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Perfluoroalkyl acids in serum of Korean children: Occurrences, related sources, and associated health outcomes

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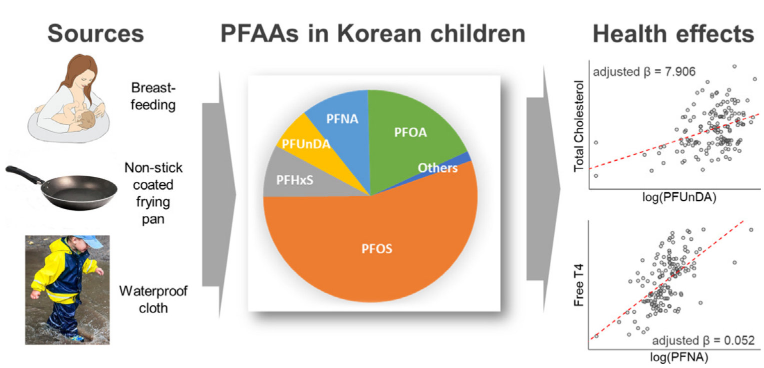
HIGHLIGHTS

- Sixteen perfluoroalkyl acids (PFAAs) were analyzed in Korean children serum.
- Serum PFUnDA concentration was higher than previous reports from other countries.
- Sex difference in exposure level was observed in older child groups.
- PFUnDA was positively associated with total cholesterol and LDL-cholesterol.
- Several predictors of serum PFAAs (e.g. non-stick frying pan use) were identified.

GRAPHICAL ABSTRACT



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ABSTRACT

Perfluoroalkyl acids (PFAAs) have been widely used in human environment, and their exposure among general population has been frequently reported. However, extent of PFAAs exposure and their potential effects among children are not well characterized. In this study, children of between 3 and 18 years of age ($n = 150$) were recruited in Seoul and Gyeonggi, Korea, and the serum levels of 16 PFAAs along with lipids and thyroid hormones were measured. Questionnaire survey was conducted for dietary and behavioral characteristics of the children. Among the measured PFAAs, PFOA, PFNA, PFHxS, and PFOS were detected in all the samples, and PFUnDA and PFDA were detected in over 75% of the samples. PFOS was detected at the highest concentration with a median of 5.68 ng/mL. PFUnDA was detected at higher levels (median of 0.652 ng/mL) compared to those reported for children in USA. Serum PFAA levels were not different by sex among the children of <10 years of age, but in older children, those of boys are significantly higher than girls. Physiological characteristics like menstruation may explain lower PFAAs levels of the girls. In addition, breastmilk consumption, fish/shellfish consumption, non-stick frying pan use, and waterproof cloth use were identified as potential sources of PFAAs exposure. Serum PFUnDA level was positively associated with total cholesterol and low-density lipoprotein level of the children. PFNA was positively associated with free T4 level. High levels of PFUnDA among children and its association with serum lipids warrant replication and confirmation in other populations and/or supports by experimental studies.

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1. Introduction

Perfluoroalkyl acids (PFAAs) and/or their derivatives are a group of compounds which have been widely used in various applications and products, such as surfactants, coating materials, and fabrics (Z. Wang et al., 2013). Because of their persistent, bioaccumulative, and toxic properties, perfluorooctane sulfonic acid (PFOS), its salts, and perfluorooctane sulfonyl fluoride have been regulated under Stockholm Convention on the persistent organic pollutants (POPs) since 2009 (Wang et al., 2009). Moreover, there have been efforts to reduce the use of other PFAAs, such as perfluorootanoic acid (PFOA), and perfluorohexane sulfonate (PFHxS), worldwide (Gomis et al., 2017). Nevertheless, many compounds belonging to this group of chemicals persist in various media such as air, dust, food, consumer products, and drinking water (Jian et al., 2017), consequently enter human body and cause potential harms such as adverse effects on reproduction, development, endocrine, lipid regulation, and immune system (Houde et al., 2006; Kennedy et al., 2010; Ji et al., 2012; Rappazzo et al., 2017).

Several attempts have been made to identify major determinants of PFAAs exposure among humans. Demographic characteristics (Kim et al., 2014; Schecter et al., 2012; Zhang et al., 2010), lifestyle, and dietary habits (Cariou et al., 2015; Cho et al., 2015; Kang et al., 2016; Rylander et al., 2010) have been associated with PFAAs exposure. Use of non-stick frying pan and personal care products, and smoking habit were identified as important determinants of exposure in Korea (Cho et al., 2015; Kang et al., 2016). In addition, various dietary characteristics including fish, potato, milk, and snack consumption have been reported to be associated with PFAAs exposure (Kang et al., 2016; Ji et al., 2012; S. Lee et al., 2018). However, among children, such information is generally limited, except breastfeeding duration (Y.A. Lee et al., 2018).

