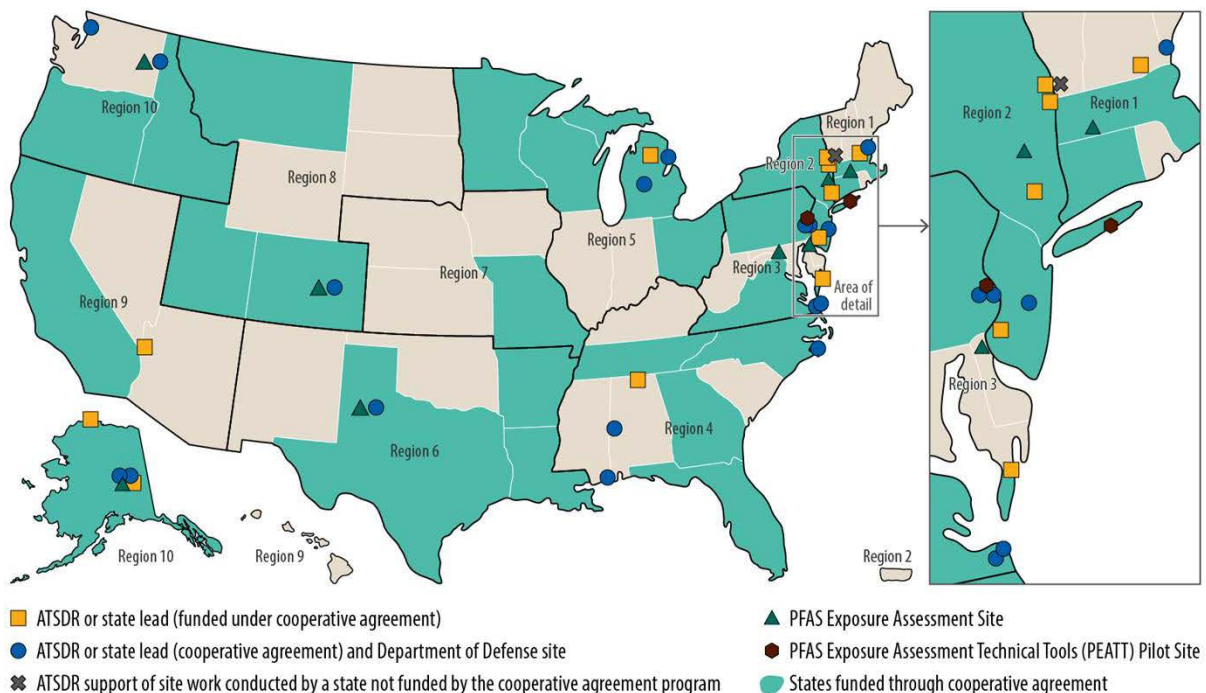


CHAPTER 5. POTENTIAL FOR HUMAN EXPOSURE

5.1 OVERVIEW

Perfluoroalkyls have been identified in at least 4 of the 1,854 hazardous waste sites that have been proposed for inclusion on the EPA National Priorities List (NPL) (ATSDR 2017). However, the number of sites in which substance perfluoroalkyls have been evaluated is not known. Figure 5-1 illustrates perfluorinated compound (PFAS) sites with ATSDR, state health department, EPA, or Department of Defense involvement.

Figure 5-1. Perfluorinated Compound (PFAS) Sites with ATSDR, State Health Department, U.S. Environmental Protection Agency, or Department of Defense Involvement



Source: ATSDR 2018

- The general population is exposed to the perfluoroalkyls through food and water ingestion, dust ingestion, inhalation exposure, and hand-to-mouth transfer of materials containing these substances.
- PFOA, PFOS, and their precursor substances are no longer produced or used in the United States or most other industrialized nations; however, these substances are persistent in the environment and exposure near highly contaminated sites may continue to occur.
- Serum levels of PFOA and PFOS in the general population of the United States have declined dramatically since 2000.

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Perfluoroalkyls have been released to air, water, and soil in and around fluorochemical facilities located within the United States (3M 2007b, 2008a, 2008b; Barton et al. 2007; Davis et al. 2007; DuPont 2008; EPA 2008a; Post et al. 2013). Since the early 2000s, eight companies in the fluorochemical industry have been working in concert with the EPA to phase out the production and use of long-chain perfluoroalkyls and their precursors (3M 2008a; DuPont 2008; EPA 2007a, 2008a, 2016a). Perfluorocarboxylic acids containing seven or more perfluorinated carbon groups and perfluoroalkyl sulfonic acids containing six or more perfluorinated carbon units are considered long-chain substances. Perfluorinated carboxylic acids and sulfonic acids containing less than seven and six perfluorinated carbons, respectively, are considered short-chain substances. PFOA, PFOA precursors, and higher homologues have been phased out by the eight corporations in the perfluorotelomer/fluorotelomer industry (Arkema, Asahi, BASF [successor to Ciba], Clariant, Daikin, 3M/Dyneon, DuPont, and Solvay Solexis) as part of the EPA's PFOA Stewardship Program (DuPont 2008; EPA 2008a, 2016a). Industrial releases of these compounds in the United States have declined or have been totally eliminated based on company reports submitted to EPA (EPA 2008a, 2016a). It is noted that PFOA and PFOS may still be produced domestically, imported, and used by companies not participating in the PFOA Stewardship Program. PFOA and PFOS may also be present in imported articles. Although the United States and most industrialized nations have stopped producing PFOA and PFOS, China remains a major producer and user of both substances, and its production has increased as production in the rest of the world has declined (HAES 2017; Li et al. 2015; Lim et al. 2011).

Perfluoroalkyl carboxylic acids and sulfonic acids are expected to dissociate in the environment based on their low pKa values (Kissa 2001; SPARC 2008), and anions will not volatilize from water or soil surfaces (Prevedouros et al. 2006). The unique surfactant properties of these substances may prevent total dissociation of perfluoroalkyls in water (EPA 2005a; Kissa 2001; Prevedouros et al. 2006); therefore, some volatilization of perfluoroalkyls may occur since the neutral forms of these substances are considered to be volatile (Barton et al. 2007; EPA 2005a; Kim and Kannan 2007). Perfluoroalkyls have been detected in air both in the vapor phase and as adsorbed to particulates (Kim and Kannan 2007). Perfluoroalkyls are very stable compounds and are resistant to biodegradation, direct photolysis, atmospheric photooxidation, and hydrolysis (3M 2000; EPA 2008a; OECD 2002, 2007; Schultz et al. 2003). Perfluoroalkyls released to the atmosphere are expected to adsorb to particles and settle to the ground through wet or dry deposition (Barton et al. 2007; Hurley et al. 2004; Prevedouros et al. 2006). The chemical stability of perfluoroalkyls and the low volatility of these substances in ionic form indicate that perfluoroalkyls will be persistent in water and soil (3M 2000; Prevedouros et al. 2006). Soil

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adsorption coefficient data as well as monitoring studies suggest that perfluoroalkyls such as PFOA are mobile in soil and can leach into groundwater (Davis et al. 2007; Prevedouros et al. 2006).

Perfluoroalkyls have been detected in environmental media and biota of the Arctic region and in other remote locations such as open ocean waters (Barber et al. 2007; Brown et al. 2018; Prevedouros et al. 2006; Wei et al. 2007a; Yamashita et al. 2005, 2008). Proposed source pathways include long-range atmospheric transport of precursor compounds followed by photooxidation to form perfluoroalkyls, direct long-range transport of perfluoroalkyls via oceanic currents, and transport of perfluoroalkyls in the form of marine aerosols (Armitage et al. 2006; Barber et al. 2007; Brown et al. 2018; Prevedouros et al. 2006; Wania 2007). Direct transport of perfluoroalkyls in the atmosphere has also been proposed as a source pathway since these substances have been detected in the vapor phase in outdoor air samples (CEMN 2008; Prevedouros et al. 2006). The actual source of perfluoroalkyls in remote locations is likely to be a combination of these pathways.

The highest concentrations of PFOA and PFOS were in apex predators, such as polar bears, which indicates that these substances biomagnify in food webs (de Vos et al. 2008; Houde et al. 2006b; Kannan et al. 2005; Kelly et al. 2007; Smithwick et al. 2005a, 2005b, 2006). The bioaccumulation potential of perfluoroalkyls is reported to increase with increasing chain length (de Vos et al. 2008; Furdui et al. 2007; Martin et al. 2004b). In living organisms, perfluoroalkyls bind to protein albumin in blood, liver, and eggs and do not accumulate in fat tissue (de Vos et al. 2008; Kissa 2001).

The levels of PFOA and PFOS in serum samples of U.S. residents have decreased appreciably since the phase out of these substances in the United States. The geometric mean serum levels of PFOS have declined over 84% from NHANES survey years 1999–2000 (30.4 ng/mL) to 2013–2014 (4.72 ng/mL) and the geometric mean serum levels of PFOA have declined 70% over the same temporal period, decreasing from 5.2 ng/mL in years 1999–2000 to 1.56 ng/mL for 2015–2016 (CDC 2018).

Mean concentrations of PFHpA, PFNA, PFDA, PFUnA, PFDoDA, PFBS, PFBA, and FOSA are generally <1 ng/mL (Calafat et al. 2006b, 2007a, 2007b; CDC 2015; De Silva and Mabury 2006; Kuklennyik et al. 2004; Olsen et al. 2003a, 2003b, 2004c, 2005, 2007a). Major PFOS exposure pathways proposed for the general population include food and water ingestion, dust ingestion, and hand-to-mouth transfer from mill- or home-treated carpets (Trudel et al. 2008). For PFOA, the major exposure pathways are proposed to be oral exposure resulting from general food and water ingestion, inhalation from impregnated clothes, and dust ingestion. While migration of residual PFOA in paper packaging and

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wrapping into food is also a potential route of exposure (Trudel et al. 2008), polyfluoroalkyl phosphoric acids in food packaging can also be metabolized in the body to PFOA (D'eon and Mabury 2007; D'eon et al. 2009). Polyfluoroalkyl phosphoric acids are fluorinated surfactant substances used to greaseproof food-containing paper products. Biotransformation of the 8:2 polyfluoroalkyl phosphoric acid and the 8:2 fluorotelomer alcohol into PFOA has been demonstrated (D'eon et al. 2009). Based on these proposed exposure pathways, Trudel et al. (2008) estimated that adult uptake doses for high-exposure scenarios were approximately 30 and 47 ng/kg body weight/day for PFOS and PFOA, respectively. The estimated dosage for children under the age of 12 under a high-exposure scenario were estimated to be 101–219 and 65.2–128 ng/kg body weight/day for PFOS and PFOA, respectively (Trudel et al. 2008). It is noted that the Trudel et al. (2008) study used older monitoring data and thus, may not be an accurate reflection of current intakes. Estimated daily doses for the general population were also estimated by Vestergren et al. (2008) to range from 3.9 to 520 ng/kg body weight/day for PFOS and from 0.3 to 150 ng/kg body weight/day for PFOA. Infants and toddlers had the highest estimated dosages due to greater hand-to-mouth contact with treated carpeting, mouthing activities of clothes, and greater dust ingestion. While conversion of precursor compounds to PFOA and PFOS was generally considered as a minor contribution to the total exposure, under certain scenarios, it was estimated that up to 80% of the intake could be attributable to exposure to precursor substances followed by subsequent metabolism to PFOS or PFOA (Vestergren et al. 2008).

Perfluoroalkyls have been detected in human breast milk and umbilical cord blood. The reported maximum concentrations of PFOS and PFOA measured in human breast milk samples were 0.360–0.685 and 0.210–0.609 ng/mL, respectively (Kärman et al. 2007; Llorca et al. 2010; So et al. 2006b; Völkel et al. 2008). Maximum concentrations of other perfluoroalkyls were <0.18 ng/mL (Kärman et al. 2007). PFOS and PFOA have been detected in most umbilical cord blood samples with reported concentrations of 4.9–11.0 and 1.6–3.7 ng/mL, respectively (Apelberg et al. 2007a, 2007b; Fei et al. 2007; Inoue et al. 2004; Midasch et al. 2007). Other perfluoroalkyls have been detected less frequently, with maximum concentrations of <2.6 ng/mL.

Individuals who perform jobs that require frequent contact with perfluoroalkyl-containing products, such as individuals who install and treat carpets or firefighters, are expected to have occupational exposure to these substances. Individuals who work at fluorochemical facilities generally have had higher perfluoroalkyl serum levels than the general population based on exposures in the work environment (3M 2007b, 2008b, 2008c; Barton et al. 2006; Davis et al. 2007). Studies of individuals living near fluorochemical facilities indicate that drinking water is the major exposure pathway (Emmett et al. 2006a;

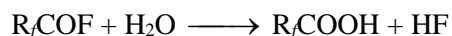
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Hölzer et al. 2008; Wilhelm et al. 2009). 3M conducted an exposure assessment to estimate the cumulative exposure to PFOA due to activities at the Decatur, Alabama facility. On-site exposure to groundskeepers, maintenance workers, construction workers, and on-site trespassers were considered. Off-site exposures to anglers, boaters, and residential individuals were also estimated. Various plausible exposure scenarios were considered, and the highest PFOA exposure doses by receptor and pathway occurred for local residents from groundwater followed by residents consuming drinking water from the West Morgan/East Lawrence (WM/EL) public drinking water supply (3M 2008c).

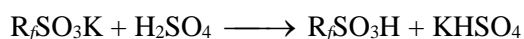
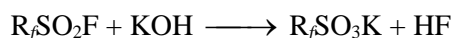
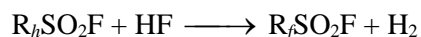
5.2 PRODUCTION, IMPORT/EXPORT, USE, AND DISPOSAL

5.2.1 Production

Perfluoroalkyls have been manufactured industrially by electrochemical fluorination (ECF), fluorotelomer iodide oxidation, fluorotelomer olefin oxidation, and fluorotelomer iodide carboxylation (Prevedouros et al. 2006; Schultz et al. 2003). During the ECF process, an organic acyl or sulfonyl fluoride backbone structure is dissolved in a solution of aqueous hydrogen fluoride (Savu 1994b; Siegemund et al. 2005). A direct electrical current is then passed through the solution, which replaces all of the hydrogens on the molecule with fluorines. Perfluoroacyl fluorides produced by ECF are hydrolyzed to form the perfluorocarboxylic acid, which is then separated via distillation. This method was used extensively by 3M in the production of perfluoroalkylsulfonates such as PFOS (3M 1999; Hekster et al. 2003; Schultz et al. 2003).



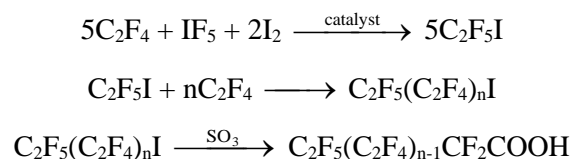
Perfluoroalkanesulfonyl fluorides produced by ECF are hydrolyzed under alkaline conditions to form the corresponding salt (Savu 1994b; Siegemund et al. 2005). Acidification followed by distillation yields the anhydrous perfluoroalkanesulfonic acid.



Perfluorosulfonamide compounds, such as FOSA, can be formed by reacting the perfluoroalkanesulfonyl fluoride with a primary or secondary amine (3M 1999; Hekster et al. 2003; Siegemund et al. 2005).

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The fluorotelomer iodide oxidation process was developed by DuPont and has served as the basis for their fluoropolymer production chemistry (Buck et al. 2011; Hekster et al. 2003; Savu 1994a; Siegemund et al. 2005). It begins with the preparation of pentafluoroiodoethane from tetrafluoroethene. Tetrafluoroethene is then added to this product at a molar ratio that gives a product of desired chain length. Finally, the product is oxidized to form the carboxylic acid. The process produces linear perfluorocarboxylic acids of even carbon numbers as illustrated below.

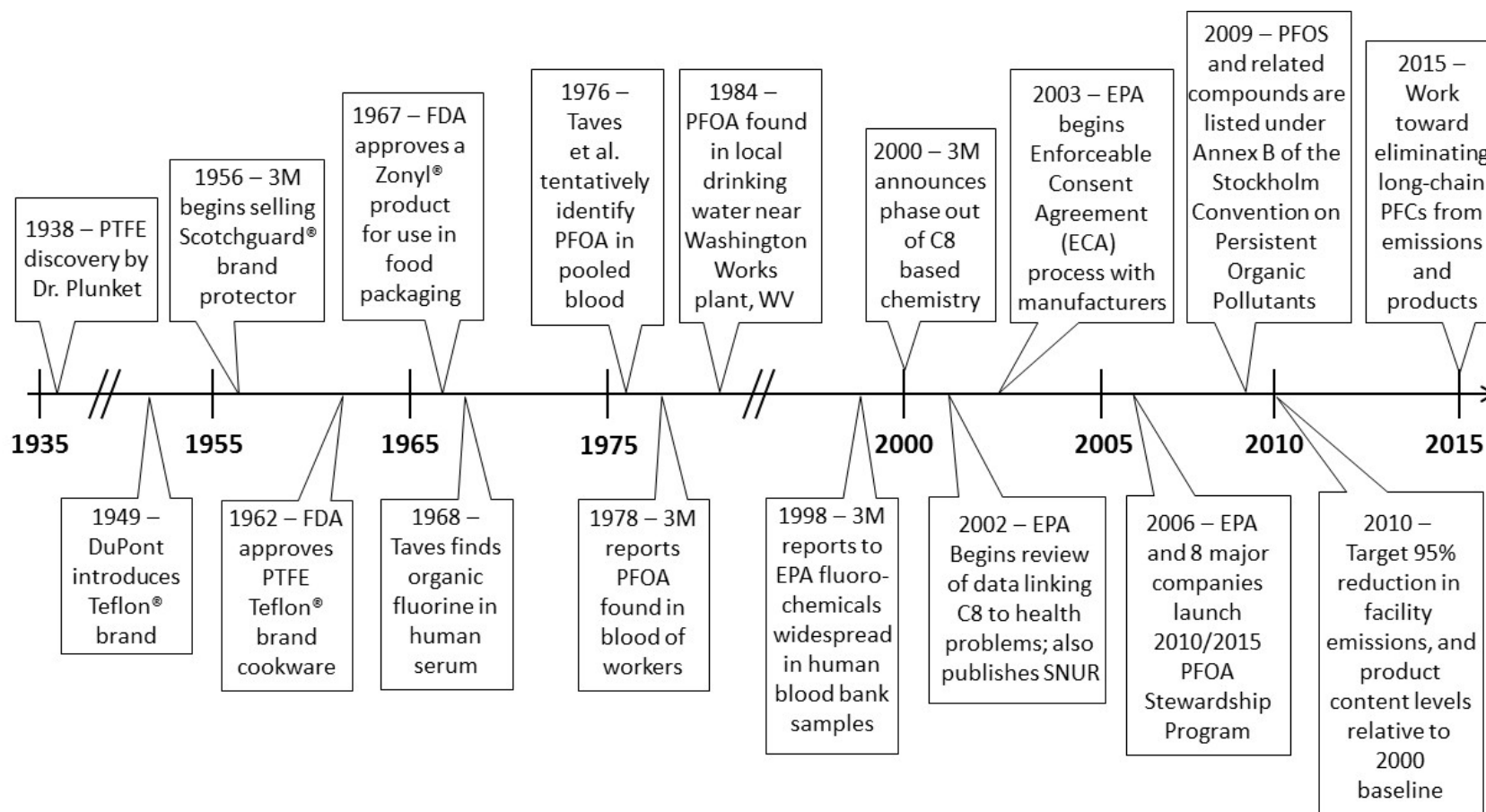


Fluorotelomer alcohols are created similarly, but the perfluoroalkyl iodide (telomer A) is reacted with ethylene to create $\text{F}(\text{CF}_2)_n\text{CH}_2\text{CH}_2\text{I}$ (telomer B), which is converted to the alcohol. The ECF process resulted in a mixture of linear and branched isomers, whereas the telomerization processes yielded predominantly linear products. It has been reported that the 3M ECF process resulted in approximately 70% linear and 30% branched isomers for PFOS and 78% linear and 22% branched isomers for PFOA (Benskin et al. 2009).

No information is available in the TRI database on facilities that manufacture, process, or otherwise use perfluoroalkyls because this class of substances is not required to be reported under Section 313 of the Emergency Planning and Community Right-to-Know Act (Title III of the Superfund Amendments and Reauthorization Act of 1986) (EPA 2005a, 2016g). The Chemical Data Reporting (CDR) rule, under the Toxic Substances Control Act (TSCA), requires manufacturers (including importers) to provide EPA with information on the production and use of chemicals in commerce in large quantities. Information on perfluoroalkyls can be found at (<https://www.epa.gov/chemical-data-reporting/basic-information-chemical-data-reporting#what>).

Perfluoroalkyls have been manufactured for their direct use in commercial products as well as for their use in industrial process streams. Two important chemicals that have resulted from manufacturing involving perfluoroalkyls, namely PFOS and PFOA, are of worldwide interest given their detection in multiple media in the environment. However, these substances and related long-chain perfluoroalkyls have been essentially phased out as a joint effort by EPA and industry (Lindstrom et al. 2011). The timeline for history of perfluorinated compound production, use, and phase out is presented in Figure 5-2.

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Figure 5-2. Timeline of Important Events in the History of Polyfluorinated Compounds

EPA = U.S. Environmental Protection Agency; PFC = perfluorinated compound; PFOA = perfluorooctanoic acid; PTFE = polytetrafluoroethylene; SNUR = significant new use rule; WV = West Virginia

Source: Reprinted (adapted) with permission from Lindstrom et al. 2011 (Environ Sci Technol 45:7954-7961). Copyright 2011 American Chemical Society.

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Given their unique properties, certain narrow exceptions exist for specific applications. Additionally, many of the substances that were used in the production of stain resistant or anti-sticking products that could break down into PFOA and PFOS have also been replaced.

The 3M Company was the principal worldwide manufacturer of PFOS and related chemicals. As a result of its phase-out decision in May 2000, 3M no longer manufactures perfluorooctanyl compounds (PFOA and PFOS). The company ceased manufacturing and using the vast majority of these compounds within approximately 2 years of the phase-out announcement, and ceased all manufacturing and the last significant use of this chemistry by the end of 2008 (3M 2008a; EPA 2007a). In 2000, EPA finalized the SNUR for 88 perfluoroalkyl sulfonate compounds, which requires manufacturers to notify EPA 90 days prior to commencing manufacture or import of these substances for a significant new use to allow time for evaluation (EPA 2002, 2007a, 2008a). The purpose of this rule was to limit future manufacturing and importation of these substances. According to EPA, the rule allowed for the continuation of a few limited, highly technical uses for which no alternatives are available, and which are characterized by very low volume, low exposure, and low releases. The SNUR was amended in 2007 to include 183 additional perfluoroalkyl sulfonate compounds (EPA 2007a, 2008a).

Included on the current list are PFOS, PFHxS, and FOSA. EPA believed that the perfluoroalkyl sulfonate compounds listed under the SNUR were no longer manufactured in the United States; however, during the comment period of the 2007 amendment, EPA learned of the ongoing use of tetraethylammonium perfluorooctanesulfonate as a fume/mist suppressant in metal finishing and plating baths (EPA 2007a). EPA has since excluded this from the list of significant uses. This rule has been amended again by the EPA to designate the processing, use, or importation of long-chain perfluoroalkyls as a significant new use if there are no current ongoing uses, or for uses that were scheduled to end December 31, 2015 (EPA 2015). As part of this amendment, EPA proposed to amend a SNUR for perfluoroalkyl sulfonate chemical substances that would make the exemption inapplicable for persons who import perfluoroalkyl sulfonate chemical substances as part of carpets or any articles that contain long-chain perfluoroalkyls.

In 2006, the eight major companies of the perfluoropolymer/fluorotelomer industry agreed to participate in EPA's PFOA Stewardship Program (EPA 2008a). All public documents and reports from the PFOA Stewardship Program may be reviewed at the EPA docket (EPA-HQ-OPPT-2006-0621). This program included voluntary commitments from these companies to reduce facility emissions and product content of PFOA and related chemicals on a global basis by 95% no later than 2010, and to work toward elimination of these substances in products by 2015. Progress reports have been submitted annually

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Table 5-1. Content (ppm) and Percent Reduction of PFOA, PFOA Homologues, or PFOA Precursors in Products from 2006 and 2013 U.S. Operations of Fluoropolymer/Fluorotelomer Companies

Company	Chemicals	Dispersions		Other fluoropolymers		Telomers	
		Content	Percent reduction ^a	Content	Percent reduction ^a	Content	Percent reduction ^a
Solvay Solexis	PFOA, PFOA salts, and higher homologues	Not applicable	>99.999%	Not applicable	>99.999%	Not applicable	>99.999%
	Precursors	Not applicable	Not applicable	Not applicable	Not applicable	Not applicable	Not applicable

^aPercent reduction in product content of these compounds from baseline year levels. The baseline year is the year nearest to the year 2000 for which company data are available.

PFOA precursors include: octane, 1,1,1,2,2,3,3,4,4,5,5,6,6,7,7, 8,8-heptadecafluoro-8-iodo- (CAS 507-63-1); 1-decanol, 3,3,4,4,5,5,6,6,7,7,8,8,9,9,10,10,10-heptadecafluoro-(CAS 678-39-7); 1-decene, 3,3,4,4,5,5,6,6,7,7,8,8,9,9,10,10,10-heptadecafluoro- (CAS 21652-58-4); 2-propenoic acid, 3,3,4,4,5,5,6,6,7,7,8,8,9,9,10,10,10-heptadecafluorodecyl ester (CAS 27905-45-9); 2-propenoic acid, 2-methy 1-, 3,3,4,4,5,5,6,6,7,7,8,8,9,9,10,10,10-heptadecafluorodecyl ester (CAS 1996-88-9); 2-decenoic acid, 3,4,4,5,5,6,6,7,7,8,8,9,9,10,10,10-hexadecafluoro- (CAS 70887-84-2); and decanoic acid, 3,3,4,4,5,5,6,6,7,7,8,8,9,9,10,10,10-heptadecafluoro- (CAS 27854-31-5).

Higher homologues include: dodecane, 1,1,1,2,2,3,3,4,4,5,5,6,6,7,7,8,8,9,9,10,10,11,11,12,12-pentacosafuoro-12-iodo (CAS 307-60-8); decane, 1,1,1,2,2,3,3,4,4,5,5,6,6,7,7,8,8,9,9,10,10-heneicosafuoro-10-iodo- (CAS 423-62-1); nonanoic acid, heptadecafluoro- (CAS 375-95-1); decanoic acid, nonadecafluoro- (CAS 335-76-2); 1-Decanol, 3,3,4,4,5,5,6,6,7,7,8,8,9,9,10,10,10-heptadecafluoro- (CAS 678-39-7); decane, 1,1,1,2,2,3,3,4,4,5,5,6,6,7,7,8,8-heptadecafluoro-10-iodo- (CAS 2043-53-0); dodecane, 1,1,1,2,2,3,3,4,4,5,5,6,6,7,7,8,8,9,9,10,10-heneicosafuoro-12-iodo- (CAS 2043-54-1); 2-propenoic acid, 3,3,4,4,5,5,6,6,7,7,8,8,9,9,10,10,10-heptadecafluorodecyl ester (CAS 4980-53-4); and 2-propenoic acid, 2-methyl-, 3,3,4,4,5,5,6,6,7,7,8,8,9,9,10,10,11,11,12,12,12-heneicosafuorododecyl ester (CAS 17741-60-5).

CAS = Chemical Abstracts Service; PFOA = perfluorooctanoic acid

China is one of the few remaining producers and consumers of PFOA and its salts, with a total of 480 metric tons produced from 2004 to 2012 (Li et al. 2015). China also continues to be a producer of PFOS. Growth in production volumes in China have coincided with decreases in production in the west. For example, China produced approximately 30 metric tons of PFOS in 2001; however, as 3M ceased production, China's production of PFOS increased to 91, 165, and 247 metric tons in 2004, 2005, and 2006, respectively (Lim et al. 2011). PFOA production in China was approximately 30 metric tons in 2004, but increased to approximately 90 metric tons in 2012 (Li et al. 2015). According to a report from the Hubei Academy of Environmental Sciences (HAES), China is planning to gradually phase out the production of some PFOS uses before 2019 and conduct a best available technology (BAT)/best environmental practice (BEP) analysis with the ultimate goal of completely phasing out the production and use of PFOS and potential precursors (HAES 2017).

