Environmental Research 154 (2017) 145–151

Contents lists available at ScienceDirect

Environmental Research

journal homepage: www.elsevier.com/locate/envres

Perfluoroalkyl substances and fish consumption

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ARTICLE INFO

Keywords:
PFAS
PFOA
PFOS
Fish
Shellfish
NHANES

ABSTRACT

Background: Perfluoroalkyl substances (PFAS) are an emerging class of contaminants. Certain PFAS are regulated or voluntarily limited due to concern about environmental persistence and adverse health effects, including thyroid disease and dyslipidemia. The major source of PFAS exposure in the general population is thought to be consumption of seafood.
Objectives: In this analysis we examine PFAS levels and their determinants, as well as associations between PFAS levels and self-reported fish and shellfish consumption, using a representative sample of the U.S. population.
Methods: Data on PFAS levels and self-reported fish consumption over the past 30 days were collected from the 2007–2008, 2009–2010, 2011–2012, and 2013–2014 cycles of the National Health and Nutrition Examination Survey. Twelve different PFAS were measured in serum samples from participants. Ordinary least squares regression models were used to identify factors (demographic characteristics and fish consumption habits) associated with serum PFAS concentrations. Additional models were further adjusted for other potential exposures including military service and consumption of ready-to-eat and fast foods.
Results: Seven PFAS were detected in at least 30% of participants and were examined in subsequent analyses (PFDA, PFOA, PFOS, PFHxS, MPAH, PFNA, PFUA). The PFAS with the highest concentrations were PFOS, followed by PFOA, PFHxS and PFNA (medians of 8.3, 2.7, 1.5 and 1.0 ng/mL). Fish consumption was generally low, with a median of 1.2 fish meals and 0.14 shellfish meals, reported over the past 30 days. After adjusting for demographic characteristics, total fish consumption was associated with reduced MPAH, and with elevated PFDE, PFNA and PFuDA. Shellfish consumption was associated with elevations of all PFAS examined except MPAH. Certain specific fish and shellfish types were also associated with specific PFAS. Adjustment for additional exposure variables resulted in little to no change in effect estimates for seafood variables.
Conclusions: PFAS are emerging contaminants with widespread exposure, persistence, and potential for adverse health effects. In the general population, fish and shellfish consumption are associated with PFAS levels, which may indicate an avenue for education and outreach.

1. Introduction

Perfluoroalkyl substances (PFAS) are emerging chemical pollutants which have been used for a wide range of consumer products due to their non-stick/non-stain properties (ATSDR, 2009; Steenland et al., 2010). The primary non-occupational route of exposure to PFAS is through the diet, namely seafood from contaminated water bodies. PFAS levels in seafood vary by location, age and type of seafood, and other factors (EPA, 2009b). Human biomonitoring studies have demonstrated that exposure to PFAS is widespread (e.g. (CDC, 2015)) with the most common being perfluorooctane sulfonate (PFOS), perfluorooctanoic acid (PFOA) and perfluorohexane sulfonate (PFHxS). However, in the United States (U.S.), PFOS and other PFAS with six or more carbon atoms were voluntarily phased out of production (reviewed in Buck et al. (2011)) and the U.S. Environmental Protection Agency (EPA) PFOA Stewardship Program was designed to reduce production and use of PFOA as well (EPA, 2009a). Despite these restrictions, PFAS continue to contaminate environmental media due to their persistence in the environment and in humans (Buck et al., 2011; Wang et al., 2013). Human exposure

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http://dx.doi.org/10.1016/j.envres.2016.12.032
Received 17 November 2016; Received in revised form 28 December 2016; Accepted 29 December 2016
0013-9351/

to PFAS is of concern because of the potential for adverse health effects observed in both animal (toxicology) and human (epidemiology) studies. For example, PFOA has been reported to be associated with thyroid disease (Melzer et al., 2010) and higher levels of cholesterol (Eriksen et al., 2013; Steenland et al., 2009) and uric acid (Gleason et al., 2015) in multiple human studies, and there is some evidence for an association between PFOA and elevation of liver enzymes (Gleason et al., 2015), and testicular and renal cancers (Benbrahim-Tallaa et al., 2014). While the evidence for health effects is not conclusive based on human epidemiology studies, investigation of exposure sources is still warranted.

Both in the U.S. and worldwide, fish are an increasingly important part of the human diet and offer many important nutritional benefits. Over the past few decades, fish consumption has increased by about 30% in the United States (Loke et al., 2012). Fish consumption may also be associated with health benefits; for example, epidemiologic studies suggest that increased fish consumption is associated with reduced risk of cardiovascular disease and coronary death, in part due to selenium and omega-three fatty acid content (He et al., 2004; Whelton et al., 2004). However, the presence of environmental contaminants such as PFAS in fish necessitates that risks and benefits both be considered when advising individuals about fish consumption (reviewed in Domingo (2016)). In the U.S. specifically, national fish tissue monitoring data have demonstrated widespread occurrence of many PFAS, with PFOS (median levels of 10.7 ng/g) being the most predominant, in the Great Lakes and in urban rivers across the country (Stahl et al., 2014). Although levels of certain PFAS are declining over time in the U.S. population, possibly reflecting limitation or elimination of certain exposure sources, levels of other PFAS are steady or increasing over time (CDC, 2015), pointing to the need to consider fish as an important ongoing source of exposure.

