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Per- and polyfluoroalkyl substances in human serum and urine samples from a residentially exposed community

Rachel Rogers Worley[a,*], Susan McAfee Moore[a], Bruce C. Tierney[a], Xiaoyun Ye[b], Antonia M. Calafat[b], Sean Campbell[c], Million B. Woudneh[c], Jeffrey Fisher[d]

[a] Division of Community Health Investigations, Agency for Toxic Substances and Disease Registry, Atlanta, GA, USA
[b] Division of Laboratory Sciences, National Center for Environmental Health, Atlanta, GA, USA
[c] AXYS Analytical, Sidney, British Columbia, Canada
[d] National Center for Toxicological Research, Food and Drug Administration, Jefferson, AR, USA

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ABSTRACT

Background: Per- and polyfluoroalkyl substances (PFAS) are considered chemicals of emerging concern, in part due to their environmental and biological persistence and the potential for widespread human exposure. In 2007, a PFAS manufacturer near Decatur, Alabama notified the United States Environmental Protection Agency (EPA) it had discharged PFAS into a wastewater treatment plant, resulting in environmental contamination and potential exposures to the local community.

Objectives: To characterize PFAS exposure over time, the Agency for Toxic Substances and Disease Registry (ATSDR) collected blood and urine samples from local residents.

Methods: Eight PFAS were measured in serum in 2010 (n = 153). Eleven PFAS were measured in serum, and five PFAS were measured in urine (n = 45) from some of the same residents in 2016. Serum concentrations were compared to nationally representative data and change in serum concentration over time was evaluated. Biological half-lives were estimated for perfluorooctanoic acid (PFOA), perfluorooctane sulfonic acid (PFOS), and perfluorohexane sulfonic acid (PFHxS) using a one-compartment pharmacokinetic model.

Results: In 2010 and 2016, geometric mean PFOA and PFOS serum concentrations were elevated in participants compared to the general U.S. population. In 2016, the geometric mean PFHxS serum concentration was elevated compared to the general U.S. population. Geometric mean serum concentrations of PFOA, PFOS, and perfluorononanoic acid (PFNA) were significantly ($p \leq 0.0001$) lower (49%, 53%, and 58%, respectively) in 2016 compared to 2010. Half-lives for PFOA, PFOS, and PFHxS were estimated to be 3.9, 3.3, and 15.5 years, respectively. Concentrations of PFOA in serum and urine were highly correlated (r = 0.75) in males.

Conclusions: Serum concentrations of some PFAS are decreasing in this residentially exposed community, but remain elevated compared to the U.S. general population.

1. Introduction

Per- and polyfluoroalkyl substances (PFAS) are used in industrial applications and consumer products, including certain fire-fighting foams and stain, grease, and water repellent coatings on carpet, leather, and paper (ATSDR, 2015). The toxicity of and human exposure to perfluorooctanoic acid (PFOA) and perfluorooctane sulfonic acid (PFOS) have been extensively studied (Gilliland and Mandel, 1993; Butenhoff et al., 2002; Alexander et al., 2003; Butenhoff et al., 2004; Kennedy et al., 2004; Lau et al., 2006; Butenhoff et al., 2009; Frisbee et al., 2009; Butenhoff et al., 2012a; Butenhoff et al., 2012b). Information on the toxicity of other PFAS, particularly those with fewer than eight carbon atoms, is limited.

Production of PFOA and PFOS peaked between 1970 and 2002 and has diminished since then (DeWitt, 2015). PFOS is no longer manufactured in the United States (USEPA, 2014a). In January 2006, the United States Environmental Protection Agency (EPA) initiated the 2010/15 PFOA Stewardship Program, in which eight major companies in the PFAS industry committed voluntarily to eliminate emissions and product content of PFOA by 2015 (USEPA, 2014b). PFOA, PFOS, and other PFAS continue to be found in the environment, in wildlife, and in the blood of the general population, with accumulating evidence that human exposures are in decline (Taniyasu et al., 2003; Kannan et al., 2004; Calafat et al., 2006; Kato et al., 2011b; CDC, 2017).

* Corresponding author at: ATSDR Division of Community Health Investigations, 4770 Buford Highway, MS F59, Atlanta, GA 30341, USA.
E-mail address: RWorley@cdc.gov (R.R. Worley).

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The scientific evidence linking PFOA and PFOS exposures with adverse health effects is mixed and inconclusive. Human studies of people exposed to PFOA and PFOS occupationally, residentially, and at background levels have found associations with changes in lipid and cholesterol concentrations (Frisbee et al., 2010; Nelson et al., 2010; Fletcher et al., 2011; Steenland et al., 2015), increased uric acid levels (Costa et al., 2009; Steenland et al., 2010; Shankar et al., 2011; Geiger et al., 2013; Gleason et al., 2015), changes in the concentrations of thyroid and sex hormones (Olsen and Zobel, 2007; Knox et al., 2011; Jain, 2013; Wen et al., 2013; Winquist and Steenland, 2014), changes in liver enzymes (Olsen et al., 2000; Sakr et al., 2007; Lin et al., 2010; Gallo et al., 2012; Gleason et al., 2015), immune effects (Grandjean et al., 2012; Granum et al., 2013; Dalsager et al., 2016), reduced birth weight (Apelberg et al., 2007; Fei et al., 2007; Chen et al., 2012; Darrow et al., 2013), reproductive effects (Joensen et al., 2013; Kristensen et al., 2013; Crawford et al., 2017), and some cancers (Alexander and Olsen, 2007; Barry et al., 2013; Bonefeld-Jorgensen et al., 2014; Hardell et al., 2014; Steenland et al., 2015). Other studies have demonstrated no association between PFAS exposure and these health effects (Inoue et al., 2004; Alexander and Olsen, 2007; Fisher et al., 2013; Chang et al., 2014).