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Historical U.S. production volume data for PFOA, PFBA, and PFOS reported by manufacturers under the EPA Inventory Update Rule (IUR) are provided in Table 5-2. Production volume ranges for the ammonium salt of PFOA, APFO, are also listed. During the reporting year 2002, manufacturers reported that the production volumes were within the range of 10,000–500,000 pounds (6–227 metric tons) for PFOS and PFOA and within the range of 500,000–1,000,000 pounds (227–454 metric tons) for APFO (EPA 2008b). PFBA was reported as having a production volume within the range of 10,000–500,000 pounds (6–227 metric tons) during 1986; however, PFBA production volumes were not reported for subsequent years (EPA 2008b). None of the other perfluoroalkyls were listed in EPA’s IUR database. Current U.S. production volume data for perfluoroalkyls are limited. The IUR database has been superseded by the Chemical Data Reporting (CDR) database. Data for 2012 indicated that PFOA was not imported into the United States, but any use or production volume data were reported as confidential business information. No data were located in the CDR for the other substances listed in Table 5-2. Nonconfidential emission reports from 2015 obtained from the EPA docket indicate that there is no current production of PFOA or PFOS in the United States.

Table 5-2. U.S. Production Volume Ranges for Perfluoroalkyls (1986–2002) Reported under the EPA Inventory Update Rule

Perfluoro-alkyl	Reporting year production volume range (pounds)				
	1986	1990	1994	1998	2002
PFOA	10,000–500,000	Not reported	10,000–500,000	10,000–500,000	10,000–500,000
APFO	10,000–500,000	10,000–500,000	10,000–500,000	10,000–500,000	500,000–1,000,000
PFBA	10,000–500,000	Not reported	Not reported	Not reported	Not reported
PFOS	Not reported	Not reported	10,000–500,000	Not reported	10,000–500,000

APFO = ammonium perfluorooctanoate; EPA = Environmental Protection Agency; PFBA = perfluorobutanoic acid; PFOA = perfluorooctanoic acid; PFOS = perfluorooctane sulfonic acid

Source: EPA 2008b

5.2.2 Import/Export

The SNURs cited in Section 5.2.1 severely limits the production, import, or export of the long-chain perfluoroalkyls. There were no reported imports of chemicals listed in Table 4-1 in 2012 from the CDR database. Production volumes and import volumes of the 8:2 fluorotelomer alcohol were listed as confidential business information by DuPont. Shaw Industries Group reported that they imported 37,478 pounds of 6:2 fluorotelomer alcohol into the United States in 2012, but DuPont declared both production volume and import volumes as confidential business information (EPA 2016d).

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5.2.3 Use

Applications of perfluoroalkyls have made use of their unique surfactant properties (Schultz et al. 2003). The alkyl tails of perfluoroalkyls make these substances both hydrophobic (water-repelling) and oleophobic (oil-repelling) (3M 1999; Kissa 2001; Schultz et al. 2003). Because of these properties, perfluoroalkyls have been used extensively in surface coating and protectant formulations (Kissa 2001). Major applications have included protectants for paper and cardboard packaging products, carpets, leather products, and textiles that enhance water, grease, and soil repellency (Hekster et al. 2003; Schultz et al. 2003). These compounds have been widely used in industrial surfactants, emulsifiers, wetting agents, additives, and coatings as well (3M 1999; Schultz et al. 2003). Perfluoroalkyls have been used in fire-fighting foams since they are effective in extinguishing hydrocarbon fueled fires (Schultz et al. 2003). Perfluoroalkyls have also been used as processing aids in the manufacture of fluoropolymers such as nonstick coatings on cookware, membranes for clothing that are both waterproof and breathable, personal care products (such as dental floss, cosmetics, sunscreens), electrical wire casing, fire and chemical resistant tubing, and plumbing thread seal tape (DuPont 2008; EPA 2008a).

5.2.4 Disposal

Information concerning disposal of individual perfluoroalkyl products may be found on Material Safety Data Sheets (MSDS) or Safety Data Sheets (SDS) from the manufacturers of the chemicals. Two methods are generally recommended for the disposal of fluoropolymer dispersions. The first method involves precipitation, decanting, or filtering to separate solids from liquid waste. The dry solids are then disposed of in an approved industrial solid waste landfill or incinerated, while the liquid waste is discharged to a waste water treatment facility (Plastics Europe 2012). The second method involves incineration at temperatures $>800^{\circ}\text{C}$ using a scrubber to remove hydrogen fluoride (Plastics Europe 2012). According to perfluorochemical facility assessment reports, historical disposal of perfluoroalkyl containing waste has been through on- and off-site landfills, through sludge incorporation (subsurface injection), and through incineration (3M 2007b, 2008a; ATSDR 2005). Pilot scale studies in which carpet samples were incinerated using a rotary kiln incinerator indicated that most perfluoroalkyls were effectively destroyed in combustors (Lemieux et al. 2007). Similar conclusions were reached by Yamada et al. (2005) when studying the incineration of textiles and paper treated with fluorotelomer-based acrylic polymers. Incineration at conventional temperatures is a proven technology for treating wastes containing perfluoroalkyls.

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5.3 RELEASES TO THE ENVIRONMENT

There is no information listed in EPA's Toxic Release Inventory (TRI) on releases of perfluoroalkyls to the environment from facilities manufacturing, processing, or otherwise using perfluoroalkyls because these releases are not required to be reported within this program (EPA 2005b, 2016g).

Perfluoroalkyls are man-made compounds that are not naturally occurring in the environment. Perfluoroalkyls such as PFOS and PFOA have been widely used in the manufacturing of many consumer products (Hekster et al. 2003; Schultz et al. 2003). These substances are still detected in both environmental and biological media around the world as well as in serum samples collected from the general population (Calafat et al. 2006b, 2007a, 2007b; CDC 2018, 2019; De Silva and Mabury 2006; Kuklenyik et al. 2004; Olsen et al. 2003b, 2003c, 2004b, 2004c, 2005, 2007a; Prevedouros et al. 2006).

In 2006, the eight major companies of the perfluoropolymer/perfluorotelomer industry agreed to participate in EPA's PFOA Stewardship Program (EPA 2008a). This included voluntary commitments from these companies to reduce facility emissions and product content of PFOA and related chemicals on a global basis by 95% no later than 2010, and to work toward elimination of these substances by 2015 (EPA 2008a). Data from 2007 and 2013 progress reports regarding releases of PFOA, PFOA precursors, and higher PFOA homologues to all media as well as percent reduction in releases are listed in Table 5-3.

Table 5-3. Reported Emissions of PFOA, PFOA Homologues, or PFOA Precursors in Products from the 2006 and 2013 U.S. Operations of Fluoropolymer/Fluorotelomer Companies

Company	Chemicals	Releases to all media from fluorotelomer and telomer manufacturing		
		kg	kg of release/100 kg of product produced	Percent reduction in emissions ^a
2006 Data (EPA 2008a)				
Arkema, Inc.	PFOA and higher homologues	>1,000–10,000	For fluorotelomer production: >0.1–1	22%
	Precursors	Not applicable	Not applicable	Not applicable
Asahi Glass Company	PFOA, PFOA salts, and higher homologues	4,922	For fluorotelomer production: <1	6%
	Precursors	Not applicable	Not applicable	Not applicable
BASF (Ciba Specialty Chemicals Corporation)	PFOA	0.05 ^b		>99%
	Higher homologues	0.05 ^b		>99%
	Precursors	0 ^b		>99%

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Table 5-3. Reported Emissions of PFOA, PFOA Homologues, or PFOA Precursors in Products from the 2006 and 2013 U.S. Operations of Fluoropolymer/Fluorotelomer Companies

Company	Chemicals	Releases to all media from fluoro- telomer and telomer manufacturing		
		kg	kg of release/100 kg of product produced	Percent reduction in emissions ^a
Clariant International Ltd.	Not applicable	Not applicable	Not applicable	Not applicable
Daikin America, Inc.	PFOA	Confidential business information	For fluorotelomer production: 8.0×10^{-3} ; for telomer production: 6.4×10^{-7}	94% for FP production; 92% for telomer production
	Precursors and higher homologues	Confidential business information	For production: 6.4×10^{-7}	22% for telomer production
E.I. DuPont de Nemours and Company	PFOA, PFOA salts	1,100	Not reported	98%
	Direct precursors	Confidential business information	Not reported	Confidential business information
3M/Dyneon	PFOA	0	0	100%
Solvay Solexis	PFOA and PFOA salts	Not applicable	Not applicable	Not applicable
	Higher homologues	>1,000–10,000	For fluorotelomer production: 0.161	28%
	Precursors	Not applicable	Not applicable	Not applicable
2013 data (EPA 2016a)				
Arkema, Inc.	PFOA and higher homologues	>500–2,000	>0.001–0.005	91%
	Precursors	Not applicable	Not applicable	Not applicable
Asahi Glass Company	PFOA, PFOA salts, and higher homologues	0	Not applicable	100%
	Precursors	Not applicable	Not applicable	Not applicable
BASF (Ciba Specialty Chemicals Corporation)	PFOA	Not applicable	Not applicable	100%
	Precursors	Not applicable	Not applicable	100%
Clariant International Ltd.	PFOA, PFOA salts	Not applicable	Not applicable	Not applicable
	Precursors	Not applicable	Not applicable	Not applicable
Daikin America, Inc.	PFOA	0	0	100%

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Table 5-3. Reported Emissions of PFOA, PFOA Homologues, or PFOA Precursors in Products from the 2006 and 2013 U.S. Operations of Fluoropolymer/Fluorotelomer Companies

Company	Chemicals	Releases to all media from fluorotelomer and telomer manufacturing		
		kg	kg of release/100 kg of product produced	Percent reduction in emissions ^a
E.I. DuPont de Nemours and Company	Precursors and higher homologues	0	0	100%
	PFOA, PFOA salts	90	None reported	99.8%
	Higher homologues	None reported	None reported	None reported
	Precursors	Confidential business information	None reported	Confidential business information
3M/Dyneon	PFOA, PFOA salts, and higher homologues	0	0	100%
	Precursors	0	0	Not applicable
Solvay Solexis	PFOA, PFOA salts, and higher homologues	0	0	>99.999%
	Precursors	0	0	Not applicable

^aPercent reduction in product content of these compounds from baseline year levels. The baseline year is the year nearest to the year 2000 for which company data are available.

^bTotal for emissions and product content

PFOA = perfluorooctanoic acid

While the United States and most industrialized countries around the world have ceased production of PFOS and PFOA, China is still a major producer of both substances (Li et al. 2015; Lim et al. 2011). Over the period from 2004 to 2012, it was estimated that 250 metric tons of PFOA were released to the environment from production in China (Li et al. 2015). Fluoropolymer manufacturing and processing was considered the dominant source of environmental releases, accounting for >80% of the total, while PFOA releases related to end use consumer products accounted for 6% of the total. Brazil produced 379 metric tons of N-EtFOSA for use in the pesticide, Sulfuramid, from 2004 and 2015, which contribute to PFOS releases to the environment (Wang et al. 2017).

Prevedouros et al. (2006) estimated the total global historical emissions of perfluoroalkyl carboxylates into the environment from both direct and indirect sources from the time period of 1951–2004. These data are provided in Table 5-4. Based on these estimations, direct emissions (3,200–6,900 metric tons) have far exceeded indirect emissions (30–350 metric tons). The largest direct emissions identified are

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from industrial processes such as the manufacture of perfluoroalkyl carboxylates (470–900 metric tons), fluoropolymer manufacture (2,400–5,400 metric tons), and fluoropolymer processing (210–320 metric tons). Direct release of perfluoroalkyl carboxylates from use of aqueous firefighting foams and consumer and industrial products were estimated to be 50–100 and 40–200 metric tons, respectively. The largest indirect emissions identified were from perfluoroalkyl carboxylate residual impurities in perfluorooctyl-sulfonyl fluoride products (20–130 metric tons) and fluorotelomer-based precursor degradation (6–130 metric tons). Wang et al. (2014) expanded upon the work of Prevedouros et al. (2006) by considering additional emission sources of these substances and estimating emissions from 2003 to 2015 and projecting future emissions. These authors estimated emissions of 820–7,180 metric tons for 2003–2015 and projected between 20 and 6,420 metric tons for years 2016–2030. The estimates by Prevedouros et al. (2006) and Wang et al. (2014) contain a great degree of uncertainty as demonstrated by the wide range of values presented in the data. Wang et al. (2014) stated that uncertainty analysis using Monte Carlo methods is not possible because there is insufficient information available with respect to the range or distribution of the emissions. Instead, they introduced a scoring system to provide a qualitative description of the accuracy of the estimates that ranged from 0–1 (low uncertainty) to 2–3 (high uncertainty where estimates were based on crude assumptions or extrapolations).

Table 5-4. Global Historical PFCA Production and Emissions Estimates from 1951 to 2004^a

Environmental input source	Historical time period (years)	Estimated total global historical PFCA emissions (tonnes)
Direct PFCA sources		
PFCA manufacture		
PFO/APFO	1951–2004	400–700
PFN/APFN	1975–2004	70–200
Total manufactured		470–900
Industrial and consumer uses		
Fluoropolymer manufacture (APFO)	1951–2004	2,000–4,000
Fluoropolymer dispersion processing (APFO)	1951–2004	200–300
Fluoropolymer manufacture (APFN)	1975–2004	400–1,400
Fluoropolymer processing (APFN)	1975–2004	10–20
Aqueous firefighting foams (AFFF)	1965–1974	50–100
Consumer and industrial products	1960–2000	40–200

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Table 5-4. Global Historical PFCA Production and Emissions Estimates from 1951 to 2004^a

Environmental input source	Historical time period (years)	Estimated total global historical PFCA emissions (tonnes)
Total direct		3,200–6,900
Indirect PFCA sources		
POSF-based products		
PFCA residual impurities ^b	1960–2002	20–130
POSF-based precursor degradation	1960–2002	1–30
POSF-based AFFF	1970–2002	3–30
Fluorotelomer-based products		
PFCA residual impurities ^b	1974–2004	0.3–30
Fluorotelomer-based precursor degradation	1974–2004	6–130
Fluorotelomer-based AFFF	1975–2004	<1
Total indirect		30–350
Total source emissions (direct and indirect)		3,200–7,300

^aLow and high estimated values as well as the period of use/production for each source are based upon publicly available information cited in the text.

^bSome authors classify residual impurities as a direct emission source rather than indirect emission source (Buck et al. 2011).

AFFF = aqueous firefighting foams; APFN = ammonium perfluorononanoate; APFO = ammonium perfluorooctanoate; PFCA = perfluorinated carboxylic acid; PFN = perfluorononanoate; PFO = perfluorooctanoate; POSF = perfluorooctanesulfonyl fluoride

Source: Prevedouros et al. 2006

Wang et al. (2017) estimated the global emissions in 1958–2015 as 1,228–4,930 tonnes of PFOS and 1,230–8,738 tonnes of PFOS precursors, with most emissions occurring from 1958 to 2002. It was estimated that PFOS emissions from 2016 to 2030 will be 8–153 tonnes.

5.3.1 Air

There is no information listed in the TRI on releases of perfluoroalkyls to the atmosphere from facilities manufacturing, processing, or otherwise using perfluoroalkyls because these releases are not required to be reported (EPA 2005b, 2016g).

According to 3M, low levels of PFOA were released to air during manufacturing processes at the Decatur, Alabama facility until use of this substance ceased in 2004 (3M 2008b). This company states that there are currently no process-related air emissions of PFOA at this facility (3M 2008b). PFOA concentrations as high as 75,000–900,000 pg/m³ were measured at the fence line of the DuPont Washington Works

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facility near Parkersburg, West Virginia in 2004 (Barton et al. 2006; Davis et al. 2007; Prevedouros et al. 2006). High volume air samples collected at several monitoring stations near the Washington Works facility during nine events between August and October of 2005 contained PFOA at reported concentrations ranging from 10 to 75,900 pg/m³ (EPA 2007b). The mean and median of these reported concentrations are 5,500 and 240 pg/m³, respectively.

The presence of perfluoroalkyls in indoor air and dust indicates that perfluoroalkyl-containing consumer products such as treated carpets and textiles may be sources of release to air (Barber et al. 2007; Jahnke et al. 2007b; Kubwabo et al. 2005; Moriwaki et al. 2003; Prevedouros et al. 2006; Shoeib et al. 2004; Strynar and Lindstrom 2008). Perfluoroalkyls have also been identified on both indoor and outdoor window films (Gewurtz et al. 2009). Disposal of perfluoroalkyl-containing consumer products is also expected to be a source of release to air (Prevedouros et al. 2006). Harada et al. (2005a, 2006) proposed that automobiles may be a source of PFOA in urban air based on elevated levels measured near heavy traffic areas and the widespread use of this substance in automobile materials.

Perfluoroalkyl carboxylic acids and perfluoroalkyl sulfonic acids are formed by the atmospheric photooxidation of precursor compounds such as fluorotelomer alcohols and perfluoroalkyl sulfonamides (D'eon et al. 2006; Ellis et al. 2004; Martin et al. 2006; Wallington et al. 2006; Wania 2007).

Perfluoroalkyl carboxylic acids including PFOA, PFNA, PFHpA, and PFBA were observed as products during a laboratory study involving the photooxidation of 4:2, 6:2, and 8:2 fluorotelomer alcohols (Ellis et al. 2003, 2004). D'eon et al. (2006) observed both perfluoroalkyl carboxylic acids and perfluorobutane sulfonate among products of the photooxidation of N-methyl perfluorobutane sulfonamidoethanol.

5.3.2 Water

There is no information listed in the TRI on releases of perfluoroalkyls to water from facilities manufacturing, processing, or otherwise using perfluoroalkyls because these releases are not required to be reported (EPA 2005b, 2016g).

There are a number of sources of perfluoroalkyl release to surface water and groundwater, including release from manufacturing sites, industrial use, use and disposal of perfluoroalkyl-containing consumer products, fire/crash training areas, waste water treatment facilities, and from the use of contaminated biosolids (3M 2008b; Clara et al. 2009; Davis et al. 2007; Eggen et al. 2010; EPA 2009a; Kelly and

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Solem 2009; Moody and Field 1999; Moody et al. 2003; Sinclair and Kannan 2006; Prevedouros et al. 2006).

Waste water discharge was identified as a release pathway for APFO from the DuPont Washington Works facility in West Virginia (Davis et al. 2007). The average monthly concentrations of APFO measured in surface water from three outlets at the facility during 2007 and early 2008 ranged from 3.65 to 377 $\mu\text{g/L}$ (EPA 2008d). Reported concentrations of APFO and PFOA measured in surface water from four separate outlets at this facility during the same period were 3–64 and 2.3–61 $\mu\text{g/L}$, respectively. During perfluorochemical operations at the 3M Cottage Grove facility in Minnesota, waste water treatment plant effluent containing perfluoroalkyls was discharged to the Mississippi River. Discharge into Bakers Creek from the waste water treatment plant at the 3M Decatur facility was a principal source of PFOA release from this facility (3M 2008b).

Elevated levels of perfluoroalkyls, such as PFOA, PFOS, and PFHxS, measured in groundwater near fire-training areas are attributed to the use of these substances in aqueous firefighting foams (Moody and Field 1999; Moody et al. 2003). The concentrations of these three perfluoroalkyls in groundwater near a military fire-training site in Michigan were 8–105, 4.0–110, and 9–120 $\mu\text{g/L}$, respectively (Moody et al. 2003). A study of landfill leachates showed that perfluoroalkyls were primarily distributed to the water phase of leachates, which could eventually contaminate ground water (Eggen et al. 2010). Lang et al. (2017) estimated that between 563 and 638 kg of perfluoroalkyl substances were released from landfill leachate in the United States in 2013.

Waste water treatment plants have been shown to be significant contributors to perfluoroalkyls contamination of surface and ground water (Clara et al. 2009; EPA 2009a; Kelly and Solem 2009; Loganathan et al. 2007; Sinclair and Kannan 2006; Yu et al. 2009c). Influent, effluent, and sludge samples from 28 public and private waste water treatment plants in Minnesota were analyzed for 13 perfluoroalkyls; detectable concentrations of perfluoroalkyls were found in several facilities, primarily urban treatment plants (Kelly and Solem 2009). Elevated levels of PFOS at one facility (1.51 $\mu\text{g/L}$ in effluent) were attributed to a chrome plating facility using a surfactant containing fluorosulfonate to control hexavalent chromium emissions. Another study of chromium electroplating facilities in Chicago, Illinois and Cleveland, Ohio also found them to be significant sources of PFOS and other perfluoroalkyls in the environment (EPA 2009a). It was determined that perfluoroalkyls were being discharged from all 11 facilities at quantifiable levels and that PFOS was detected in waste water from 10 out of 11 facilities at levels of 0.0314–39 $\mu\text{g/L}$. PFOA and PFOS were detected in effluents of six waste water treatment

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plants located in New York at levels of 0.058–1.05 and 0.003–0.068 $\mu\text{g/L}$, respectively (Sinclair and Kannan 2006). PFOS and PFOA were detected in effluents of two waste water treatment plants located in Singapore at levels of 0.0053–0.5609 and 0.0112–1.057 $\mu\text{g/L}$, respectively (Yu et al. 2009c). PFOA, PFOS, and several other perfluoroalkyls were detected in effluent samples of 21 waste water treatment plants and 9 industrial point sources; PFOA and PFOS were reportedly identified in the effluents of all of the facilities monitored at an average level of 0.060 $\mu\text{g/L}$ for both substances (Clara et al. 2009).

Studies comparing perfluoroalkyl levels in influent and effluent from municipal waste water treatment facilities have found higher levels of some perfluoroalkyls, such as PFNA, PFOA, PFOS, and FOSA, in the effluent, as compared to the influent (Loganathan et al. 2007; Schultz et al. 2006b). For others, such as PFHxS and PFDA, waste water treatment resulted in lower concentrations or no change in the concentrations. Increases in perfluoroalkyl concentrations are likely due to the breakdown of perfluoroalkyl precursors such as polyfluoroalkyl phosphoric acids or fluorotelomer alcohols (D'eon et al. 2009; Gauthier and Mabury 2005; Wang et al. 2005a, 2005b).

A study of eight waste water treatment plants that discharge effluent to the San Francisco Bay found increases in PFBA and PFHxA levels between 2009 and 2014 and declines in longer-chain legacy substances (Houtz et al. 2016). Average concentrations of PFBA were 7.4 ± 4.7 ng/L in 2009 and 16 ± 5.8 ng/L in 2014 and the average concentration of PFHxA rose from 17 ± 4.0 ng/L in 2009 to 25 ± 5.1 ng/L in 2014. The average PFOA concentration decreased from 32 ± 30 ng/L in 2009 to 21 ± 13 ng/L in 2014 and the average PFOS concentration decreased from 24 ± 32 ng/L in 2009 to 13 ± 4.4 ng/L in 2014.

Land application of biosolids (treated sewage sludge) can also result in the release of perfluoroalkyls to surface and groundwater (Clark and Smith 2011; Lindstrom et al. 2011; Sepulvado et al. 2011). There appears to be some differences in the distribution of PFOA and PFOS in waste water effluent and biosolids, with higher levels of PFOA in waste water and higher PFOS levels in biosolids (Guo et al. 2010).

5.3.3 Soil

There is no information listed in the TRI on releases of perfluoroalkyls to soil from facilities manufacturing, processing, or otherwise using perfluoroalkyls because these releases are not required to be reported (EPA 2005b, 2016g).

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Perfluoroalkyls can be inadvertently released to soils through the use of biosolids applied as fertilizer to help maintain productive agricultural soils and stimulate plant growth. PFOA and PFOS were detected in both biosolids and biosolid-amended soils (Sepulvado et al. 2011). Six samples of biosolids obtained from the Metropolitan Water Reclamation District of Greater Chicago had levels of PFOS and PFOA of 80–219 and 8–68 ng/g, respectively (Sepulvado et al. 2011). The mean sum (\pm SD) of all perfluoroalkyls in the biosolids was 433 ± 121 ng/g, with PFOS being most prominent. Perfluoroalkyls can also be released into soil due to atmospheric transport and wet/dry deposition (Rankin et al. 2016; Strynar et al. 2012).

Liu et al. (2007) measured PFOA as a product of the biodegradation of 8:2 fluorotelomer alcohol in soil. This result, along with similar findings in activated sludge tests, indicates that biodegradation of fluorotelomer alcohols may result in the formation of perfluoroalkyl carboxylic acids in soil (Liu et al. 2007; Rankin et al. 2014; Wang et al. 2005a, 2005b).

5.4 ENVIRONMENTAL FATE

5.4.1 Transport and Partitioning

Air. Barton et al. (2007) investigated the atmospheric partitioning of PFOA during rain events near an industrial facility and concluded that this substance will be primarily adsorbed to particles in the air since PFOA was not detected in the vapor phase (detection limit of 0.2 ng/m³). Concentrations of PFOA in raindrops and as particulates were 11.3–1,660 ng/L and 0.09–12.40 ng/m³, respectively. The authors proposed that PFOA or APFO released into air from industrial facilities will be scavenged by atmospheric particles (including aqueous aerosols and raindrops) and dissociate to form the perfluorooctanoate anion. Although Barton et al. (2007) did not detect PFOA in the vapor phase during rain events, low concentrations (<0.12 – 3.16 pg/m³) of vapor-phase perfluoroalkyls measured by Kim and Kannan (2007) in urban air provide evidence of a partitioning equilibrium. Wet and dry deposition are expected to be the principal removal mechanisms for perfluoroalkyl carboxylic acids and sulfonic acids in particulate form from the atmosphere. Residence times with respect to these processes are expected to be days to weeks (Barton et al. 2007; Hurley et al. 2004; Kim and Kannan 2007).

Long-range atmospheric transport of precursor compounds such as fluorotelomer alcohols and perfluoroalkyl sulfonamides followed by the atmospheric photooxidation of these substances to form perfluoroalkyl carboxylic acids and perfluoroalkyl sulfonic acids resulted in PFOA and PFOS

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contamination in remote locations with no direct point sources for these compounds (Barber et al. 2007; D'eon et al. 2006; Dinglasan-Panlilio and Mabury 2006; Ellis et al. 2004; Martin et al. 2006; Simcik 2005; Small 2009; Wallington et al. 2006; Wania 2007). Fluorotelomer alcohols and perfluoroalkyl sulfonamides are volatile and possess long enough atmospheric residence times for long-range transport to occur (Barber et al. 2007; Yarwood et al. 2007). The presence of fluorotelomer alcohols and perfluoroalkyl sulfonamides in urban and Arctic air offers evidence of long-range atmospheric transport (Loewen et al. 2005; Shoeib et al. 2006; Stock et al. 2004). Photooxidation studies have demonstrated the conversion of these substances to perfluoroalkyl carboxylic acids and sulfonates. According to Young et al. (2007), the presence of perfluorodecanoic acid and perfluoroundecanoic acid in an Arctic ice cap indicates atmospheric oxidation of precursors as a source. Yeung et al. (2017) collected samples of snow and water from the Central Arctic region in 2012 and observed that perfluorinated alkyl substances were only detectable in ocean waters above a depth of 150 m. Atmospheric deposition from precursors was estimated to account for approximately 34–59% of the PFOA input to the ocean. Gawor et al. (2014) studied the distribution of fluorotelomer alcohols, fluorinated sulfonamides, and fluorinated sulfonamidoethanols in polar, remote, and urban regions of the world during four sampling campaigns covering the years 2005–2011. They observed higher levels of fluorotelomer alcohols as compared to the fluorinated sulfonamides and sulfonamidoethanols, with the 8:2 fluorotelomer alcohol being the predominant species in most samples.

Water. The pKa range of perfluoroalkyls indicates that these substances will exist primarily as the dissociated conjugate base (anion) when in contact with water at environmental pH (pH 5–9). Volatilization will not be an important environmental fate process when the substances exist as anions; however, under acidic conditions, undissociated perfluoroalkyls may volatilize into the atmosphere (Martin et al. 2006). Perfluoroalkyls may be transported to remote areas by direct oceanic advection of these substances (Armitage et al. 2006; Barber et al. 2007; Simcik 2005; Wania 2007; Yamashita et al. 2005, 2008). Perfluoroalkyls may also be transported over long distances in the form of marine aerosols (Barber et al. 2007; CEMN 2008; Prevedouros et al. 2006). This transport mechanism may be especially relevant since surfactants have been shown to accumulate in upper sea layers and at water surfaces (Prevedouros et al. 2006).

