatography (GC) with electron capture detector and confirmed by GC/Mass Spectrometry. The sum-DDT concentration (range: 0.92–2.05 mg/kg fat) was age-dependent and indicated a decreasing trend since the 1970s. Mothers who recently came from mainland China had higher *p,p′*-DDT to *p,p′*-DDE ratios, suggesting a more recent exposure compared to women mainly residing in Hong Kong. The average Hong Kong sum-DDT level (1.50 mg/kg fat) was among the highest of the contemporary levels (range: 0.12–1.97 mg/kg fat; median: 0.40 mg/kg fat) in the 16 countries/regions participating in the 2002–03 WHO/EURO exposure study. This is probably due to the previous extensive DDT exposure and continuing use of DDT in agriculture in mainland China. Despite the apparent decrease of DDT body load over 30 years, the environmental health hazard from DDT contamination in the Pearl River Delta region remains a concern. Measures to eradicate illegal use of DDT in mainland China and regular food monitoring programs are needed in the region. Despite the presence of DDT and other persistent organic pollutants in human breast milk, breastfeeding should continue to be strongly supported for its life-long benefits to infants.

1. Introduction

DDT (Dichlorodiphenyltrichloroethane) was extensively used worldwide in the 1960–80s, with large quantities released for both agricultural and vector control applications. China has been one of the nations with the highest production and consumption of DDT. Between the 1950s and the 1980s, the amount of DDT production in China was 0.4 million tons, accounting for 20% of the total world production (Hua and Shan, 1996). The high lipid solubility combined with persistence leads to the retention of DDT and its metabolites in fatty tissue of organisms along the food chain and food is the major source of human exposure. They are also present in the optimal food for infants, human breast milk, and their levels had been measured since 1960s. The main metabolite in human milk is p,p'-DDE with levels up to a factor of 20 times higher than *p,p′*-DDT levels. Following the international bans on DDT since the 1970s because of its ecological consequences, a drop in the levels of DDT in human tissues or breast milk was observed worldwide (Smith, 1999). The same was observed in the Special Administrative Region of Hong Kong, adjacent to the southern aspect of mainland China where widespread use of DDT in agriculture was common before the bans. Since the ban of DDT in China in 1983 and Hong Kong in 1987, the *p,p'*-DDT and *p,p'*-DDE levels in breast milk have fallen from a high level (respectively, 5.2 and 13.67 mg/kg fat) in the 1970s (Ip, 1983) to, respectively, 2.17 and 11.67 mg/kg fat in 1980s (Ip and Phillips, 1989) and much lower levels (0.39 and 2.48 mg/kg fat) in the 2000s (Wong et al., 2002).

However, recently concerns for environmental contamination by DDT in the Pearl River Delta were raised by reports of increased DDT levels found in the riverine and estuarine sediments (Mai

Abbreviations: DDT, dichlorodiphenyltrichloroethane; DDE, dichlorodiphenyldichloroethylene; DDD, dichlorodiphenyldichloroethane; GC/MS, gas chromatography/mass spectrometry; HCH, hexachlorocyclohexanes; IARC, International Agency for Research on Cancer; WHO, World Health Organisation.

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et al., 2002) and water column (Luo et al., 2004; Zhou et al., 2001). There is speculation that environmental contamination by DDT in the Pearl River Delta region is still continuing (Fu et al., 2003). As new source, dicofol contributed up to 93%, 81% and 22% to the current DDT pollution in atmosphere, water and sediments, respectively, in the Taihu Lake region, China (Qiu, 2005). This contamination was correlated with a particularly high level of o,p′-DDT.

IARC (1991) concluded that there is insufficient evidence in humans but sufficient evidence in experimental animals to classify DDT as a possible carcinogenic to humans (Group 2B). However, body loads of DDT also raise concerns about potential effects on developing infants and children because DDT transfers across the placenta from mother to fetus and exposure continues through breastfeeding after birth (Shen et al., 2007). Inconclusive findings have been reported for the effect of DDT exposure on growth (Karmaus et al., 2002; Gladen et al., 2000; Rogan et al., 1986; Gladen et al., 2003; Longnecker et al., 2001), mental and psychomotor development (Ribas-Fito et al., 2003; Rogan and Gladen, 1991; Gladen et al., 1988; Dorner and Plagemann, 2002) and other health outcomes including neurological development, immunity and infections (Dewailly et al., 2000; Karmaus et al., 2003; Hardell et al., 2002; Rogan et al., 1987) of infants and children. Increases in the odds of preterm birth and small-for-gestational-age (Longnecker et al., 2001), adverse effects on psychomotor development at two years (Rogan and Gladen, 1991) and brain development in school age children (Dorner and Plagemann, 2002) have been suggested. As a precautionary approach to protect the health of the developing fetus and children from possible adverse effects of DDT, reinforcement of the restrictions on the illegal use of DDT is needed. Monitoring DDT levels in human breast milk, which indicates the degree of its exposure, is also of importance. Such monitoring will now become more important since in 2006, after a ban for almost 30 years, WHO recommended the use of indoor residue spraying of DDT for controlling malaria in areas with high rates of transmission (Rehwagen, 2006).

