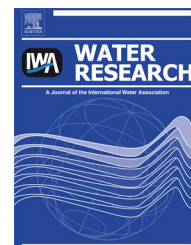


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Treatment of poly- and perfluoroalkyl substances in U.S. full-scale water treatment systems

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ABSTRACT

The near ubiquitous presence of poly- and perfluoroalkyl substances (PFASs) in humans has raised concerns about potential human health effects from these chemicals, some of which are both extremely persistent and bioaccumulative. Because some of these chemicals are highly water soluble, one major pathway for human exposure is the consumption of contaminated drinking water. This study measured concentrations of PFASs in 18 raw drinking water sources and 2 treated wastewater effluents and evaluated 15 full-scale treatment systems for the attenuation of PFASs in water treatment utilities throughout the U.S. A liquid-chromatography tandem mass-spectrometry method was used to enable measurement of a suite of 23 PFASs, including perfluorocarboxylic acids (PFCAs) and perfluorosulfonic acids (PFSAs). Despite the differences in reporting levels, the PFASs that were detected in >70% of the source water samples ($n = 39$) included PFSAs, perfluorobutane sulfonic acid (74%), perfluorohexane sulfonic acid (79%), and perfluorooctane sulfonic acid (84%), and PFCAs, perfluoropentanoic acid (74%), perfluorohexanoic acid (79%), perfluoroheptanoic acid (74%), and perfluorooctanoic acid (74%). More importantly, water treatment techniques such as ferric or alum coagulation, granular/micro-/ultra-filtration, aeration, oxidation (i.e., permanganate, ultraviolet/hydrogen peroxide), and disinfection (i.e., ozonation, chlorine dioxide, chlorination, and chloramination) were mostly ineffective in removing PFASs. However, anion exchange and granular activated carbon treatment preferably removed longer-chain PFASs and the PFSAs compared to the PFCAs, and reverse osmosis demonstrated significant removal for all the PFASs, including the smallest PFAS, perfluorobutanoic acid.

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1. Introduction

Poly- and perfluoroalkyl substances (PFASs) are a group of chemicals that have been used directly in or as part of the manufacturing of a wide variety of industrial and consumer products including, but not limited to, firefighting foams, paper and cardboard coating materials employed in food packaging, ScotchGard™, and Teflon™. One class of PFASs are the perfluoroalkyl acids (PFAAs), which are stable chemicals made of a carbon backbone surrounded by fluorine atoms and a terminal acid group, such as carboxylic or sulfonic acid. These PFAAs are extremely resistant to degradation and thus stable in both water and soil and highly persistent in the environment (Buck et al., 2011; ATSDR, 2009).

Unlike most other persistent and bioaccumulative organic pollutants (e.g. dioxin, polychlorinated biphenyls), PFAAs are water soluble. A number of PFAAs have been detected in U.S. surface waters, including lakes, rivers, and tributaries, in the ng/L range or lower (Furdui et al., 2008; Mak et al., 2009; Nakayama et al., 2007; Plumlee et al., 2008; Post et al., 2012; ATSDR, 2009), and they have also been detected in ground waters at similar concentrations (Plumlee et al., 2008; Post et al., 2009). In one study (Sinclair and Kannan, 2006), several PFAAs were detected in effluents from wastewater treatment plants in New York, with concentrations in the low ng/L range for all of the chemicals tested, except for perfluorooctanoic acid (PFOA), which had median concentrations for each utility ranging from 67 ng/L to 697 ng/L. The high persistence of PFAAs in the environment and widespread detection has created a concern for the possible exposure to animals and humans. PFOA, perfluorononanoic acid (PFNA), perfluorooctane sulfonic acid (PFOS), and perfluorohexane sulfonic acid (PFHxS) are found in the serum of virtually all U.S. residents (Kato et al., 2011). Some of these chemicals, including PFOA and PFOS, have been detected in the blood of animals in remote regions of the world (Houde et al., 2006). Due to concerns about widespread occurrence and potential health effects, major U.S. manufacturers of PFOS and PFHxS stopped production in 2002 (Butenhoff et al., 2009), while the worldwide use of PFOA and longer chain PFCA homologues is currently being phased out by their major manufacturers (Lindstrom et al., 2011). Replacements for these phased-out compounds include less extensively studied shorter chain PFASs, such as PFHxA and other fluorinated compounds, which generally are more rapidly excreted in humans and animals than the longer chain-length compounds (DuPont™, 2011; Gordon, 2011). However, these shorter-chain PFASs are still highly persistent in the environment (Ritter, 2010; Gordon, 2011).

