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Serum concentrations of per- and poly-fluoroalkyl substances and factors associated with exposure in the general adult population in South Korea

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ABSTRACT

Per- and polyfluoroalkyl substances (PFASs) are ubiquitous contaminants found worldwide, including in South Korea. As a result, they are frequently detected in Koreans. However, there is limited representative data and information on potential sources in Korea. Therefore, we measured the serum concentrations of ten PFASs in nationally representative samples of the Korean population (n = 1874, 18–69 years) and evaluated the factors associated with their exposure. Serum PFOS, PFDA, PFOA, and PFNA were detected in nearly all participants (83.1–99.9%). However, serum PFPA, PFHxA, and PFHpA were almost undetected (<0.5% of participants). PFOS had the highest population-weighted geometric mean of 10.23 ng/mL (95% CI: 9.99–10.47), which was followed by PFOA with 2.85 ng/mL (95% CI: 2.73–2.97) and PFDA with 2.17 ng/mL (95% CI: 2.12–2.23). PFNA, PFDA, PFHxS, PFOA, and PFOS concentrations were higher in males (p < 0.001) and older adults (p < 0.001). PFNA was higher in those who used wax, polish, and water-resistant materials (adjusted proportional change = 1.14; 95% CI: 1.08–1.22), and those who ate cooked fish (1.16; 95% CI: 1.03–1.31) compared to those who ate nearly no fish. PFDA was higher in those who used herbicides and pesticides (1.05; 95% CI: 1.02–1.09), those who drank beverages in a plastic bag on a daily basis (1.10; 95% CI: 1.03–1.19), and those who ate raw fish (1.15; 95% CI: 1.03–1.29) or cooked fish (1.13; 95% CI: 1.05–1.23) compared to those who ate nearly no fish. PFHxS was higher in those who used traditional Korean health supplement foods (1.08; 95% CI: 1.01–1.15). PFOA was higher in those who used plastic wrap in a microwave daily or weekly (1.08; 95% CI: 1.00–1.16), and those who used disposable paper cups (1.07; 95% CI: 1.01–1.13). PFOS was lower in underweight participants (0.84; 95%CI: 0.75–0.93) compared to those who were obese, and higher in those who exercised regularly (1.08; 95% CI: 1.03–1.14) or irregularly (1.06, 95% CI: 1.01–1.12) compared to those who did not exercise. Subjects who used severely damaged Teflon appliances had lower concentrations of PFOA (0.78, 95% CI: 0.65–0.95), while regular use of Gore-Tex goods was related to higher PFNA (1.15, 95% CI: 1.03–1.28) and PFDA (1.11; 95% CI: 1.02–1.20) levels. These findings suggest that most Koreans are frequently exposed to PFASs, and that serum concentrations of PFASs vary with age, sex, and exposure factors.

1. Introduction

Per- and poly-fluoroalkyl substances (PFASs) are synthetic chemicals that have been used widely since the late 1940s in industrial and commercial applications to provide water, oil, and stain repellency to textiles, carpets, and leather, create grease- and water-proof coatings on paper plates and food packaging, and as processing aids in fluoropolymer manufacturing, chrome plating, firefighting foams, and in many other applications (Trier et al., 2011; Wang et al., 2013; SGP meeting, 2015).

Studies have focused on PFASs as emerging environmental contaminants, and a large body of research is available on their environmental contamination and human exposure. PFASs are highly persistent and recalcitrant to typical environmental degra-

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dation processes due to their carbon-fluorine bonds (Blake et al., 1997; Falandysz et al., 2006). Several PFASs, including perfluorooctanesulfonic acid (PFOS) and perfluorooctanoic acid (PFOA), have been detected frequently in wildlife and humans worldwide, with greater exposure in areas near urbanized and industrialized regions (Houde et al., 2006), as well as in non-occupationally exposed humans (Lau et al., 2007; Gloria et al., 2012).

Concerns over the potential environmental and toxicological effects of PFASs led to the phasing out of PFOS and PFOA production by their major manufacturers in 2002 based on a voluntary stewardship agreement between the United States Environmental Protection Agency (US EPA) and global leading companies to work toward their elimination by 2015 (US EPA, 2016a). Additionally, PFOS has been included in the Stockholm Convention on Persistent Organic Pollutants as an Annex substance (UNEP, 2013). However, environmental contamination and human exposure to PFASs are anticipated to continue for the foreseeable future due to their persistence, formation from precursor compounds, and potential for continued production by other manufacturers worldwide, including in South Korea (Lindstrom et al., 2011; Gloria et al., 2012; US EPA, 2016a,b).

Exposure to PFASs, including PFOS and PFOA, is widespread in the general population, which may have potential health consequences, although demographic, geographic, and temporal differences may exist (Hansen et al., 2001; Taniyasu et al., 2003; Kannan et al., 2004; Calafat et al., 2007; Fromme et al., 2007; Jessica et al., 2009; Patricia et al., 2012; Sarah et al., 2014; Kristina et al., 2015; Su et al., 2016). In Korea, for example, total PFAS emissions from wastewater treatment plants are estimated to be 0.78 ton/year (Cho et al., 2010). Additionally, 21.4–328.0 and 3.8–329.2 ng/L of PFOS and PFOA, respectively, were detected in a major river basin in Korea (Son et al., 2013), while average PFOA, PFOS, perfluorohexanesulfonic acid (PFHxS), and perfluorononanoic acid (PFNA) concentrations of 22.64, 12.67, 8.16, and 7.41 ng/L, respectively, were reported in southern coastal waters of Korea (Paik and Kam, 2015).