Perfluoroalkyls have been measured in invertebrates, fish, amphibians, reptiles, birds, bird eggs, and mammals located around the world (Brown et al. 2018; Dai et al. 2006; Giesy and Kannan 2001; Houde et al. 2005, 2006a, 2006b; Keller et al. 2005; Kannan et al. 2001a, 2001b, 2002a, 2002b, 2002c, 2002d, 2005, 2006; Sinclair et al. 2006; So et al. 2006a; Wang et al. 2008). The highest concentrations of PFOA

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and PFOS in animals are measured in apex predators, such as polar bears, which indicates that these substances biomagnify in food webs (de Vos et al. 2008; Houde et al. 2006b; Kannan et al. 2005; Kelly et al. 2007). Loi et al. (2011) also provided evidence of the biomagnification potential of several perfluoroalkyl substances in tropical food webs. Evidence for biomagnification of chlorinated polyfluoroalkyl ether sulfonic acids, which have been used in the plating industry, has recently been reported (Liu et al. 2017; Shi et al. 2015). Table 5-5 shows levels of PFOA and PFOS measured in Arctic organisms. The bioaccumulation potential of perfluoroalkyls increases with increasing chain length from 4 to 8 carbon units and then declines with further increases in chain length (Conder et al. 2008; de Vos et al. 2008; Furdui et al. 2007; Martin et al. 2004b). In living organisms, perfluoroalkyls bind to protein albumin in blood, liver, and eggs and do not accumulate in fat tissue, which may explain why bioconcentration factors (BCFs) are lower than expected in aquatic organisms (de Vos et al. 2008; Kissa 2001). The Arctic Monitoring and Assessment Programme December 2017 report contains a large summary of environmental monitoring data for perfluoroalkyl substances in the Arctic (AMAP 2017). Temporal trends indicated declining residues of PFOA in the liver of ringed seals at several Arctic monitoring sites; however, PFNA and PFDA showed increasing levels at some locations and declining levels at others (AMPA 2017). Declining levels of PFOA and PFOS were observed for polar bears in Canadian Arctic and Greenland; however, total perfluoroalkyl levels remained relatively constant as precursor substances appear to continue to be transported and degraded in the Arctic food web. Moreover, newer perfluorinated substances such as perfluoroethylcyclohexane sulfonate (used in hydraulic fluids) and perfluorobutane sulfonamide (a PFBS precursor) have been detected in some environmental media.

Table 5-5. Biological Monitoring of PFOA and PFOS in the Arctic

Location and organism	Concentration (ng/g)		Reference
	PFOA	PFOS	
Northeastern Canada, 1996–2002; wet weight ^a			Tomy et al. 2004
Zooplankton (n=5)	2.6	1.8	
Clams (n=5)	ND	0.28	
Shrimp (n=7)	0.17	0.35	
Arctic cod (n=6)	0.16	1.3	
Redfish (n=7)	1.2	1.4	
Walrus (n=5)	0.34	2.4	
Narwhal (n=5)	0.9	10.9	
Beluga (n=5)	1.6	12.6	
Black-legged kittiwake (n=4)	ND	10.0	
Glaucous gulls (n=5)	0.14	20.2	

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Table 5-5. Biological Monitoring of PFOA and PFOS in the Arctic

Location and organism	Concentration (ng/g)		Reference
	PFOA	PFOS	
Northern Canada, 1992–2002 ^a			Martin et al. 2004a
Polar bear (n=7)	8.6	3,100	
Arctic fox (n=10)	<2	250	
Ringed seal (n=9)	<2	16	
Mink (n=10)	<2	8.7	
Common loon (n=5)	<2	20	
Northern fulmar (n=5)	<2	1.3	
Black guillemot (n=5)	<2	ND	
White sucker (n=3)	<2	7.6	
Brook trout (n=2)	<2	39	
Lake whitefish (n=2)	<2	12	
Lake trout (n=1)	<2	31	
Northern pike (n=1)	<2	5.7	
Arctic sculpin (n=1)	<2	12	
Northwestern Canada, 2004			Powley et al. 2008
Zooplankton (n=3)	ND	ND–0.2	
Arctic cod (n=5)	ND	0.3–0.7	
Ringed seal (n=5)		2.5–8.6	
Bearded seal (n=1)	ND	1.3	
Northern Norway; ng/g wet weight ^a			Verreault et al. 2005, 2007
Herring gull eggs	<0.091–0.652	21.4–42.2	
Glaucous gulls			
Eggs (n=10)	<0.70	104	
Plasma (n=20)	<0.70–0.74	134	
Nanavut, Canada			Butt et al. 2007a, 2007b
Thick-billed murres	<MDL ^b –0.16	<0.40–0.76	
Northern fulmars	<MDL ^b –0.09	<0.40–0.60	
Ringed seals	<0.85–6.2	2–20	
Northern Canada, 2002–2005			Butt et al. 2008
Ringed seal livers (n=110)	<0.7–13.9	0.89–189	
Greenland			Bossi et al. 2005
Ringed seals	<1.2	12.5–95.6	
North American and European Arctic, 1999–2002			Smithwick et al. 2005a
Polar bears (n>72)	<2.3–57.1	263–6,340	
Greenland, 1999–2001			Smithwick et al. 2005b
Polar bears (n=29) ^a	10	2,470	
Greenland, 1972–2002			Smithwick et al. 2006
Polar bears	1.6–4.4	120–1,400	
Arviat, 2010–2012			AMAP 2017
Ringed seal liver (n=28)	0.62±0.6	No data	

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Table 5-5. Biological Monitoring of PFOA and PFOS in the Arctic

Location and organism	Concentration (ng/g)		Reference
	PFOA	PFOS	
Pangnirtung, 2009–2011			AMAP 2017
Ringed seal liver (n=7)	0.22±0.15	No data	
Resolute, 2010–2013			AMAP 2017
Ringed seal liver (n=46)	0.23±0.16	No data	
Sachs Harbor, 2011–2013			AMAP 2017
Ringed seal liver (n=29)	0.54±0.36	No data	
Ulukhaktok, 2010–2013			AMAP 2017
Ringed seal liver (n=9)	1.4±0.11	No data	

^aReported as mean values

^bMinimum detection limits for study analytes ranged from 0.03 to 2.3 ng/g. To calculate means, concentrations less than the MDL were replaced with a random value that was less than half the MDL.

MDL = minimum detection limit; ND = not detected; PFOA = perfluorooctanoic acid; PFOS = perfluorooctane sulfonic acid

Sediment and Soil. K_{oc} values of 17–230 measured for PFOA in soils of various organic carbon content indicate that PFOA will be mobile in soil and will not adsorb to suspended solids and sediment in the water column (Davis et al. 2007; Prevedouros et al. 2006); rather, it tends to remain in solution and migrate with groundwater (Davis et al. 2007). This is supported by the presence of PFOA in groundwater at the Decatur, Cottage Grove, and Washington Works fluorochemical industrial facilities (3M 2007b, 2008b; Davis et al. 2007). Other sources of perfluoroalkyls in soil include air emissions followed by atmospheric deposition to soils and subsequent leaching (Davis et al. 2007). Low volatility, high water solubility (9,500 mg/L at 25°C), and low sorption to solids indicate that the perfluorooctanoate anion will accumulate in surface waters, especially oceans (Armitage et al. 2006; Kauck and Diesslin 1951; Prevedouros et al. 2006; Wania 2007). McGuire et al. (2014) reported field-based K_{oc} values for several perfluoroalkyl substances (see Table 4-2).

Perfluoroalkyls can be taken up by plants in contaminated soils. Laboratory studies have suggested that short-chain perfluoroalkyls such as PFBA are more concentrated in edible portions of plants when compared to longer carbon chain substances such as PFOA or PFOS (Blaine et al. 2013, 2014a, 2014b; MDH 2014). Yoo et al. (2011) studied the accumulation of perfluoroalkyl carboxylic acids, perfluorosulfonic acids, and fluorotelomer alcohols in grass samples collected near Decatur, Alabama and calculated the grass-soil accumulation factor (GSAF), which is the concentration of perfluoroalkyl in grass divided by the concentration of perfluoroalkyl in soil. The shortest chain compounds had the largest GSAFs, and accumulation factors decreased rapidly with chain length. The mean (\pm SD) GSAF values

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were 3.4 ± 2.6 , 0.90 ± 0.66 , 0.25 ± 0.23 , 0.12 ± 0.08 , 0.10 ± 0.08 , 0.11 ± 0.09 , and 0.10 ± 0.09 for PFHxA, PFHpA, PFOA, PFNA, PFDA, PFUnA, and PFDoDA, respectively. The GSAF for PFOS was 0.07 ± 0.04 . Increasing salinity and temperature was shown to increase uptake and transport from the roots into the shoots by the wheat plants grown in hydroponic systems spiked with perfluorocarboxylic acids (Zhao et al. 2016). Transport into the shoots also increased with decreasing carbon chain length. Concentrations in the shoots of the wheat plants increased in the following order: PFBA > PFHpA > PFOA > PFDoDA. Stahl et al. (2009) demonstrated that both PFOA and PFOS were taken up from soil by five cultivated plants (spring wheat, oats, potatoes, maize, and perennial ryegrass) and the amount of uptake was generally linearly dependent upon the concentration of PFOA or PFOS spiked to the soil. Lysimeter studies suggested that PFOA and short-chain perfluoroalkyls such as PFBA pass through the soil and get taken up by plants more quickly as compared to PFOS (Stahl et al. 2013).

Krippner et al. (2014) analyzed the uptake of perfluoroalkyl substances with chain length of C4–C10 over a pH range of 5–7. They observed that short-chain perfluoroalkyls partition predominantly and at higher concentrations to the shoot. Longer-chain substances such as PFOA, PFNA, and PFDA, as well as PFHxS and PFOS, accumulated at higher concentrations in the roots of maize plants. Maize grown in soil pots containing perfluoroalkyls only accumulated compounds with a chain length $\leq C8$ in the kernels (Krippner et al. 2015).

Other Media. Data are not available regarding the transport and partitioning of perfluoroalkyls in other media.

5.4.2 Transformation and Degradation

Perfluoroalkyls are considered to be environmentally persistent chemicals (EPA 2008a; OECD 2002, 2007; Schultz et al. 2003). The carbon atoms of the perfluoroalkyl chain are protected from attack by the shielding effect of the fluorine atoms; furthermore, environmental degradation processes generally do not possess the energy needed to break apart the strong fluorine-carbon bonds (3M 2000; Hekster et al. 2003; Schultz et al. 2003). Perfluoroalkyls are resistant to biodegradation, direct photolysis, atmospheric photooxidation, and hydrolysis (OECD 2002, 2007; Prevedouros et al. 2006).

Air. Although transport and partitioning information indicates that air will not be a sink for perfluoroalkyls in the environment, low concentrations of perfluoroalkyl carboxylic acids, sulfonic acids, and sulfonamides have been measured in air both in the vapor phase and as bound to particulates (Barton

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et al. 2007; Kim and Kannan 2007). Available information indicates that photodegradation will not compete with wet deposition as an atmospheric removal process for perfluoroalkyls (Barton et al. 2007; Hurley et al. 2004; Prevedouros et al. 2006). However, photooxidation may be an important degradation mechanism for perfluoroalkyl sulfonamides (D'eon et al. 2006; Martin et al. 2006).

PFOA does not absorb UV light at environmentally relevant wavelengths (>290 nm); Hori et al. (2004a) reported a weak absorption band for PFOA that ranged from 220 to 270 nm. Based on the measured absorption wavelength of PFOA, perfluoroalkyl carboxylic acids are not expected to undergo direct photolysis. Following irradiation of the potassium salt of PFOS with light of wavelength 290–800 nm for 67–167 hours, it was concluded that there was no evidence of direct photolysis of PFOS under any of the test conditions (OECD 2002). Based on these test results for PFOS, perfluoroalkyl sulfonic acids are not expected to undergo direct photolysis in the atmosphere.

A measured photooxidation rate constant is not available for PFOA. Hurley et al. (2004) measured the reaction of short-chain (C1–C4) perfluoroalkyl carboxylic acids with photochemically generated hydroxyl radicals. The proposed mechanism begins with abstraction of the carboxyl hydrogen, which is followed by the removal of the carboxyl group and generation of a perfluoroalkyl radical. Finally, the perfluoroalkyl chain is broken down one carbon atom at a time through an unzipping sequence. The same rate constant, 1.69×10^{-13} cm³/molecule-second, was measured for the photooxidation of the C2, C3, and C4 molecules, indicating that the chain length may have little effect on the reactivity of perfluoroalkyls with hydroxyl radical. According to the authors, this rate constant corresponds to a half-life of 130 days. Based on the data for the short-chain structures, the authors concluded that atmospheric photooxidation of perfluoroalkyl carboxylic acids is not expected to compete with wet and dry deposition, which is predicted to occur on a time scale of the order of 10 days.

Atmospheric photooxidation data are not available for perfluoroalkyl sulfonic acids. Atmospheric photooxidation studies involving n-methyl perfluorobutane sulfonamidoethanol (Me-FBSE) and n-ethyl perfluorobutanesulfonamide (Et-FBSA) indicate possible mechanisms for the reaction of these substances with atmospheric hydroxyl radicals (D'eon et al. 2006; Martin et al. 2006). Products observed from the photooxidation of these compounds indicate the following pathways: removal of an alkyl from the amide (cleavage of the N-C bond); removal of the amido group (cleavage of the S-N bond); and removal of the sulfonamido group (cleavage of the S-C bond) (D'eon et al. 2006; Martin et al. 2006). The last two pathways indicate that FOSA may be photooxidized through removal of the amido or sulfonamido group. The third pathway, cleavage of the S-C bond, also indicates a photooxidation mechanism for

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perfluoroalkyl sulfonic acids. Martin et al. (2006) proposes an unzipping sequence for the perfluoroalkyl chain following removal of the sulfonyl group.

Measured rate constants for the reaction of Me-FBSE and Et-FBSA with atmospheric hydroxyl radicals are 5.8×10^{-12} and 3.74×10^{-13} $\text{cm}^3/\text{molecule-second}$, respectively (D'eon et al. 2006; Martin et al. 2006). Atmospheric half-lives calculated using these rate constants were 2 days for Me-FBSE and 20–50 days for Et-FBSA.

Water. PFOS and PFOA are expected to be stable to hydrolysis in the environment based on half-lives of 41 and 92 years, respectively, calculated from experimental hydrolysis data that were measured at pH 5, 7, and 9 (OECD 2002, 2006b). Based on the data for PFOS and PFOA, hydrolysis is not expected to be an important degradation process for perfluorinated carboxylates and sulfonates in the environment. Hydrolysis data were not located for perfluoroalkyl sulfonamides.

Available information indicates that perfluoroalkyls are resistant to aerobic biodegradation. PFOA and PFNA were not biodegraded using an Organisation for Economic Co-operation and Development (OECD) guideline (301F) manometric respirometry screening test for ready biodegradability; 0% of the theoretical oxygen demand was reached after 28 days (Stasinakis et al. 2008). Meesters and Schröder (2004) reported that PFOA and PFOS were not degraded from an initial concentration of 5 mg/L in aerobic sewage sludge in a laboratory scale reactor.

Substances such as fluorotelomer alcohols and perfluoroalkyl sulfonamides are degraded to other substances such as PFOA and PFOS in water and can be considered a source of these substances in the environment (Liu et al. 2007).

Sediment and Soil. Data are not available regarding the transformation and degradation of perfluoroalkyls in sediment and soil. Based on the chemical stability of these substances and their resistance to biodegradation in screening tests, environmental degradation processes are not expected to be important removal mechanisms for perfluoroalkyls in sediment and soil (3M 2000; EPA 2008a; Hekster et al. 2003; OECD 2002, 2007; Prevedouros et al. 2006; Schultz et al. 2003).

Substances such as fluorotelomer alcohols and perfluoroalkyl sulfonamides are degraded to other substances such as PFOA and PFOS in soil and sediment and can be considered a source of these substances in the environment (Liu et al. 2007; Washington and Jenkins 2015; Washington et al. 2015).

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Other Media. Data are not available regarding the transformation and degradation of perfluoroalkyls in other media.

5.5 LEVELS IN THE ENVIRONMENT

Reliable evaluation of the potential for human exposure to perfluoroalkyls depends, in part, on the reliability of supporting analytical data from environmental samples and biological specimens.

Concentrations of perfluoroalkyls in unpolluted atmospheres and in pristine surface waters are often so low as to be near the limits of current analytical methods. In reviewing data on perfluoroalkyls levels monitored or estimated in the environment, it should also be noted that the amount of chemical identified analytically is not necessarily equivalent to the amount that is bioavailable.

Table 5-6 shows the limits of detection typically achieved by analytical analysis in environmental media. An overview summary of the range of concentrations detected in environmental media is presented in Table 5-7.

Detections of perfluoroalkyls in air, water, and soil at NPL sites are summarized in Table 5-8.

Table 5-6. Lowest Limit of Detection Based on Standards^a

Media	Detection limit	Reference
Air	0.1 pg/m ³	Harada et al. 2006
Drinking water	0.5–6.5 ng/L	EPA 2009c (Method 537)
Surface water and groundwater	0.2 ng/L	Nakayama et al. 2007
Soil	0.11–0.75 µg/kg (median reporting limits)	Anderson et al. 2016
Sediment	0.21–1.2 µg/kg (median reporting limits)	Anderson et al. 2016
Whole blood	0.1–2 ng/mL	Kärrman et al. 2005
Serum	0.082–0.2 ng/mL	CDC 2015

^aDetection limits based on using appropriate preparation and analytics. These limits may not be possible in all situations.

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Table 5-7. Summary of Environmental Levels of Perfluoroalkyls^a

Media	Low	High ^b	For more information
Outdoor air (pg/m ³)			Table 5-9
PFOA	1.22	900,000	
PFOS	0.46	9.8	
Indoor air (pg/m ³)			Table 5-10
PFOA		4.4 (mean)	
PFOS		<47.4 (mean)	
Dust (ng/g)			Table 5-11
PFOA	<2.29	9,818	
PFOS	<4.56	18,071	
Surface water (ppb)			Tables 5-12, 5-14, 5-17
PFOA	0.00051	598	
PFOS	<0.00008	8,970	
Groundwater (ppb)			Tables 5-14, 5-17
PFOA	0.083	619	
PFOS	0.0404	4,300	
Drinking water (ppb)			EPA 2010
PFOA	<0.0023	~100	
Ocean water (pg/L)			Table 5-15
PFOA	88	192,000	
PFOS	8	57,700	
Food (ppb)			Fromme et al. 2007b
PFOA	0.025	118.29	
PFOS	0.025	1.03	
Soil (ppb)			Tables 5-16, 5-17
PFOA	<0.00017	140	
PFOS	0.0002	9700	

^aFor PFOA or PFOS only.

^bHigh levels are representative of monitoring data at localized contaminated sites and are not reflective of background environmental levels.

Table 5-8. Perfluoroalkyls Levels in Water, Soil, and Air of National Priorities List (NPL) Sites

Medium	Median ^a	Geometric mean ^a	Geometric standard deviation ^a	Number of quantitative measurements	NPL sites
PFOA					
Water (ppb)	0.35	0.25	6,064	5	4
Soil (ppb)	18,050	18,050	1,000	2	2
Air (ppbv)	No data				
PFOS					
Water (ppb)	0.91	0.35	9,089	4	3
Soil (ppb)	108,000	108,000	1,000	2	2
Air (ppbv)	No data				

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Table 5-8. Perfluoroalkyls Levels in Water, Soil, and Air of National Priorities List (NPL) Sites

Medium	Median ^a	Geometric mean ^a	Geometric standard deviation ^a	Number of quantitative measurements	NPL sites
PFBA					
Water (ppb)	2.15	1.03	28,192	3	3
Soil (ppb)	1,600	1,600	1,000	2	2
Air (ppbv)	No data				
PFBS					
Water (ppb)	0.05	0.02	6,770	2	2
Soil (ppb)	224	224	1,000	2	2
Air (ppbv)	No data				
PFHpA					
Water (ppb)	0.07	0.04	3,169	3	2
Soil (ppb)	1,275	1,275	1,000	2	2
Air (ppbv)	No data				
PFHxA					
Water (ppb)	0.25	0.10	8,444	2	2
Soil (ppb)	1,175	1,175	1,000	2	2
Air (ppbv)	No data				
PFHxS					
Water (ppb)	0.26	1.12	52,496	4	3
Soil (ppb)	5,585	5,585	1,000	2	2
Air (ppbv)	No data				
PFNA					
Water (ppb)	No data				
Soil (ppb)	27.2	27.2	1,000	2	2
Air (ppbv)	No data				
PFPeA					
Water (ppb)	0.18	0.11	3,465	3	3
Soil (ppb)	178	178	1,000	2	2
Air (ppbv)	No data				

^aConcentrations found in ATSDR site documents from 1981 to 2017 for 1,854 NPL sites (ATSDR 2017). Maximum concentrations were abstracted for types of environmental media for which exposure is likely. Pathways do not necessarily involve exposure or levels of concern.

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5.5.1 Air

Perfluoroalkyl levels have been measured in outdoor air at locations in the United States, Europe, Japan, and over the Atlantic Ocean (Barber et al. 2007; Barton et al. 2006; Harada et al. 2005a, 2006; Kim and Kannan 2007). Concentrations reported in these studies are provided in Table 5-9.

Mean PFOA levels ranged from 1.54 to 15.2 pg/m³ in air samples collected in the urban locations in Albany, New York; Fukuchiyama, Japan; and Morioka, Japan and in the rural locations in Kjeller, Norway and Mace Head, Ireland. Higher mean concentrations (101–552 pg/m³) were measured at the urban locations in Oyamazaki, Japan and Manchester, United Kingdom, and semirural locations in Hazelrigg, United Kingdom. Maximum reported concentrations at Oyamazaki and Hazelrigg were 919 and 828 pg/m³, respectively. The authors attributed the elevated concentrations at the Hazelrigg location to emissions from a fluoropolymer production plant located 20 km upwind of this semirural community.

Elevated levels of PFOA were observed in air samples collected along the fence line of the DuPont Washington Works fluoropolymer manufacturing facility, which is located near Parkersburg, West Virginia, in the Ohio River valley (Barton et al. 2006).

Table 5-9. Concentrations of Perfluoroalkyl in Outdoor Air

Location	Mean (range) concentration (pg/m ³)			Reference
	PFOA	PFHpA	PFNA	
Urban				
Albany, New York				
Gas phase (n=8)	3.16 (1.89–6.53)	0.26 (0.13–0.42)	0.21 (0.16–0.31)	Kim and Kannan 2007
Particulate phase (n=8)	2.03 (0.76–4.19)	0.37 (<0.12–0.81)	0.13 (<0.12–0.40)	Kim and Kannan 2007
Oyamazaki, Japan (n=12)	262.7 (72–919); 3,412.8 ng/g in dust	—	—	Harada et al. 2005b
Fukuchiyama, Japan	15.2; 314 ng/g in dust	—	—	Harada et al. 2006
Morioka, Japan (n=8)	2.0 (1.59–2.58)	—	—	Harada et al. 2005b
Manchester, United Kingdom (n=2,1) ^a	341, 15.7	8.2, 0.2	<26.6, 0.8	Barber et al. 2007

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Table 5-9. Concentrations of Perfluoroalkyl in Outdoor Air

Rural				
Kjeller, Norway (n=2)	1.54	0.87	0.12	Barber et al. 2007
Mace Head, Ireland (n=4)	8.9	<0.001	<3.3	Barber et al. 2007
Hazelrigg, United Kingdom (semi-rural) (n=10)	101, 552 ^{b,c}	1.6, 14.4 ^b	0.9	Barber et al. 2007
Marine air				
Near Europe (northwest) (n=3)	1.22 (0.5–2.0)	<0.6 (ND–<0.6)	0.3 (ND–0.5)	Jahnke et al. 2007a
Near Africa (east coast) (n=5)	<0.5 (ND–0.7)	ND	<0.2 (ND–0.3)	Jahnke et al. 2007a
Source dominated				
DuPont Washington Works Facility; Parkersburg, West Virginia (n=28)	430,000 (75,000–900,000) ^d	—	—	Barton et al. 2006
DuPont Washington Works Facility; Parkersburg, West Virginia (n=90)	5,500 (10–75,900)	—	—	EPA 2007b
Mean (range) concentration (pg/m ³)				
Location	PFDA	PFUnA	PFDODA	Reference
Urban				
Albany, New York Gas phase (n=8)	0.63 (0.24–1.56)	<0.12 (ND–0.16)	0.27 (0.14–0.43)	Kim and Kannan 2007
Albany, New York Particulate phase (n=8)	0.27 (0.13–0.49)	ND	0.12 (<0.12–0.38)	Kim and Kannan 2007
Oyamazaki, Japan (n=12)	—	—	—	Harada et al. 2005b
Fukuchiyama, Japan	—	—	—	Harada et al. 2006
Morioka, Japan (n=8)	—	—	—	Harada et al. 2005b
Manchester, United Kingdom (n=2,1) ^a	5.4, <0.8	<0.01, <0.4	<0.01, <0.01	Barber et al. 2007
Rural				
Kjeller, Norway (n=2)	<0.15	<0.12	<0.12	Barber et al. 2007
Mace Head, Ireland (n=4)	<2.8	<0.002	<0.003	Barber et al. 2007
Hazelrigg, United Kingdom (semi-rural) (n=10)	1.0, 8.3 ^b	0.7	<0.01	Barber et al. 2007

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Table 5-9. Concentrations of Perfluoroalkyl in Outdoor Air

Table 5-9. Concentrations of Perfluoroalkyl in Outdoor Air					
Marine air					
Near Europe (northwest) (n=3)	<0.6 (ND–0.6)	ND		<0.14 (ND–0.17)	Jahnke et al. 2007a
Near Africa (east coast) (n=5)	ND	0.03 (ND–0.2)		ND	Jahnke et al. 2007a
Source dominated					
DuPont Washington Works Facility; Parkersburg, West Virginia (n=28)	—	—		—	Barton et al. 2006
DuPont Washington Works Facility; Parkersburg, West Virginia (n=90)	—	—		—	EPA 2007b
Location	Mean (range) concentration (pg/m ³)				Reference
	PFOS	PFBS	PFHxS	FOSA	
Urban					
Albany, New York					
Gas phase (n=8)	1.70 (0.94–3.0)	—	0.31 (0.13–0.44)	0.67 (0.22–2.26)	Kim and Kannan 2007
Particulate phase (n=8)	0.64 (0.35–1.16)	—	<0.12	0.29 (<0.12–0.79)	Kim and Kannan 2007
Oyamazaki, Japan (n=12)	5.2 (2.51–9.80); 72.2 ng/g in dust	—	—	—	Harada et al. 2005b
Fukuchiyama, Japan	2.2; 46.0 ng/g in dust	—	—	—	Harada et al. 2006
Morioka, Japan (n=8)	0.7 (0.46–1.19)	—	—	—	Harada et al. 2005b
Manchester, United Kingdom (n=2,1) ^a	46, 7.1	2.2, <1.6	1.0, 0.1	<1.6, <0.2	Barber et al. 2007
Rural					
Kjeller, Norway (n=2)	1.0	<0.09	0.05	0.78	Barber et al. 2007
Mace Head, Ireland (n=4)	<1.8	<1.0	0.07	<0.56	Barber et al. 2007
Hazelrigg, United Kingdom (semi-rural) (n=10)	1.6	2.6	0.04	0.2	Barber et al. 2007
Marine air					
Near Europe (north west) (n=3)	1.36 (0.4–2.5)	ND	0.12 (0.02–0.3)	ND	Jahnke et al. 2007a
Near Africa (east coast) (n=5)	0.544 (0.05–1.9)	ND	0.013 (ND–0.05)	ND	Jahnke et al. 2007a

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Table 5-9. Concentrations of Perfluoroalkyl in Outdoor Air

Source dominated					
DuPont Washington Works Facility; Parkersburg, West Virginia (n=28)	—	—	—	—	Barton et al. 2006
DuPont Washington Works Facility; Parkersburg, West Virginia (n=90)	—	—	—	—	EPA 2007b

^aMean values were reported for separate sampling sessions.

^bThe second concentration reported was measured during an earlier sampling session (n=2).

^cA maximum PFOA concentration of 828 pg/m³ was measured in air at Hazelrigg, United Kingdom.

^dAverage and range of concentrations in 6 out of 28 samples that contained PFOA.

