

History and Use of Per- and Polyfluoroalkyl Substances (PFAS)

1 Introduction

The unique physical and chemical properties of per- and polyfluoroalkyl substances (PFAS) impart oil and water repellency, temperature resistance, and friction reduction to a wide range of products used by consumers and industry. For example, PFAS have been used in coatings for textiles, paper products, and cookware and to formulate some firefighting foams, and have a range of applications in the aerospace, photographic imaging, semiconductor, automotive, construction, electronics, and aviation industries (KEMI 2015; USEPA 2017b). USEPA has compiled a web-based resource for PFAS information. The information includes topics such as Policy and Guidance, Chemistry and Behavior, Occurrence, Toxicology, Site Characterization and Remediation Technologies (USEPA 2017h).

The scientific community is rapidly recognizing and evolving its understanding of the environmental and health impacts associated with the release of PFAS. Certain PFAS, most notably some of the perfluoroalkyl acids (PFAAs), such as perfluorooctanoate (PFOA) and perfluorooctane sulfonate (PFOS), are mobile, persistent, and bioaccumulative, and are not known to degrade in the environment (USEPA 2003b; ATSDR 2015; NTP 2016; Concawe 2016).

Understanding the manufacturing history of PFAS, as well as past and current uses, allows for the identification of potential environmental sources of PFAS, possible release mechanisms, and associated pathway-receptor relationships.

ITRC has developed a series of six fact sheets to summarize the latest science and emerging technologies regarding PFAS. The purpose of this fact sheet is to:

- provide an overview of the discovery and development of PFAS and the subsequent detection of PFAS in the environment
- describe emerging concerns of potential adverse human health effects, and efforts to reduce use or replace with alternate formulations, or both
- identify the major sources of PFAS in the environment, as well as other sources of PFAS to the environment that may be of interest

2 Discovery and Manufacturing History

PFAS are a complex family of more than 3,000 manmade fluorinated organic chemicals (Wang et al. 2017) that have been produced since the mid-20th century, although not all of these may be currently in use or production. Table 2-1 provides a general timeline of initial synthesis and commercial production of some of the more well-known PFAS, along with some of the more frequently associated products.

PFAS are produced using several different processes. Two major processes have been used to manufacture fluorosurfactants (includes PFAAs) and side-chain fluorinated polymers: electrochemical fluorination (ECF) and telomerization (KEMI 2015). ECF was licensed by 3M in the 1940s (Banks, Smart, and Tatlow 1994), and used by 3M until 2001. ECF produces a mixture of even- and odd- numbered carbon chain lengths of approximately 70% linear and 30% branched substances (Concawe 2016). Telomerization was developed in the 1970s (Benskin 2011), and yields mainly even numbered, straight carbon chain isomers (Kissa 2001; Parsons et al. 2008).

Table 2-1. Discovery and manufacturing history of select PFAS

PFAS ¹	Development Time Period							
	1930s	1940s	1950s	1960s	1970s	1980s	1990s	2000s
PTFE	Invented	Non-Stick Coatings			Waterproof Fabrics			
PFOS		Initial Production	Stain & Water Resistant Products	Firefighting foam				U.S. Reduction of PFOS, PFOA, PFNA (and other select PFAS ²)
PFOA		Initial Production	Protective Coatings					
PFNA					Initial Production	Architectural Resins		
Fluoro-telomers					Initial Production	Firefighting Foams		
Dominant Process ³		Electrochemical Fluorination (ECF)						Fluoro-telomerization (shorter chain ECF)
Pre-Invention of Chemistry /			Initial Chemical Synthesis / Production			Commercial Products Introduced and Used		
Notes: 1. This table includes fluoropolymers, PFAAs, and fluorotelomers. PTFE (polytetrafluoroethylene) is a fluoropolymer. PFOS, PFOA, and PFNA (perfluorononanoic acid) are PFAAs. 2. Refer to Section 3.4. 3. The dominant manufacturing process is shown in the table; note, however, that ECF and fluorotelomerization have both been, and continue to be, used for the production of select PFAS.								
Sources: Prevedouros et al. 2006; Concawe 2016; Chemours 2017; Gore-Tex 2017; US Naval Research Academy 2017								