Associations between consumption of seafood and body burdens of PFAS have been observed in several studies across different countries. A cross-sectional study in Japan found significant associations between fish consumption (both raw and cooked) with increased PFOS serum concentrations (Yamaguchi et al., 2013). Similar observations were confirmed in studies conducted in Norway, where fish consumption is common in their traditional diets (Haug et al., 2010) (Hansen et al., 2016; Rylander et al., 2009, 2010). A study of fresh water anglers in Germany established a dose-response relationship between fish consumption and PFOS body burden (Holzer et al., 2011), and fresh water fish consumption was also found to be a significant contributor to PFAS body burden among anglers from a French metropolitan population (Denys et al., 2014). In the U.S., Egeghy and Lorber used a pharmacokinetic model to identify sources of PFOS exposure, and found that for the adult population the major source of exposure was indeed dietary; however, they noted the lack of occurrence data for the U.S. and indeed relied upon Canadian data for this purpose (Egeghy and Lorber, 2011). These concerns may be amplified for certain populations at increased risk for adverse health effects of PFAS exposure due to demographic characteristics or greater exposure via high fish consumption, including: pregnant women and women who are breast feeding, sport-anglers, subsistence anglers, and tribal communities.

PFAS concentrations and demographic characteristics in the U.S. general population have previously been studied using National Health and Nutrition survey (NHANES) data (Calafat et al., 2007; Kato et al., 2011). However, associations between specific seafood consumption and PFAS levels among the U.S. general population have not been explored and established. Due to their persistence in the environment and in the body, as well as the potential for adverse health effects due to exposure, it is important to monitor the levels of PFAS in the general population and in potentially vulnerable and susceptible subgroups. In this analysis we examine PFAS levels and associations with self-reported seafood consumption among a representative sample of the U.S. population.

2. Materials and methods

2.1. Study population

The National Health and Nutrition Examination Survey (NHANES) is a cross-sectional survey, designed to provide a representative sample of the US non-institutionalized civilian population (CDC, 2016). PFAS are measured in a random one-third subsample of NHANES participants 12 years of age and older. For this study, the four most recent NHANES cycles with PFAS information were combined: 2007/2008, 2009/2010, 2011/2012, and 2013/2014. Laboratory methods are described in detail in the NHANES documentation (CDC, 2014b); in brief, PFAS were measured in serum using solid phase extraction coupled to high performance liquid chromatography-turbo ion spray ionization-tandem mass spectrometry. As stated in the laboratory documentation, values below the limit of detection (LOD) are replaced with the value ($LOD/\sqrt{2}$). The list of PFAS analyzed is given in Table 1; PFAS which were not detected in at least 30% of samples (shaded in grey in Table 1) were not carried through further analyses. Due to inconsistency between PFAS acronyms used by NHANES and those generally accepted by the scientific community (Buck et al., 2011), chemical names, formulas, and acronyms are provided in Supplementary Table 1. Fish and shellfish consumption over the past 30 days was ascertained during the dietary interview (CDC, 2014c). Participants aged 12 years and older answered questions for themselves, and interviews were conducted in the participant's choice of either English or Spanish.

2.2. Statistical analysis

All data analysis was performed using SAS/STAT software version 9.4.[1] Ordinary least squares regression models were used to identify factors associated with PFAS serum levels; these factors included demographic characteristics as well as fish consumption. Demographic characteristics included: sex, age (years), body mass index (BMI), and race/ethnicity (Mexican American, Other Hispanic, Non-Hispanic white, Non-Hispanic Black, Other/multiracial). Fish and shellfish consumption were evaluated using self-reported meals over the past 30 days. This included total number of fish and shellfish meals consumed, as well as number of meals broken out by specific type of shellfish (clams, crabs, crayfish, lobsters, mussels, oysters, scallops, shrimp, other shellfish) and fish (breaded fish, tuna, bass, catfish, cod, flatfish, haddock, mackerel, perch, pike, pollock, porgy, salmon, sardines, sea bass, shark, swordfish, trout, walleye, and other fish). Additional models included family income and other suspected PFAS exposure sources, including history of military service and foreign-born versus U.S. born. The additional models included variables designed to capture potential non-seafood sources of PFAS exposure. PFAS have been used in firefighting substances, with subsequent detection at military (hence the inclusion of military service), firefighting and aviation sites (e.g., (Bhavsar et al., 2016; Hu et al., 2016)). The inclusion of country of birth is a proxy for differential exposure due to different sources and levels in non-U.S. countries.

Associations of demographic factors with PFAS levels and fish consumption were evaluated using Kruskal-Wallis tests. Each demographic factor evaluated was associated with at least one fish consumption parameter and one PFAS, and thus were included in multiple linear regression models as potential confounders. Due to non-normality of the data, PFAS levels were natural logarithm transformed. Effect estimates were exponentiated for easier interpretation of results, and represent proportional changes in the

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