“—” indicates no available data; FOSA = perfluorooctane sulfonamide; ND = not detected; PFBS = perfluorobutane sulfonic acid; PFDA = perfluorodecanoic acid; PFDoDA = perfluorododecanoic acid; PFHpA = perfluoroheptanoic acid; PFHxS = perfluorohexane sulfonic acid; PFNA = perfluorononanoic acid; PFOA = perfluorooctanoic acid; PFOS = perfluorooctane sulfonic acid; PFUnA = perfluoroundecanoic acid

The reported concentrations in these samples ranged from 75,000 to 900,000 pg/m³. The highest concentrations were measured at locations downwind of the facility. High volume air samples collected at several monitoring stations near the Washington Works facility contained PFOA at reported concentrations ranging from 10 to 75,900 pg/m³ (EPA 2007b). The mean and median of these reported concentrations are 5,500 and 240 pg/m³, respectively.

PFOS was detected above quantitation limits in most of the studies, but concentrations were generally below 5 pg/m³. A concentration of 46 pg/m³ was reported in samples from Manchester, United Kingdom. Reported concentrations of other perfluoroalkyls (PFHpA, PFNA, PFDA, PFUnA, PFDoDA, PFBS, PFHxS, and FOSA) were generally <1 pg/m³ in these studies. PFHpA was detected at slightly higher concentrations (8.2 and 14.4 pg/m³) at Manchester and Hazelrigg, United Kingdom, respectively.

Jahnke et al. (2007a) collected eight marine air samples during a cruise between Germany and South Africa (53°N to 33°S). Perfluoroalkyl concentrations steadily declined as the sampling moved further from Europe and toward less industrialized regions. Only PFOS was detected in the two samples collected over the Atlantic Ocean east of southern Africa.

Measurements of perfluoroalkyls in snow samples collected from Canadian Arctic ice caps suggest that volatile precursors, such as fluorinated telomer alcohols, may oxidize in the atmosphere at these locations (Young et al. 2007). Reported concentrations in these snow samples were 2.6–86 pg/L for PFOS, 12–147 pg/L for PFOA, 5.0–246 pg/L for PFNA, <8–22 pg/L for PFDA, and <6–27 pg/L for PFUnA.

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The concentration of PFOS measured in rainwater collected during a rain event in Winnipeg, Manitoba was 0.59 ng/L (Loewen et al. 2005). PFOA, PFNA, PFDA, PFUnA, and PFDoDA were not detected in the rainwater. Reported method detection limits for these compounds were 7.2, 3.7, 1.7, 1.2, and 1.1 ng/L, respectively.

Studies of perfluoroalkyl concentrations in indoor environments are available. The reported mean concentrations of perfluoroalkyls measured in four indoor air samples collected from Tromso, Norway (Barber et al. 2007) are presented in Table 5-10.

Table 5-10. Concentrations of Perfluoroalkyl in Indoor Air^a

	Mean concentration (pg/m ³)
PFOA	4.4
PFHpA	0.8
PFNA	2.7
PFDA	3.4
PFUnA	<1.3
PFDoDA	1.2
PFOS	<47.4
PFBS	<0.5
PFHxS	<4.1
FOSA	2.8

^aSamples (n=4) collected in Tromso, Norway in May–June 2005.

FOSA = perfluorooctane sulfonamide; PFBS = perfluorobutane sulfonic acid; PFDA = perfluorodecanoic acid; PFDoDA = perfluorododecanoic acid; PFHpA = perfluoroheptanoic acid; PFHxS = perfluorohexane sulfonic acid; PFNA = perfluorononanoic acid; PFOA = perfluorooctanoic acid; PFOS = perfluorooctane sulfonic acid; PFUnA = perfluoroundecanoic acid

Source: Barber et al. 2007

Several studies have measured perfluoroalkyl levels in indoor dust samples (Table 5-11). Kubwabo et al. (2005) measured the concentrations of selected perfluoroalkyls in dust samples from 67 Canadian homes. PFOA, PFOS, and PFHxS were each detected in 37, 33, and 15% of these samples, respectively (detection limits of 2.29, 4.56, and 4.56 ng/g, respectively). FOSA was only detected above 0.99 ng/g in 10% of the samples and PFBS was not detected in any of the samples. Moriwaki et al. (2003) measured PFOS and PFOA concentrations in vacuum cleaner dust samples collected from 16 Japanese homes. PFOS and PFOA were detected in every sample with reported concentrations of 11–140 and 69–380 ng/g, respectively, in 15 of the 16 samples. One of the samples contained 2,500 ng/g PFOS and 3,700 ng/g PFOA. The geometric means of PFOA and PFOS in pooled indoor air sample meta data analysis were

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reported as 37.34 and 38.91 ng/g, respectively (Mitro et al. 2016). Precursor substances present in dust can be biotransformed to perfluoroalkyl substances and may be a further source of human exposure. Fluorotelomer alcohols were detected in hotel dust in China at levels ranging from 24.8 to 678 ng/g (Yao et al. 2018). Makey et al. (2017) reported precursor substances such as dipolyfluoroalkyl phosphates (diPAPs), fluorotelomer alcohols, perfluorooctyl sulfonamides, and sulfonamidoethanols in airborne and dust samples.

Table 5-11. Concentrations of Perfluoroalkyls in Indoor Dust

Location	Concentration (ng/g): mean (range); median			Reference	
	PFOA	PFHpA	PFNA		
Ottawa, Canada (n=67)	106.00 (<2.29–1,234); 19.72 ^a	—	—	Kubwabo et al. 2005	
Japan (n=16)	380 (70–3,700); 165	—	—	Moriwaki et al. 2003	
North Carolina and Ohio (n=112)	296 (<10.2–1,960); 142 ^b	109 (<12.5–1,150); 50.2 ^b	22.1 (<11.3–263); 7.99 ^b	Strynar and Lindstrom 2008	
United Kingdom, Australia, German, and United States (n=39) ^c	96.5 (9,818)	97.3 (5,195)	<LOQ (832)	Kato et al. 2009a	
Stockholm, Sweden ^d		—	—	Björklund et al. 2009	
Houses (n=10)	54 (15–98)				
Apartments (n=38)	93 (17–850)				
Daycare centers (n=10)	41 (31–110)				
Offices (n=10)	70 (14–510)				
Cars (n=5)	33 (12–96)				
Location	Concentration (ng/g): mean (range) median			Reference	
	PFDA	PFUnA	PFDODA		
Ottawa, Canada (n=67)	—	—	—	Kubwabo et al. 2005	
Japan (n=16)	—	—	—	Moriwaki et al. 2003	
North Carolina and Ohio (n=112)	15.5 (<9.40–267); 6.65 ^b	30.4 (<10.7–588); 7.57 ^b	18.0 (<11.0–520); 7.78 ^b	Strynar and Lindstrom 2008	
United Kingdom, Australia, German, and United States (n=39) ^c	<LOQ (1,965)	<LOQ (732)	<LOQ (1,048)	Kato et al. 2009a	
Stockholm, Sweden ^d	—	—	—		
Location	Concentration (ng/g): mean (range); median				Reference
	PFOS	PFBS	PFHxS	FOSA	
Ottawa, Canada (n=67)	443.68 (<4.56–5,065); 37.8 ^a	ND ^a	391.96 (<4.56–4,305); 23.1 ^a	<0.99 ^a	Kubwabo et al. 2005
Japan (n=16)	200 (11–2,500); 24.5	—	—	—	Moriwaki et al. 2003
North Carolina and Ohio (n=112)	761 (<8.93–12,100); 201 ^b	41.7 (<12.5–1,150); 9.11 ^b	874 (<12.9–35,700); 45.5 ^b	—	Strynar and Lindstrom 2008

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Table 5-11. Concentrations of Perfluoroalkyls in Indoor Dust

United Kingdom, Australia, German, and United States (n=39) ^c	479.6 (18,071)	359.0 (7,718)	185.5 (43,765)	<LOQ (18.4)	Kato et al. 2009a
Stockholm, Sweden ^d	—	—	—	—	Björklund et al. 2009
Houses (n=10)	39 (15–120)				
Apartments (n=38)	85 (8–1,100)				
Daycare centers (n=10)	31 (23–65)				
Offices (n=10)	110 (29–490)				
Cars (n=5)	12 (8–33)				

^aMethod detection limits (MDL) and percent below MDL are as follows: PFOA (2.29 ng/g, 37%), PFOS (4.56 ng/g, 33%), PFBS (1.38 ng/g, 100%), PFHxS (4.56, 15%), and FOSA (0.99 ng/g, 90%).

^bLOQ and percent above LOQ are as follows: PFHpA (12.5 ng/g, 74.1%), PFOA (10.2 ng/g, 96.4%), PFNA (11.3 ng/g, 42.9%), PFDA (9.40 ng/g, 30.4%), PFUnA (10.7 ng/g, 36.6%), PFDODA (11.0 ng/g, 18.7%), PFOS (8.93 ng/g, 94.6%), PFHxS (12.9 ng/g, 77.7%), PFBS (12.5 ng/g, 33.0%). Values below the LOQ were assigned a value of LOQ/1.412 when calculating the median and mean.

^cReported values are the 50th percentile and (maximum) values; all minimum values were <LOQ.

^dReported values are the median and range.

“—” indicates no available data; FOSA = perfluorooctane sulfonamide; LOQ = limit of quantification; ND = not detected; PFBS = perfluorobutane sulfonic acid; PFDA = perfluorodecanoic acid; PFDODA = perfluorododecanoic acid; PFHpA = perfluoroheptanoic acid; PFHxS = perfluorohexane sulfonic acid; PFNA = perfluorononanoic acid; PFOA = perfluorooctanoic acid; PFOS = perfluorooctane sulfonic acid; PFUnA = perfluoroundecanoic acid

Strynar and Lindstrom (2008) measured perfluoroalkyl levels in 112 indoor dust samples collected from homes and daycare centers in North Carolina and Ohio. These authors detected PFHpA, PFOA, PFNA, PFDA, PFUnA, PFDODA, PFOS, PFHxS, and PFBS. PFOS and PFOA were detected in 94.6 and 96.4% of the samples, respectively. Maximum detections in the samples were as high as 12,100 ng/g for PFOS and 35,700 ng/g for PFHxS. Household dust samples collected from the United Kingdom, Australia, Germany, and the United States showed the presence of perfluoroalkyls (Kato et al. 2009a). PFOS, PFBS, and PFHxS were detected in 74.4, 92.3, and 79.5% of the samples, respectively, whereas PFOA, PFNA, and PFDA were detected in 64.1, 25.6, and 38.5% of the samples, respectively. Björklund et al. (2009) measured PFOA and PFOS in dust samples collected in houses, apartments, daycare centers, offices, and cars in Sweden. PFOA and PFOS were detected in 100 and 79% of the apartment samples and in 100 and 60% of the car samples. The authors concluded that while dietary intake was the major PFOA/PFOS exposure pathway for adults and toddlers in the general population, dust ingestion could become an important pathway under a worst-case scenario (e.g., high dust ingestion and maximum dust levels).

Perfluoroalkyls in indoor and outdoor environments in urban, suburban, and rural locations near Toronto, Ontario, Canada were assessed using window films for passive sampling. The sum of perfluoroalkyls concentrations on outdoor window films ranged from 0.04 to 0.75 pg/cm² in winter and from 0.04 to

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0.92 pg/cm² in summer, with higher values found in urban and suburban locations than in rural locations. Indoors, concentrations on window film ranged from less than the detection limit (which ranged from 0.033 to 0.06 pg/cm²) to 2.1 pg/cm² in winter and from 0.08 to 4.3 pg/cm² in summer, although there were no distinct trends between urban and rural for indoor concentrations (Gewurtz et al. 2009).

5.5.2 Water

PFOS and PFOA have been widely detected in surface water samples collected from various rivers, lakes, and streams in the United States (Boulanger et al. 2004; DRBC 2013; Kannan et al. 2005; Kim and Kannan 2007; Nakayama et al. 2007; Simcik and Dorweiler 2005; Sinclair et al. 2004, 2006). Levels of these substances in surface water appear to be declining since the phase out of these two substances. Zhang et al. (2016) measured surface water levels of perfluoroalkyls in 2014 at 37 sites across the northeastern United States. Detectable levels of PFOA and PFNA were found at all sites, and PFHxS, PFOS, and PFDA were detectable at >90% of the sites. The respective maximum concentrations of PFOA, PFOS, PFHxS, and PFNA were 56 ng/L measured in the Passaic River, New Jersey, 27.5 ng/L measured in the Woonasquatucket River, Rhode Island, 43 ng/L measured at Mill Cove, Rhode Island, and 14 ng/L measured at Mill Cove, Rhode Island (Zhang et al. 2016).

Less data are available regarding the concentrations of other perfluoroalkyls in surface water. PFHpA and PFHxS were commonly detected in the few studies that analyzed surface water for these compounds (DRBC 2013; Kim and Kannan 2007; Nakayama et al. 2007; Simcik and Dorweiler 2005).

Concentrations of PFOA and PFOS measured in surface water are presented in Table 5-12 and other perfluoroalkyls are summarized in Table 5-13. Maximum concentrations of PFOS, PFOA, PFHpA, PFNA, PFDA, PFUnA, PFDoDA, PFBS, and PFHxS measured in surface water collected from the Cape Fear Basin, North Carolina were 287, 132, 329, 194, 120, 52.1, 4.46, 9.41, and 35.1 ng/L, respectively (Nakayama et al. 2007). Much higher concentrations of PFOS (198–1,090 ng/L) have been measured in Onondaga Lake in Syracuse, New York (Sinclair et al. 2006). Onondaga Lake is a Superfund site that has become contaminated through industrial activity along its banks.

Table 5-12. Concentrations of PFOA and PFOS in Surface Water (ng/L)

Location	Concentration		Reference
	PFOA	PFOS	
Great Lakes			Boulanger et al. 2004
Lake Ontario (n=8)	15–70	6–121	

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Table 5-12. Concentrations of PFOA and PFOS in Surface Water (ng/L)

Location	Concentration		Reference
	PFOA	PFOS	
Lake Erie (n=8)	21–47	11–39	
New York State waters			Sinclair et al. 2006
Lake Ontario (n=13)	18–34	2.9–30	
Niagara River (n=3)	18–22	3.3–6.7	
Lake Erie (n=3)	13–27	2.8–5.5	
Finger Lakes (n=13)	11–20	1.3–2.6	
Onondaga Lake (n=3)	39–64	198–1,090 (median=756)	
Oneida Lake (n=1)	19	3.5	
Erie Canal (n=3)	25–59	5.7–13	
Hudson River (n=8)	22–173 (median=35)	1.5–3.4	
Lake Champlain (n=4)	10–46	0.8–7.7	
Albany, New York			Kim and Kannan 2007
Lake water (n=11)	3.27–15.8 (median=7.20)	ND–9.30 (median=2.88)	
Surface water runoff (n=14)	0.51–29.3 (median=3.80)	<0.25–14.6 (median=0.81)	
New York/New Jersey/Rhode Island metropolitan region	56 (maximum) 3.5 (median)	27.5 (maximum) 0.96 (median)	Zhang et al. 2016
Michigan water regions			Sinclair et al. 2004
Detroit (n=10)	<8–16.14	<0.08–6.13	
Flint (n=4)	<8–23.01	1.50–12.31	
Saginaw Bay (n=5)	<8–24.08	3.10–12.69	
Northeastern Michigan (n=2)	<8	0.87–6.34	
Upper Peninsula (n=7)	<8–13.77	<0.8–3.09	
Northwestern Michigan (n=2)	11.96	<0.8–4.48	
Western Michigan (n=6)	<8–15.17	<0.8–5.32	
Southwestern Michigan (n=5)	8.74–35.86	7.22–29.26	
Lansing (n=3)	<8–13.37	1.04–4.96	
Minnesota Waters and Lake Michigan			Simcik and Dorweiler 2005
Remote (n=4)			
Loiten	0.7	ND	
Little Trout	0.3	1.2	
Nipisiquit	0.1	ND	
Tettegouche	0.5	0.2	
Urban (n=4)			
Calhoun	20	47	
Lake Harriet	3.5	21	
Lake of the Isles	0.5	2.4	

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Table 5-12. Concentrations of PFOA and PFOS in Surface Water (ng/L)

Location	Concentration		Reference
	PFOA	PFOS	
Minnesota River	1.2	9	
Lake Michigan (n=4)	<0.6–0.5	1–3.2	
Cape Fear Basin, North Carolina			Nakayama et al. 2007
80 Sites (n=100)			
Mean	43.4	31.2	
Median	12.6	28.9	
Minimum	ND	<1	
Maximum	287	132	
Percent not detected ^a	7.6	0	
Raisin and St. Clair Rivers, Michigan			Kannan et al. 2005
Raisin River	14.7	3.5	
St. Clair River (n=3)	4.0–5.0	1.9–3.9	
Conasauga River, Georgia	253–1,150	192–318	Konwick et al. 2008
Dalton, Georgia	49.9–299	15.8–120	Konwick et al. 2008
Tidal Delaware River (six locations)			DRBC 2013
2007	3.54–75.40	2.70–8.42	
2008	3.99–48	3.53–11.7	
2009	3.29–27.7	2.86–7.97	
Several rivers in Japan	0.1–67,000	0.3–59	Harada and Koizumi 2009
Lake Victoria Gulf, Kenya	0.4–96.4 (rivers) 0.4–11.6 (lakes)	<0.4–13.23 (rivers); <0.4–2.53 (lakes)	Orata et al. 2009
River Po, Italy	1–1,270	1–25	Loos et al. 2008

^aDetection limit is 0.05 ng/L

ND = not detected; PFOA = perfluorooctanoic acid; PFOS = perfluorooctane sulfonic acid

Table 5-13. Concentrations of Other Perfluoroalkyls in Surface Water

Location (reference) ^a	Concentration (ng/L)							
	PFHpA	PFNA	PFDA	PFUnA	PFDoDA	PFBS	PFHxS	FOSA
Great Lakes (Boulanger et al. 2004)								
Lake Ontario (n=8)	—	—	—	—	—	—	—	—
Lake Erie (n=8)	—	—	—	—	—	—	—	—
New York State waters (Sinclair et al. 2006)								
Onondaga Lake (n=3)	—	—	—	—	—	—	4.2–8.5	—

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Table 5-13. Concentrations of Other Perfluoroalkyls in Surface Water

Location (reference) ^a	Concentration (ng/L)							
	PFHpA	PFNA	PFDA	PFUnA	PFDoDA	PFBS	PFHxS	FOSA
Erie Canal (n=3)	—	—	—	—	—	—	2.5–5.6	—
Other lakes and rivers	—	—	—	—	—	—	0.5–2.8	—
Albany, New York (Kim and Kannan 2007)								
Lake water (n=11)	1.15–12.7	ND–3.51	0.25–3.58	ND–1.45	ND–<0.25	—	<0.25– 4.05	ND–0.47
Surface water runoff (n=14)	<0.25– 6.44	<0.25– 5.90	ND–8.39	ND–1.99	ND–1.60	—	ND–13.5	ND–2.14
New York/New Jersey/Rhode Island metropolitan region (Zhang et al. 2016)								
	48.2 (max); 0.9 (median)	14 (max); 0.4 (median)	5.8 (max); 0.2 (median)	1.9 (max); 0.1 (median)	2.6 (max); 0.0 (median)	6.2 (max); 0.4 (median)	43 (max); 0.7 (median)	—
Minnesota waters and Lake Michigan (Simcik and Dorweiler 2005)								
Remote (n=4)								
Loiten	10	ND	ND	—	—	—	—	—
Little Trout	4.8	ND	ND	—	—	—	—	—
Nipisiquit	0.9	<0.3	ND	—	—	—	—	—
Tettegouche	3.1	ND	ND	—	—	—	—	—
Urban (n=4)								
Calhoun	11	0.6	0.5	—	—	—	—	—
Lake Harriet	2.6	ND	ND	—	—	—	—	—
Lake of the Isles	0.4	ND	ND	—	—	—	—	—
Minnesota River	0.7	1.9	ND	—	—	—	—	—
Lake Michigan (n=4)	<0.6–4.1	<0.6–3.1	ND	—	—	—	—	—
Cape Fear Basin, North Carolina (Nakayama et al. 2007)								
80 Sites (n=100)								
Mean	38.7	33.6	22.1	10.4	2.17	2.58	7.29	—
Median	14.8	5.70	13.2	5.67	1.95	2.46	5.66	—
Maximum	329	194	120	52.1	4.46	9.41	35.1	—
Percent not detected ^b	32.9	10.1	15.2	17.7	53.2	38.0	45.6	—
Raisin and St. Clair Rivers, Michigan (Kannan et al. 2005)								
Raisin River	—	—	—	—	—	—	<1	<10
St. Clair River (n=3)	—	—	—	—	—	—	<1	<10

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Table 5-13. Concentrations of Other Perfluoroalkyls in Surface Water

Location (reference) ^a	Concentration (ng/L)							
	PFHpA	PFNA	PFDA	PFUnA	PFDoDA	PFBS	PFHxS	FOSA
Delaware River (6 locations) (DRBC 2013)								
2007	ND–24.30	1.71–976	ND-9.97	ND-26	ND	—	ND-4.48	ND
2008	2.03–16.3	3.24–650	ND-5.25	ND-11.8	ND	—	BD-3.55	ND
2009	1.14–47.4	1.65–546	ND-1.75	ND-8.3	ND	—	ND-3.62	ND

^aSee Table 5-12 for numbers of samples collected at these locations.

^bDetection limit = 0.05 ng/L.

“—” indicates no available data; ND = not detected; FOSA = perfluorooctane sulfonamide; PFBS = perfluorobutane sulfonic acid; PFDA = perfluorodecanoic acid; PFDoDA = perfluorododecanoic acid; PFHpA = perfluoroheptanoic acid; PFHxS = perfluorohexane sulfonic acid; PFNA = perfluorononanoic acid; PFUnA = perfluoroundecanoic acid

Higher levels of perfluoroalkyls are expected in surface water and groundwater surrounding perfluorochemical industrial facilities. Paustenbach et al. (2007) estimated PFOA concentrations in environmental media for communities located near the DuPont Washington Works chemical manufacturing facility. From this analysis, the authors concluded that much of the PFOA detected in groundwater near the facility was attributed to deposition to soil surfaces following atmospheric emissions from the plant followed by subsequent leaching into groundwater. DuPont entered into an agreement with the EPA to collect monitoring data for PFOA in the Ohio River around the DuPont Washington Works facility. In its final phase III assessment, it was reported that levels of PFOA downstream from the facility in 2011 were about an order of magnitude lower than when monitoring began in 2002, while levels upstream essentially remained unchanged (URS 2012). The maximum concentration measured in 2011 was 200 ng/L and was obtained from a monitoring location adjacent to the site.

PFOA, PFOS, PFBA, PFHxS, and PFBS have been detected in the municipal drinking water of communities located near the 3M Cottage Grove fluorochemical facility (ATSDR 2008). Xiao et al. (2015) summarized soil and groundwater monitoring data of select perfluoroalkyls near this facility. Groundwater samples for PFBA, PFHxA, PFOA, PFHxS, and PFOS in wells surrounding the site obtained from 2009 to 2013 showed little or no change in concentration over this time period, even though 3M had stopped producing perfluoroalkyls at this facility in 2002. Levels in groundwater were shown to decrease exponentially with distance from the source. A measured PFOA concentration of approximately 20,000 ng/L was observed from a well near the historical unlined disposal site of this

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facility, but levels decreased to <100 ng/L measured at a distance of 1.4 km away. Some other examples are provided in Table 5-14.

Table 5-14. Concentrations of Perfluoroalkyls in Surface Water and Groundwater at Fluorochemical Industrial Facilities

Location	Percent detection and concentration ($\mu\text{g/L}$)					Reference
	PFOA	PFBA	PFOS	PFHxS	PFBS	
DuPont Washington Works Facility, West Virginia						
<i>Groundwater</i>						
Borings (n=18)						Davis et al. 2007
Percent detected	89% ^a	—	—	—	—	
Minimum	0.0912 ^a	—	—	—	—	
Maximum	78 ^a	—	—	—	—	
Wells (n=14)						Davis et al. 2007
Percent detected	100% ^a	—	—	—	—	
Minimum	0.081 ^a	—	—	—	—	
Maximum	37.1 ^a	—	—	—	—	
Wells (n=3)						EPA 2008d
Percent detected	100%	—	—	—	—	
Minimum	2.8	—	—	—	—	
Maximum	100	—	—	—	—	
<i>Surface water</i>						
Outlets (n=4)						EPA 2008d
Percent detected	100%	—	—	—	—	
Minimum	2.3	—	—	—	—	
Maximum	61	—	—	—	—	
3M Cottage Grove Facility, Minnesota						
<i>Groundwater</i>						
Wells (n=1–7)						3M 2007b
Percent detected	100%	100%	100%	100%	100%	
Minimum	24.6	23.3	26.0	6.47	2.11	
Maximum	619	318	26.0	40.0	26.1	
<i>Surface water</i>						
East and West Cove (n=3–9)						3M 2007b
Percent detected	100%	100%	100%	100%	78%	
Minimum	0.172	0.803	0.227	0.0936	0.304	
Maximum	2.79	1.01	3.12	4.58	9.69	
Mississippi River Shoreline (n=52–80)						3M 2007b
Percent detected	60%	52%	43%	28%	56%	
Maximum	0.760	6.92	0.539	1.04	3.05	

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Table 5-14. Concentrations of Perfluoroalkyls in Surface Water and Groundwater at Fluorochemical Industrial Facilities

Location	Percent detection and concentration (µg/L)					Reference
	PFOA	PFBA	PFOS	PFHxS	PFBS	
Mississippi River Transect (n=34–44)						3M 2007b
Percent detected	14%	12%	0%	0%	0%	
Maximum	0.0501	0.0530	ND	ND	ND	
3M Decatur Facility, Alabama						
<i>Groundwater</i>						
Off-site groundwater (n=18)						3M 2008c
Percent detected	94%	—	—	—	—	
Mean	1.87	—	—	—	—	
Range	0.083–19.8	—	—	—	—	
Off-site groundwater						3M 2010
Range			0.0404– 2.41	0.0396– 0.622	0.0615– 0.480	
Groundwater range (n=51)	0.149–6.41	0.0104– 1.26	0.012– 0.151	0.0127– 0.0875	0.0101– 0.0766	Lindstrom et al. 2011
<i>Surface water</i>						
On-site surface water (n=7)						3M 2008c
Percent detected	100%	—	—	—	—	
Median	2.66	—	—	—	—	
Range	0.32–127	—	—	—	—	
Off-site surface water (n=60)						3M 2008c
Percent detected	98%	—	—	—	—	
Range	0.026–27.7	—	—	—	—	
Tennessee River						Hansen et al. 2002
Upstream of facility (n=19)						
Percent detected	0%	—	100%	—	—	
Range	<25	—	16.8–52.6	—	—	
Downstream of facility (n=21)						
Percent detected	0%	—	100%	—	—	
Median	355	—	107	—	—	
Range	<25–598	—	30.3–144	—	—	

^aAnalyte was reported as APFO.

“—” indicates no available data; APFO = ammonium perfluorooctanoate; ND = not detected; PFBA = perfluorobutanoic acid; PFBS = perfluorobutane sulfonic acid; PFHxS = perfluorohexane sulfonic acid; PFOA = perfluorooctanoic acid; PFOS = perfluorooctane sulfonic acid

Groundwater sampling of PFOA and PFOS was conducted at 401 military installations (DoD 2018). The PFOS/PFOA levels exceeded EPA’s lifetime health advisories of 70 ppt at 22% of the installations (9/64 Army, 40/127 Navy/U.S. Marine Corp, 39/203 Air Force, and 2/7 Defense Logistics Agency

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installations). The PFOS/PFOA levels exceeded the EPA lifetime health advisories in 1,621 of the 2,668 groundwater wells sampled (61%).

Yamashita et al. (2005) measured PFOA, PFOS, PFNA, and PFHxS concentrations in ocean water collected from locations in the Atlantic Ocean, Pacific Ocean, and areas near China, Korea, and Japan. These concentrations are listed in Table 5-15. Wei et al. (2007a) measured perfluoroalkyl concentrations in surface seawaters from the western Pacific Ocean, Indian Ocean, and near-Antarctic region. PFOS and PFOA were detected in 60 and 40% of the samples, respectively, with maximum concentrations of 71.7 and 441.6 pg/L, respectively. Concentrations of other perfluoroalkyls (PFHxS, PFBS, PFDoDA, PFDA, PFNA, PFHpA) were generally below detection in most samples, with the exceptions being in samples collected near Shanghai, the Philippines, and Indonesia. Maximum concentrations of these perfluoroalkyls ranged from 3.1 to 70.2 pg/L near Shanghai. PFOA, PFOS, and other perfluoroalkyl species were monitored in waste water effluents and 20 rivers located in Japan (Murakami et al. 2008). Perfluoroalkyls were ubiquitous in the river water samples, with concentrations of PFOA as large as 0.054–0.192 µg/L in seven of the river samples with low waste water effluent sources.