3 Health and Environmental Investigations

3.1 Initial Studies

Studies that found some PFAS in the blood of occupationally exposed workers in the 1970s and reported detections in the blood of the general human population in the 1990s (Buck et al. 2011) led to increased awareness of PFAAs in the environment, associated human exposure, and the potential for health effects. PFAAs (such as PFOS and PFOA) are found in the blood and serum of most people whether exposed in the workplace or not. This is attributed to widespread use, ability to bind to blood proteins and long half-lives in humans (Kannan et al. 2004; Karrman et al. 2006; Olsen et al. 2003). Laboratory studies using animals and epidemiological studies of human populations show that exposure to some PFAS may be associated with a wide range of adverse human health effects (USEPA 2016b, c; ATSDR 2017).

Although some PFAS have been manufactured for more than 50 years, PFAS were not widely documented in environmental samples until the early 2000s. Early detection at low reporting limits was hindered due to analytical capability challenges arising from the unique surface-active properties of PFAS (Giesy and Kannan 2001; 3M 2000). Many manuscripts have since been published showing widespread distribution of certain PFAS, such as PFAAs, in various matrices including sediments, surface and groundwater, wildlife, and human blood (whole, plasma, and serum) (Kannan et al. 2004; Yamashita et al. 2005; Higgins et al. 2005; Rankin et al. 2016). Some PFAS (such as PFAAs) are found in many places throughout the globe, even in areas well beyond where they were initially used or manufactured (Houde et al. 2011).

3.2 Emerging Awareness

The awareness and emphasis on various PFAS have evolved. Early focus was on the longer-chain (see Section 3.4), perfluoroalkyl acids (PFAAs). In 2016, USEPA issued a Lifetime Health Advisory (LHA) for two of the most widely detected PFAAs, PFOA and PFOS. Set at 70 nanograms per liter (ng/L, equivalent to parts per trillion [ppt]), the LHA applies for each PFAA, as well as in combination, in drinking water (USEPA 2016d).

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Six additional PFAAs have recently gained attention after their inclusion in the USEPA Unregulated Contaminant Monitoring Rule (UCMR). The third round of monitoring, or UCMR3, was promulgated in 2012 for tracking chemicals suspected to be present in drinking water, but do not have health-based standards set by the Safe Drinking Water Act. A summary of the occurrence data is included in the *Regulations, Guidance, and Advisories Fact Sheet*.

Many state regulatory agencies now request or require an expanded list of perfluoroalkyl substances (short and long chain), and fluorotelomers and polyfluoroalkyl substances are also receiving increased attention. This progression is illustrated in Figure 3-1.

