Environmental Pollution 159 (2011) 524–531

Contents lists available at ScienceDirect

Environmental Pollution

journal homepage: www.elsevier.com/locate/envpol

National survey of the levels of persistent organochlorine pesticides in the breast milk of mothers in China

Pingping Zhou [a], Yongning Wu [a,*], Shian Yin [a], Jingguang Li [a], Yunfeng Zhao [a], Lei Zhang [a], Huijing Chen [a], Yinping Liu [b], Xin Yang [a], Xiaowei Li [a]

[a] Key Laboratory of Chemical Safety and Health, and National Institute of Nutrition and Food Safety, Chinese Center for Disease Control and Prevention, 7 Panjiayuan nanli Road, Chaoyang District, Beijing 100021, China
[b] Hebei Provincial Center for Disease Control and Prevention, 97 Huaian East Road, Shijiazhuang, Hebei 050021, China

A survey of concentrations of OCPs in breast milk helps identify background concentrations in the Chinese population.

ARTICLE INFO

Article history:
Received 19 May 2010
Received in revised form
4 October 2010
Accepted 10 October 2010

Keywords:
Organochlorine pesticides
DDTs
HCHs
Human milk
Infant health risk
China

ABSTRACT

The occurrence of persistent organochlorine pesticides (OCPs) in breast milk samples collected from mothers from twelve provinces in mainland China was investigated. Dichlorodiphenyltrichloroethanes (DDTs) were the most prevalent agent, followed by HCHs and HCB, whereas levels of chlordane compounds, drins and mirex were lower. The relatively lower DDE/DDT ratio in the Fujian rural area suggested more recent exposure to DDT than in other areas. The mean level of DDTs in breast milk from the southern China was higher than those from northern China ($p < 0.05$). A positive correlation was observed between concentration of DDTs in human milk and consumption of animal-origin food, suggesting that this parameter could play an important part in influencing OCPs burdens in lactating women. The mean estimated daily intakes of different OCPs for breastfed infants were lower than the tolerable daily intake.

© 2010 Elsevier Ltd. All rights reserved.

1. Introduction

Organochlorine pesticides (OCPs) are widely used for agriculture, forestry, building protection, and insect control. Most OCPs are lipophilic, very persistent, and highly stable. They can accumulate in ecosystems. Many toxic effects on the reproduction, development and immunological function of animals from the use of OCPs have been reported (Cooper et al., 2004). Several OCPs have weakly estrogenic or anti-estrogenic effects (Dalvie et al., 2004) and also harm the nervous system (Langer et al., 2003). OCPs such as aldrin, chlordane (CHL), dichlorodiphenyltrichloroethane (DDT), dieldrin, endrin, heptachlor, hexachlorobenzene (HCB), mirex and toxaphene were listed in the initial "dirty dozen" agents in the Stockholm Convention in 2001 (WHO, 2007). In 2009, hexachlorocyclohexane (HCH) isomers such as α-HCH, β-HCH and γ-HCH were added to the list of persistent organic pollutants (POPs) of the Stockholm Convention for their potential adverse effects on humans and ecosystems (UNEP, 2010). Now, most nations and regions have restricted or banned the use of persistent OCPs.

Compared with blood or adipose tissue, breast milk is a unique biological matrix for investigating certain environmental contaminants because it can provide exposure information about the mother and breastfed infant (WHO, 2009). In addition, breast milk offers a convenient sampling specimen for monitoring the residues of OCPs in human tissues through a non-invasive method of collection. The levels of OCPs in breast milk have been used to assess the trend of pollution by OCPs in the environment since the early 1970s and to evaluate the effect of banning OCPs in many countries (Smith, 1999; Zietz et al., 2008). A general declining trend of concentrations of OCPs in breast milk after the limited use or banning of OCPs was reported (Johnson-Restrepo et al., 2007). Similarly, a downward trend in the concentrations of HCHs and DDTs were observed in the breast milk from mothers in some regions of China (Cao et al., 2000; Su et al., 2001; Yu et al., 2005). However, the surveys focused only on DDTs, HCHs and HCB in smaller districts. The background exposure to OCPs across the country and regional differences such as north–south or urban–rural gradients was not investigated.