Table 5-15. Concentrations of PFOA and PFOS in Ocean Water^a

Location	Concentration (pg/L)			
	PFOA	PFOS	PFNA	PFHxS
North Atlantic (n=9)	160–338	8.6–36	15–36	4.1–6.1
Mid Atlantic (n=7)	100–439	37–73	—	2.6–12
Central to Eastern Pacific (n=14)	15–62	1.1–20	1.0–16	0.1–1.6
Western Pacific (n=2)	136–142	54–78	—	2.2– ^a 2.8
Tokyo Bay (n=8)	1,800–192,000	338–57,700	163–71,000	17–5,600
Offshore Japan (n=4)	137–1,060	40–75	—	3.0–6.1
Coastal Hong Kong (n=12)	673–5,450	70–2,600	22–207	<5–311
Coastal China (n=14)	243–15,300	23–9,680	2.0–692	<5–1,360
Coastal Korea (n=10)	239–11,350	39–2,530	15–518	<5–1,390
Sulu Sea (n=5)	88–510	<17–109	—	<0.2
South China Sea (n=2)	160–420	8–113	—	<0.2

^aIncludes samples of coastal and open ocean water.

“—” indicates no available data; PFHxS = perfluorohexane sulfonic acid; PFNA = perfluorononanoic acid; PFOA = perfluorooctanoic acid; PFOS = perfluorooctane sulfonic acid

Source: Yamashita et al. 2005

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The presence of PFOA and PFOS in surface water in the United States indicates that drinking water taken from these sources may contain detectable levels of these substances. The most vulnerable drinking water systems are those in close proximity to sites that are contaminated with perfluoroalkyls. PFOA was detected in 12 out of 13 samples collected from four municipal drinking water treatment plants that draw water from the Tennessee River and are located downstream from the 3M Decatur Facility in Alabama. Reported concentrations ranged from 0.025 to 0.16 µg/L (3M 2008c). PFOA was not detected in any samples collected from a fifth plant located upstream of the 3M Decatur facility (3M 2008c). Sampling conducted in October 2015 continued to show a presence of perfluoroalkyls in water samples collected at nine sites along the Tennessee River near Decatur, with maximum PFOS, PFBS, and PFOA levels reported as 0.220, 0.160, and 0.120 µg/L, respectively (Newton et al. 2017). Waterways adjacent to areas where aqueous film forming foams (AFFFs) used to fight fires have been shown to have high levels of perfluoroalkyls. For example, a PFOS concentration of 2,210 µg/L was detected in the Etobicoke Creek, Canada following the use of AFFF at the Toronto Pearson Airport (D'agostino and Mabury 2017). The vertical distribution of 15 perfluoroalkyl substances was studied in concrete samples obtained from a location in which AFFFs were used in firefighter training (Baduel et al. 2015). At the surface of the concrete pad, PFOS was observed to be the dominant substance measured; however, shorter-chain compounds were observed to a depth of 12 cm from the surface, suggesting vertical transport of the shorter-chain compounds and the potential for movement into groundwater.

Hu et al. (2016) presented geospatial monitoring data for six perfluoroalkyls (PFBS, PFHxS, PFHpA, PFOA, PFOS, and PFNA) in U.S. drinking water from information contained in the EPA third Unregulated Contaminant Monitoring Rule (UCMR 3) program. Frequency of detection and concentrations in drinking water was correlated with proximity to industrial facilities using perfluoroalkyls, military fire training areas, and the number of waste water treatment plants. Note that UCMR 3 required all large water systems (4,120 PWSs, serving >10,000 people) and a representative sample of 800 small water systems (serving ≤10,000 people) to monitor for PFOA and PFOS from 2013 through 2015.

It was reported that 66 public drinking water systems that serve 6 million U.S. residents had at least one sample that exceeded the current EPA health advisory level of 0.07 µg/L for PFOA and PFOS (Hu et al. 2016). The dataset used by Hu et al. (2016) has since been updated by the EPA (EPA 2017). The most recent report dated January 2017 showed that PFOS, PFOA, PFNA, PFHxS, PFHpA, and PFBS were detected above their respective minimal reporting level in 95, 117, 14, 55, 86, and 8 out of 4,920 public water systems (EPA 2017). PFOA and PFOS were identified above the health advisory level of

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0.07 µg/L in 13 and 46 out of the 4,920 public water supplies, respectively (EPA 2017). It is possible that combined concentrations of PFOA and PFOS may exceed the advisory level while individual concentrations may still be below 0.07 µg/L.

Based on a memorandum of understanding with the EPA, DuPont began collecting water monitoring data of both public and private wells near the Washington Works chemical plant. The quarterly reports and monitoring data affiliated with these reports may be obtained from the regulations.gov portal (<http://www.regulations.gov>). In samples of water collected at 17 public water facilities from 2002 to 2009 in West Virginia and Ohio, PFOA levels ranged from below the detection limit (0.0023 µg/L) to nearly 100 µg/L in a few test wells in Little Hocking, Ohio (EPA 2010). In the final phase III summary report, PFOA concentrations were reported to range from below the detection limit to 0.79 µg/L in 34 wells located approximately 3 miles upstream and 82 miles downstream from the facility (URS 2012).

Rumsby et al. (2009) reviewed the presence of PFOS and PFOA in drinking waters worldwide and discussed treatment methods for removing these substances from public water supplies. Conventional waste water treatment does not always efficiently remove perfluoroalkyls, and effluent may contain higher levels of some perfluoroalkyls than influent due to degradation of precursor substances during the treatment process (Schultz et al. 2006a, 2006b). While granulated activated carbon and reverse osmosis followed by nanofiltration have been shown to be effective methods of removing perfluoroalkyls, conventional methods such as chlorination, ozonolysis, and slow sand filtration may not be as effective. As a consequence, public drinking water systems impacted by effluent from waste water treatment plants often contain higher levels of perfluoroalkyls than systems that are not impacted by waste water treatment plant effluent. Quinones and Snyder (2009) analyzed raw and finished water at seven different public water systems in the United States for the presence of perfluoroalkyls. Water systems that were heavily impacted by waste water treatment plant effluents had greater frequency and higher levels of perfluoroalkyls when compared to water systems that were not highly impacted by waste water treatment plants. For example, no perfluoroalkyls were detected in either influent or finished water from a public water system in Aurora, Colorado with no impact from waste water treatment plant effluent; however, PFHxA, PFOA, PFNA, PFDA, PFUnA, PFHxS, and PFOS were detected in all samples of a Los Angeles, California public water system that was highly impacted by waste water effluent. Perfluoroalkyls were commonly detected in the influent and effluent of 10 waste water treatment plants across the United States (Schultz et al. 2006a). PFBS was detected in 100% of both influent and effluent samples of the 10 plants, while other perfluoroalkyls like PFOA, PFOS, PFNA, PFHxA, PFHpA, and PFHxS were detected in 80% of the influent and effluent samples at the 10 plants. In a national study of

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10 perfluoroalkyls in raw and treated drinking water of France, Boiteux et al. (2012) observed that several perfluoroalkyl carboxylic acids had greater concentrations in treated water than the raw water. In eight drinking water treatment plants, PFBA, PFHxA, and PFHpA were not detected in raw water, but were detected in treated water, indicating that these substances were released from saturated activated carbon used to treat raw waters or were formed by the degradation of precursor substances. Perfluoroalkyl sulfonates appeared to be removed more efficiently than the carboxylates. PFHxS, PFBS, and PFOS were detected less frequently in treated water as compared to raw water influent. These three compounds comprised 53% of the total concentration of perfluoroalkyls in the raw water samples, but only 37% of the total concentration of the perfluoroalkyls in the treated water. The summed concentration of 10 perfluoroalkyls was analyzed in the raw water and treated water of two drinking water treatment plants downstream from a fluoropolymer manufacturing facility located in France (Dauchy et al. 2012). The total concentration of perfluoroalkyls in the raw water at four sampling locations of the first plant ranged from 0.140 to 0.287 $\mu\text{g/L}$, while the summed concentration in the treated water was 0.179 $\mu\text{g/L}$. The total concentration of the 10 perfluoroalkyls in raw water at the second plant was 0.132 $\mu\text{g/L}$, while the total concentration in the treated water was 0.130 $\mu\text{g/L}$. Levels of PFHxA were greater in the treated water than the raw water at three of the four raw water sampling points, and levels of PFNA were greater in treated water than raw water at all sampling points of the first plant, but were slightly lower in the treated water of the second drinking water plant even though both systems used simple chlorination to treat the water.

PFOA was detected in 65% of the public drinking water systems tested in New Jersey in 2006 at concentrations ranging from 0.005 to 0.039 $\mu\text{g/L}$ (Post et al. 2009). In a follow-up study conducted in 2009, PFOA was detected in 57% of raw water samples from 29 additional public drinking water systems in New Jersey at a maximum concentration of 0.100 $\mu\text{g/L}$ (Post et al. 2013). Nine other perfluoroalkyls were also tested for, with PFOS and PFNA being the most frequently detected compounds (30% detection frequency each) after PFOA. PFOA and PFOS were detected in tap water from 21 cities located in China at concentrations of <0.0001–0.0459 and <0.0001–0.0148 $\mu\text{g/L}$, respectively (Jin et al. 2009). Mak et al. (2009) published a study comparing detections of perfluoroalkyls including PFOA and PFOS in tap water collected in China, Japan, India, Canada, and the United States. PFOA and PFOS were the predominant species measured, accounting for 40–50% of the total perfluoroalkyls present in water, with the exception of certain location of India where PFOS or PFOA may not have been present or were present at low levels.

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Gellrich et al. (2013) analyzed 119 samples of mineral water, 26 samples of tap water, 18 spring water samples, and 14 raw water samples from Germany for the presence of perfluoroalkyls. Perfluoroalkyls were detected in 58% of all of the samples tested, with the greatest summed total concentration observed in tap water at 0.0427 $\mu\text{g/L}$. The maximum concentration of individual perfluoroalkyls occurring in bottled water, spring water, untreated water, and tap water were observed for PFBS (0.0133 $\mu\text{g/L}$), PFOA (0.0074 $\mu\text{g/L}$), PFBS (0.010 $\mu\text{g/L}$), and PFHxS (0.0121 $\mu\text{g/L}$), respectively. Perfluoroalkyls were widely detected in drinking water samples collected in 2008 at 40 different locations of Catalonia, Spain (Ericson et al. 2009). Median concentrations ranged from 0.00002 $\mu\text{g/L}$ (FOSA) to 0.00098 $\mu\text{g/L}$ (PFOA). The most frequently detected compounds were PFOS and PFHxS, which were detected in 35 and 31 samples, respectively. PFOS, PFOA, and PFHxS were detected in all samples collected in a study of drinking water contamination of perfluoroalkyls in Rio de Janeiro, Brazil (Quinete et al. 2009). Concentration ranges were 0.00058–0.00670 $\mu\text{g/L}$ (PFOS), 0.00035–0.00282 $\mu\text{g/L}$ (PFOA), and 0.00015–0.001 $\mu\text{g/L}$ (PFHxS) respectively.

Perfluoropolyethers (PFPEs) such as perfluoroether carboxylic and sulfonic acids containing one or more ether oxygens in the carbon backbone are expected to be less persistent than the legacy substances. However, hexafluoropropylene oxide trimer and dimer acid (HFPO-TA and HFPO-DA) were detected in downstream water samples near a fluoropolymer facility in China at levels of 5.2–68.5 $\mu\text{g/L}$ (Pan et al. 2017). Pan et al. (2018) also provided data of HFPO-DA and HFPO-TA in surface waters in China, United Kingdom, United States, Sweden, Germany, Netherlands, and South Korea. The substances were frequently detected in surface waters in all countries with median levels of 0.00095 $\mu\text{g/L}$ (HFPO-DA) and 0.00021 $\mu\text{g/L}$ (HFPO-TA). Gebbink et al. (2017) also reported that the GenX (HFPO-DA) was detected in all sampling sites downstream from a fluoropolymer facility in the Netherlands and at three out of four drinking water facilities located near the facility. Short-chain perfluoroalkyl compounds and legacy perfluoroalkyls were analyzed for in 97 drinking water samples from Canada and other nations in 2015–2016 (Kabore et al. 2018). PFOA and PFOS levels did not exceed 0.005 $\mu\text{g/L}$ in any of the 97 samples; however, high detection frequencies ranging from 64 to 92% were observed in tap water for some short-chain perfluoroalkyls.

5.5.3 Sediment and Soil

Concentrations of perfluoroalkyls in soils are expected to be greater in the vicinity of fluorochemical plants that produced or used these substances as processing aids in the manufacture of fluoropolymers than in the environment at large. Levels of some perfluoroalkyls measured in soil and sediment

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surrounding perfluorochemical industrial facilities are listed in Table 5-16. PFOA was detected in most soil and sediment samples collected on- and off-site at the 3M Decatur facility in Alabama in monitoring studies conducted between October 2004 and December 2006. Maximum soil concentrations were as high as 14,750 ng/g on-site and 7.85 ng/g off-site, and maximum sediment concentrations were as high as 347 ng/g on-site and 2,385 ng/g off-site (3M 2008c). The highest levels of PFOA were measured in soil from on-site fields formerly amended with PFOA-containing sludge. In its final project report for this location, six on-site soil samples were analyzed in December 2012 for the presence of PFOA, PFOS, PFBS, and PFHxS. Average levels were 3.86–3,890 ng/g (PFOS), 3.56–270 ng/g (PFHxS), 0.423–64.8 ng/g (PFBS), and 17.0–1,410 ng/g (PFOA) (3M 2012).

Table 5-16. Concentrations of Perfluoroalkyls in Soil and Sediment at Fluorochemical Industrial Facilities

Location	Percent detection and concentration (ng/g)					Reference
	PFOA	PFBA	PFOS	PFHxS	PFBS	
DuPont Washington Works Facility, West Virginia						
Soil						
Boring samples (n=22)						Davis et al. 2007
Percent detected	36% ^a	—	—	—	—	
Minimum	<0.17 ^a	—	—	—	—	
Maximum	170 ^a	—	—	—	—	
3M Cottage Grove Facility, Minnesota						
Soil						
Boring samples (n=50–108)						3M 2007b
Percent detected	100%	—	95%	90%	60%	
Maximum	21,800	—	104,000	3,470	139	
Fire training area (n=8–11)						3M 2007b
Percent detected	91%	82%	100%	100%	73%	
Maximum	262	11.5	2,948	62.2	24.6	
Sediment						
East and West Cove (n=21–28)						3M 2007b
Percent detected	100%	93%	100%	96%	65%	
Minimum	0.764	ND	40.0	ND	ND	
Maximum	1,845	94.6	65,450	126	9.14	
Mississippi River shoreline (n=84–92)						3M 2007b
Percent detected	70%	44%	80%	28%	29%	
Maximum	341	124	79.0	11.5	29.4	
Mississippi River transect (n=38–40)						3M 2007b
Percent detected	18%	0%	82%	0%	0%	
Maximum	1.09	ND	3.16	ND	ND	

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Table 5-16. Concentrations of Perfluoroalkyls in Soil and Sediment at Fluorochemical Industrial Facilities

Location	Percent detection and concentration (ng/g)					Reference
	PFOA	PFBA	PFOS	PFHxS	PFBS	
3M Decatur Facility, Alabama						
Soil						
On-site former sludge incorporation area (n=357)						3M 2008c
Percent detected	99%	—	—	—	—	
Mean	885–929					
Range	2.91–14,750	—	—	—	—	
On-site background (n=18)						3M 2008c
Percent detected	100%	—	—	—	—	
Mean	3.53–4.1					
Range	1.61–6.03	—	—	—	—	
Off-site soil (n=23)						3M 2008c
Percent detected	100%	—	—	—	—	
Mean	3.68–4.6					
Range	0.72–7.85	—	—	—	—	
Sediment						
On-site sediment (n=8)						3M 2008c
Percent detected	88%	—	—	—	—	
Median	16.8					
Range	1.64–347	—	—	—	—	
Off-site sediment (n=30)						3M 2008c
Percent detected	93%	—	—	—	—	
Range	0.39–2,385	—	—	—	—	
3M Decatur Facility, Alabama December 2012						
On-site former sludge incorporation area (n=6)						3M 2012
Percent detected	100%	—	100%	86%	86%	
Mean	17.0–1,410	—	3.86–3,890	3.56–270	0.423–64.8	
3M Cottage Grove Facility, Minnesota						
Surface soil along U.S. Highway 10 near facility						Xiao et al. 2015
Percent detected	100%	—	100%	—	—	
Median	8.0		12.2			
Range	5.5–125.7		0.2–28.2			

^aAnalyte was reported as APFO.

“—” indicates no available data; APFO = ammonium perfluorooctanoate; ND = not detected; PFBA = perfluorobutanoic acid; PFBS = perfluorobutane sulfonic acid; PFHxS = perfluorohexane sulfonic acid; PFOA = perfluorooctanoic acid; PFOS = perfluorooctane sulfonic acid

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PFOA, PFOS, and PFHxS were detected in 90–100% of soil samples collected from a former tar neutralization area, a former sludge disposal area, a former solids burn pit area, a former waste water treatment plant area, and a former fire training area at the 3M Cottage Grove facility in Minnesota (3M 2007b). PFBS was detected in 60–73% of these samples. Maximum concentrations for these substances were 21,800, 104,000, 3,470, and 139 ng/g, respectively. Levels of PFBA were only reported for soil in the fire training area; it was detected in 9 out of 11 samples from this location at 0.306–11.5 ng/g. The percent detection of these compounds in sediment from the East and West Cove sites was similar to that in soil. Maximum concentrations of PFOA and PFOS were 1,845 and 65,450 ng/g, respectively. These perfluoroalkyls were also analyzed in Mississippi River sediment near the Cottage Grove Facility. Levels of these compounds were much greater along the facility shoreline compared to levels in transect samples collected at points crossing the river. Maximum shoreline concentrations for PFOA, PFBA, PFOS, PFHxS, and PFBS were 341, 124, 79.0, 11.5, and 29.4 ng/g, respectively. PFHxS, PFBS, and PFBA were not detected in any of the transect samples, and PFOA was found in only 18%. Although the maximum concentration of PFOS was 3.16 ng/g, it was still detected in 82% of the transect samples.

PFOA and PFOS were detected in all surface soils (top 10 cm) samples collected at 28 sites in September and October of 2012 along U.S. Highway 10 running from Cottage Grove, Minnesota (where the former 3M perfluoroalkyl manufacturing facility was located) to Big Lake, Minnesota (Xiao et al. 2015). Measured levels of PFOS and PFOA ranged from 0.2 to 28.2 and from 5.5 to 125.7 ng/g, respectively. Subsurface soils up to a depth of 65 cm were collected at four sites as well. Levels of PFOA and PFOS generally increased with increasing depth at each of the locations, suggesting a downward movement of the contaminants and the potential to contaminate groundwater.

The use of aqueous firefighting foams at fire training areas of military installations has resulted in widespread contamination of perfluoroalkyls in the soil and groundwater at these facilities. Monitoring data obtained from 40 sites at 10 U.S. military installations in the continental United States and Alaska were collected for several perfluoroalkyls (Anderson et al. 2016). These data are summarized in Table 5-17.

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Table 5-17. Summary of Perfluoroalkyls Detected in Soil, Sediment, Surface Water, and Groundwater at 10 Military Installations^a

Compound	Parameter	Surface soil (µg/kg)	Subsurface soil (µg/kg)	Sediment (µg/kg)	Surface water (µg/L)	Groundwater (µg/L)
PFBA	DF	38.46	29.81	24.24	84.00	85.51
	Median	1.00	0.960	1.70	0.076	0.180
	Maximum	31.0	14.0	140	110	64.0
PFBS	DF	35.16	34.62	39.39	80.00	78.26
	Median	0.775	1.30	0.710	0.106	0.200
	Maximum	52.0	79.0	340	317	110
PFHxA	DF	70.33	65.38	63.64	96.00	94.20
	Median	1.75	1.04	1.70	0.320	0.820
	Maximum	51.0	140	710	292	120
PFHxS	DF	76.92	59.62	72.73	88.00	94.93
	Median	5.70	4.40	9.10	0.710	0.870
	Maximum	1,300	520	2,700	815	290
PFHpA	DF	59.34	45.19	48.48	84.00	85.51
	Median	0.705	0.660	1.07	0.099	0.235
	Maximum	11.4	17.0	130	57.0	75.0
PFOA	DF	79.12	48.08	66.67	88.00	89.86
	Median	1.45	1.55	2.45	0.382	0.405
	Maximum	58.0	140	950	210	250
FOSA	DF	64.84	29.81	75.76	52.00	48.55
	Median	1.20	0.470	1.30	0.014	0.032
	Maximum	620	160	380	15.0	12.0
PFOS	DF	98.90	78.85	93.94	96.00	84.06
	Median	52.5	11.5	31.0	2.17	4.22
	Maximum	9,700	1,700	190,000	8,970	4,300
PFNA	DF	71.43	14.42	12.12	36.00	46.38
	Median	1.30	1.50	1.10	0.096	0.105
	Maximum	23.0	6.49	59.0	10.0	3.00
PFDA	DF	67.03	12.50	48.48	52.00	34.78
	Median	0.980	1.40	1.90	0.067	0.023
	Maximum	15.0	9.40	59.0	3.20	1.80
PFUnA	DF	45.05	9.62	24.24	20.00	8.70
	Median	0.798	1.15	160	0.021	0.025
	Maximum	10.0	2.00	14.0	0.210	0.086

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Table 5-17. Summary of Perfluoroalkyls Detected in Soil, Sediment, Surface Water, and Groundwater at 10 Military Installations^a

Compound	Parameter	Surface soil (µg/kg)	Subsurface soil (µg/kg)	Sediment (µg/kg)	Surface water (µg/L)	Groundwater (µg/L)
PFDoDA	DF	21.98	6.73	45.45	20.00	4.35
	Median	1.95	2.40	2.80	0.058	0.022
	Maximum	18.0	5.10	84.0	0.071	0.062

^aWater concentrations are ppb (µg/L); soil and sediment levels are ppb (µg/kg).

DF = detection frequency as a percentage; FOSA = perfluorooctane sulfonamide; PFBA = perfluorobutanoic acid; PFBS = perfluorobutane sulfonic acid; PFDoDA = perfluorododecanoic acid; PFDA = perfluoro-n-decanoic acid; PFHpA = perfluoroheptanoic acid; PFHxA = perfluorohexanoic acid; PFHxS = perfluorohexane sulfonic acid; PFNA = perfluorononanoic acid; PFOS = perfluorooctane sulfonic acid; PFUnA = perfluoroundecanoic acid

Source: Anderson et al. (2016)

Perfluoroalkyls have been detected in soils that were amended with biosolids (Sepulvado et al. 2011). Several perfluoroalkyls were detected in biosolid-amended soils, with PFOS being the predominant compound with levels ranging from 5.5 to 483 ng/g, depending upon the loading rate.

5.5.4 Other Media

In a study conducted by the Food and Drug Administration (FDA) of 91 food samples collected during the 2017 Total Diet Study, PFOS was detected in 10 meat/seafood samples; the levels ranged from 0.134 ng/g in a boiled frankfurter to 0.865 ng/g in baked tilapia (FDA 2019). PFBA was detected in one sample of raw/frozen pineapple (0.068 ng/g). PFOA, PFHxS, PFNA, PFDA, PFBS, and PFHxA levels were below the lower limit of quantitation. It is important to note that FDA states that the sample size is limited and cannot be used to draw definitive conclusions. Levels of PFOS, PFOA, PFBS, PFHxS, PFHxA, PFHpA, PFDA, PFNA, and PFDoDA were analyzed in 31 food items collected from 5 grocery stores located in Texas in 2009 (Schechter et al. 2010). PFOA was the most frequently detected item (detected in 17 of 31 of the food samples), with levels ranging from 0.07 ng/g in potatoes to 1.80 ng/g in olive oil. PFOS, PFHxA, PFHpA, PFNA, PFDA, and PFDoDA were not detected in any samples. PFBS and PFHxS were detected in cod at 0.12 and 0.07 ng/g, respectively. The data for PFOA are summarized in Table 5-18. Several studies have evaluated the levels of perfluoroalkyls in fish from lakes and rivers in the United States; these data are summarized in Table 5-19.

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Table 5-18. Detections of PFOA in 31 U.S. Food Items

Food	PFOA concentration in ng/g (LOD for non-detects)
Hamburger	0.15
Bacon	0.24
Sliced turkey	ND (0.02)
Sausage	0.09
Ham	0.02
Sliced chicken breast	0.02
Roast beef	ND (0.02)
Canned chili	0.02
Salmon	0.23
Canned tuna	ND (0.05)
Fresh catfish fillet	0.30
Tilapia	0.10
Cod	0.10
Canned sardines	0.19
Frozen fish sticks	0.21
Butter	1.07
American cheese	ND (0.04)
Other cheese	ND (0.04)
Whole milk	ND (0.02)
Ice cream	ND (0.03)
Frozen yogurt	ND (0.02)
Whole milk yogurt	ND (0.02)
Cream cheese	ND (0.03)
Eggs	ND (0.04)
Olive oil	1.80
Canola oil	ND (0.05)
Margarine	0.19
Cereals	ND (0.04)
Apples	ND (0.02)
Potatoes	0.07
Peanut butter	0.10

LOD = limit of detection; ND = not detected; PFOA = perfluorooctanoic acid

Source: Schechter et al. 2010

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Table 5-19. Detections of Perfluoroalkyls in Fish from U.S. Lakes and Rivers

Type of seafood (location)	Perfluoroalkyl concentration in ng/g										Reference
	PFOA	PFOS	PFHxS	PFNA	PFDA	PFUnA	PFHpA	PFBS	PFDODA	FOSA	
Lake trout (Lake Superior)	1.1	4.8	<0.01– 0.43	1.0	0.72	0.90	<0.02– 0.87		0.37	0.25	Furdui et al. 2007
Lake trout (Lake Superior)	<0.42	2.3	<0.10	0.70	0.39	1.1			0.97		DeSilva et al. 2011
Lake trout (Lake Michigan)	4.4	16	<0.01– 0.87	0.57	0.76	0.74	<0.02– 0.97		0.41	0.99	Furdui et al. 2007
Lake trout (Lake Huron)	1.6	39	<0.01– 6.2	2.8	2.2	2.7	<0.02– 1.43		0.88	1.6	Furdui et al. 2007
Lake trout (Lake Huron)	<0.42	17	<0.10	1.4	1.3	1.8	NE		0.74		DeSilva et al. 2011
Lake trout (Lake Erie)	1.6	121	<0.01– 1.2	2.9	4.9	3.5	<0.02– 0.71		0.97	2.1	Furdui et al. 2007
Lake trout (Eastern Lake Erie)	<0.42	96	1.4	2.6	6.1	5.7	NE		2.0		DeSilva et al. 2011
Walleye (Western Lake Erie)	0.50	1.1	<0.10	1.2	3.6	3.1	NE		1.1		DeSilva et al. 2011
Lake trout (Lake Ontario)	1.5	46	0.65	1.1	1.8	1.6	<0.02– 1.39		0.70	0.82	Furdui et al. 2007
Lake trout (Lake Ontario)	0.88	2.5	0.70	0.90	1.4	2.	0.64		0.32		DeSilva et al. 2011
Mixture of whole fish (Missouri River)	<1.00	84.7	1.89	0.43	0.25	<1.00	1.53	<0.40	0.49		Ye et al. 2008
Mixture of whole fish (Mississippi River)	<0.20	83.1	0.42	0.78	1.24	3.38	0.27	<0.20	<0.40		Ye et al. 2008
Mixture of whole fish (Ohio River)	<1.00	147	0.52	1.03	3.88	6.57	<4.00	<0.40	1.72		Ye et al. 2008
Smallmouth bass (Raisin River)	<2	2.0–41.3	<1							<1–4.1	Kannan e al. 2005

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Table 5-19. Detections of Perfluoroalkyls in Fish from U.S. Lakes and Rivers

Type of seafood (location)	Perfluoroalkyl concentration in ng/g										Reference	
	PFOA	PFOS	PFHxS	PFNA	PFDA	PFUnA	PFHpA	PFBS	PFDODA	FOSA		
Smallmouth bass (St Clair River)	<2	<2–2.7	<1								1.1–6.3	Kannan et al. 2005
Smallmouth bass (Calumet River)	<2	2.5–7.6	<1								<1	Kannan et al. 2005
Carp ^a (Saginaw Bay)	<36	124	<34								<19	Kannan et al. 2005
Carp ^a (Saginaw Bay)		120										Giesy and Kannan 2001
Lake whitefish ^a (Michigan waters)		130										Giesy and Kannan 2001
Chinook salmon ^a (Michigan waters)		110										Giesy and Kannan 2001
Brown trout ^a (Michigan waters)		<6–46										Giesy and Kannan 2001
Bluegill (St Croix River, Minnesota)		2.87			<LOQ	<LOQ			<LOQ			Delinsky et al. 2009
Bluegill (Lake Calhoun, Minnesota)		272			5.82	4.18			4.72			Delinsky et al. 2009
Bluegill (Haw River, North Carolina)		29.8			10.3	26.9			7.25			Delinsky et al. 2009
Mixture of fish fillet (Zumbrol Lake, Minnesota)		52.4			3.24							Delinsky et al. 2010
Mixture of fish fillet (McCarrons Lake, Minnesota)		47.3			1.97							Delinsky et al. 2010

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Table 5-19. Detections of Perfluoroalkyls in Fish from U.S. Lakes and Rivers

Type of seafood (location)	Perfluoroalkyl concentration in ng/g										Reference
	PFOA	PFOS	PFHxS	PFNA	PFDA	PFUnA	PFHpA	PFBS	PFDODA	FOSA	
Mixture of fish fillet (Pickereel Lake, Minnesota)		10.0			1.23						Delinsky et al. 2010
Mixture of fish fillet (Carlos Lake, Minnesota)		12.3									Delinsky et al. 2010
Mixture of fish fillet (other Minnesota lakes)		1.08– 5.13									Delinsky et al. 2010
Mixture of fish fillet (Mississippi River sites, Minnesota)		3.06– 20.00	0.47		2.94–15.0	2.13– 6.72			3.74– 4.42		Delinsky et al. 2010

^aPerfluoroalkyl levels measured in muscle.