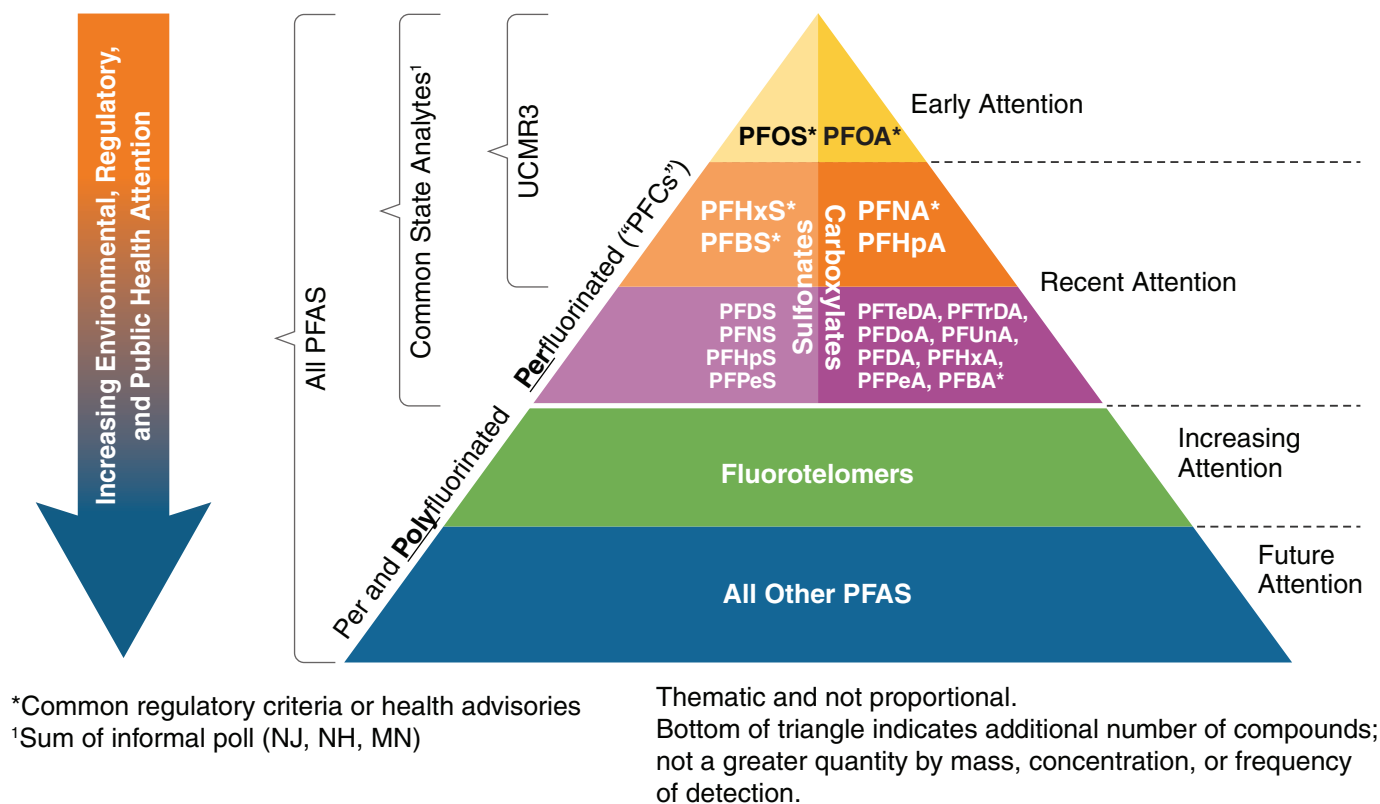


Figure 3-1. Emerging awareness and emphasis on PFAS occurrence in the environment

(Source: J. Hale, Kleinfelder, used with permission)

3.3 Stockholm Convention on Persistent Organic Pollutants

The Stockholm Convention on Persistent Organic Pollutants (POPs) is a United Nations treaty signed in 2001 aimed at reducing or eliminating the production, use, and release of key POPs. POPs are defined as synthetic, organic compounds that, to varying degrees, resist photolytic, biological, and chemical degradation (KEMI 2004 and 2005; USEPA 2017k).

In 2009, *Annex B of the Stockholm Convention* was amended to include PFOS, because it is persistent in the environment and is not known to degrade at any environmental condition. At this time, the U.S. has not ratified the amendment (KEMI 2017). According to the Stockholm Convention website, PFOA and PFHxS (perfluorohexane sulfonic acid) are currently proposed for listing.

3.4 Phase-out of Long-Chain PFAS

Due to industry and regulatory concerns about the potential health and environmental impacts, there has been a reduction in the manufacture and use of long-chain PFAAs. Long-chain PFAAs include perfluorinated carboxylates (PFCAs) with eight or more fully fluorinated carbons (for example, PFOA) and perfluorinated sulfonates (PFSAs) with six or more fully fluorinated carbons (for example, PFHxS and PFOS), their salts, and precursor compounds capable of forming long-chain PFAAs (USEPA 2009a; Buck et al. 2011; OECD 2013; Wang et al. 2015).