We completed a national project for biomonitoring the background level of persistent OCPs in the breast milk of Chinese mothers based on the WHO-coordinated Fourth Survey of Breast Milk for Persistent Organic Pollutants in cooperation with the United Nations Environment Program (UNEP). The present survey

* Corresponding author.
E-mail address: wuyncdc@yahoo.com.cn (Y. Wu).

0269-7491/$ – see front matter © 2010 Elsevier Ltd. All rights reserved.
doi:10.1016/j.envpol.2010.10.014

included eight OCPs (DDTs, HCHs, HCB, drins, CHLs and mirex) except toxaphene. All breast milk samples were collected in 2007 according to the fourth WHO survey protocol, in which a Chinese total diet study (TDS) was carried out simultaneously.

2. Materials and methods

2.1. Collection of breast milk and sample preparation

Samples of breast milk were collected from August to November 2007. The location of the twelve provinces of China and domiciles of the donors are shown in Fig. 1. Information regarding donor selection, sample collection, and sample pooling are detailed in our previous study on dioxin-like compounds in breast milk from China (Li et al., 2009). In brief, 1237 individual milk samples were collected from twelve provinces of China: Heilongjiang, Liaoning, Hebei, Henan, Shanxi, Ningxia, Jiangxi, Fujian, Shanghai, Hubei, Sichuan and Guangxi. Before analysis, the individual samples were thawed at ambient temperature and pooled according to their collection regions. For each province, the individual samples from the urban areas and rural areas were pooled separately, resulting in 24 pooled samples of breast milk.

2.2. Reagents and chemicals

The standards were purchased from Cambridge Isotope Laboratories (Andover, MA, USA). They were used for the calibration, quantification and recovery of OCPs: ES-5348 CS1-6 calibration and verification solutions; ES-5399-10x-0.5 unlabeled compound surrogate solution (concentration: 2000 ng/mL); and ES-5349-L $^{13}C_{12}$-labeled compound surrogate solution (concentration: 1000 ng/mL). All pesticide residue analysis-grade solvents used in the extraction and analysis procedures were purchased from Fisher Scientific Company (Pittsburgh, PA, USA).

2.3. Sample analysis

The methodology used for OCPs was based on Soxhlet extraction, gel permeation chromatography (GPC) cleanup, and gas chromatography–negative chemical ionization-mass spectrometry (GC–NCI-MS) detection. Briefly, 20 mL of pooled breast milk were freeze-dried. After spiking with 10 ng of $^{13}C_{12}$-labeled internal standard and equilibrating for 5 h, samples were grounded with anhydrous sodium sulfate and extracted in a Soxhlet apparatus with a mixture of *n*-hexane and dichloromethane (1:1, v/v) for 24 h. After Soxhlet extraction, the bulk lipid was removed by an auto GPC system (J2 Scientific CoF85 express column ethyl acetate/cyclohexane 50:50, US) with a low-pressure column (GPC, Bio-Beads S-X3, 200–400 beads). Samples were cleaned up using a Florisil Solid-Phase Extraction Column (Supelclean™ LC-Florisil® SPE Tube bed weight 1 g, volume 6 mL; Supelco, Bellefonte, PA, USA) (Ministry of Agriculture Minister of China Industry Standard NY/T761-2004). The final eluate was evaporated under nitrogen to dryness and reconstituted to 500 μL using hexane. The 23 analytes were quantified using the isotope dilution method.

The analysis was undertaken by GC-NCI-MS (Shimadzu, Tokyo, Japan) with a VF5-5MS capillary column (30 m × 0.25 mm i.d. × 0.25 μm; Varian, Las Vegas, NV, USA). High-pressure injection mode was selected and the pressure set at 230.0 kPa. Helium was used as the carrier gas at a flow rate of 2.0 mL/min. The column oven temperature was initially maintained at 100 °C for 1 min and increased to 160 °C at a rate of 40 °C/min. It was then increased to 200 °C at a rate of 2 °C/min and held for 5 min. It was then increased to 260 °C at a rate of 8 °C/min and held for 2 min. The injection temperature and ion source temperature were set at 260 °C and 170 °C, respectively. The injection port was operated in splitless mode and the injection volume was 1 μL. The pressure of methane was 250 kPa.