As far as potential human exposure is concerned, some of these chemicals have also been detected in U.S. finished/tap waters in the low ng/L range (Post et al., 2009; Quiñones and Snyder, 2009), and higher levels (hundreds of ng/L to several µg/L) of some PFASs have been found in finished drinking water, particularly near sites of industrial use (Emmett et al., 2006; Rumsby et al., 2009; Skutlarek et al., 2006). Drinking water exposure is one pathway that may have contributed to increased concentrations in the serum of humans in most developed countries (Post et al., 2012; ATSDR, 2009) and serum

levels of PFOA, PFOS, and PFHxS were increased in communities with contaminated drinking water (ATSDR, 2013; Hölzer et al., 2008; Post et al., 2009). Other potential routes of human exposure include ingestion from food, food packaging, treated carpets, upholstery, and clothing, house dust, protective sprays, and waxes sold as consumer products (Post et al., 2012).

Human epidemiological studies have found associations with numerous health endpoints resulting from exposure, including drinking water exposure (Post et al., 2012; Saikat et al., 2013). An independent panel of prominent environmental epidemiologists concluded that probable links exist between six adverse health conditions and PFOA exposure in a community with contaminated drinking water, including kidney cancer and testicular cancer (C8 2012, Vieira et al., 2013). A number of associations of PFAA exposure with health effects have been reported in the general population, including a study that found an association between PFOA and PFOS exposures and a reduced humoral immune response to routine childhood immunizations in children aged 5 and 7 years (Grandjean et al., 2012).

In 2009, the U.S. Environmental Protection Agency (EPA) established Provisionary Health Advisory (PHA) values for PFOA and PFOS of 0.4 and 0.2 µg/L, respectively, in response to an emergency situation in Decatur, Alabama, which were meant to protect from short term exposure (USEPA, 2013). In addition, PFOS and PFOA have been added to the EPA's Contaminant Candidate List 3 of chemicals under consideration for future drinking water regulation in the U.S. (USEPA, 2009). These two chemicals as well as perfluoroheptanoic acid (PFHpA), PFNA, perfluorobutane sulfonic acid (PFBS), and PFHxS were also added to the EPA's Unregulated Contaminant Monitoring Rule 3 (UCMR 3) which requires nationwide monitoring by public water suppliers to provide occurrence data needed for regulatory decision making (Post et al., 2012; USEPA, 2012). As some PFAAs have been shown to have a potential to harm humans, it is imperative that treatment options be examined for their ability to remove these chemicals from contaminated water sources.

Previous studies assessed the occurrence of PFAAs in raw and finished waters of full-scale conventional drinking water treatment trains (Post et al., 2009; Quiñones and Snyder, 2009), where the latter study examined treatment systems that consisted of coagulation/flocculation, filtration, ozonation, chlorination, and chloramination processes (Quiñones and Snyder, 2009). These studies suggested these treatment systems were ineffective towards PFAA removal, but confirmation of these results has not been shown at other types of full-scale systems or for a wider spectrum of PFASs. To date, some less commonly employed processes, such as anion exchange (AIX), granular activated carbon (GAC), nanofiltration (NF), and reverse osmosis (RO) have been evaluated at the bench scale and showed promise in the removal of some of these chemicals (Deng et al., 2010; Lampert et al., 2007; Steinle-Darling and Reinhard, 2008; Tang et al., 2006; Appleman et al., 2013). GAC and RO have also been evaluated at the full-scale in a few studies performed outside of the U.S. (Eschauzier et al., 2012; Flores et al., 2013; Takagi et al., 2011; Thompson et al., 2011), where GAC was somewhat ineffec-

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