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- In May 2000, 3M, the principal worldwide manufacturer and sole U.S. manufacturer of PFOS, announced a voluntary phase-out of perfluorooctanyl chemistries, which included PFOS, PFHxS, PFOA, and related precursors. 3M reportedly completed most of the phase-out by the end of 2002, with the remaining phase-out completed by 2008 (USEPA 2003b; USEPA 2017e; 3M 2017b).
- USEPA issued Significant New Use Rules (SNURs) under the Toxic Substances Control Act (TSCA) to restrict any future use or production of 183 PFAS, which include 88 of the PFOS-related chemicals phased-out by 3M. However, the SNURs allowed for continued, low-volume use of some of these PFAS in the photographic/imaging, semiconductor, etching, metal plating, and aviation industries. Also, due to the long shelf-life of PFOS-based fire-fighting foams, they may still be stored and in use at various facilities (see Section 4.2) (USEPA 2007). SNURs for some long-chain PFCAs and PFASs have been proposed (USEPA 2015b).
- In January 2006, USEPA initiated the PFOA Stewardship Program (USEPA 2006b). The eight major manufacturing companies committed to reducing PFOA, other longer-chain PFCAs (such as perfluorononanoic acid [PFNA] and perfluoroundecanoic acid [PFUnA]), and related precursors (for example, 8:2 fluorotelomer alcohol [FTOH]) that could be converted to these PFCAs from their global facility emissions and product content. USEPA indicates all eight companies successfully satisfied the program goals, meeting a 95% reduction by 2010, and elimination by 2015 (USEPA 2017e). Even though the program goals were met, materials imported to the United States may contain these PFCAs and related precursors.

PFAS are manufactured globally. Recently increased production of PFOA and related PFAS in China, India, and Russia have potentially offset the global reduction anticipated with the U.S. phase-out (OECD 2015b). PFAS manufacture began in China in the 1980s (World Bank 2017a, b), and PFOS production in China increased with the long-chain PFAA phase-out in the United States (Concawe 2016). In 2016, PFOS and its derivatives were still being produced in Germany, Italy, and China (Witteveen+Bos and TTE 2016), but by early 2017, China was the only known producer of PFOS. China has ratified the Stockholm Convention on POPs and a grant from Global Environment Facility (GEF) was approved in 2017 to support the reduction of PFOS in China (World Bank 2017a).

3.5 Replacement Chemistry

Manufacturers have been developing replacement technologies, including reformulating or substituting longer-chain substances with shorter-chain perfluoroalkyl or polyfluorinated substances that include, but are not limited to, compounds produced with ECF and fluorotelomerization, such as: FTOH, perfluorobutane sulfonyl fluoride (PBSF)-based derivatives (for example, perfluorobutane sulfonate [PFBS] in lieu of PFOS), polyfluoroethers (for example, GenX and ADONA used in the manufacture of fluoropolymers) and other types of PFAS (Hori et al. 2006; OECD 2007; Herzke, Olson, and Posner 2012; Wang et al. 2013; Wang et al. 2015; Holmquist et al. 2016).

Many long-chain PFAS alternatives are structurally similar to their predecessors and manufactured by the same companies (Concawe 2016; Wang et al. 2015). However, it is not yet clear if some of these chemicals can achieve the same performance effectiveness of some of their predecessors. For example, a 2015 study concluded that there are no non-fluorinated alternatives that provide equivalent technical performance in textiles (Danish EPA 2015).

Several studies suggest some of the replacement PFAS may or may not be less hazardous than the long-chain predecessors, although publicly available information on most replacement chemicals is limited (Wang et al. 2015; RIVM 2016). Documentation regarding the USEPA's review of hundreds of "shorter chain-length PFAS telomeric" substitutes for long-chain PFAS is available under the TSCA New Chemicals Program (OECD 2013; USEPA 2017g). Other documentation regarding replacement chemistries is available from the FluoroCouncil (2017).

Information on environmental contamination by replacement PFAS is limited, and most are not detected by standard analytical methods (Wang et al. 2013). Treatment processes used to remove these chemicals from waste streams may not be as effective as with longer-chain PFAS (Sun et al. 2016).

4 Major Sources of PFAS in the Environment

PFAS are used in many industrial and consumer applications. Major sources may have released PFAS into the environment and impacted drinking water supplies in many areas of the United States (Environmental Working Group and Northeastern University Social Science Environmental Health Research Institute 2017).

