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Determinants of plasma concentrations of perfluoroalkyl and polyfluoroalkyl substances in pregnant women from a birth cohort in Shanghai, China

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ABSTRACT

Perfluoroalkyl and polyfluoroalkyl substances (PFASs) are widely used in commercial applications and have been commonly detected in pregnant women in Europe and North America. However, data on PFAS concentrations in pregnant women in China are limited. Additionally, the determinants of maternal PFAS concentrations with respect to diet habits have been less extensively described, especially in Asian countries. In the present study, we aimed to measure PFAS concentrations in pregnant women and evaluate sociodemographic, lifestyle, and dietary factors as potential determinants of PFAS concentrations. We analyzed eleven PFASs in maternal blood samples (N = 981) collected at 12–16 weeks of gestation between April and December 2012 at Maternal and Child Health Hospital of Minhang District in Shanghai, China. Multivariate linear regression models were used to examine the associations of PFAS concentrations with maternal sociodemographic, lifestyle, and dietary factors. Eight PFASs, including perfluorohexane sulfonate (PFHxS), perfluorooctane sulfonate (PFOS), perfluorooctanoic acid (PFOA), perfluorononanoic acid (PFNA), perfluorodecanoic acid (PFDA), perfluoroundecanoic acid (PFUdA), perfluorododecanoic acid (PFDoA), and perfluorotridecanoic acid (PFTrDA), were detected in > 85% of the samples. PFOA and PFOS were the predominant PFASs with high median concentrations (19.97 ng/mL and 10.81 ng/mL, respectively). Pregnant women who were older, multiparous, well educated, passive smokers, with lower per capita household incomes, and had lived in rooms decorated within the past two years had higher PFAS concentrations, after mutual adjustment for maternal sociodemographic characteristics and lifestyles. With regard to dietary factors, intake of red meat, poultry, animal offal, fish, pastries and fried food, and drinking tap water during pregnancy contributed to higher concentrations of most PFASs, after adjustment for sociodemographic characteristics and lifestyles. Furthermore, higher intake of wheat, coarse cereals, tubers, and soy products was associated with lower maternal PFAS concentrations. Our findings indicate that PFASs were ubiquitous among pregnant women in Shanghai. We provide new evidence for the association between dietary factors and maternal PFAS exposure in China.

1. Introduction

Perfluoroalkyl and polyfluoroalkyl substances (PFASs), a class of synthetic chemicals consisting of a fully fluorinated carbon chain, have been widely used since the 1950s in commercial applications, such as surfactants, lubricants, paints, textiles, fire retardants, non-stick cookware, and food packaging (Lindstrom et al., 2011). With strong carbon-fluorine covalent bonds, PFASs are highly resistant to thermal desorption, chemical degradation, and biodegradation, and therefore persist in the environment, accumulating in animal and human bodies (Lau et al., 2007). Generally, the half-lives of PFASs in human are approximately 2–9 years (Bartell et al., 2010; Lau et al., 2007). In general, longer-chain PFASs are more bioaccumulative than shorter-chain ones (Conder et al., 2008).

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Due to the persistency and bioaccumulation of PFASs, concerns about their adverse effects including hepatotoxicity, immunotoxicity, and developmental toxicity, have been raised for decades (Lau et al., 2007). PFASs are readily transferred from mother to fetus via the placenta (Chen et al., 2017; Zhang et al., 2013), and in utero exposure to PFASs has been linked with adverse birth outcomes (Darrow et al., 2013; Fei et al., 2007), pediatric neurodevelopment problems (Goudarzi et al., 2016; Y. Wang et al., 2015), and poor semen quality in offspring (Vested et al., 2013). PFASs, especially perfluorooctane sulfonate (PFOS, C8) and perfluorooctanoic acid (PFOA, C8), have been commonly detected in pregnant women in Europe (Bjerregaard-Olesen et al., 2016; Cariou et al., 2015; Manzano-Salgado et al., 2016), North America (Kato et al., 2014; Sagiv et al., 2015), Asia (Okada et al., 2013; Y. Wang et al., 2015), Australia (Callan et al., 2016) and Africa (Hanssen et al., 2010). However, information on concentrations of PFASs in pregnant women is limited in China (Liu et al., 2011), especially in the most intensively industrialized regions, such as the eastern coast, with 1.91 μg/day per capita discharge load of PFOS equivalents (Xie et al., 2013).

Several studies have reported that maternal sociodemographic characteristics, including maternal age, educational level, household income, and pre-pregnancy body mass index (BMI), might be associated with PFAS concentrations in pregnant women, but the results have been inconsistent across different countries or regions (Bjerregaard-Olesen et al., 2016; Sagiv et al., 2015). Moreover, most previous studies evaluated the determinants of maternal PFAS concentrations using samples before PFOS was categorized as persistent organic pollutants (POPs) under Annex B of the Stockholm convention in 2009 (Stockholm Convention, 2009). Since the 3M Company, one of the largest PFAS manufacturers, announced the phasing out PFOS production by 2002 (Paul et al., 2009), global production volumes of PFOS have decreased dramatically, whereas other long-chain PFASs and new alternative fluorinated products have been manufactured in increasing volumes in several countries, including China (Land et al., 2015; Wang et al., 2013). The alteration in production patterns has led to corresponding changes in the distributions of long-chain PFAS concentrations in pregnant women (Glynn et al., 2012; Okada et al., 2013). For example, maternal concentrations of perfluorononanoic acid (PFNA, C9) and perfluorodecanoic acid (PFDA, C10) have increased by 4.7% and 2.4% per year, respectively, between 2003 and 2011 in Japan (Okada et al., 2013). The determinants of these long-chain (> 8 carbon atoms) PFAS concentrations in pregnant women are much less described (Bjerregaard-Olesen et al., 2016; Manzano-Salgado et al., 2016). Diet has been considered as a major route of non-occupational exposure to PFASs (Haug et al., 2010). Several studies from Europe found that the intake of fish, red meat, animal fats, snacks, or cereals was associated with higher PFAS concentrations in pregnant women (Cariou et al., 2015; Halldorsson et al., 2008; Manzano-Salgado et al., 2016). However, the association between PFAS concentrations in pregnant women and dietary factors has not been well described in Asian countries.

