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Perfluoroalkyl substances (PFASs) in breast milk from Korea: Time-course trends, influencing factors, and infant exposure

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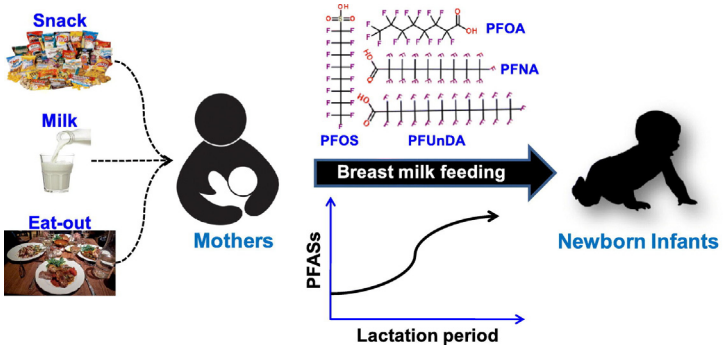
HIGHLIGHTS

- PFOS, PFOA, PFUnDA, and PFNA were the predominant compounds in breast milk.
- Concentrations of PFASs were significantly correlated with maternal age, BMI, and parity.
- Increased levels of PFASs were found in breast milk after the first month of nursing.
- Snack consumption and frequency of eating-out were significantly associated with increased PFAS levels.
- The infant exposure levels of PFOS and PFOA via breast milk were lower than the TDI.

GRAPHICAL ABSTRACT



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ABSTRACT

Breastfeeding is an important exposure pathway to perfluoroalkyl substances (PFASs) for newborn infants. Nevertheless, reports are limited on the occurrence and time-course of PFASs in breast milk, and most studies have focused on the analysis of perfluorooctane sulfonate (PFOS) and perfluorooctanoic acid (PFOA). In this study, 16 PFASs were analyzed in breast milk samples ($n = 293$) collected from 128 mothers in Korea during various lactation periods to assess maternal exposure levels, contamination profiles, time-course variations, and infant health risks. The total concentrations of PFASs (ΣPFAS) ranged from 31.7 to 1004 (median: 188) ng/L, which was within the ranges recently reported for Asian and European populations. After a month of nursing, the concentrations of PFOS, PFOA, perfluorononanoic acid (PFNA), and ΣPFAS significantly increased. This could be due to changes in the dietary and behavior patterns of the mothers after the first month of lactation. The concentrations of PFOS and PFOA were significantly correlated with maternal age, body mass index, and parity. Certain types of diet (e.g. consuming snacks and milk) and eating-out frequency were significantly associated with increasing levels of PFAS. Significant correlations and similar time-course trends were found between PFASs and PCBs/DDTs, implying similar exposure sources and biokinetics for these contaminants. The estimated daily

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intakes of PFOS and PFOA via the consumption of breast milk were below the tolerable daily intakes for infants suggested by the European Food Safety Authority (EFSA).

1. Introduction

Perfluoroalkyl substances (PFASs) have been widely used in a variety of consumer products and industrial applications over several decades, due to their ability to repel both water and oil (Giesy and Kannan, 2001; Paul et al., 2009). Associated with the widespread use of these contaminants, PFASs have been detected in various environmental sectors such as air, water, sediment, and wildlife (Shoeib et al., 2004; Ahrens et al., 2009, 2010; Moon et al., 2010). Toxicological and epidemiological studies have reported adverse health outcomes of PFASs such as developmental toxicity, hepatoxicity, renal toxicity, immunotoxicity, and endocrine disrupting effects (Fei et al., 2008; Hu and Hu, 2009; Dong et al., 2013; Vieira et al., 2013). Based on the evidences of persistence and health effects for PFASs, human biomonitoring has been conducted on several biological matrices such as serum, breast milk, and adipose tissue (Maestri et al., 2006; Mondal et al., 2014; Kang et al., 2016). Although the PFAS family comprises several perfluorinated carbons (Wang et al., 2017), most studies have focused on detection of perfluorooctane sulfonate (PFOS) and perfluorooctanoic acid (PFOA) in humans.

A variety of foodstuffs, such as seafood and dairy products, are the major exposure pathways of PFASs to the general population (Haug et al., 2010; Domingo and Nadal, 2017). However, newborn infants are primarily exposed to PFASs via breastfeeding due to exposure of the mothers during the pregnancy, as well as throughout their lives (Thomsen et al., 2010; Kang et al., 2016). In addition, the developing fetus has been exposed to PFSAs via placental transfer from their mothers (Kim et al., 2011). Previous studies have reported that decreasing levels of PFOS and PFOA in maternal serum correlate with increasing serum levels in newborn infants during the lactation period (Mondal et al., 2014; Mogensen et al., 2015). Thus, breastfeeding is the major elimination and exposure pathway of PFASs for mothers and infants, respectively (Thomsen et al., 2010; Kang et al., 2016). The World Health Organization (WHO) maintains that breastfeeding is nutritionally essential for newborn infants for the first six months (WHO, 2012). However, limited time-dependent information on PFASs in breast milk is available to identify a time-point for minimizing PFAS exposure to newborn infants. Two studies have reported time-course trends of PFASs in breast milk (Thomsen et al., 2010; Kang et al., 2016), but these studies either had limited sample size ($n = 10$) or analyzed samples from different populations for the lactation periods investigated (Thomsen et al., 2010; Kang et al., 2016). Therefore, it is essential to collect data from a sufficiently sized population over time to better understand the time-course variations in PFASs during the lactation period.