4.1 Production and Manufacturing Facilities

Both in the United States and abroad, primary manufacturing facilities produce PFAS and secondary manufacturing facilities use PFAS to produce goods. Due to the solubility and persistence of many PFAS, environmental release mechanisms associated with these facilities include air emission and dispersion, spills, and disposal of manufacturing wastes and wastewater. Potential impacts to air, soil, surface water, stormwater, and groundwater are present not only at release areas but potentially over the surrounding area (Shin et al. 2011). Table 4-1 summarizes potential major sources of PFAS releases to the environment based on the manufacturing sector. Manufacturing of commercial products and consumer goods summarized in Section 5 may also be environmental sources but are not included in this table.

Table 4-1. Potential major manufacturing sources of PFAS releases to the environment

Sector	Example Uses	References
Textiles & Leather	Factory- or consumer-applied coating to repel water, oil, and stains. Applications include protective clothing and outerwear, umbrellas, tents, sails, architectural materials, carpets, and upholstery.	Rao and Baker 1994; Hekster, Laane, and de Voogt 2003; Brooke, Footitt, and Nwaogu 2004; Poulsen et al. 2005; Prevedouros et al. 2006; Walters and Santillo 2006; Trudel et al. 2008; Guo et al. 2009; USEPA 2009a; Ahrens 2011; Buck et al. 2011; UNEP 2011; Herzke, Olsson, and Posner 2012; Patagonia 2015; Kotthoff et al. 2015; ATSDR 2015
Paper Products	Surface coatings to repel grease and moisture. Uses include non-food paper packaging (for example, cardboard, carbonless forms, masking papers) and food-contact materials (for example, pizza boxes, fast food wrappers, microwave popcorn bags, baking papers, pet food bags).	Rao and Baker 1994; Kissa 2001; Hekster, Laane, and de Voogt 2003; Poulsen et al. 2005; Trudel et al. 2008; Buck et al. 2011; UNEP 2011; Kotthoff et al. 2015; Schaidler et al. 2017
Metal Plating & Etching	Corrosion prevention, mechanical wear reduction, aesthetic enhancement, surfactant, wetting agent/fume suppressant for chrome, copper, nickel and tin electroplating, and post-plating cleaner.	USEPA 1996; USEPA 1998; Kissa 2001; Prevedouros et al. 2006; USEPA 2009b; UNEP 2011; OSHA 2013; KEMI 2015; Danish EPA 2015
Wire Manufacturing	Coating and insulation.	Kissa 2001; van der Putte et al. 2010; ASTSWMO 2015
Industrial Surfactants, Resins, Molds, Plastics	Manufacture of plastics and fluoropolymers, rubber, and compression mold release coatings; plumbing fluxing agents; fluoroplastic coatings, composite resins, and flame retardant for polycarbonate.	Kissa 2001; Renner 2001; Poulsen et al. 2005; Fricke and Lahl 2005; Prevedouros et al. 2006; Skutlarek, Exner, and Farber 2006; van der Putte et al. 2010; Buck et al. 2011; Herzke, Olsson, and Posner 2012; Kotthoff et al. 2015; Miteni 2016; Chemours 2017
Photolithography, Semiconductor Industry	Photoresists, top anti-reflective coatings, bottom anti-reflective coatings, and etchants, with other uses including surfactants, wetting agents, and photo-acid generation.	SIA 2008; Choi et al. 2005; Rolland et al. 2004; Brooke, Footitt, and Nwaogu 2004; van der Putte et al. 2010; UNEP 2011; Herzke, Olsson, and Posner 2012

4.2 Class B Fluorine-Containing Firefighting Foams

Class B fluorine-containing firefighting foams (firefighting foam) for extinguishing flammable liquid fires include aqueous film forming foam (AFFF), fluoroprotein (FP), and film forming fluoroprotein foam (FFFP) (Concawe 2016). These foams have been stored and used for fire suppression, fire training, and flammable vapor suppression at hundreds of military installations and civilian airports (Hu et al. 2016), as well as at petroleum refineries and storage facilities, and chemical manufacturing plants throughout the United States. Additionally, local fire departments in communities have used and maintained quantities of firefighting foam in their inventories. Despite the phase-out of longer-chain PFAAs, these products still have long-chain PFAA constituents in firefighting foam due to the long shelf-life of these products. Facilities that manufactured firefighting foams are also potential sources.

