Chemosphere 93 (2013) 1578–1585

Contents lists available at ScienceDirect

Chemosphere

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

Contamination of polychlorinated biphenyls and organochlorine pesticides in breast milk in Korea: Time-course variation, influencing factors, and exposure assessment

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HIGHLIGHTS

- The levels of PCBs and OCPs in breast milk significantly increased within the first month of lactation.
- Seafood and noodle consumption was associated with the concentrations of PCBs and OCPs in breast milk.
- OC exposure to breast-fed infants was below the thresholds proposed by US EPA and Health Canada.
- Chlordane has a potential health risk to Korean infants via breast milk.
- This is the first comprehensive study to assess infant exposure to OCs via breast milk in Korea.

ARTICLE INFO

Article history:
Received 27 February 2013
Received in revised form 30 July 2013
Accepted 4 August 2013
Available online 7 October 2013

Keywords:
Lactation
Estimated daily intake
Health risk
Threshold value
Chlordane

ABSTRACT

Breast milk is a noninvasive specimen to assess maternal and infant exposure to polychlorinated biphenyls (PCBs) and organochlorine pesticides (OCPs). In this study, 206 breast milk samples were collected from 87 participants during lactation, at <7, 15, 30, or 90 days postpartum in four cities in Korea. The total concentrations of PCBs (ΣPCB) and OCPs (ΣOCP) ranged from <LOQ to 84.0 (median: 12.1) ng g^{-1} lipid weight and from <LOQ to 559 (median: 144) ng g^{-1} lipid weight, respectively. The residue levels of these contaminants measured in our study were relatively lower than those reported for European, African and Asian populations. Within a month postpartum typically after day seven the levels of ΣPCB and ΣOCP significantly increased. Some OCP compounds were correlated with maternal age, BMI, parity, and delivery mode. Certain types of dietary habits such as seafood and noodle consumption were significantly associated with ΣPCB and ΣOCP. The estimated daily intakes (EDIs) of ΣPCB and ΣOCP were 45.2–127 ng kg^{-1} bw day^{-1} and 625–1259 ng kg^{-1} bw day^{-1} during lactation, respectively, which are lower than the threshold values proposed by the US EPA and Health Canada. The exposure of Korean infants to chlordanes via breast milk had a potential health risk which deserves further investigation.

1. Introduction

Polychlorinated biphenyls (PCBs) and organochlorine pesticides (OCPs) are persistent organic pollutants (POPs), which have been regulated worldwide since 2001 by the Stockholm Convention. Because of their persistence and lipophilicity, high levels of organochlorines (OCs) including PCBs and OCPs have been widely detected in wildlife (Nakata et al., 2005; Kannan et al., 2008) and in human such as serum, breast milk and adipose tissue (Bergonzi et al., 2009; Tsang et al., 2011; Moon et al., 2012). A number of studies have suggested that exposure to PCBs and OCPs can cause adverse health effects such as nervous system damage,

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http://dx.doi.org/10.1016/j.chemosphere.2013.08.011

reproductive disorders, immunological effects and cancer (Schantz et al., 2001; Martin et al., 2002; McGlynn et al., 2006; Noakes et al., 2006). Levels of OCs in human specimens have decreased considerably during the past three decades as a consequence of a ban on their production and use (Norén and Meironyté, 2000; Johnson-Restrepo et al., 2007). In Korea, approximately 4300 tons of PCBs had been used until the 1990s (Breivik et al., 2002), and approximately 3600 tons of OCPs had been used for agricultural purposes until the 1980s (Kim and Smith, 2001). Despite the ban on these compounds in Korea, exposure to PCBs and OCPs via seafood consumption is still significant, and such exposure has been associated with cancer risk in Korean populations (Moon et al., 2009).

Dietary intake is a major exposure route for OCs in general populations (Polder et al., 2010; Fromberg et al., 2011; Su et al., 2012). Unlike adults, infants have additional exposure pathways to OCs such as placental transfer and breast-feeding (Park et al., 2008; Johnson-Restrepo and Kannan, 2009). Several studies have reported the partitioning and maternal transfer for POPs in blood, milk, placenta and adipose tissue (Schecter et al., 1998, 2007). Moreover, infants are sensitive human population who is more vulnerable to the exposure to toxic contaminants. Continuous monitoring of OCs for infants is therefore important in order to manage the risks of these contaminants.

Breast milk is an important OC exposure medium (e.g. DDTs) for infants, due to its high fat content as well as initial food for nursing infants (Lakind, 2004; Bordajandi et al., 2008; Shen et al., 2011). Measuring OC levels in breast milk is necessary not only to estimate the maternal body burden but also to determine the potential health risks from exposure to these contaminants. To date, a few studies have measured PCB and OCP levels in breast milk from Korea, but with a limited sample size ($n < 30$) from small sampling regions (Yang et al., 2002; Haraguchi et al., 2009). Thus, a comprehensive study with a large sample size collected from geographically representative locations is warranted to identify the current contamination status of PCBs and OCPs in breast milk, and the associated risks for infants.