Grayed cells indicate that study did not evaluate compound; FOSA = perfluorooctane sulfonamide; LOQ = limit of quantification; NE = not evaluated; PFBS = perfluorobutane sulfonic acid; PFDODA = perfluorododecanoic acid; PFDA = perfluorodecanoic acid; PFHpA = perfluoroheptanoic acid; PFHxS = perfluorohexane sulfonic acid; PFNA = perfluorononanoic acid; PFOS = perfluorooctane sulfonic acid; PFUnA = perfluoroundecanoic acid

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Concentrations of perfluoroalkyls have been reported in foods sampled in Canada, the United Kingdom, and Germany (Food Standards Agency 2006; Fromme et al. 2007b; Tittlemier et al. 2007).

Perfluoroalkyls were detected in only 9 out of 54 food composites collected during Canadian Total Diet studies from 1992 to 2004 (Tittlemier et al. 2007). PFOS was detected in beef steak, ground beef, luncheon meats, marine fish, freshwater fish, and microwave popcorn at concentrations ranging from 0.98 to 2.7 ng/g, wet weight. PFOA was detected in roast beef, pizza, and microwave popcorn at 0.74–3.6 ng/g, wet weight. PFHpA was detected in pizza and microwave popcorn at 1.5–2.0 ng/g, wet weight. PFNA was detected only in beef steak at 4.5 ng/g, wet weight. PFDA, PFUnA, and PFDoDA were analyzed for but not detected in any of the food composites. During the U.K. Food Standards Agency Total Diet Study, PFOS was detected in eggs, sugars and preserves, potatoes, and canned vegetables at 1, 1, 10, and 2 µg/kg, respectively (Food Standards Agency 2006). PFOA was detected only in potatoes at 1 µg/kg. Neither substance was detected in the bread, miscellaneous cereals, carcass meats, offal, meat products, poultry, fish, oils and fats, green vegetables, other vegetables, fresh fruit, fruit products, beverages, milk, dairy products, or nuts categories. Fromme et al. (2007b) detected PFOS, PFOA, and PFHxS in 33, 45, and 3%, respectively, of 214 daily duplicate food portions for 31 adults in the city of Munich, Germany. Concentrations were 0.025–1.03 ng/g fresh weight for PFOS, 0.025–118.29 ng/g fresh weight for PFOA, and 0.05–3.03 ng/g fresh weight for PFHxS. Reported 90th percentile values were 0.11 and 0.21 ng/g fresh weight for PFOS and PFOA, respectively (Fromme et al. 2007b).

The temporal trend of perfluoroalkyl residues in eggs, milk, and farmed rainbow trout from Sweden were studied from 1999 to 2010 (Johansson et al. 2014). Over this period, the mean annual decreases in levels of PFOS were 18 and 31% in rainbow trout and eggs, respectively. The mean annual decreases of PFOA and PFHxS were 12 and 11%, respectively, in eggs. The detection frequency of PFOA and PFHxS was too low in milk samples to assess changes in levels over the time period, and decreases in the levels of PFOS were found to be not statistically significant over the temporal period. The mean annual decrease in levels of PFHxS in rainbow trout was 4.3% annually.

Elevated levels of PFOS were measured in water and fish samples obtained from 2009 and 2012 at six sampling locations along the Welland River and Lake Niapenco in Ontario, Canada (Gewurtz et al. 2014). These locations were downstream from the Hamilton International airport where PFOS containing AFFF was used until the mid-1990s at a firefighting training facility at the airport. PFOS concentrations were generally highest in benthic feeding fish collected at the sampling locations nearest to the airport. The maximum level of PFOS was observed in common carp collected at a site near the airport at a concentration of 2,300 ng/g. Maximum levels of PFOS in smallmouth bass, largemouth bass, and

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channel catfish were 640, 450, and 430 ng/g, respectively. Freshwater fish and seafood obtained from China had detectable levels of nine perfluoroalkyls, with PFOS being detected in 62% of the samples at a concentration range of <0.10–26.2 ng/g and PFOA detected in 70% of the samples at levels of <0.10–1.99 ng/g (Zhang et al. 2011).

PFOA was detected in the packaging paper of two microwave popcorn bags at 0.3–4.7 ng/cm² uncooked and 0.5–4.3 ng/cm² cooked (Sinclair et al. 2007). The mean mass of PFOA in the gas phase of popcorn vapors following popping was 16–17 ng/cm². PFHpA, PFNA, PFDA, PFUnA, and PFDoDA were detected in one of the bags at 0.4–3.2 ng/cm² uncooked and 0.5–4.3 ng/cm² cooked; however, these perfluoroalkyls were not detected (<0.2 ng/cm²) in the second bag. Begley et al. (2005) measured PFOA concentrations of 6–290 µg/kg in microwave popcorn bags. These authors also tested a hamburger wrapper, sandwich wrapper, French fry box, and soak-proof paper plates and did not find PFOA above the detection limit in these products. The concentration of PFOA measured in undiluted perfluoro paper coating formulations ranged from 88,000 to 160,000 µg/kg (Begley et al. 2005).

A study of perfluorinated and polyfluorinated substances in food packaging from U.S. fast food restaurants found perfluoroalkyl carboxylates (such as PFOA and PFHxA), perfluoroalkyl sulfonates (such as PFBS), fluorotelomer sulfonates, and unknown polyfluorinated compounds (Schaidler et al. 2017). PFOA was detected in 6 of the 20 samples collected in 2014–2015. Fluorotelomer alcohols that can degrade or metabolize to perfluoroalkyl carboxylic acids such as PFOA continue to be detected in food packaging materials. Yuan et al. (2016) analyzed 69 food contact materials (paper tableware, paper cups, cupcake cups, paper boxes, paper bags, and microwave popcorn bags) produced in China and 25 materials (paper tableware, microwave popcorn bags, and paper cups) produced in the United States. The median concentration of total fluorotelomer alcohols in food contact materials produced in China (sum of 6:2, 8:2, 10:2, 12:2, 14:2, 16:2, and 18:2 fluorotelomer alcohol) ranged from 2 to 18,200 ng/g, with the highest levels observed in microwave popcorn bags. The detection frequencies of 6:2, 8:2, 10:2, 12:2, 14:2, 16:2, and 18:2 fluorotelomer alcohol in all food contact materials were reported as 38, 65, 77, 70, 58, 35, and 30%, respectively. The only fluorotelomer detected in paper tableware produced in the United States was 6:2 fluorotelomer alcohol, with a detection frequency of 11%; however, all fluorotelomer alcohols, with the exception of 18:2 fluorotelomer alcohol, were detected in microwave popcorn bags from the United States, although the levels were much lower than those produced in China. Table 5-20 shows the median concentration of the fluorotelomer alcohols in microwave popcorn bags produced in the United States versus those produced in China. In contrast to the products produced in China, the predominant fluorotelomer alcohol currently detected in food contact materials produced in the

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United States appears to be 6:2 fluorotelomer alcohol. The authors concluded that the results of the PFOA Stewardship Program had effectively reduced the usage of long-chain fluorotelomer alcohols in the United States; however, they were still widely present in products produced in China.

Table 5-20. Fluorotelomer Alcohols Detected in Microwaveable Popcorn Bags Produced in China and the United States

Fluorotelomer alcohol	Median concentration of FTOH in microwave popcorn bag produced in China (ng/g)	Median concentration of FTOH in microwave popcorn bag produced in the United States (ng/g)
6:2 FTOH	80	485
8:2 FTOH	4,810	1.36
10:2 FTOH	6,700	0.73
12:2 FTOH	5,650	0.55
14:2 FTOH	384	0.12
16:2 FTOH	61	<MQL
18:2 FTOH	7.5	Not detected

FTOH = fluorotelomer alcohol; MQL = method quantitation limit

Source: Yuan et al. 2016

Washburn et al. (2005) measured the concentration of the perfluorooctanoate anion in fluorotelomer-treated consumer articles as well as the fluorotelomer formulations used for the treatments. PFOA was detected in mill-treated carpeting (0.2–0.6 mg/kg), carpet-care solution-treated carpeting (0.2–2 mg/kg), treated apparel (<0.02–1.4 mg/kg), treated home textiles (<0.02–1.4 mg/kg), industrial floor waxes and wax removers (0.0005–0.06 mg/kg), latex paint (0.02–0.08 mg/kg), and home and office cleaners (0.005–0.05 mg/kg). The concentrations of PFOA measured in the formulations used for these applications were 30–80, 1–50, <1–40, <1–40, 5–120, 50–150, and 50–150 mg/L, respectively. PFOA was not detected in treated upholstery (<0.034 mg/kg), treated technical textiles (<0.034 mg/kg), treated nonwoven medical garments (<0.034 mg/kg), or stone, tile, and wood sealants (<0.1 mg/kg).

Liu et al. (2014) measured levels of perfluoroalkyls in 35 consumer products that are typically used indoors, such as treated home textiles, food contact paper, carpet care products, and floor waxes. All products were obtained from retail stores in the United States between March 2007 and September 2011. The general trend was that these products contained decreasing quantities of perfluorocarboxylic acids, including PFOA, over the temporal period studied; however, there was an increase in use of PFBS, presumably as a replacement of PFOS in consumer products. Levels of perfluoroalkyls were analyzed in 115 random samples of consumer products obtained in Germany in 2010, including textiles, carpets,

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cleaning and impregnating agents, leathers, food contact materials, baking and sandwich paper, and ski waxes (Kotthoff et al. 2015). Cleaning agents and some baking and sandwich papers had nondetectable or negligible amounts of perfluoroalkyls; however, PFOA and PFOS were frequently detected in outdoor textiles, ski wax, leather products, gloves, awning cloths, nanosprays, impregnation sprays, and food contact materials. Cleaning agents, nanosprays, and impregnation sprays tended to have the highest level of fluorotelomer alcohols. EPA (2009b) analyzed 116 articles of commerce obtained from retail outlets in the United States from March 2007 and May 2008 for the presence of perfluorocarboxylic acids ranging in carbon number from C5 (perfluoropentanoic acid) to C12 (perfluorododecanoic acid). Total C5–C12 perfluorocarboxylic acid levels ranged from below the detection limit (1.5 ng/g) to 47,100 ng/g, with levels of PFOA ranging from below the detection limit to 6,750 ng/g.

PTFE is a fluoropolymer used in applications such as nonstick cookware coatings and plumbing sealant tape. In the past, PFOA had been used as a processing aid in the emulsion polymerization of PTFE (DuPont 2008). PFOA was largely removed from the fluoropolymer material during the baking and curing step of nonstick cookware coatings in a high temperature oven; however, residual PFOA could be found in the final coatings (DuPont 2008). Begley et al. (2005) measured PFOA concentrations of 4–75 µg/kg in PTFE cookware, 3 µg/kg in PTFE-based dental floss, 4 µg/kg in PTFE-based dental tape, and 1,800 µg/kg in PTFE film/sealant tape. PFOA was not detected in tubing made of a fluoro-ethylene-propene copolymer (Begley et al. 2005).

Studies have been conducted that investigated the release of PFOA from PTFE cookware when heated. Sinclair et al. (2007) reported PFOA release concentrations ranging from 19 to 287 pg/cm² measured using four new nonstick frying pans. These concentrations were measured at normal cooking temperatures within the range of 180–229°C. PFOA was detected in water (7 and 75 ng) boiled for 10 minutes in two out of five non-stick pans (Sinclair et al. 2007). PFOA was not found above the detection limit (0.1 ng/cm²) during 40 extraction tests on PTFE cookware using an ethanol/water mixture (Washburn et al. 2005). Likewise, Powley et al. (2005) conducted extraction tests on commercial fluoropolymer-treated cookware using water and water/ethanol mixtures at 100 and 125°C. Under simulated cooking conditions, PFOA was not identified above the detection limit of 100 pg/cm². Begley et al. (2005) reported that additional PFOA was not generated in the PTFE coating of three empty pans heated to 320°C (DuPont 2008). According to DuPont, the non-stick coating on a pan may begin to deteriorate if the pan is accidentally heated above 348°C, which is well above the maximum recommended cooking temperature of 260°C (DuPont 2008). Although it is possible for an unattended

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empty pan to reach these high temperatures, overheating non-stick cookware is expected to be prevented in most cases because food oils begin to generate smoke around 190°C (Begley et al. 2005).

A comprehensive study that examined 116 articles of commerce (AOC) found perfluorocarboxylic acids, including PFOA, in many commercially available substances, such as carpet care products and waxes (EPA 2009b). Levels of PFOA ranged from nondetectable to 6,750 ng/g, and levels of total perfluorocarboxylic acids (the sum of C5–C12 acids) ranged from nondetectable to 47,100 ng/g. Perfluoroalkyls, including PFOA, have been detected at low levels in personal care products such as cosmetics and sunscreens (Fujii et al. 2013).

5.6 GENERAL POPULATION EXPOSURE

Levels of perfluoroalkyls have been measured in indoor air, outdoor air, dust, food, surface water, and various consumer products. Possible exposure pathways have been proposed; however, the relative importance of these pathways, including their association with the accumulation of perfluoroalkyls in blood, remains unclear (Apelberg et al. 2007b; Begley et al. 2005; Calafat et al. 2006b; Trudel et al. 2008; Washburn et al. 2005). For populations that have elevated levels of perfluoroalkyls in water supplies, the primary route of exposure is expected to be ingestion of contaminated drinking water.

Trudel et al. (2008) provides a thorough analysis of general population exposure to PFOS and PFOA based on the available information and proposes the following possible exposure pathways: food and water consumption, ingestion of house dust, hand-to-mouth transfer from treated carpets, migration into food from PFOA-containing paper or cardboard, inhalation of indoor and ambient air, and inhalation of impregnation spray aerosols. Other pathways proposed to be less significant included oral exposure from hand-to-mouth contact with clothes and upholstery, migration into food prepared with PTFE-coated cookware, dermal exposure from wearing treated clothes, deposition of spray droplets on skin while using impregnation sprays, skin contact with treated carpet and upholstery, and deposition of dust onto skin (Trudel et al. 2008). The strong correlation between PFOA and PFOS concentrations in human serum samples indicates that common exposure pathways for these two substances are possible (Calafat et al. 2007a).

In order to estimate human uptake and the major pathways for human exposure to PFOS and PFOA, reported levels of these compounds in various environmental media, including food and consumer products, were analyzed with respect to product use patterns, personal activity patterns, and personal

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intake rates (Trudel et al. 2008). For PFOS, the major exposure pathways in a high-exposure scenario were proposed to be food and water ingestion, dust ingestion, and hand-to-mouth transfer from mill-treated carpets. Relative contributions of these pathways to the total uptake of PFOS in adults were estimated to be approximately 80, 15, and 5%, respectively (Trudel et al. 2008). For PFOA, the major exposure pathways in a high-exposure scenario were proposed to be oral exposure resulting from migration from paper packaging and wrapping into food, general food and water ingestion, inhalation from impregnated clothes, and dust ingestion. Relative contributions of these pathways to the total uptake of PFOA in adults were estimated to be approximately 60, 15, 15, and 10%, respectively (Trudel et al. 2008). Major exposure pathways for the intermediate and low exposure scenarios were proposed to be through food and drinking water (PFOA and PFOS) and ingestion of house dust (PFOA only). Based on these proposed exposure pathways, adult uptake doses estimated for low, medium, and high exposure scenarios were approximately 7, 15, and 30 ng/kg body weight/day, respectively, for PFOS and approximately 0.4, 2.5, and 41–47 ng/kg body weight/day, respectively, for PFOA (Trudel et al. 2008). The estimated uptake values were similar for men and women. Trudel et al. (2008) used older monitoring data and the estimated intakes may not be reflective of current exposure since there has been a downward trend in PFOA and PFOS exposure.

Fromme et al. (2009) assessed human exposure to perfluoroalkyls for adults in the general population of western countries. Based on measurements of indoor and outdoor air, house dust, drinking water, and dietary PFOS and PFOA levels, the investigators estimated average daily exposure levels of 1.6 ng/kg body weight/day for PFOS and 2.9 ng/kg body weight/day for PFOA. Upper daily exposure levels were determined to be 8.8 ng/kg body weight/day for PFOS and 12.6 ng/kg body weight/day for PFOA. The investigators concluded that the oral route, especially diet, was the primary route of exposure to perfluoroalkyls (Fromme et al. 2007a, 2007b, 2009).

As a group of compounds, perfluoroalkyls appear to be ubiquitous in human blood based on the widespread detection of these substances in human serum samples (Byrne et al. 2017; Calafat et al. 2006b, 2007a, 2007b; De Silva and Mabury 2006; Kuklennyik et al. 2004; Olsen et al. 2003b, 2003c, 2004b, 2004c, 2005, 2007a). Tables 5-21 and 5-22 list concentrations of perfluoroalkyls measured in serum samples collected from a representative sample of the general population in the United States. Most studies have reported that PFOA and PFOS levels have been detected in over 90% of subjects (Calafat et al. 2006b, 2007a, 2007b; Olsen et al. 2003b, 2007b, 2004c, 2005, 2008). PFHxS, PFNA, and PFDA are also typically detected in over 90% of the subjects (Calafat et al. 2006b, 2007a, 2017a, 2007b; Kuklennyik et al. 2004; Olsen et al. 2017a, 2017b).

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Table 5-21. Concentrations of PFOA and PFOS in Human Serum Collected in the United States

Location	Detection and concentration (ng/mL [ppb]) ^a		
	PFOA	PFOS	Reference
U.S. Residents—NHANES			
<i>1999–2000 (n=1,562)</i>			
Percent >LOD	100%	100%	Calafat et al. 2007a
LOD	0.1	0.2	
Geometric mean	5.2	30.4	
95th percentile	11.9	75.6	
<hr/>			
<i>2003–2004 (n=2,094)</i>			
Percent >LOD	99.7%	99.9%	Calafat et al. 2007b
LOD	0.1	0.4	
Geometric mean	3.95	20.7	
95th percentile	9.80	54.6	
<hr/>			
<i>2005–2006 (n=2,120)</i>			
Percent >LOD	NR	NR	CDC 2018
LOD	0.1	0.2	
Geometric mean	3.92	17.1	
95th percentile	11.3	47.5	
<hr/>			
<i>2007–2008 (n=2,100)</i>			
Percent >LOD	NR	NR	CDC 2018
LOD	0.1	0.2	
Geometric mean	4.12	13.2	
95th percentile	9.60	40.5	
<hr/>			
<i>2009–2010 (n=2,233)</i>			
Percent >LOD	NR	NR	CDC 2018
LOD	0.1	0.2	
Geometric mean	3.07	9.32	
95th percentile	7.50	32.0	
<hr/>			
<i>2011–2012 (n=1,904)</i>			
Percent >LOD	NR	NR	CDC 2018
LOD	0.1	0.2	
Geometric mean	2.08	6.31	
95th percentile	5.68	21.7	
<hr/>			
<i>2013–2014 (n=2,165)</i>			
Percent >LOD	NR	NR	CDC 2018
LOD	0.1	0.2	
Geometric mean	1.94	4.99	
95th percentile	5.57	18.5	
<hr/>			

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Table 5-21. Concentrations of PFOA and PFOS in Human Serum Collected in the United States

Location	Detection and concentration (ng/mL [ppb]) ^a		Reference
	PFOA	PFOS	
<i>2015–2016 (n=1,993)</i>			CDC 2019
Percent >LOD	NR	NR	
LOD	0.1	0.2	
Geometric mean	1.56	4.72	
95th percentile	4.17	18.3	
U.S. blood donors			
<i>2000–2001 (n=645)</i>			Olsen et al. 2003b
Percent >LLOQ ^b	92%	99.8%	
Geometric mean	4.6	34.9	
95th percentile ^c	12.1	88.5	
Maximum	52.3	1,656.0	
<i>2006 (n=600)</i>			Olsen et al. 2008, 2017b
Geometric mean	3.44	14.5	
95th percentile	7.9	31.5	
<i>2010 (n=600)</i>			Olsen et al. 2017b
Geometric mean	2.44	8.3	
95th percentile	5.6	21.8	
<i>2015 (n=616)</i>			Olsen et al. 2017b
Geometric mean	1.09	4.3	
95th percentile	3.2	8.6	
U.S. Regional			
Minneapolis-St. Paul blood donors (plasma)			
<i>2005 (n=40)</i>			Olsen et al. 2007b
Percent >LLOQ	95%	100%	
LLOQ	NR	3.4	
Geometric mean	2.2	15.1	
75th percentile	3.5	20.2	
Maximum	4.7	36.9	
Atlanta, Georgia			
<i>2003 (n=20)</i>			Kuklenyik et al. 2004
Percent >LOD	100%	100%	
LOD	0.1	0.4	
Mean	4.9	55.8	
Minimum	0.2	3.6	
Maximum	10.4	164.0	

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Table 5-21. Concentrations of PFOA and PFOS in Human Serum Collected in the United States

Location	Detection and concentration (ng/mL [ppb]) ^a		Reference
	PFOA	PFOS	
Seattle, Washington elderly individuals			
<i>No data reported (n=238)</i>			Olsen et al. 2004c
Percent >LLOQ ^b	99.2%	99.5%	
Geometric mean	4.2	31.0	
95th percentile ^b	9.7	84.1	
Maximum	16.7	175.0	
Washington County, Maryland			
<i>1974 (n=178)</i>			Olsen et al. 2005
Percent >LLOQ	71%	100%	
LLOQ	1.9	3.9	
Geometric mean	2.1	30.1	
75th percentile	3.0	40.2	
<i>1989 (n=178)</i>			Olsen et al. 2005
Percent >LLOQ	99% ^c	100% ^p	
Geometric mean	5.5	33.3	
LLOQ	1.9	3.9	
75th percentile	6.7	44.0	
Pease Tradeport, Portsmouth, New Hampshire			
<i>2015–2016 (n=1,578)</i>			NH HHS 2016
Percent >LLOQ	99.2	99.8	
LOD	0.1	0.1	
Geometric mean	8.59	3.09	
95th percentile	8.28–8.91	2.99–3.19	
St. Lawrence Island, Alaska			
<i>2013–2014 (n=85)</i>			Byrne et al. 2017
Percent >LOD	92	99	
50th percentile	1.01	4.55	
95th percentile	2.14	12.32	

^a"Less than" values indicate that the concentration was reported as below the LOD or LLOQ. For cases where samples had concentrations below the limit of detection or lower limit of quantification, a value between zero and the LOD or LLOQ was assigned when calculating the mean concentration.

^bExperimental LLOQs not determined.

^cReported as bias-corrected estimates.

^dLLOQ, LOQ, or LOD not reported.

CI = confidence interval; LLOQ = lower limit of quantification; LOD = limit of detection; LOQ = limit of quantification; NR = not reported; PFOA = perfluorooctanoic acid; PFOS = perfluorooctane sulfonic acid

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Table 5-22. Concentrations of Other Perfluoroalkyls in Human Serum Collected in the United States

Sample population	Detection and concentration (ng/mL [ppb]) ^a							
	PFHpA	PFNA	PFDA	PFUnA	PDoDA	PFBS	PFHxS	FOSA
U.S. Residents NHANES								
<i>1999–2000 (n=1,562) (Calafat et al. 2007a)</i>								
Percent >LOD	10%	95%	25%	12%	<1%		100%	100%
LOD	0.4	0.1	0.2	0.2	0.2		0.1	0.05
Geometric mean	<0.4	0.5	<0.2	<0.2	<0.2		2.1	0.4
95 th percentile	NR	1.7	0.5	NR	NR		8.7	1.4
<i>2003–2004 (n=2,094) (Calafat et al. 2007b)</i>								
Percent >LOD	6.2%	98.8%	31.3%	9.7%	<0.1%	<0.4%	98.3%	22.2%
LOD	0.3	0.1	0.3	0.3	1.0	0.4	0.3	0.2
Geometric mean	<0.3	1.0	<0.3	<0.3	<1.0	<0.4	1.9	<0.2
95 th percentile	0.4	3.2	0.8	0.6	<1.0	<0.4	8.3	0.2
<i>2005–2006 (n=2,120) (CDC 2018)</i>								
Percent >LOD	NR	NR	NR	NR	NR	NR	NR	NR
LOD	0.4	0.1	0.2	0.2	0.2	0.1	0.1	0.1
Geometric mean	—	1.09	0.355	—	—	—	1.67	—
95 th percentile	0.700	3.60	1.50	0.700	<LOD	0.1	8.30	0.300
<i>2007–2008 (n=2,100) (CDC 2018)</i>								
Percent >LOD	NR	NR	NR	NR	NR	NR	NR	NR
LOD	0.4	0.082	0.2	0.2	0.2	0.1	0.1	0.1
Geometric mean	—	1.22	0.286	—	—	—	1.95	—
95 th percentile	0.500	3.28	0.900	0.600	<LOD	<LOD	9.80	<LOD
<i>2009–2010 (n=2,233) (CDC 2018)</i>								
Percent >LOD	NR	NR	NR	NR	NR	NR	NR	NR
LOD	0.1	0.082	0.1	0.1	0.1	0.1	0.1	0.1
Geometric mean	—	1.26	0.279	0.172	—	—	1.66	—
95 th percentile	0.200	3.77	0.900	0.900	<LOD	<LOD	6.90	<LOD
<i>2011–2012 (n=1,904) (CDC 2018)</i>								
Percent >LOD	NR	NR	NR	NR	NR	NR	NR	NR
LOD	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1
Geometric mean	—	0.881	0.199	—	—	—	1.28	—
95 th percentile	0.220	2.54	0.690	0.620	0.140	<LOD	5.44	<LOD

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Table 5-22. Concentrations of Other Perfluoroalkyls in Human Serum Collected in the United States

Sample population	Detection and concentration (ng/mL [ppb]) ^a							
	PFHpA	PFNA	PFDA	PFUnA	PDoDA	PFBS	PFHxS	FOSA
<i>2013–2014 (n=2,168) (CDC 2018)</i>								
Percent >LOD	NR	NR	NR	NR	NR	NR	NR	NR
LOD	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1
Geometric mean	—	0.675	0.185	—	—	—	1.35	NR
95 th percentile	0.200	2.00	0.700	0.500	0.200	<LOD	5.60	NR
<i>2015–2016 (n=1,993) (CDC 2019)</i>								
Percent >LOD		NR	NR	NR	NR		NR	
LOD		0.1	0.1	0.1	0.1		0.1	
Geometric mean		0.577	0.154	—	—		1.18	
95 th percentile		1.90	0.700	0.400	<LOD		4.90	
U.S. blood donors								
<i>2000–2001 (n=645) (Olsen et al. 2003b, 2017a, 2017b)</i>								
Percent >LLOQ ^c	62%	100%	97.7%	93.8%	22.6%		52%	2%
Geometric mean	0.13	0.57	0.16	0.10	NR		1.9	NR
95 th percentile ^b	0.5	1.4	0.4	0.3	0.04		9.5	NR
<i>2006 (n=600) (Olsen et al. 2008, 2017a, 2017b)</i>								
Percent >LLOQ	62%	100%	99.8%	99.8%	68%	1.2%	95.7%	
LLOQ ^d	<0.5	NR	<0.05	<0.05	<0.05	<0.5	<0.5	
Geometric mean	0.09	0.97	0.34	0.18	0.04	LLOQ	1.52	
95 th percentile	0.4	2.2	0.8	0.5	0.07	LLOQ	5.7	
<i>2010 (n=600) (Olsen et al. 2017a, 2017b)</i>								
Percent >LLOQ	79.7%	100%	100%	99.8%	46.2%	22.5%	95.5%	
LLOQ ^d	<0.05	NR	NR	<0.025	<0.025	<1.0	<0.05	
Geometric mean	0.05	0.83	0.27	0.14	0.03	LLOQ	1.34	
95 th percentile	0.2	2.3	0.8	0.5	0.06	0.3	5.3	
<i>2015 (n=616) (Olsen et al. 2017a, 2017b)</i>								
Percent >LLOQ	3.3%	100%	96.9%	47.9%	0.6%	8.4%	99.7%	0%
LLOQ ^d	<0.09	<0.093	<0.093	<0.466	<0.932	<0.047	<0.04	<0.093
Geometric mean	NR	0.43	0.15	NR	NR	LLOQ	0.87	LLOQ
95 th percentile	0.16	1.1	0.49	0.25	LLOQ	0.02	3.5	LLOQ

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Table 5-22. Concentrations of Other Perfluoroalkyls in Human Serum Collected in the United States

Sample population	Detection and concentration (ng/mL [ppb]) ^a							
	PFHpA	PFNA	PFDA	PFUnA	PDoDA	PFBS	PFHxS	FOSA
Pease Tradeport, Portsmouth, New Hampshire (NH HHS 2016)								
2015–2016 (n=1,578)								
Percent >LOD	0.9	85.2	42.1	30.0	4.7	20.0	94.2	2.0
LOD	NA	0.1	0.1	0.1	NA	NA	0.1	0.1
Geometric mean	0.07	0.73	0.22	0.19	0.08	0.04	4.12	0.13
95 th percentile	0.07	0.70– 0.75	0.21– 0.23	0.18–0.19	0.08	0.04	3.92–4.33	0.12–0.14
St. Lawrence Island, Alaska (Byrne et al. 2017)								
2013–2014 (n=85)								
Percent >LOD		99	39	72			32	
50 th percentile		2.21	<LOD	0.72			<LOD	
95 th percentile		7.35	1.06	1.72			2.74	

^a"Less than" values indicate that the concentration was reported as below the LOD or LLOQ. For cases where samples had concentrations below the LOD or LLOQ, a value between zero and the LOD or LLOQ was assigned when calculating the mean concentration.