Firefighting foams are a complex mixture of both known and unidentified PFAS of differing molecular structures present in varying proportions. Foams were produced to meet firefighting specifications, rather than formulated to contain a specified mixture of PFAS. These types of firefighting foams have been in use since the 1960s. The United States Naval Research Laboratory began research on the development of firefighting foams in the 1960s, which led to advancements in performance and increased safety (U.S. Naval Research Laboratory 2017). Fluorotelomer foams have been in use since the 1970s and became the predominant foam after 2001 when long-chain ECF-based foams were discontinued.

Firefighting foams are released into the environment through a variety of practices and mechanisms (Anderson et al. 2016; Hale 2016):

- low volume releases of foam concentrate during storage, transfer or equipment calibration
- moderate volume discharge of foam solution for apparatus testing
- occasional, high-volume, broadcast discharge of foam solution for firefighting and fire suppression/prevention
- periodic, high volume, broadcast discharge for fire training
- leaks from foam distribution piping between storage and pumping locations

Firefighting foam is applied by mixing foam concentrate and water to make foam solution. When applied to a fire, the foam solution is aerated at the nozzle, yielding finished foam. Thousands of gallons of foam solution may be applied during a given event. Figure 4-1 illustrates the use of firefighting foam, how it may be released to the environment, and potentially affected media.