In this study, we aimed to determine the current levels and compound-specific profiles of PCBs and OCPs in breast milk across Korea and time-course variation of these contaminants during lactation. We also aimed to investigate associations between individual variability in OC levels in breast milk and demographic parameters such as maternal age, parity, body mass index (BMI) and dietary habits. To assess the potential health risks of OCs via breast-feeding, the daily intakes of these contaminants were calculated and compared to the threshold values suggested by the United States Environmental Protection Agency (US EPA) and Health Canada. The results of this study will be particularly helpful for establishing environmental health policy for OCs among sensitive populations.

2. Materials and methods

2.1. Sample collection

Pregnant women were recruited from five Korean university hospitals located in Seoul, Pyungchon, Ansan and Jeju from February to December in 2011. These cities are representative regions for residential (Seoul and Pyungchon), industrial (Ansan), and rural (Jeju) in Korea. For representative sample collection, we excluded breast milk samples associated with occupational exposure, gestational diabetes, thyroid disease, surgical disease, and congenital deformity. A total of 206 breast milk samples were collected from 87 lactating women at <7, 15, 30, or 90 days postpartum. Shortly before delivery, participants completed a detailed questionnaire about current and previous pregnancy histories, medical history, and demographic parameters. Demographic information, including age, BMI, parity, gestational age at delivery, and delivery mode, are summarized in Table 1. Breast milk samples were collected in polypropylene tubes, and were frozen and transported on ice to the laboratory. Samples were stored in the laboratory at −70 °C until analysis. The present study was approved by the Institutional Review Board of the School of Public Health, Seoul National University, as well as all of the participating university hospitals. Informed consent was obtained from all participants.

The amount of breast milk consumption was measured on every participating infants by weighing before and after a feeding at days of breast milk collection (i.e., <7, 15, 30 or 90 days). The estimated average breast milk consumption amount used in our study is summarized in Table S1. The details of measurement methods for daily milk breast milk intake for the participating infants are presented in the Supporting information.

2.2. Chemical analysis

The analytical procedures for measuring PCB and OCP levels in breast milk were optimized with some modifications from previous studies (Dmitrovic et al., 2002; Kang et al., 2008). In brief, breast milk samples (2 mL) were fortified with formic acid (90%; Merck, Darmstadt, Germany) and Milli-Q water for protein denaturation, after ^{13}C-labeled internal standards for PCBs (CBs 28, 52, 101, 138, 153, 180 and 209; EC-9605-SS; Wellington Laboratories, Guelph, ON, Canada) and OCPs (*o,p′*-DDE, *p,p′*-DDE, *o,p′*-DDD, *p,p′*-DDD, *o,p′*-DDT, *p,p′*-DDT, α-HCH, β-HCH, γ-HCH, δ-HCH, *oxy*-CHL, *trans*-CHL, *trans*-nonaCHL, *cis*-nonaCHL, HCB, heptachlor, heptachlor epoxide and mirex; ES-5349L; Cambridge Isotope Laboratories, Andover, MA, USA) were spiked. The samples were extracted by solid phase extraction (SPE) using a Sep-Pak Vac C_{18} cartridge (500 mg/6 cc; Waters, Milford, MA, USA), which was pre-washed with methanol (ultra-trace residue analysis; J.T. Baker, Phillipsburg, NJ, USA) and conditioned with Milli-Q water. The extracted cartridge was rinsed with Milli-Q water and then dried. A Sep-Pak Plus NH_2 cartridge (360 mg; Waters), pre-washed with 6 mL of hexane (ultra-trace residue analysis; J.T. Baker) and was connected to the lower end of the C_{18} cartridge. Eight milliliters of hexane were passed through the combined NH_2–C_{18} cartridge and were collected. After removing the C_{18} cartridge, 6 mL of 5% DCM (ultra-trace residue analysis; J.T. Baker) in hexane was passed through the NH_2 cartridge and combined with a previous fraction. The pooled eluants (5 mL) were cleaned up on a silica gel/florisil SPE cartridge (waters), using 12 mL of 50% DCM in hexane. The purified eluants were concentrated and dissolved in 100 μL of nonane (pesticide analysis grade; Sigma–Aldrich, St. Louis, MO, USA) for instrumental analysis.

A high-resolution gas chromatography interfaced with a high-resolution mass spectrometer (HRGC/HRMS; JMS 800D, JEOL, Tokyo, Japan) was used for the identification and quantification of target compounds. Nineteen PCB congeners (CBs 18, 28, 33, 44, 52, 70, 101, 105, 118, 128, 138, 153, 170, 180, 187, 194, 195, 199 and 206: EC9605-CVS, Wellington) and dichlorodiphenyltrichloroethanes (DDTs), chlordanes (CHLs), hexachlorocyclohexanes (HCHs), heptachlors, hexachlorobenzene (HCB), and mirex were analyzed in breast milk samples. Details of instrumental parameters have been reported elsewhere (Moon et al., 2010, 2012). In brief, PCBs and OCPs were quantified using the isotope dilution method based on relative response factors of individual compounds. The HRMS was operated under positive electron impact mode, and ions were monitored by selected ion monitoring using the molecular ions of PCBs and OCPs. A DB5-MS capillary column (30 m length, 0.25 mm inner diameter, 0.25 μm film thickness; J&W Scientific, Palo Alto, CA, USA) was used for the separation of PCB and OCP compounds.

The recoveries spiked ^{13}C-labeled CBs 28, 52, 101, 138, 153, 180 and 209 were 64 ± 21% (average ± standard deviation), 66 ± 10%,