The analytes were α-HCH, β-HCH, γ-HCH (lindane), δ-HCH, *p,p'*-DDT, *o,p'*-DDT, *p,p'*-DDE, *o,p'*-DDE, *p,p'*-DDD, *o,p'*-DDD, HCB, aldrin, dieldrin, endrin, *trans*-chlordane, *cis*-chlordane, *trans*-nonachlor, *cis*-nonachlor, oxychlordane, heptachlor, *trans*-heptachlor epoxide, *cis*-heptachlor epoxide, and mirex.

2.4. Quality control and quality assurance

Blank samples were used every five samples to check for interference or contamination from solvents and glassware. For recovery tests, a matrix (cow milk) spiking test was conducted. Two spiked levels (2.0 and 10.0 ng/g) were used. Recoveries of the 23 analytes ranged from 85% to 130%; the relative standard deviations (RSDs) were <10%. The reported concentrations were not corrected by recovery. Limits of detection (LODs) were defined as three-times that of the noise. The LODs for α-, β-, γ- and δ-HCH in breast milk were 1.2, 1.8, 2.3 and 3.0 ng/g lipid, respectively. The LODs for *pp'*-DDT, *op'*-DDT, *pp'*-DDE, *op'*-DDE, *pp'*-DDD, *op'*-DDD were 3.0, 1.2, 0.5, 0.8, 1.2 and 1.0 ng/g lipid, respectively. The LOD for HCB was 0.05 ng/g lipid. The LODs for other OCPs were 0.5–3.0 ng/g lipid. If a proportion of non-quantified results (value < LOD) exceeded 60%, the upper bound was calculated by setting all non-detectable results equal to the LOD and the lower bound was calculated by setting all non-detectable equal to zero (Vlachonikolis and Marriott, 1995). The upper bound of values was used to describe the level of OCPs in breast milk in the present study unless otherwise indicated.

Laboratory performance was validated by participating in the Food Analysis Performance Assessment Scheme (FAPAS). The proficiency test for OCPs in fish oil and milk powder was organized by the Central Science Laboratory of the UK in 2007 and 2008, respectively. The Z-score for dieldrin, oxychlrodane, endrin, *pp'*-DDE and *pp'*-DDD in fish oil were 1.1, 0.4, 0.3, 0.4 and 0.2, respectively. The Z-score for heptachlor in milk powder was 0.6. The laboratory also participated in the First Worldwide UNEP Intercalibration Study on Persistent Organic Pollutants in October 2009. Good results were obtained for the test solutions, fish samples, and samples of human milk.

2.5. Statistical analyses

Statistical analyses were undertaken using the Statistical Package for Social Science (SPSS) 12.0 version (SPSS, Chicago, IL, USA). The Student's *t*-test was subsequently used to compare the differences among each group. $P < 0.05$ was considered significant. Spearmen's rank correlation coefficients were used to test the correlations between OCPs and dietary consumption.

Fig. 1. Map of China showing locations of residence of breast milk donors.

3. Results and discussion

3.1. Contamination

A wide range of concentrations for 23 OCPs compounds were detected in the 24 pooled samples of breast milk from <LOD to 1660 ng/g lipid (Table 1). Among the OCPs analyzed, DDTs, HCHs and HCB were detectable in every pooled sample. CHLs, drins and mirex could be detected in, respectively, 75.0%, 29.2% and 20.8% of the samples. DDTs were found at the highest mean concentration in 24 pooled samples, followed by (in order) HCHs, HCB, CHLs, drins and mirex.

The contamination level of DDTs, HCHs and HCB in breast milk from 24 pooled samples varied from 153.6 ng/g lipid to 1756.3 ng/g lipid (mean, 527.2 ng/g lipid); 55.8 ng/g lipid to 536.4 ng/g lipid (mean, 231.8 ng/g lipid); and 18.4 ng/g lipid to 56.8 ng/g lipid (mean, 32.8 ng/g lipid), respectively (Table 1). The mean concentrations of DDTs, HCHs and HCB in breast milk from rural and urban

Download English Version:

https://daneshyari.com/en/article/6319326

Download Persian Version:

https://daneshyari.com/article/6319326

Daneshyari.com