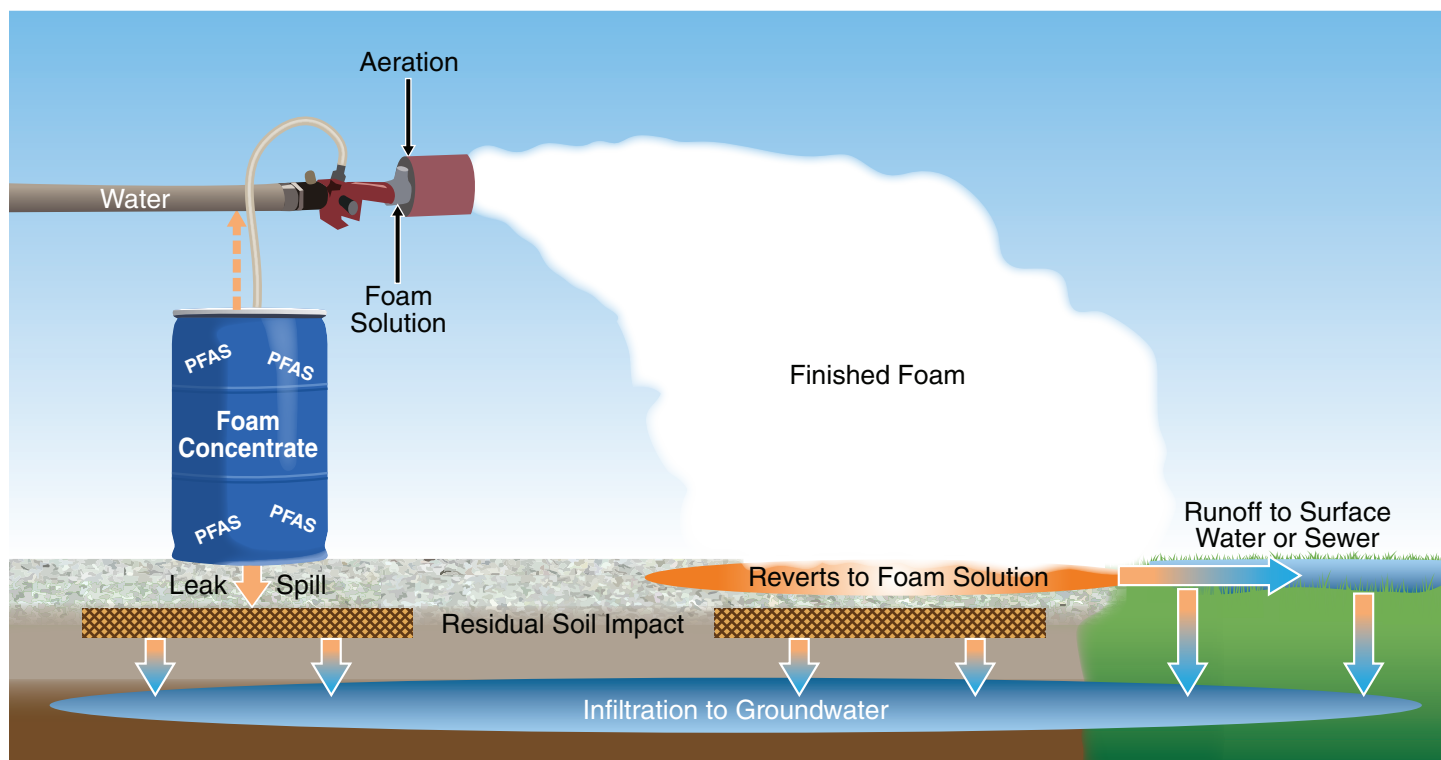


Figure 4-1. Release of firefighting foam

(Source: Adapted from figure by J. Hale, Kleinfelder, used with permission)

The U.S. Department of Defense (USDOD) has undertaken an evaluation of potential firefighting foam contamination at its facilities nationwide (Anderson et al. 2016). Similar efforts have been undertaken by some states. For example, the Minnesota Pollution Control Agency (MPCA) conducted a state-wide survey of firefighting foam use at training sites. Working with the State Fire Chief Association, the MPCA identified more than two dozen locations where Class B foams were likely used in firefighting training (Antea Group 2011).

4.3 Waste Disposal

Disposal of wastes generated during primary PFAS production and secondary manufacturing using PFAS can be sources of PFAS environmental contamination. As PFAS manufacturing processes change with time, the resulting type and composition of waste streams also change. Given that PFAS production and use began several decades before the enactment of federal and state regulations governing waste disposal (for example, Resource Conservation and Recovery Act [RCRA] in 1976 [USEPA 2017d]), environmental impacts, including impacted drinking water supplies, from disposal of legacy PFAS industrial waste have been documented (Shin et al. 2011; MPCA 2017).

Leachate from some municipal solid waste landfills has been shown to be a source of PFAS release to the environment, with the presence of some PFAS reportedly due to the disposal of consumer goods treated with hydrophobic, stain-resistant coatings (Busch et al. 2010; Eggen, Moeder, and Arukwe 2010). PFAS composition and concentration in leachates vary depending on waste age, climate, and waste composition (Allred et al. 2015; Lang et al. 2017).

The evolution of waste reduction and landfill technology has provided significant protection to human health and the environment (Hickman 1999). Leachate collection systems are essential to providing systematic transport of leachate to a central location for recirculation, treatment, or offsite treatment (Arabi and Lugowski 2015). Leachate treatment by wastewater treatment plants (WWTPs) is common prior to discharge to surface water, or distribution for agricultural or commercial use (Lang 2016). Standard WWTP technologies may do little to reduce or remove PFAS and discharge of landfill leachate treated at WWTPs represents a secondary source of certain PFAS release to the environment (Ahrens et al. 2015; CRC Care 2017).

4.4 Wastewater Treatment

Consumer and industrial use of PFAS-containing materials, including disposal of landfill leachate and firefighting foam, results in the discharge of PFAS to WWTPs. WWTPs, particularly those that receive industrial wastewater, are possible sources of PFAS release. (Lin, Panchangam, and Lo 2009; Ahrens et al. 2009).

4.4.1 WWTP Operations

Conventional sewage treatment methods do not efficiently remove PFAAs (Ahrens et al. 2011; Schultz et al. 2006). Evaluation of full-scale WWTPs has indicated that conventional primary (sedimentation and clarification) and secondary (aerobic biodegradation of organic matter) treatment processes, can result in changes in PFAS concentrations and classes (for example, an increase in the concentrations of PFAAs in effluent, presumably from degradation of precursor PFAS) (Schultz et al. 2006).