Adverse effects of PFAAs on lipid regulation have been reported in many experimental studies, not only in a cellular level (Watkins et al., 2015; Wolf et al., 2012) but also in an organism level (Kennedy et al., 2010; Qazi et al., 2010; L. Wang et al., 2013). Imbalance of lipid profile has been associated with cardiovascular diseases (Lobstein et al., 2004), cognitive impairment (Anstey et al., 2008), and diabetic nephropathy (Ravid et al., 1998). A number of epidemiological studies have also reported associations between PFAAs and cholesterol levels in blood, but directions of the associations are often controversial. Positive (Chateau-Degat et al., 2010; Fisher et al., 2013; Nelson et al., 2010), but often negative associations (Starling et al., 2014; Wang et al., 2012) have been reported between PFAAs exposure and cholesterol levels among adult populations. In children populations, similarly, both positive (Frisbee et al., 2010; Geiger et al., 2014) and negative associations (Lin et al., 2009; Zeng et al., 2015) were reported depending on the population.

Thyroid hormone disrupting effects of PFAAs have been demonstrated in experimental studies (Ren et al., 2016; Yu et al., 2009). PFAAs have been also associated with thyroid hormones in human populations, however the directions of such association varied. For example, positive (Ji et al., 2012; Wang et al., 2014) and often negative associations (Kim et al., 2014) were observed in epidemiological studies (reviewed by Lee and Choi (2017)). Among children, positive associations between PFOS or PFNA and total thyroxine (T4) were reported (Lopez-Espinosa et al., 2012), but a report suggesting otherwise is also available (Rappazzo et al., 2017). Thyroid disruption among children is important, as this hormone is crucial for proper growth and development of children (Rappazzo et al., 2017).

In this study, we aimed 1) to characterize current exposure profile of 16 PFAAs among Korean children, 2) to identify potential exposure sources among children, and 3) to examine associations with lipid and thyroid hormone related effect biomarkers. The results of this study will help identify priority PFAAs among children, and understand potential health effects of such exposure during this vulnerable period of life stage.

Table 1
Participant characteristics.

Characteristic		N (%)
Total		150 (100%)
Sex	Boys	80 (53%)
	Girls	70 (47%)
Age group	3–5 yr	30 (20%)
	7–9 yr	30 (20%)
	10–12 yr	30 (20%)
	13–15 yr	30 (20%)
	16–18 yr	30 (20%)
Monthly household income[a]	<3000 USD	40 (27%)
	3000–5000 USD	54 (37%)
	>5000 USD	53 (36%)
Paternal education[a]	≤High school	49 (33%)
	University (undergraduate level)	81 (55%)
	University (graduate level)	18 (12%)
Maternal education[a]	≤High school	60 (41%)
	University (undergraduate level)	80 (54%)
	University (graduate level)	7 (5%)
Second-hand smoking at home[a]	No	111 (76%)
	Yes	36 (24%)

[a] This information was missing for three observations.

2. Materials and methods

2.1. Study population and sample collection

The study subjects employed in the present study were a subgroup extracted from the population participating in Korea Environmental Health Survey in Children and Adolescents (KorEHS-C) of 2012–2014 (Fig. S1). KorEHS-C was the first nationwide survey on environmental health for nationally representative children and adolescent population of Korea. In 2012–2013, children with 7–18 years of age ($n = 1817$) were recruited, and subsequently in 2014, those with 3–5 years of age ($n = 577$) were added. Details of the study design and recruitment method are described previously (Ha et al., 2014; Won et al., 2016; Kang et al., 2017).

Questionnaire survey was conducted for demographic factors (e.g. sex and age), socioeconomic status (e.g. household income), information on chemical exposure (e.g. second-hand smoking), breast-feeding history, dietary characteristics (e.g. fish/shellfish consumption), behavioral characteristics (e.g. non-stick frying pan use and water-proof clothes use), and health status of the participating children. In case of elementary school students and younger (age 12 years and younger), all questionnaire was answered by their parents or legal guardians. For older participating children (age 13 and older), the questionnaire was answered by both participating children and their parents or legal guardians.

Fasting blood samples were collected. Vacutainer® EDTA tubes (3 mL volume, BD, Franklin Lakes, NJ, USA) and Vacuette® tubes SST (5 mL volume, Greiner Bio-One, Monroe, NC, USA) were used for collection of whole blood and serum, respectively. For serum separation, blood samples were centrifuged at 3000 rpm for 10 min. Both whole blood and serum samples were stored at −70 °C until the analyses. Among the archived serum samples of the KorEHS-C population, those collected from Seoul or Gyeonggi in 2012 (for age from 7 to 18) and 2014 (for age from 3 to 5) were stratified by age and sex, and randomly chosen for chemical analyses ($n = 150$) (Tables 1 and S1). However, among age group of 16–18 years old, only boys were chosen mistakenly for the age of 16 years old ($n = 10$), while those of age 17 and 18 years were evenly chosen ($n = 5$ for each year of age for each sex). The protocol of the present study was reviewed and approved by the Institutional Review Board of Hanyang University (IRB# HYI-15-067-2).