The first study on PFAS exposure in South Korea reported on PFOS and PFOA levels in inhabitants of Daegu city, and found relatively high levels compared to other regions worldwide (n = 25; Kannan et al., 2004). Since then, Korean PFAS exposure has been investigated in several other cities, including Seoul and Gumi (n = 20 pregnant women >25 years old; Kim et al., 2011a,b), Siheung, an industrial city near Seoul (n = 633, age range 12–78; Ji et al., 2012a), Daegu (n = 140, > 20 years old; Ji et al., 2012b), and Busan (n = 150, ages 20–40; Suh et al., 2012). However, because of limitations in sample size and regional scope, these observations are insufficient for generalization to all of South Korea, although several results have been useful for comparing concentrations and considering covariates that might influence PFAS exposure.

Various exposure pathways of PFASs in humans have been suggested, including diet, drinking water, household dust, and outdoor and indoor air (Fromme et al., 2009; Haug et al., 2011). Among them, diet is considered one of the major exposure pathways for PFASs (Fromme et al., 2009; Kärrman et al., 2009; Haug et al., 2010). However, the contribution of specific food types to PFAS concentrations varies by geographical region, population characteristics, and between specific PFAS compounds (Zhang et al., 2011; Jain, 2014). Additionally, migration from food packaging and non-stick cookware has been identified as a possible source of human exposure to PFASs (Tittlemier et al., 2006). In one Korean city, vegetable, potato, fish/shellfish, and popcorn consumption were reported to be significantly related with blood concentrations of several PFASs (Ji et al., 2012a). However, information on dietary predictors in Korea is limited. In this study, we examined representative exposure levels of ten PFASs in Korean adults. We also determined factors associated with exposure that affected the serum concentrations of PFASs, including individual habits, lifestyle, and diet, as well as cooking utensils.

2. Methods

2.1. Study population

In this study, we performed a population-based, cross-sectional survey from 2009 to 2010 representing adult inhabitants (18–69 years of age) of South Korea. Participants were recruited from census unit-stratified random samples based on the Korea National Census Registry, composed of 264,183 units (Statistics Korea, 2005). The number of census units was determined based on the population of each city and province using square root proportion sampling, and 100 census units were selected randomly. Initially, 2157 people were included as subjects in the census units, of which 1874 individuals completed the interviews without missing data and provided serum samples, equivalent to a usable response rate of 86.9%. The study was supervised by the Korean Food and Drug Administration, and the study protocol was approved by the Asan Medical Center Institutional Review Board and conducted in accordance with the ethical principles for medical research involving human subjects, as defined by the Helsinki Declaration. All study participants provided written informed consent. The data used in this study are publicly available by request from the Korean Ministry of Food and Drug Safety.

2.2. Data and blood sample collection

Subjects were invited to a public health center in their designated census unit for an interview and serum sample collection. First, participants received an explanation from trained staff and the data were collected in an interview with pre-trained community surveyors who provided guidance during the questionnaire, which included several demographic characteristics (e.g., age, sex, and body mass index), lifestyle factors and dietary habits (e.g., smoking, exercise, use of traditional Korean health supplement foods, and consumption of processed milk products, fish, and organic agricultural products), exposure factors (e.g., use of wax/polish/water-resistant materials for cleaning of furniture and leather goods, household pesticides, plastic wrap use in microwaves, disposable paper cups, and Teflon appliances, as well as wearing Gore-Tex fabric, and consumption of beverages in a plastic bag), and socioeconomic status (i.e., income). Then, a pathologist collected blood into a 10-mL red top Vacutainer (Becton-Dickinson, Rutherford, NJ, USA) with no anticoagulant and transported the blood tubes on ice for processing and storage. Blood was allowed to clot for at least 2 h after collection before processing. Then, it was centrifuged at 2000 rpm for 15 min. To obtain the maximum serum yield, a second centrifugation was performed for 10 min, if necessary. Serum was divided into aliquots for analysis. First, 0.5 mL was placed a 2-mL Nalgene cryovial (Sigma-Aldrich, St Louis, MO, USA) for the PFAS measurements. A blank sample was made with artificial cerebrospinal fluid (ACSF, Showa, Tokyo, Japan) confirmed from a source with no PFAS exposure. All sample preparation and processing procedures were conducted under a chemical fume hood. Aliquots were stored at −20 °C until overnight shipment on dry ice to the Kyunghee University laboratory for analysis. In total, 1874 (90.8%) usable serum samples were obtained from the initial 2063 participants.

2.3. Sample analysis

The concentrations of ten PFASs (perfluoropentanoic acid (PFPA), perfluorohexanoic acid (PFHxA), perfluoroheptanoic acid