^bReported as bias-corrected estimates.

^cExperimental LLOQs not determined.

^dHighest LLOQ listed.

^eLOD not reported.

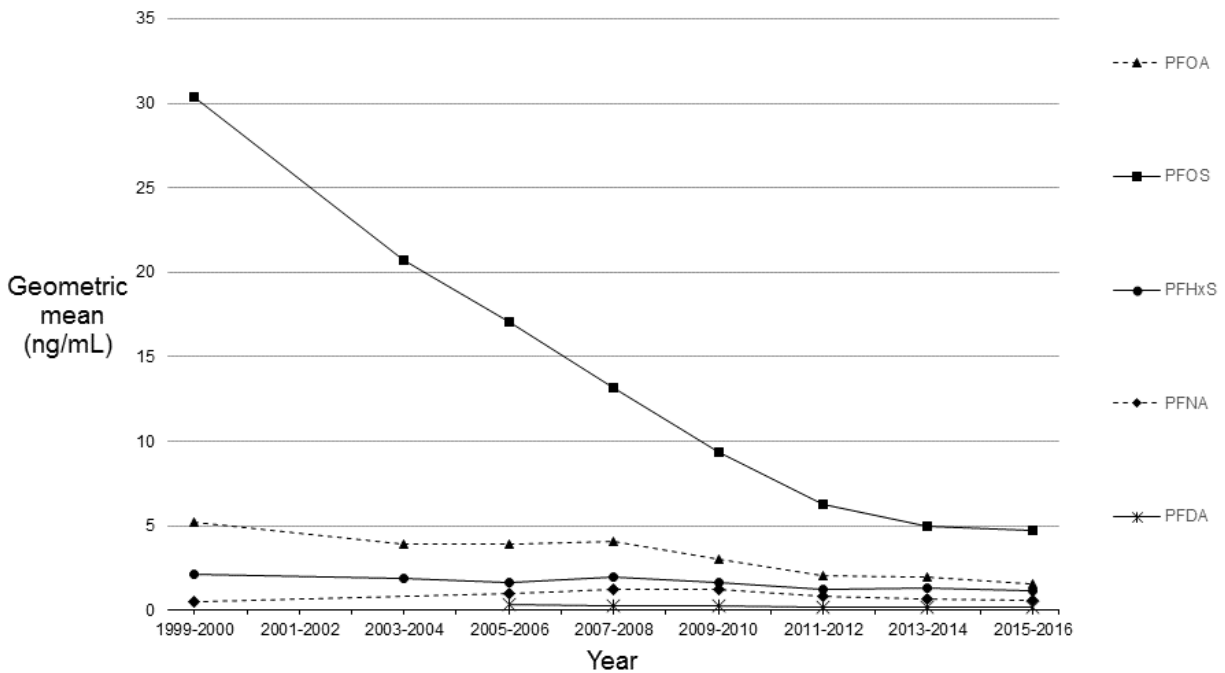
Grayed cells indicate that study did not evaluate compound; "—" indicates no available data;

FOSA = perfluorooctane sulfonamide; LLOQ = lower limit of quantification; LOD = limit of detection; NA = not applicable; NR = not reported; PFBS = perfluorobutane sulfonic acid; PFDA = perfluorodecanoic acid; PFDODA = perfluorododecanoic acid; PFHpA = perfluoroheptanoic acid; PFHxS = perfluorohexane sulfonic acid; PFNA = perfluorononanoic acid; PFUnA = perfluoroundecanoic acid

As illustrated in Figure 5-3, there has been a clear trend in decreasing serum levels of both PFOS and PFOA in the general population of the United States since 2000 as these substances were phased out; from 1999–2000 to 2015–2015 (CDC 2018, 2019), the geometric mean serum PFOS and PFOA levels in the general population have declined approximately 84 and 70%, respectively. Serum concentrations for PFHxS and PFDA (Figure 5-3) have also been declining over time, whereas serum PFNA concentrations increased from 1999–2000 to 2009–2010 and then started to decrease (Figure 5-3).

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Figure 5-3. Geometric Mean Concentrations of PFOA, PFOS, PFHxS, PFNA, and PFDA in U.S. Residents from 1999 to 2016



NHANES = National Health and Nutrition Examination Survey; PFDA = perfluorodecanoic acid; PFHxS = perfluorohexane sulfonic acid; PFNA = perfluorononanoic acid; PFOA = perfluorooctanoic acid; PFOS = perfluorooctane sulfonic acid

Source: CDC 2018

Serial studies of American Red Cross blood donors also allow for temporal evaluations of serum perfluoroalkyl levels. Olsen et al. (2008) reported a nearly 60% decline in PFOS blood levels when comparing data from 2001 to 2006; from 2006 to 2015, the serum PFOS levels dropped another 70% (Olsen et al. 2017b). From 2000 to 2015, the PFOS levels dropped 88%. Serum PFOA levels among the blood donors decreased 76% from 2000 to 2015 (Olsen et al. 2003b, 2017b). Geometric mean serum PFHxS levels went from 1.9 ng/mL in 2000 to 0.87 ng/mL in 2015, a 54% decrease (Olsen et al. 2003b, 2017a, 2017b). From 2006 to 2015, both serum PFNA and PFDA levels decreased by 56% (Olsen et al. 2017a, 2017b).

Several studies have evaluated sex- and age-related differences in serum perfluoroalkyl levels in the general population. In an analysis of NHANES data from 1999 to 2008, Kato et al. (2011) found that males had significantly higher levels of PFOA, PFOS, and PFNA than females and that PFOS levels increased with age, especially in females. Fu et al. (2014b) analyzed the effects of sex and age on levels of perfluoroalkyls in a study of 133 (79 male, 54 female) participants. In general, higher levels of PFOA,

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PFOS, PFNA, and PFDA were observed in male subjects; however, differences were only statistically significant for PFOA and PFDA. Sex differences in other perfluoroalkyls were not observed. For both male and female subjects, increasing levels of PFOA, PFNA, and PFOS were positively correlated with increasing age. Age- and sex-specific differences have been incorporated into a regression model developed by Jain (2015), which uses measured serum PFOA and PFOS levels to predict total serum perfluoroalkyl levels.

A small number of studies have measured serum perfluoroalkyl levels in U.S. children (Tables 5-23 and 5-24). NHANES has included adolescents (ages 12–19 years) in the 1999–2000 through 2013–2014 surveys (Calafat et al. 2007a, 2007b; CDC 2018). Analysis of NHANES data from younger children were conducted for the 2001–2002 survey (ages 6–11 years) (Kato et al. 2009b) and 2013–2014 survey (ages 3–5 and 6–11 years) (CDC 2018). Pinney et al. (2014) measured serum PFOA, PFOS, PFHxS, PFNA, and PFDA levels in girls (ages 6–8 years) living in Cincinnati, Ohio and San Francisco, California; and Olsen et al. (2004b) measured PFOA, PFOS, PFHxS, and FOSA in the serum of children ages 2–12 years from various locations in the United States who were diagnosed with group A streptococcal infections. Serum levels of several perfluoroalkyls were examined in children exposed to the World Trade Center (WTC) disaster (n=123) and a sociodemographically-matched comparison group (n=185) (Trasande et al. 2017). Children exposed during the WTC disaster were identified from the WTC Health Registry (WTCHR). All participants were ≤ 8 years of age on September 11, 2001. Blood serum levels of PFOA, PFOS, PFHxS, PFNA, PFDA, and PFUnA were collected during 2014–2016. For all perfluoroalkyls, serum levels in WTCHR children were significantly ($p < 0.01$) higher than in matched controls, with percentage increases above control as follows: PFOA 29%; PFOS 34%; PFHxS 26%; PFNA 24%; PFDA 27%; and 200% PFUnA.

Geometric mean serum perfluoroalkyl levels in adolescents (ages 12–19 years) included in NHANES are similar to geometric mean concentrations reported for adults (Calafat et al. 2007a, 2007b; CDC 2018). For example, geometric mean concentrations of PFOA and PFOS measured during the 1999–2000 and 2003–2004 NHANES surveys were 3.9–5.5 and 19.3–29.1 ng/mL, respectively, in adolescent serum and 3.9–5.2 and 20.7–30.4 ng/mL, respectively, in serum of the total population. The most recent NHANES survey (2013–2014) also included serum levels for children aged 3–5 and 6–11 years; the geometric mean concentrations in the younger children were similar to those in adolescents and the total population.

Olsen et al. (2004b) also found that the geometric mean serum PFOA, PFOS, PFHxS, and PFNA levels in older children (12–19 years of age) were similar to those measured in adults (Olsen et al. 2003b).

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However, estimated 95th percentile values of PFHxS measured in children were noted to be higher than values estimated for adults. Olsen et al. (2004b) reported bias-corrected 95th percentile estimates of 65 ng/mL for PFHxS in the serum of children ages 2–12 years. This value is higher than bias-corrected 95th percentile estimates of 9.5 and 8.3 ng/mL based on PFHxS measurements in the serum of adult blood donors (Olsen et al. 2003b) and elderly individuals (Olsen et al. 2004c), respectively (see Table 5-22). The difference is less extreme in the NHANES data, with PFHxS 95th percentile values of 12.9–13.1 ng/mL reported for children compared to values of 8.3–8.7 ng/mL reported for the total population. Reasons for the observed differences of PFHxS levels in childhood serum samples compared to adult samples have not been determined. Olsen et al. (2004b) stated that different exposure and activity patterns between children and adults should be considered. For example, children may have a higher exposure than adults to PFHxS, a substance that has been used in carpet treatment applications, since they are lower to the ground and have increased contact with carpeted floors (Calafat et al. 2007a; Olsen et al. 2004b).

Table 5-23. Percent Detection and Levels of PFOA and PFOS in Children’s Serum

Location	Detection and concentration (ng/mL [ppb]) ^a		Reference
	PFOA	PFOS	
U.S. Adolescents—NHANES (ages 12–19)			
1999–2000 (n=543)			Calafat et al. 2007a
Percent >LOD	100%	100%	
LOD	0.1	0.2	
Geometric mean	5.5	29.1	
95th percentile	11.2	56.8	
2003–2004 (n=640)			Calafat et al. 2007b
Percent >LOD	99.7% ^b	99.9% ^b	
LOD	0.1	0.4	
Geometric mean	3.9	19.3	
95th percentile	8.6	42.2	
2005–2006 (n=640)			CDC 2018
Percent >LOD	NR	NR	
LOD	0.1	0.2	
Geometric mean	3.59	15.0	
95th percentile	8.40	38.5	
2007–2009 (n=357)			CDC 2018
Percent >LOD	NR	NR	
LOD	0.1	0.2	
Geometric mean	3.91	11.3	
95th percentile	7.30	28.0	

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Table 5-23. Percent Detection and Levels of PFOA and PFOS in Children's Serum

Location	Detection and concentration (ng/mL [ppb]) ^a		Reference
	PFOA	PFOS	
2009–2010 (n=364)			CDC 2013
Percent >LOD	NR	NR	
LOD	0.1	0.2	
Geometric mean	2.74	6.84	
95th percentile	5.00	18.1	
2011–2012 (n=344)			CDC 2018
Percent >LOD	NR	NR	
LOD	0.1	0.2	
Geometric mean	1.80	4.16	
95th percentile	3.59	10.8	
2013–2014 (n=401)			CDC 2018
Percent >LOD	NR	NR	
LOD	0.1	0.2	
Geometric mean	1.66	3.54	
95th percentile	3.47	9.30	
2015–2016 (n=353)			CDC 2019
Percent >LOD	NR	NR	
LOD	0.1	0.2	
Geometric mean	1.25	2.94	
95th percentile	2.47	6.60	
U.S. Children—NHANES			
2001–2002; ages 6–11 (n=936)			Kato et al. 2009b
Least square mean	6.1–7.6	30.45–42.45	
2013–2014; ages 3–5 (n=181)			
Percent >LOD	100%	100%	Ye et al. 2018a; CDC 2018
LOD	0.1	0.1	
Geometric mean	2.00	3.38	
95th percentile	5.58	8.82	
2013–2014; ages 6–11 (n=458)			
Percent >LOD	100%	100%	Ye et al. 2018a; CDC 2018
LOD	0.1	0.1	
Geometric mean	1.89	4.15	
95th percentile	3.84	12.4	
U.S. Regional			
Girls ages 6–8; Cincinnati, Ohio (n=353)			Pinney et al. 2014
Percent >LOD ^c	99.7%	99.7%	
Geometric mean	7.8	13.2	
Median	7.3	13.6	

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Table 5-23. Percent Detection and Levels of PFOA and PFOS in Children's Serum

Location	Detection and concentration (ng/mL [ppb]) ^a		Reference
	PFOA	PFOS	
Girls ages 6–8; San Francisco, California (n=351)			Pinney et al. 2014
Percent >LOD ^c	100%	100%	
Geometric mean	5.7	13.2	
Median	5.8	12.5	
Children ages 6–10; Project Viva, Boston Massachusetts 2007–2010 (n=653)			Harris et al. 2017
Percent >LOD	99.5%	99.5%	
Geometric mean	4.2	6.2	
90 th percentile	7.9	13.7	
Children ages 2–12; 23 states and District of Columbia 1994–1995 (n=598)			Olsen et al. 2004b
Percent >LLOQ ^c	96%	100%	
Geometric mean	4.9	37.5	
95 th percentile ^d	10	89	
Children ages 1–19; West Virginia and Ohio 2005–2006 (n=4,943)			Mondal et al. 2012
Geometric mean	31.2	19.2	
90 th percentile	201	36.8	
Children (DOB: 9/11/1993–9/10/2001); New York City, WTCHR 2014–2016 (n=123)			Trasande et al. 2017
Percent >LOD	100%	100%	
Median	1.81	3.72	
Children (DOB: 9/11/1993–9/10/2001); New York City, not eligible for WTCHR 2014–2016 (n=185)			Trasande et al. 2017
Percent >LOD	100%	100%	
Median	1.39	2.78	

^a"Less than" values indicate that the concentration was reported as below the LOD or LLOQ. For cases where samples had concentrations below the LOD or LLOQ, a value between zero and the LOD or LLOQ was assigned when calculating the mean concentration.

^bPercent detection for the adolescent age group was not specified for the 2003–2004 NHANES samples. Percentages listed here are for the total sample population.

^cLOD or LLOQ not reported.

"—" indicates no available data; DOB = date of birth; LLOQ = lower limit of quantification; LOD = limit of detection; LOQ = limit of quantification; NR = not reported; PFOA = perfluorooctanoic acid; PFOS = perfluorooctane sulfonic acid; WTCHR = World Trade Center Health Registry

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Table 5-24. Percent Detection and Levels of Other Perfluoroalkyls in Children's Serum

Sample population	Detection and concentration (ng/mL [ppb]) ^a							
	PFHpA	PFNA	PFDA	PFUnA	PFDoDA	PFBS	PFHxS	FOSA
U.S. Adolescents NHANES (ages 12–19)								
1999–2000 (n=543) (Calafat et al. 2007a)								
Percent >LOD	10% ^b	96%	15%	12% ^b	<1% ^b		100%	100%
Geometric mean	—	0.5	<0.2	—	—		2.7	0.4
95th percentile	—	1.1	0.5	—	—		12.9	1.5
2003–2004 (n=640) (Calafat et al. 2007b)								
Percent >LOD	6.2% ^b	98.8% ^b	31.3% ^b	9.7% ^b	<0.1% ^b	<0.4% ^b	98.3% ^a	22.2% ^b
Geometric mean	<0.3	0.9	<0.3	<0.3	<1.0	<0.4	2.4	<0.2
95th percentile	0.5	2.7	0.7	<0.3	<1.0	<0.4	13.1	0.3
2005–2006 (n=640) (CDC 2018)								
Percent >LOD	NR	NR	NR	NR	NR	NR	NR	NR
Geometric mean	—	0.929	0.295	—	—	—	2.09	—
95th percentile	1.10	2.70	0.800	0.500	<LOD	0.100	14.1	0.300
2007–2008 (n=357) (CDC 2018)								
Percent >LOD	NR	NR	NR	NR	NR	NR	NR	NR
Geometric mean	—	1.16	0.231	—	—	—	2.40	—
95th percentile	0.600	2.54	0.800	0.300	<LOD	<LOD	15.9	0.300
2009–2010 (n=364) (CDC 2018)								
Percent >LOD	NR	NR	NR	NR	NR	NR	NR	NR
Geometric mean	—	1.10	0.220	—	—	—	2.03	—
95th percentile	0.400	2.62	0.600	0.400	<LOD	<LOD	12.3	<LOD
2011–2012 (n=344) (CDC 2018)								
Percent >LOD	NR	NR	NR	NR	NR	NR	NR	NR
Geometric mean	—	0.741	0.146	—	—	—	1.28	—
95th percentile	0.190	2.06	0.360	0.250	<LOD	<LOD	6.45	<LOD
2013–2014 (n=402) (CDC 2018)								
Percent >LOD	NR	NR	NR	NR	NR	NR	NR	NR
Geometric mean	—	0.599	0.136	—	—	—	1.27	—
95th percentile	0.200	2.00	0.400	0.200	0.200	<LOD	6.30	<LOD
2015–2016 (n=353) (CDC 2019)								
Percent >LOD		NR	NR	NR	NR		NR	
Geometric mean		0.481	—	—	—		0.918	
95th percentile		1.20	0.300	0.200	<LOD		3.10	
U.S. Children NHANES								
2001–2002; ages 6–11 (n=936) (Kato et al. 2009b)								
Least square mean		0.7-1.2						
2013–2014; ages 3–5 (n=181) (Ye et al. 2018a, 2018b; CDC 2018)								
Percent >LOD	27%	100%	50%	27%	0%	1%	100%	7%
LOD	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1
Geometric mean	<LOD	0.764	<LOD	<LOD	<LOD	<LOD	0.715	<LOD
95th percentile	0.310	3.49	0.370	0.370	<LOD	<LOD	1.62	0.110

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Table 5-24. Percent Detection and Levels of Other Perfluoroalkyls in Children's Serum

Sample population	Detection and concentration (ng/mL [ppb]) ^a							
	PFHpA	PFNA	PFDA	PFUnA	PFDoDA	PFBS	PFHxS	FOSA
2013–2014; ages 6–11 (n=458) (Ye et al. 2018a, 2018b; CDC 2018)								
Percent >LOD	15%	100%	46%	28%	0%	7%	100%	2%
LOD	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1
Geometric mean	<LOD	0.809	<LOD	<LOD	<LOD	<LOD	0.913	<LOD
95th percentile	0.170	3.19	0.350	0.250	<LOD	0.130	4.14	<LOD
U.S. Regional								
Girls ages 6–8; Cincinnati, Ohio (n=353) (Pinney et al. 2014)								
Percent >LOD ^c		99.9	75.8				99.7	19
Geometric mean		1.4	0.3				5.1	<LOD
Median		1.4	0.3				5.2	<LOD
Girls ages 6–8; San Francisco, California (n=351) (Pinney et al. 2014)								
Percent >LOD ^c		100	78.7				100	10
Geometric mean		1.7	0.3				3.0	<LOD
Median		1.6	0.3				2.3	<LOD
Children ages 6–10; Project Viva, Boston Massachusetts 2007–2010 (n=653) (Harris et al. 2017)								
Percent >LOD ^c		99.5%	88.2%				99.5%	0.9%
Geometric mean		1.7	0.3				2.2	<LOD
90 th percentile		3.8	0.6				7.0	<LOD
Children ages 2–12; 23 states and District of Columbia 1994–1995 (n=598) (Olsen et al. 2004b)								
Percent >LLOQ							78%	14%
LLOQ							NR	1.0
Geometric mean							4.5	<2.0
95th percentile ^b							65	<2.0
Children (DOB: 9/11/1993–9/10/2001); New York City, WTCHR 2014–2016 (n=123) (Trasande et al. 2017)								
Percent >LOD	99.7 ^d	75 ^d	53 ^d				100	
Median	0.61	0.14	0.12				0.67	
Children (DOB: 9/11/1993–9/10/2001); New York City, not eligible for WTCHR 2014–2015 (n=185) (Trasande et al. 2017)								
Percent >LOD	99.7 ^d	75 ^d	53 ^d				100	
Median	0.49	0.11	0.04				0.53	

^a"Less than" values indicate that the concentration was reported as below the LOD or LLOQ. For cases where samples had concentrations below the LOD or LLOQ, a value between zero and the LOD or LLOQ was assigned when calculating the mean concentration.

^bPercent detection for the adolescent age group was not specified for these samples. Percentages listed here are for the total sample population.

^cLOD or LLOQ not reported.

^d%>LOD reported for combined WTCHR and non-WTCHR groups.

Gray cells indicate that chemical was not evaluated; "—" indicates no available data; DOB = date of birth; FOSA = perfluorooctane sulfonamide; LLOQ = lower limit of quantification; LOD = limit of detection; LOQ = limit of quantification; NC = not calculated; ND = no data; NR = not reported; PFBS = perfluorobutane sulfonic acid; PFDA = perfluorodecanoic acid; PFDoDA = perfluorododecanoic acid; PFHpA = perfluoroheptanoic acid; PFHxS = perfluorohexane sulfonic acid; PFNA = perfluorononanoic acid; PFUnA = perfluoroundecanoic acid

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When estimating PFOS and PFOA uptake doses for children, Trudel et al. (2008) assumed the same exposure pathways for children as were proposed for adults, but considered exposure from hand-to-mouth transfer from treated carpets to be much larger in children. This pathway was estimated to contribute 40–60% of the total uptake of both PFOS and PFOA in infants (0–1 years), toddlers (1–4 years), and children (5–11 years) in the high-exposure scenario. Exposure via human breast milk was included in the food consumption pathway for infants. Exposure via mouthing of clothes, carpet, and upholstery was also considered for children <12 years old; however, this was considered to be a minor pathway of exposure. PFOS uptake doses estimated for the low-, medium-, and high-exposure scenarios were 18.1–219 ng/kg body weight/day for infants, 14.8–201 ng/kg body weight/day for toddlers, and 9.7–101 ng/kg body weight/day for children. PFOA uptake doses estimated for the low-, medium-, and high-exposure scenarios were 2.2–121 ng/kg body weight/day for infants, 1.2–128 ng/kg body weight/day for toddlers, and 0.8–65.2 ng/kg body weight/day for children. In contrast to the estimates for children under age 12, relative exposure pathways and uptake doses estimated for teenagers (12–20 years old) were approximately the same as for adults.

Perfluoroalkyls have been measured human breast milk and umbilical cord blood; reported concentrations are listed in Tables 5-25 and 5-26. Measurements of perfluoroalkyls in amniotic fluid, meconium, neonatal blood, or other tissues have not been located.

Table 5-25. Percent Detection and Levels of PFOA and PFOS in Umbilical Cord Blood and Breast Milk

Location	Detection and concentration (ng/mL [ppb]) ^a		Reference
	PFOA	PFOS	
Umbilical cord blood			
San Francisco, California			Morello-Frosch et al. 2016
Percent >LOD ^c	56%	100%	
Geometric mean	–	2.27	
95th percentile	1.68	4.35	

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Table 5-25. Percent Detection and Levels of PFOA and PFOS in Umbilical Cord Blood and Breast Milk

Location	Detection and concentration (ng/mL [ppb]) ^a		Reference
	PFOA	PFOS	
Baltimore THREE Study			Apelberg et al. 2007a, 2007b
Cord serum (n=299)			
Percent >LOD	100%	99%	
LOD	0.1–0.2	0.2	
Geometric mean	1.6	4.9	
Minimum	0.3	<0.2	
Maximum	7.1	34.8	
Maternal serum (n=293)			
Median	1.4–1.6	4.1–5.0	
Germany			Midasch et al. 2007
Cord plasma (n=11)			
Percent detected	100%	100%	
LOQ	0.5	0.5	
Median	3.4	7.3	
Maternal plasma (n=11)			
Percent detected	100%	100%	
LOQ	0.5	0.5	
Median	2.6	13.0	
Spain			Manzano-Salgado et al. 2015
Cord serum (n=66)			
Percent >LOD	100%	100%	
Minimum	0.60	0.53	
Maximum	10.56	4.71	
Maternal serum (n=53)			
Percent >LOD	100%	100%	
Minimum	0.86	1.17	
Maximum	14.54	23.14	
Danish National Birth Cohort			Fei et al. 2007
Cord blood (n=50)			
Percent >LLOQ	98%	100%	
LLOQ	1.0	1.0	
Mean	3.7	11.0	
Maternal blood (n=200)			
Percent >LLOQ	98%	100%	
LLOQ	1.0	1.0	
Mean	4.5	29.9	

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Table 5-25. Percent Detection and Levels of PFOA and PFOS in Umbilical Cord Blood and Breast Milk

Location	Detection and concentration (ng/mL [ppb]) ^a		Reference
	PFOA	PFOS	
Japan			Inoue et al. 2004
Cord serum (n=15)			
Percent >LOD	0%	100%	
LOD	0.5	0.5	
Range		1.6–5.3	
Maternal serum (n=15)			
Percent >LOD	20%	100%	
LOD	0.5	0.5	
Range	0.5–2.3	4.9–17.6	
Breast milk			
Massachusetts (n=45)			Tao et al. 2008b
Milk			
Percent >LOQ ^b	89%	96%	
Mean	0.0438	0.131	
Minimum	<0.0301	<0.032	
Maximum	0.161	0.617	
Sweden (n=12)			Kärman et al. 2007
Milk			
Percent >LOD	8% ^c	100%	
LOD	0.01	0.005	
Mean	–	0.201	
Range	<0.209–0.492	0.060–0.470	
Maternal serum			
Percent >LOD	100%	100%	
LOD	0.01	0.005	
Mean	3.8	20.7	
Range	2.4–5.3	8.2–48.0	
Sweden (n=20)			Sundstrom et al. 2011
Milk			
LOD	0.005	0.005	
1997 Mean	0.138	0.237	
2007 Mean	0.086	0.122	
Norway (n=9)			Thomsen et al. 2010
Milk			
LOD	0.008	0.003	
Median	0.05	0.11	
Range	0.016–0.19	0.028–0.36	

5. POTENTIAL FOR HUMAN EXPOSURE

Table 5-25. Percent Detection and Levels of PFOA and PFOS in Umbilical Cord Blood and Breast Milk

Location	Detection and concentration (ng/mL [ppb]) ^a		Reference
	PFOA	PFOS	
China (n=19)			So et al. 2006b
Percent >LOD	100%	100%	
LOD	0.021–0.027	0.001–0.0036	
Range	0.047–0.210	0.045–0.360	
Middle East, including Jordan (n=19)			Al-sheyab et al. 2015
Percent >LOQ	100%	94%	
LOQ	0.01	0.01	
Mean	0.14	0.035	
Range	0.024–1.22	0.006–0.18	
France (n=48)			Antignac et al. 2013
Percent >LOD	90%	98%	
LOD	0.05	0.05	
Mean	0.082	0.092	
Range	<LOD-0.22	<LOD-0.33	
France (n=61)			Cariou et al. 2015
Percent >LOD	77%	82%	
LOD	0.05	0.04	
Mean	0.041	0.04	
Range	<LOD-0.31	<LOD-0.376	
Belgium			Croes et al. 2012
Percent >LOD	100%	100%	
LOD	0.01	0.01	
Mean	0.08	0.13	
Italy (n=49)			Guerranti et al. 2013
Percent >LOD	2% (1 sample)	27%	
LOD	0.5 ^e	0.5 ^d	
Mean	8.04 ^e	0.85 ^e	
Italy			Barbarossa et al. 2013
Primiparous (n=21)			
Percent >LOQ	81%	90%	
LOQ	0.024	0.015	
Mean	0.076	0.057	
Range	<LOQ–0.24	<LOQ–0.29	
Multiparous (n=16)			
Percent >LOQ	69%	62%	
LOQ	0.024	0.015	
Mean	0.043	0.036	
Range	<LOQ–0.1	<LOQ–0.12	

5. POTENTIAL FOR HUMAN EXPOSURE

Table 5-25. Percent Detection and Levels of PFOA and PFOS in Umbilical Cord Blood and Breast Milk

Location	Detection and concentration (ng/mL [ppb]) ^a		Reference
	PFOA	PFOS	
Korea			Kang et al. 2016
Percent >LOD	98%	98%	
LOD	0.019	0.007	
Median	0.072	0.05	
Czech Republic (n=50)			Lankova et al. 2013
Percent >LOQ	100%	100%	
LOQ	0.006	0.005	
Mean	0.05	0.033	
Range	0.012–0.13	0.007–0.11	
Germany/Hungary (n=70)			Völkel et al. 2008
Percent >LOQ	16%	100%	
Minimum	<0.200	0.028	
Maximum	0.460	0.639	

^a"Less than" values indicate that the concentration was reported as below the LOD or LLOQ. For cases where samples had concentrations below the LOD or LLOQ, a value between zero and the LOD or LLOQ was assigned when calculating the mean concentration.