Some PFAS are frequently detected in WWTP effluent (for example, PFOA and PFBS), with concentrations of some PFAS ranging up to hundreds of ng/L; effluents are believed to be major point sources of these chemicals in surface water (Ahrens 2011). Hu et al. (2016) demonstrated that the presence of WWTPs in an area was predictive of the presence of PFOS and PFOA in drinking water. PFOS and PFOA are two of the most frequently detected PFAS in wastewater. (Hamid and Li 2016). Using WWTP effluent-impacted surface water as a source of tap water can, in turn, recycle the PFAS back to the WWTP, recirculating PFAS in the water cycle (Hamid and Li 2016).

At some WWTPs, studies have shown concentrations of PFAS in ambient air to be 1.5 to 15 times greater than reference sites (Ahrens et al. 2011). PFAS distribution (primarily PFAAs and FTOH, with higher concentrations of FTOH) changes based on the specific PFAS sources in the effluent and the type of treatment methods employed at the WWTP. Lagoon systems contain a greater fraction of PFAAs.

4.4.2 Biosolids

PFAS (measured as PFCAs and PFSAAs) have been found in domestic sewage sludge (Higgins et al. 2005). USEPA states that more than half of the sludge produced in the United States is applied to agricultural land as biosolids, therefore biosolids application can be a source of PFAS to the environment (USEPA 2017n). The most abundant PFAS found in biosolids (PFOS and PFOA) are the same as in WWTP effluent; however, biosolids may also contain other long-chain PFAS (Hamid and Li 2016). Application of biosolids as a soil amendment can result in a transfer of PFAS to soil

(Sepulvado et al. 2011). These PFAS can then be available for uptake by plants and soil organisms. There are indications that PFAAs can enter the food chain through the use of biosolids-amended soil (Lindstrom et al. 2011; Blaine et al. 2013; Blaine et al. 2014; Navarro et al. 2017). Further studies show that PFAS concentrations can be elevated in surface and groundwater in the vicinity of agricultural fields that received PFAS contaminated biosolids for an extended period of time (Washington et al. 2010).

5 Other Sources of PFAS—Use of Commercial and Consumer Products

PFAS are widely used in consumer products and household applications, with a diverse mixture of PFAS found in daily use in varying concentrations (Clara et al. 2008; Trier, Granby, and Christensen 2011; Fujii, Harada, and Koizumi 2013; OECD 2013; ATSDR 2015; Kotthoff et al. 2015; KEMI 2015; USEPA 2016b, c).

Environmental releases associated with the use of commercial and consumer products are primarily related to management of solid waste (for example, disposal of used items in a municipal solid waste [MSW] landfill), and wastewater disposal (for example, discharge to WWTPs, private septic systems, or other subsurface disposal systems).

As increased environmental sampling for PFAS occurs, it is likely that additional sources may emerge. Studies have shown that physical degradation of some consumer products (such as PFAS-treated paper, textiles, and carpets) may be a source of PFAS in house dust (Bjorklund, Thuresson, and de Wit 2009). Additionally, studies have also shown that professional ski wax technicians may have significant inhalation exposures to PFAS (Nilsson et al. 2013) and snowmelt and surface waters near ski areas may have measurable PFAS impacts (Kwok et al. 2013).

6 References and Acronyms

The references cited in this fact sheet, and the other ITRC PFAS fact sheets, are included in one combined list that is available on the ITRC web site. The combined acronyms list is also available on the ITRC web site.

Commercial and Consumer Products Containing PFAS:

- paper and packaging
- clothing and carpets
- outdoor textiles and sporting equipment
- ski and snowboard waxes
- non-stick cookware
- cleaning agents and fabric softeners
- polishes and waxes, and latex paints
- pesticides and herbicides
- hydraulic fluids
- windshield wipers
- paints, varnishes, dyes, and inks
- adhesives
- medical products
- personal care products (for example, shampoo, hair conditioners, sunscreen, cosmetics, toothpaste, dental floss)



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November 2017



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