In this study, we investigated the residue levels, compound-specific profiles, and time-course trends of PFASs in breast milk collected from Korean mothers. We also investigated the relationships between PFAS concentrations and demographic parameters, such as age, body mass index (BMI), and dietary habits, to identify the contributing factors affecting PFAS contamination in breast milk. The daily intakes of PFASs via the consumption of breast milk were estimated and compared with the threshold value proposed by the European Food Safety Authority (EFSA).

2. Materials and methods

2.1. Sample collection

Breast milk samples ($n = 293$) were collected from 127 mothers in the Children's Health and Environmental Chemicals in Korea (CHECK) Cohort (Kim et al., 2013, 2015a, 2015b; Lee et al., 2013a, 2013b, 2015; Choi et al., 2014; Jeong et al., 2014a, 2014b, 2016; Shin et al., 2016). The CHECK Cohort comprises pairs of pregnant women and fetuses that were recruited from four South Korean cities (Seoul, Pyungchon, Ansan, and Jeju) beginning in 2011. Certain populations related to gestational diabetes, surgical disease, occupational exposure, thyroid disease, and congenital deformity were excluded from the study. For the breast milk collection, we considered a postpartum care period of Korean mothers after delivery (Kim and Chung, 2012). During a month for the postpartum care, the Korean mothers normally stay in the hospital or at home with restrictions on diet and behavior. Based on the postpartum care period, the breast milk samples were collected at four different lactation periods (i.e., <7, 15, 30, and 90 days after delivery). The participants completed questionnaires regarding their pregnancy and medical histories, and demographic parameters such as maternal age, BMI, gestation age, newborn sex, and delivery mode (Table 1). The breast milk samples were collected in polypropylene (PP) tubes, frozen in a refrigerator, then transported to the laboratory on ice. The samples were stored in the laboratory at −70 °C until further processing. This study was approved by the Institutional Review Board (IRB) of Seoul National University, as well as the other participating university hospitals.

The breast milk consumption rate and body weight of the newborn infants were measured during each lactation period (i.e., <7, 15, 30, or 90 days). The detailed methods for determining the breast milk consumption rate and body weight for the participating infants are described in a previous study (Lee et al., 2013a). In our study, the estimated consumption rate of breast milk and body weight for <7, 15, 30, and 90 days after delivery were 445 mL/day and 3.36 kg, 521 mL/day and 3.73 kg, 578 mL/day and 4.44 kg, and 514 mL/day and 6.84 kg, respectively.

2.2. Standards and reagents

Sixteen native PFASs, including the 12 perfluoroalkyl carboxylic acids (PFCAs) such as perfluoropentanoic acid (PFPeA), perfluorohexanoic acid (PFHxA), perfluoroheptanoic acid (PFHpA), PFOA, perfluorononanoic acid (PFNA), perfluorodecanoic acid (PFDA), perfluoroundecanoic acid (PFUnDA), perfluorododecanoic acid (PFDoDA), perfluorotridecanoic acid (PFTrDA), perfluorotetradecanoic acid (PFTeDA), perfluorohexadecanoic acid (PFHxDA), and perfluorooctadecanoic acid (PFOcDA), and the four perfluoroalkane sulfonates (PFSAs) such as perfluorobutane sulfonate (PFBS), perfluorohexane sulfonate (PFHS), PFOS, and perfluorodecane sulfonate (PFDS) were measured in breast milk samples. Eight mass-labeled PFASs ($^{13}C_2$-PFHxA, $^{13}C_4$-PFOA, $^{13}C_5$-PFNA, $^{13}C_2$-PFDA, $^{13}C_2$-PFUnDA, $^{13}C_2$-PFDoDA, $^{18}O_2$-PFHS, and $^{13}C_4$-PFOS; MPFAC-MXA; Wellington Laboratories, Guelph, ON, Canada) were used as internal standards. HPLC-grade methanol (MeOH) and methyl tert-butyl ether (MTBE) were obtained from J.T. Baker (Center Valley, PA, USA). Tetrabutylammonium hydrogen sulfate (TBA; 97%), sodium hydroxide (NaOH; ≥97%), and hydrochloric acid (HCl; molecular-biology-grade) were obtained from Sigma-Aldrich (St. Louis, MO, USA). Sodium carbonate (Na_2CO_3) was obtained from Kanto (Tokyo, Japan).

2.3. Sample preparation

Breast milk samples were analyzed using the ion-pairing method described elsewhere (Taniyasu et al., 2005; Kim et al., 2011) with minor modifications. Before extraction, 5 ng of mass-labeled internal standards of PFASs were spiked into the milk samples. Approximately 2 mL of the milk sample was mixed with 2 mL of Milli-Q water, 1 mL