The pharmacokinetic behavior of many PFAS is different in humans than in animals (Andersen et al., 2006; Tatum-Gibbs et al., 2011). Human half-lives for PFAS have been determined in occupationally and residentially exposed populations; however, there are discrepancies in these estimates. These discrepancies potentially result from differences in the studied populations, including the level of exposure and the treatment of ongoing background exposures. Because of the observed variability in the estimation of serum half-lives, additional estimates of the biological half-lives of PFAS in human populations are needed to improve the understanding of PFAS pharmacokinetics.

In 2007, a PFAS manufacturer in the vicinity of Decatur, Alabama notified the EPA that it had discharged PFAS-contaminated waste water into a local wastewater treatment plant. Sewage sludge from this facility was applied to approximately 5000 acres of privately owned agricultural fields in the region between 1995 and 2008 (Lindstrom et al., 2011). Testing of soil, surface water, private drinking water wells, municipal water, and other environmental media revealed the potential for human exposures to these compounds (Hansen et al., 2002; USEPA, 2008; USEPA, 2009b; USEPA, 2009c; USEPA, 2009a; Lindstrom et al., 2011). In 2010, at EPA's request, the Agency for Toxic Substances and Disease Registry (ATSDR) collected blood samples from members of this community in order to characterize pathways of exposure. In January 2016, ATSDR conducted follow-up blood sampling, and added urine sampling, to evaluate how exposures in this community may have changed since 2010.

2. Methods

2.1. Study population

In 2009, ATSDR recruited individuals from Lawrence, Morgan and Limestone Counties, Alabama to participate in an exposure investigation. Community members with the highest likelihood of PFAS exposure were targeted for recruitment. In order to investigate the potential impact of exposure to PFAS in soil as a consequence of living or working on fields that received contaminated biosolid sludge, people who lived on or near agricultural fields that received contaminated sewage sludge were targeted for inclusion in the investigation. Because consumption of PFAS contaminated drinking water is an established exposure route, people who drank water from private wells with detectable levels of PFAS were also targeted for inclusion in the investigation. Participants were required to be 12 years of age or older, to have lived on their current property for at least one year, to be free of bleeding disorders and anemia, and to have no current or past occupational exposure to PFAS. One-hundred fifty-three people participated and sampling was conducted in 2010.

In 2015, these 153 people were contacted for recruitment into a follow up investigation. Potential participants were mailed a letter and contacted by phone in the summer and fall of 2015. Community members who agreed to be re-tested were sent a letter confirming their participation and were scheduled for an appointment to sign consent forms and receive urine collection materials (first appointment), and an additional appointment to provide a blood sample (second appointment). Seventy-eight of the original 153 agreed to be re-tested and 46 people completed all portions of the follow-up investigation. One participant reported occupational exposure to PFAS and was excluded from the analysis.

All participants in the 2010 and 2016 investigations provided written informed consent to participate. All phases of the investigation were conducted in compliance with the Centers for Disease Control and Prevention Institutional Review Board and the Office of Management and Budget Paperwork Reduction Act.

2.2. Questionnaire

In 2010 and 2016, ATSDR staff administered a questionnaire to each participant to gather information on exposure risk factors prior to blood sample collection. Participants were asked their address, how long they have lived there, how long they have lived in the Morgan, Lawrence, or Limestone county area, and to identify their primary source of drinking water. Participants were asked about their occupational history, and the frequency with which they work in the soil at work or home, consume locally grown vegetables, and eat locally caught fish.

In 2016, participants were asked to identify any changes related to drinking water, consumption of locally caught fish and locally grown vegetables, or other changes in personal habits or behavior that may have impacted their exposure to PFAS since the 2010 investigation.

2.3. Physical measurements

Physical measurements were obtained for each participant as part of the 2016 investigation. Each participant had their height measured with a SECA 217 portable stadiometer with a measuring range of 20–205 cm and 1 mm graduations. Body weight (BW) was measured with a SECA 869 scale with maximum capacity of 249.5 kg (kg), report graduations of 0.09 kg, and greater than ± 0.15% accuracy. Body fat percentage was measured with an Omron BF306 hand-held body fat analyzer (accuracy standard estimate of error: 4.1%). All information was recorded by an ATSDR staff person.

Body mass index (BMI) was calculated according to the following equation:

$$BMI = {}^{BW\ (kg)}\Big/_{height\ (m)^2}$$

Pearson's correlation test was applied to evaluate the strength of the association between body fat percentage and PFAS serum concentration and the association between BMI and PFAS serum concentration. Correlation coefficients were determined for total PFOS, total PFOA, PFNA, and PFHxS. Statistical analyses were performed with the freely available software R version 3.2.4 using the stats and NADA packages (R Core Team, 2016).

2.4. Serum sampling

Serum sampling was conducted in 2010 and 2016. In each investigation, five milliliter (mL) blood was collected by venipuncture by trained phlebotomists at a centralized sample collection location. Each sample tube was placed upright in a rack, allowed to clot for 30 min at room temperature, and then placed inside a storage box and kept at 4–5 °C. At the conclusion of sample collection the box was placed inside a plastic biohazard bag, placed inside a styrofoam shipping container

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