^bLOD or LLOQ not reported.

^cReported as bias-corrected estimates.

^dng/g wet weight.

"—" indicates no available data; DOB = date of birth; LLOQ = lower limit of quantification; LOD = limit of detection; LOQ = limit of quantification; NR = not reported; PFOA = perfluorooctanoic acid; PFOS = perfluorooctane sulfonic acid; WTCHR = World Trade Center Health Registry

Table 5-26. Percent Detection and Levels of Other Perfluoroalkyls in Children's Umbilical Cord Blood and Breast Milk

Sample population	Detection and concentration (ng/mL [ppb]) ^a							
	PFHpA	PFNA	PFDA	PFUnA	PFDoDA	PFBS	PFHxS	FOSA
Umbilical cord blood								
San Francisco, California (Morello-Frosch et al. 2016) (n=65)								
Percent >LOD ^c	56%	97%	9%	84%	0%	25%		91%
Geometric mean	—	0.29	—	0.03	—	—		0.02
95th percentile	0.23	0.93	0.49	0.16	—	0.03		0.10
Baltimore THREE Study (Apelberg et al. 2007a, 2007b)								
Cord serum (n=299)								
Percent >LOD	2%		24%	34%	5%	3%		26%
LOD	0.4		0.2	0.2	0.2	0.1		0.05
Minimum	<0.4		<0.2	<0.2	<0.2	<0.1		<0.05
Maximum	2.6		1.1	1.9	1.7	0.2		0.8

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Table 5-26. Percent Detection and Levels of Other Perfluoroalkyls in Children's Umbilical Cord Blood and Breast Milk

Sample population	Detection and concentration (ng/mL [ppb]) ^a							
	PFHpA	PFNA	PFDA	PFUnA	PFDODA	PFBS	PFHxS	FOSA
Japan (Inoue et al. 2004)								
Cord serum (n=15)								
Percent >LOD								0%
LOD								1.0
Maternal serum (n=15)								
Percent >LOD								0%
LOD								1.0
Spain (Manzano-Salgado et al. 2015)								
Cord blood (n=66)								
Percent >LOD		100%					88%	
Minimum		0.13					0.05	
Maximum		2.24					1.93	
Maternal serum (n=53)								
Percent >LOD		100%					96%	
Minimum		0.20					0.05	
Maximum		5.37					2.53	
Breast milk								
Massachusetts (n=45) (Tao et al. 2008b)								
Milk								
Percent >LOQ ^b	<1%	64%	<1%	<1%	<1%	<1%	51%	
Mean	NR	0.01726	NR	NR	NR	NR	0.0145	
Minimum	<0.010	<0.0052	<0.00772	<0.00499	<0.00440	<0.0100	<0.0120	
Maximum	0.0234	0.0184	0.0111	0.00884	0.00974	0.0198	63.8	
France (n=49) (Antignac et al. 2013)								
Percent >LOD	2%	2%					100%	
LOD	0.05	0.05					NR	
Mean	—	—					0.049	
Minimum	<LOD	<LOD					0.040	
Maximum	0.074	0.064					0.066	
France (n=61) (Cariou et al. 2015)								
Percent >LOD		0%					15%	
LOD		0.05					0.03	
Mean		0.014					0.026	
Minimum		<LOD					<LOD	
Maximum		<LOD					0.217	
Belgium (n=40) (Croes et al. 2012)								
Percent >LOD		42%					20%	
LOD		0.01					0.01	
Mean		—					—	

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Table 5-26. Percent Detection and Levels of Other Perfluoroalkyls in Children's Umbilical Cord Blood and Breast Milk

Sample population	Detection and concentration (ng/mL [ppb]) ^a							
	PFHpA	PFNA	PFDA	PFUnA	PFDODA	PFBS	PFHxS	FOSA
Sweden (n=12) (Kärman et al. 2007a)								
Milk								
Percent >LOD		17%	0%	0%			100%	67%
LOD		0.005	0.008	0.005			0.01	0.007
Mean		0.017	—	—			0.085	0.013
Range		<0.005– 0.020	—	—			0.031– 0.172	<0.007– 0.030
Maternal serum								
Percent >LOD		100%	100%	100%			100%	75%
LOD		0.005	0.008	0.005			0.01	0.007
Mean		0.80	0.53	0.40			4.7	0.24
Range		0.43–2.5	0.27–1.8	0.20–1.5			1.8–11.8	0.16–0.19
China (n=19) (So et al. 2006b)								
Percent >LOD	37%	100%	100%	100%		11%	100%	
LOD	0.005– 0.010	0.001– 0.010	0.0011– 0.0025	0.0022– 0.0050		0.001– 0.005	0.001– 0.010	
Range	<0.005– 0.0067	0.01– 0.062	0.0038– 0.011	0.0091– 0.056		<0.001– 0.0025	0.004– 0.10	

^a"Less than" values indicate that the concentration was reported as below the LOD or LLOQ. For cases where samples had concentrations below the LOD or LLOQ, a value between zero and the LOD or LLOQ was assigned when calculating the mean concentration.

^bLOD or LLOQ not reported.

Gray cells indicate that chemical was not evaluated; "—" indicates no available data; DOB = date of birth; FOSA = perfluorooctane sulfonamide; LLOQ = lower limit of quantification; LOD = limit of detection; LOQ = limit of quantification; ND = no data; NR = not reported; PFBS = perfluorobutane sulfonic acid; PFDA = perfluorodecanoic acid; PFDODA = perfluorododecanoic acid; PFHpA = perfluoroheptanoic acid; PFHxS = perfluorohexane sulfonic acid; PFNA = perfluorononanoic acid; PFUnA = perfluoroundecanoic acid

Tao et al. (2008b) measured perfluoroalkyl concentrations in 45 human breast milk samples collected from Massachusetts. PFOS, PFOA, PFHxS, and PFNA were each detected in 96, 89, 51, and 64% of the samples, respectively, with median concentrations of 106, 36.1, 12.1, and 6.97 pg/mL, respectively. PFHpA, PFDA, PFUnA, PFDODA, and PFBS were each detected in <1% of the samples. Perfluoroalkyls have also been measured in the human breast milk of individuals from Sweden, China, and Germany/Hungary (Kärman et al. 2007; So et al. 2006b; Völkel et al. 2008). PFOS was detected in all samples, while detection of PFOA ranged from 8 to 100% in these studies. The reported maximum concentrations of PFOS and PFOA measured in human breast milk samples collected during these studies were 0.360–0.639 and 0.210–0.490 ng/mL, respectively (Kärman et al. 2007; So et al. 2006b; Völkel et al. 2008). Other perfluoroalkyls detected in human breast milk included PFHpA, PFNA, PFDA, PFUnA, PFBS,

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PFHxS, and FOSA. Maximum concentrations of these compounds were reported to be <0.18 ng/mL (Kärman et al. 2007).

The presence of perfluoroalkyls in umbilical cord blood indicates that these substances can cross the placental barrier resulting in the exposure of babies *in utero* (Apelberg et al. 2007a, 2007b; Fei et al. 2007; Inoue et al. 2004; Midasch et al. 2007). In most studies, PFOS and PFOA have been detected in most umbilical cord blood samples with reported maximum (or 95th percentile) concentrations of 5.3–34.8 and 1.68–7.1 ng/mL, respectively (Apelberg et al. 2007a, 2007b; Fei et al. 2007; Inoue et al. 2004; Midasch et al. 2007; Morello-Frosch et al. 2016). Inoue et al. (2004) did not detect PFOA in 15 cord blood samples from Japan; however, this compound was only detected in the maternal serum of three mothers. Apelberg et al. (2007a) also reported concentrations of other perfluoroalkyls measured in 299 cord serum samples collected during the Baltimore THREE Study. Of these compounds, PFDA, PFUnA, and FOSA were detected most frequently (24, 34, and 26%, respectively). Maximum concentrations in these samples ranged from 1.1 to 1.9 ng/mL. PFHpA, PFDoDA, and PFBS were each detected in <6% of the samples, with maximum concentrations ranging from 0.2 to 2.6 ng/mL. Manzano-Salgado et al. (2015) studied the potential transfer of perfluoroalkyls from mothers to their children during pregnancy. Maternal blood and cord serum were collected from 66 mother-child pairs and analyzed for the presence of perfluoroalkyls. A positive correlation was found between maternal plasma and maternal serum with cord serum levels, and the authors concluded that either maternal plasma or maternal serum could be used as a method to estimate fetal exposure to perfluoroalkyls. Median concentrations of PFOS and PFOA were 6.18 and 2.85 ng/mL, respectively, in maternal plasma and 6.99 and 2.97 ng/mL, respectively, in maternal serum. PFOS and PFOA levels in cord serum were 1.86 and 1.90 ng/mL, respectively. A biomonitoring survey of 1,533 pregnant females in Denmark from 2008 to 2013 showed decreasing levels of most perfluoroalkyls in the females' blood during this time period (Bjerregaard-Olesen et al. 2016). The results of this study showed that serum levels of PFHxS, PFOS, PFOA, PFNA, and PFDA decreased at a rate of 7.0, 9.3, 9.1, 6.2, and 6.3 per year, respectively. Morello-Frosch et al. (2016) measured the levels of perfluoroalkyls in 77 maternal and 65 paired umbilical cord blood samples from pregnant females and newborn children in San Francisco, California. Perfluoroalkyls, including PFOA and PFOS, were widely detected; however, concentrations in cord blood or serum were typically equal to or lower than maternal blood levels.

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5.7 POPULATIONS WITH POTENTIALLY HIGH EXPOSURES

Potentially high exposures to perfluoroalkyls can occur in the following population categories: perfluoroalkyl production and manufacturing workers, communities located near fluorochemical facilities, and individuals with prolonged use of perfluoroalkyl-containing products. Workers at perfluoroalkyl production and manufacturing facilities and community members living near these facilities may have higher exposure to perfluoroalkyls than the general population based on elevated concentrations of these substances measured in air, soil, sediment, surface water, groundwater, and vegetation surrounding these facilities (3M 2007b, 2008b, 2008c; Barton et al. 2006; Davis et al. 2007; Olsen 2015). Additionally, children may be at risk for higher potential exposure due to hand-to-mouth transfer of chemicals from dust and the ingestion of dirt (Shoeib et al. 2004; Trudel et al. 2008). Levels of perfluoroalkyls measured in the blood of production and manufacturing workers at several facilities are listed in Table 5-27. The serum PFOA, PFOS, and PFHxS levels in workers were frequently 100–1,000 times higher than in the general population. 3M estimated PFOA doses for various on-site exposure scenarios based on monitoring information collected at the Decatur Facility in Alabama (3M 2008c). Occupational exposure scenarios included groundskeeper/maintenance worker and construction/utility worker exposed to on-site soils, surface water, and sediment. According to 3M, estimated on-site exposure to PFOA ranged from 3.2×10^{-6} to 2.4 ng/kg/day, with the highest estimated exposure corresponding to construction/utility workers engaged in projects involving contact with soil from an on-site field. Chang et al. (2008a) measured concentrations of PFBA in the serum of 127 former employees and 50 current employees of the 3M Cottage Grove Facility in Minnesota. PFBA serum concentrations were below the detection limit in 73.2% of the former employees and 68.0% of the current employees. Only 4% of the serum samples contained PFBA above 2 ng/mL, with maximum concentrations of 6.2 ng/mL for the former employees and 2.2 ng/mL for the current employees.

Table 5-27. Concentrations of PFOA, PFOS, and PFHxS in Human Serum for Occupationally Exposed Individuals

Location	Mean concentration (ng/mL [ppb])			Reference
	PFOA	PFOS	PFHxS	
3M Decatur, Alabama				
1993 (n=111)	0.00–80,000 (range)			Olsen et al. 1998b
1995 (n=80)	0.00–114,100 (range)			Olsen et al. 1998b
1995 (n=90)		2,440		Olsen et al. 1999
1997 (n=84)		1,960		Olsen et al. 1999
1998 (n=126)	1,536	1,505	345	Olsen 2015
2000 (n=215)	11,900	1,400		Olsen et al. 2003a

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Table 5-27. Concentrations of PFOA, PFOS, and PFHxS in Human Serum for Occupationally Exposed Individuals

Location	Mean concentration (ng/mL [ppb])			Reference
	PFOA	PFOS	PFHxS	
2000 (n=263)	1,780	1,320	250	Olsen 2015
1999–2004 (n=26 retired workers)				Olsen et al. 2007a
Initial	691	799	290	
Final	262	403	182	
3M Cottage Grove, Minnesota				
1993 (n=111)	5,000 (1,100 geometric mean)			Olsen et al. 2000; Olsen 2015
1995 (n=80)	6,800 (1,200 geometric mean)			Olsen et al. 2000; Olsen 2015
1997 (n=74)	6,400 (1,300 geometric mean)			Olsen et al. 2000; Olsen 2015
2000 (n=122)	4,630 (810 geometric mean)	860		Olsen and Zobel 2007; Olsen 2015
DuPont Washington Works, Ohio				
1979-2004				Woskie et al. 2012
All workers (n=1308 workers, 2125 samples)	2,050			
Direct PFOA exposure (n=170 workers; 541 samples)	5,470			
Direct PFOA exposure (n=96 workers; 208 samples)	2,530			
2004–2005				Emmett et al. 2006a
Substantial occupational exposure (n=18)	824			
2004				Sakr et al. 2007b
Current occupational exposure (n=259)	494 (median)			
Intermittent current occupational exposure (n=160)	176 (median)			
Past occupational exposure (n=264)	195 (median)			
3M Antwerp, Belgium				
1995 (n=88)		1,930		Olsen et al. 1999
1997 (n=65)		1,480		Olsen et al. 1999
2000 (n=206)	1,030	960		Olsen et al. 2003a
2000 (n=258)	840	800	170	Olsen 2015

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Table 5-27. Concentrations of PFOA, PFOS, and PFHxS in Human Serum for Occupationally Exposed Individuals

Location	Mean concentration (ng/mL [ppb])			Reference
	PFOA	PFOS	PFHxS	
Mitenei, Trissino, Italy				
2007				
Current occupational exposure (n=39)	5,710 ^c (200–47,040)			Costa et al. 2009
Former occupational exposure (n=11)	4,430 ^c (530–18,660)			Costa et al. 2009
2000 (n=25)	18,800			Costa et al. 2009
2001 (n=42)	19,700			Costa et al. 2009
2002 (n=46)	19,300			Costa et al. 2009
2003 (n=41)	13,700			Costa et al. 2009
2004 (n=34)	11,400			Costa et al. 2009
2006 (n=49)	10,800			Costa et al. 2009
2007 (n=50)	11,600			Costa et al. 2009

^aData include results from three retirees from the 3M plant in Cottage Grove, Minnesota.

Gray cells indicate that chemical was not evaluated; PFHxS = perfluorohexane sulfonic acid; PFOA = perfluorooctanoic acid; PFOS = perfluorooctane sulfonic acid

Individuals who performed jobs that require frequent and/or prolonged contact with perfluoroalkyl containing products, such as firefighters, waste handlers, individuals who install and treat carpets, or individuals with prolonged use of ski wax may also have higher occupational exposure to perfluoroalkyls. Some firefighting foams contain perfluoroalkyls, and firefighters who use these products have been shown to have greater exposures as compared to the general population. Dobraca et al. (2015) compared perfluoroalkyl serum levels of a group of firefighters in California to an adult population from the NHANES survey. Levels of PFOA and PFOS were only slightly higher in the firefighter group (geometric means 3.75 and 12.50 ng/mL, respectively) when compared to adult males in the 2009–2010 NHANES general population survey (3.61 and 12.13 ng/mL, respectively); however, PFDA serum concentrations of firefighters were up to 3 times greater than the NHANES comparison group for the 25th–95th percentiles (50th percentile in firefighters; 0.72 ng/mL compared to 0.30 ng/mL) and the geometric mean (0.90 ng/mL compared to 0.30 ng/mL). In a small-scale study of 37 firefighters participating in the C8 Health Project, significantly (adjusted for age, water district, household income, and smoking) higher levels of PFOA and PFHxS were found in the firefighters compared to 5,373 male participants with other jobs (Jin et al. 2011). Geometric mean PFOA and PFHxS levels were 37.59 and 4.77 ng/mL, respectively, in the firefighters and 31.59 and 3.62 ng/mL, respectively, in the other

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participants. No significant differences in PFOS or PFNA levels were found between the groups. A biomonitoring study of 149 firefighters in Australia showed that 100% of serum samples collected had detectable levels of PFOA, PFOS, PFHxS, and PFNA (Rotander et al. 2015). Serum levels of PFHxS were found to be approximately 10–15 times higher than levels found in the general population of Australia and Canada, while PFOS levels in the firefighters were approximately 6–10 times greater than the general population of these nations.

Elevated serum levels of PFOA, PFNA, PFHxA, PFHpA, PFDA, and PFUnA have been found in professional ski waxes containing fluorotelomers; the perfluoroalkyls were likely formed via fluorotelomer metabolism (Olsen 2015). Christensen et al. (2016) conducted a biomonitoring study of perfluoroalkyls on male fishermen from Wisconsin ≥ 50 years old with a history of sport fish consumption. Increasing age and lower BMI were generally associated with higher levels of the perfluoroalkyls; however, there were only weak correlations observed between amounts of fish consumption and perfluoroalkyl levels, with the exception of PFDA. Levels of PFOA, PFNA, and PFHxS in the blood of the male anglers were similar to the levels for a subset of the NHANES 2011–2012 survey (non-Hispanic white males ≥ 50 years old); however, levels of PFOS and PFDA were approximately 2 times greater in the anglers as compared to the NHANES survey subgroup. The median and 95th percentile concentrations of PFOS in the anglers were 19.00 and 54.00 ng/mL, respectively, as compared to 10.33 and 25.83 ng/mL, respectively, in the NHANES study group. The median and 95th percentile concentrations of PFDA in the anglers were 0.52 and 1.90 ng/mL, respectively, as compared to 0.23 and 0.53 ng/mL in the NHANES study group. Family members of occupationally exposed workers have been shown to have higher exposure to perfluoroalkyls via dust transfer as compared to family members of nonoccupationally exposed workers (Fu et al. 2015).

PFOA, PFOS, PFBA, PFBS, PFNA, and PFHxS have been detected in the municipal drinking water and private wells of some communities located near fluorochemical facilities (3M 2008c; ATSDR 2008; Emmett et al. 2006a; Hoffman et al. 2011; Hölzer et al. 2008; Post et al. 2013; Steenland et al. 2009a; Wilhelm et al. 2009). Emmett et al. (2006a) compared PFOA serum levels to various types of exposure for individuals living in the Little Hocking community (near DuPont's Washington Works facility) and concluded that residential water source was the primary determinant of serum PFOA at this location. These authors reported that the mean human serum PFOA level was 105 times higher than the residential drinking water level. Median serum PFOA levels were 371 ng/mL in residents for whom this was the only residential water source and 71 ng/mL in those who used bottled, cistern, or spring water. Increased serum PFOA was associated with increasing number of drinks of tap water daily and also with increasing

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use of water for making soups and stews and in-home canning of fruits and vegetables. Use of a carbon water filter reduced PFOA levels by about 25%. In a follow-up study, 231 study participants in the Little Hocking Water District were evaluated 15 months later with 88% using bottled water exclusively; 8% had made other changes to their ingestion of residential water including use of activated carbon water filters. PFOA levels had decreased an average of 26% from the initial levels (Emmett et al. 2009). Similarly, Bartell et al. (2010) found that serum levels of PFOA declined significantly following the implementation of GAC filtration of the public water supply. The average decrease in serum PFOA levels for Lubeck, West Virginia residents primarily consuming public water at home (n=130) was 26% 1 year after treatment began. Similar trends were reported for residents of Little Hocking, Ohio. The average decrease in PFOA serum levels for residents primarily consuming public water (n=39) was about 11% 6 months after treatment began.

Median PFOA serum levels for residents currently residing in six water districts located in the mid-Ohio Valley near the Washington Works facility ranged from 12.1 to 224.1 ng/mL, while the median concentration ranged from 10.5 to 33.7 ng/mL for residents who previously worked or resided in these districts (Steenland et al. 2009a). Former employees at the chemical plant had much higher levels (median=75 ng/mL) than people who had not worked at the plant (median=24 ng/mL), but lower levels than those who continued to be employed at the plant during the monitoring period (median=148 ng/mL). Another study of this community reported a median serum PFOA concentration of 24.3 ng/mL for 45,276 non-occupationally exposed individuals in 2005–2006 (Shin et al. 2011a). This was about 8 times greater than the median concentration (3.20 ng/mL) of 2,120 residents of the general population taken from the NHANES data for 2005–2006 (CDC 2018).

A study of residents consuming drinking water from the Ohio River and the Ohio River Aquifer reported elevated median PFOA levels of 13.8 ng/mL in blood samples collected in 1991–1993 from 139 residents (Herrick et al. 2017). In samples collected in 2011–2013 (n=133), the serum PFOA levels dropped to 4.3 ng/mL. The serum PFOS levels in samples collected in 1994–1996 (median of 32.2 ng/mL, n=189) were similar to levels in the general population (Herrick et al. 2017); similar to the decline observed in the general population, median serum PFOS levels were 6.3 ng/mL in 2011–2013.

The Minnesota Department of Health conducted a biomonitoring study of 196 residents living in Washington county, east of the Minneapolis-St. Paul metropolitan area; private and municipal drinking water wells were shown to be contaminated with perfluoroalkyls (MDH 2009). In 2008–2009, geometric mean serum PFOA, PFOS, and PFHxS levels were 15.4, 35.9, and 8.4 ng/mL, respectively; these values

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were higher than levels reported in NHANES (Landsteiner et al. 2014; MDH 2009). The investigators noted that the geometric mean serum concentrations of these three substances were greater in residents obtaining drinking water from municipal water supplies as compared to residents with private wells (Landsteiner et al. 2014). Monitoring efforts also conducted in Washington County, Minnesota near the 3M Cottage Grove Facility revealed widespread contamination of PFBA in the groundwater in 2006 (ATSDR 2008). PFBA was detected in 28% of the community members' serum samples; the 75th percentile serum concentration for PFBA was 0.135 ng/mL and the maximum concentration was 8.5 ng/mL (MDH 2000).

ATSDR performed an exposure investigation for residents of Decatur, Alabama following an accidental release of perfluoroalkyls into the Decatur waste water treatment plant (ATSDR 2013). A group of 155 residents had their blood tested for levels of eight perfluoroalkyls. Serum levels for PFNA, PFDA, and FOSA were lower or similar to levels of the general population when compared to the NHANES results. Elevated serum PFOA, PFOS, and PFHxS levels were observed; geometric mean values were 16.3, 39.8, and 6.4 ng/mL, respectively. Residents who used the West Morgan/East Lawrence public water supply had significantly higher geometric mean serum levels of PFOA (17.59 ng/mL) and PFHxS (6.68 ng/mL) as compared to the geometric mean for a similar demographic group from the NHANES survey. Serum perfluoroalkyl levels in the residents were shown to be much lower than levels found in occupationally exposed individuals who regularly worked with these substances (ATSDR 2013).

Additional blood serum levels of PFOA and PFOS for residents in selected areas of Ohio, West Virginia, New Jersey, and Minnesota whose residential source of drinking water may have been contaminated are available from the EPA docket on PFOA and related perfluoroalkyls (EPA-HQ-OPPT-2003-0012) (Bilott 2004, 2005a, 2005b, 2007).

The Emmett et al. (2006a) and Steenland et al. (2009a) studies of the community near the Washington Works facility and the 3M (2008c) study of the Decatur Facility in Alabama found age-related differences in serum PFOA levels. In a comparison of serum PFOA levels in various age groups of residents serviced by the Little Hocking Water Association district, Emmett et al. (2006a) found that 2–5-year-old children had a higher serum PFOA (median 600 ng/mL) compared with residents in all other age groups (median 321 ng/mL) except for the group aged >60 years, whose levels were similar to those in young children. Several factors may have contributed to the observed high levels in children: infants and young children proportionally drink more water per kg of body weight than adults; children (and also the elderly) tend to spend more time at home with exclusive use of residential water than other age groups; and trans-

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placental and breast milk exposures could also contribute to levels in children. In the Steenland et al. (2009a) study, PFOA serum levels tended to be highest for children aged 0–9 years and persons >50 years old. The serum levels of the 69,030 residents participating in this study categorized by age are provided in Table 5-28. 3M (2008c) estimated doses for various off-site exposure scenarios based on monitoring information collected at the Decatur Facility. Exposure scenarios include local children and adult residents exposed to PFOA in off-site soils, groundwater, municipal water, fish from the Tennessee River, and surface water and sediments in the Tennessee River. According to 3M, estimated off-site exposure of local residents to PFOA ranged from 0.011 to 260 ng/kg/day, with the highest estimated exposure corresponding to children whose source of drinking water was groundwater adjacent to the southern side of the facility.

Table 5-28. Blood Serum Levels for 69,030 Current and Former Residents of Six Water Districts in the Mid-Ohio Valley (2005–2006)

Age (years)	Number (percentage of total)	Median perfluorooctanoic acid (PFOA) level (ng/mL)
0–9	4,915 (7.1)	32.8
10–19	9,658 (14.0)	26.6
20–29	10,073 (14.6)	21.0
30–39	10,547 (15.3)	22.7
40–49	12,113 (17.6)	28.0
50–59	10,515 (15.2)	33.6
60–69	6,881 (10)	42.9
≥70	4,328 (6.3)	40.1

Source: Steenland et al. 2009a