The WHO Regional Office for Europe (WHO/EURO) initiated a series of international studies to monitor the concentrations of PCDDs, PCDFs and PCBs in breast milk of primiparous women (Malisch and van Leeuwen, 2003; Yrjanheikki, 1989; WHOEURO, 1996). Hong Kong participated for the first time in the 2002–03 third round study with analyses of dioxins (Hedley et al., 2006), and other persistent organic pollutants. We report here the levels of DDT in 10 breast milk pools from Hong Kong and their comparison with the median levels among 16 countries/regions participating in the same study.

2. Materials and methods

2.1. Sample and data collection

In December 2001 to September 2002, we recruited 316 primiparae who gave birth to a singleton in Hong Kong to donate milk samples at 2–6 weeks postpartum. Mothers were interviewed face-to-face to collect dietary and residential information. The questionnaire design, milk sampling method and pooling strategy were adapted from the protocol for the 2002–03 WHO/EURO coordinated dioxin exposure study and are detailed elsewhere (Hedley et al., 2006). The food intake of the subjects was assessed by a semi-quantitative food frequency questionnaire containing 102 potential dioxin-rich food items.

DDT and its metabolites, including *o,p′*-DDE (dichlorodiphenyldichloroethylene), *p,p′*-DDE, *o,p′*-DDT, *p,p′*-DDT, *o,p′*-DDD (dichlorodiphenyldichloroethane) and *p,p′*-DDD were determined in 10 milk pools comprising 238 individual milk samples. The pools were created to represent relatively homogenous characteristics in terms of the mothers' residential background (Hong Kong, mainland China, China immigrants with varying periods of residence in Hong Kong), dietary habits (consumption of dairy products, fish and seafood, including riverine and marine sources, in Pools 2–7) or smoking (Pool 1) (Table 1). There were three milk pools of 78 milk samples without DDT content determined due to insufficient milk fat or heterogeneous geographic characteristics of the mothers within the pools.

The mean age of donors in each of the 10 milk pools ranged from 26.5 years to 32.6 years. The mean age of babies at the time of sampling the mothers was 4.1–4.6 weeks among pools. The majority of the mothers were never-smokers ($n = 202$, 85%). The mothers who were ever-smokers (26.7 ± 5.0years) were significantly younger than the non-smokers (30.1 ± 4.6years). The mean levels of consumption of dairy products, fish and seafood were comparatively lower in the pools designed to represent mothers with low dietary exposure to dioxins and dioxin-like PCBs (Pools 4 and 6). Mothers who recently came from mainland China (Pools 6–8) were likely to be younger, less educated and with less household income compared with mothers who resided in Hong Kong for longer periods (Pools 1–5 and 9–10).

Due to the low breastfeeding rate in Hong Kong, partially breastfeeding mothers were also included. Fifty-nine percent of our subjects practiced exclusive breastfeeding (100%) at sampling while 23% predominately breastfed (>80%) their infants. Mothers from mainland China were more likely to practice exclusive breastfeeding. However, the levels of dioxin and dioxin-like PCBs concentrations in the partially breastfeeding mothers were no different to those of the exclusively breastfeeding mothers when controlled for mother's age. (Nelson et al., 2006) All participants gave written consent before taking part in the study. The study was approved by the Ethics Committees of the University of Hong Kong, the Chinese University of Hong Kong and the Department of Health, Hong Kong SAR Government.

2.2. Chemical analyses

Mass concentrations of DDT and its metabolites were determined routinely by gas chromatography (GC) with electron capture detector (ECD) and confirmed by GC with mass spectrometry (GC/MS) in the State Institute for Chemical and Veterinary Analysis of Food, Freiburg, Germany in 2003–04. The pesticide laboratory has successfully participated in 35 proficiency tests in 1994–04. The quality control procedures for pesticide residues analysis followed the Guidelines for Residues Monitoring in the European Union (Document N° SANCO/10476/2003). The recovery rates of internal standards and analyses were in the range of 70–120% which met the requirements of the Guidelines.

As part of the determination of PCDD/Fs, dioxin-like PCBs and marker PCBs, fat and other contaminants of interest were extracted from freeze-dried human milk samples by means of continuous hot extraction device (Twisselmann extractor) with Ethanol/Toluene (70/30) for 8 h. The hot extraction has a similar functional principle as Soxhlet extraction but it allows the highest possible temperature to be maintained in the sample during extraction and therefore increases the solubility of substances, allowing extraction in a faster time. The crude fat extract was purified with butyl methyl ether (Malisch and van Leeuwen, 2002). Up to 0.5 g of the fat extract was redissolved in cyclohexane/ethyl acetate and the internal standards 2,4,5-trichlorobiphenyl and Mirex, solved in cyclohexane, were added.

The applied clean up-parts of the analytical method follow the principles of the European standardized methods, Fatty food-Determination of pesticides and PCBs, EN 1528 part 1–4, 1996–10 (confirmed 2001). To remove the fat, gel permeation chromatography was performed on a chromatography column