In the present study, we aimed to measure the plasma concentrations of PFASs in pregnant women from a birth cohort established in Shanghai, China. We also evaluated sociodemographic characteristics, lifestyles, and dietary factors as potential determinants of maternal PFAS concentrations.

2. Material and methods

2.1. Study population

The present study was based on the Shanghai-Minhang Birth Cohort Study (S-MBCS), which was designed to determine the distribution of maternal environmental exposures and examine their effects on pregnant women and their children. The S-MBCS recruited pregnant women at 12–16 weeks of gestation during routine antenatal examinations between April and December 2012 at the Maternal and Child Health Hospital of Minhang district in Shanghai, China. The recruitment criteria for the pregnant women were as follows: (1) being native Chinese and residents of Shanghai; (2) having no history of major chronic diseases of the liver, kidney or other organs diagnosed by a physician; (3) intending to deliver their babies in the study hospital; (4) being willing to take part in a specified interview during pregnancy and after delivery. Pregnant women were recruited by two trained Chinese-speaking interviewers using a structured questionnaire. At enrollment, trained interviewers explained to the pregnant women the purposes, procedures, potential benefits and risks, as well as the confidentiality issues of our study. If the pregnant women were willing to participate in the study, they were asked to fill in the structured questionnaire.

A total of 1292 eligible pregnant women agreed to participate in our study and completed the questionnaire. From these, 981 (75.93%) women provided sufficient blood samples at enrollment for PFAS measurements and were then included in the present study for analyses.

Our study was approved by the ethical committees of the Shanghai Institute of Planned Parenthood Research (SIPPR). All participants gave written informed consent before participating in the study.

2.2. PFAS analyses

One single fasting blood sample from the pregnant women was collected at enrollment, typically between 7 AM and 9 AM of a given day. The blood samples were centrifuged at 4000 rpm for 10 min immediately after collection. The plasma samples were then separated and stored at −80 °C until shipment using dry ice to the Center for Disease Control and Prevention in Hubei Province for PFAS assay. The laboratory technicians had no knowledge of the characteristics and dietary habits of the study population.

We measured eleven PFASs, including perfluorohexane sulfonate (PFHxS, C6), PFOS, PFOA, PFNA, PFDA, perfluoroundecanoic acid (PFUdA, C11), perfluorododecanoic acid (PFDoA, C12), perfluorotridecanoic acid (PFTrDA, C13), perfluorotetradecanoic acid (PFTeDA, C14), perfluorodecane sulfonate (PFDS, C10), and perfluorohexadecanoic acid (PFHxDA, C16). All PFAS standard solutions (PFACMXB0913) and internal standard solutions (MPFACMXA0214) containing perfluoro-1-hexane ($^{18}O_2$) sulfonate ($^{18}O_2$-PFHxS), perfluoro-1-(1,2,3,4-$^{13}C_4$) octanesulfonate ($^{13}C_4$-PFOS), perfluoro-n-(1,2,3,4-$^{13}C_4$) octanoic acid ($^{13}C_4$-PFOA), perfluoro-n-(1,2,3,4,5-$^{13}C_5$) nonanoic acid ($^{13}C_5$-PFNA), perfluoro-n-(1,2-$^{13}C_2$) decanoic acid ($^{13}C_2$-PFDA), perfluoro-n-(1,2-$^{13}C_2$) undecanoic acid ($^{13}C_2$-PFUdA), and perfluoro-n-(1,2-$^{13}C_2$) dodecanoic acid ($^{13}C_2$-PFDoA) were purchased from Wellington Laboratories (Guelph, Canada).

The plasma samples were extracted using the ion-pair extraction method, as described elsewhere (Liu et al., 2011). Briefly, maternal plasma sample (0.8 mL) and internal standard solution (5 μL) were mixed in a 15 mL polypropylene centrifuge tube. Then, 1 mL of 0.5 mol/L tetra-n-butylammonium hydrogen sulfate (TBA) solution and 2 mL of 0.25 mol/L sodium carbonate buffer were spiked into the mixed solutions. Methyl-tertbutyl ether (MTBE, 5 mL) was added to the solution for extraction after mixture was sonicated. The organic and aqueous layers were separated by centrifugation. The aqueous layer was rinsed twice with MTBE, followed by transfer of the supernatants to a new polypropylene tube and evaporation under nitrogen gas flow, and subsequent reconstituted in 1 mL of methanol/water (1:1). The supernatant was filtered through a 0.2 μm nylon filter into a 1.5 mL autosampler vial before analysis.

PFASs were separated and quantified using high performance liquid chromatography coupled with tandem mass spectrometry (HPLC-MS-MS, Agilent Technologies Inc., USA). A 10-μL aliquot of the sample extract was injected into an Extend-C18 column (2.1 mm × 100 mm × 1.8 μm) maintained at 45 °C. A gradient of 2 mmol/L aqueous ammonium acetate solution (A) and methanol (B) were used as mobile phases at a flow rate of 0.3 mL/min. The gradient elution started at 20% A and 80% B, increasing A to 95% and