

Review

Research and Regulatory Advancements on Remediation and Degradation of Fluorinated Polymer Compounds

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Abstract: Per- and polyfluoroalkyl substances (PFAS) are a class of chemicals used in various commercial industries to include food packaging, non-stick repellent, and waterproof products. International environmental protection agencies are currently looking for ways to detect and safely remediate both solid and aqueous PFAS waste due to their harmful effects. Incineration is a technique that disposes of chemicals by breaking down the chemicals at high temperatures, upwards of 1400 °C. Incineration has been used on other related compounds, but PFAS presents a challenge during thermal degradation due to the molecular stability and reactivity of fluorine. Research on the efficacy of this method is currently limited, as the degradation byproducts of PFAS are not fully characterized. Current research is mostly focused on the development of benchtop methods for the safe remediation of solid PFAS waste. Aqueous fire fighting foams (AFFFs) have garnered significant attention due to extensive use since development in the 1960s. Numerous communities that are closely located near airports have been shown to have higher than average PFAS contamination from the repeated use. Detection and remediation of surface, subsurface, and wastewater have become a primary concern for environmental agencies. Use of electrochemical techniques to remove the PFAS contaminants has shown recent promise to help address this issue. Critical to the remediation efforts is development of standardized detection techniques and the implementation of local and international regulations to control the production and use of fluorinated products. No single solution has yet been developed, but much progress has been made in recent years in governmental regulation, detection, and remediation techniques.

Keywords: per- and polyfluoroalkyl substances (PFAS); fluoropolymers; thermal degradation analysis; incineration; combustion; electrochemical remediation

1. Introduction

Scope of Review. Per- and poly-fluoroalkyl substances (PFAS) encompass a group of several thousand compounds of different carbon chain lengths, a varying number of fluorine atoms, and different functional groups [1]. Over recent decades, fluorinated compounds of innumerable classifications have become ever more prevalent in both scope of use and emerging as environmental

hazards [2–5]. PFAS compounds are desirable in many applications because of the carbon-fluorine (C-F) bond which yields a high degree of thermal stability [6] and various other advantageous physical properties that lend their use in a variety of common commercial products from waterproof clothing, fire resistant materials, stain resistant carpets, and long-lasting rubber components [7–20]. Despite many desirable applications of fluorinated compounds, there is growing concern that they present an emerging environmental threat [21–23]. Much effort has been devoted to both the detection and environmental remediation techniques to identify and remove fluorinated compounds from aqueous and soil environments. This review focuses on classification, sources of environmental contamination, efforts to use thermal degradation for solid waste, the impact of aqueous fire fighting foam (AFFF) as a contaminant to the environment, electrochemical techniques for aqueous remediation, and environmental regulatory guidelines, with a special focus on emerging technologies developed over the last five years.

1.1. Discovery

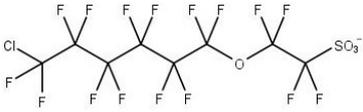
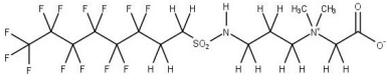
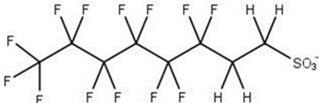
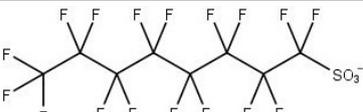
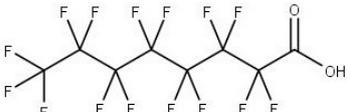
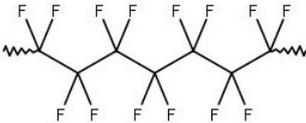
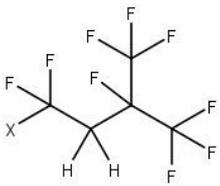
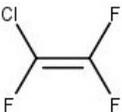
Fluoropolymers have existed for nearly a century and are popular in various commercial, industrial, and military products because of their ability to create water resistant, low friction coatings, and flame-retardant products [1]. Polymers that comprise fluorine atoms on the carbon backbone have unique physical properties when compared to a hydrocarbon polymer. The replacement of hydrogen atoms with fluorine contributes to the overall stability of the fluoropolymers due in part to the significant difference in the average bond energy between the C-F bond (565 kJ/mol) and the C-H bond (414 kJ/mol), due to the high electronegativity of fluorine [24,25]. The overall stability of the fluoropolymers arises from the strength of the C-F bond, which is one of the strongest single bonds [26]. The added bond strength and stability of the adjacent C-F and C-C bonds arises from the multiple C-F bonds on the same geminal carbons on the skeletal backbone of the polymer which induces a strong positive partial charge on the carbon atoms [25]. The intramolecular dipole moments within the structure of fluorocarbons, specifically within the individual C-F bonds, cancel each other resulting in non-polar dispersion intermolecular forces across an overall large molecular cross-sectional area that results in hydrophobicity of the fluorinated moiety [27]. Additional key properties of fluoropolymers are their tendency to possess high chemical and thermal stability and low coefficient of friction which are critical properties in the use and application of such products as polytetrafluoroethylene (PTFE). The history of the fluoropolymer industry and its widespread use began with the discovery of (PTFE) by R.J Plunkett at Dupont in 1938 [28], to advancements in characterization of physical and chemical properties of other fluorinated polymers over recent decades, and now to the modern-day uses, novel-synthesis methods, industrial viability, and general environmental concerns of the fluoropolymer industry in the 21st century [7,20,29].

1.2. Classification

Fluorinated polymers encompass all polymers for which one or more of the monomer units contains the element F; they may or may not be PFASs, depending on whether they contain perfluoroalkyl moiety [30]. PFAS are often comprised of non-polar fluorinated alkyl chain and polar functional groups that provide a reactive site for the given PFAS molecule. There are vast differences in characteristics and properties of PFAS within each of its classes of compounds. Consistent terminology has been provided to standardize PFAS for a variety of categories to include perfluoroalkyl substances, polyfluoroalkyl substances, fluoropolymers, perfluoropolyethers, side-chain fluorinated polymers, short-chain PFAS, and long-chain PFAS [30]. Specifically, the U.S. Environmental Protection Agency (EPA) and the international Organization for Economic Cooperation and Development (OECD) both define long-chain PFAS as perfluoroalkyl carboxylic acids (PFCAs) with seven or more perfluoroalkyl carbons, perfluoroalkane sulfonic acids (PFSA) with six or more perfluoroalkyl carbons, and precursors such as perfluoroalkane sulfonyl fluoride (PASF) or fluorotelomer (FT) based substances that have the potential to transform into long-chain PFAS [31]. Table 1 displays select classes of PFAS compounds

mostly used in household or commercial applications. Within the PFAS group are several different categories of compounds, one of which is fluoropolymers that consist of a carbon-only polymer backbone with fluorine atoms surrounding the carbon atoms [6].

Table 1. Relevant classes, chemical structure, and common sources of per- and poly-fluoroalkyl substances (PFAS).

Class	Molecular Structure	Sources	References
Non-polymers	 <p>Chlorinated polyfluorinated ether sulfonate (F53-B)</p>	<ul style="list-style-type: none"> • Fire Fighting Foams • Chrome Plating Waste 	[12,16]
	 <p>6:2 Fluorotelomer sulfonamide alkylbetaine (FTAB)</p>	<ul style="list-style-type: none"> • Fire Fighting Foams • Ceramic Coating 	[3,8,9,11]
	 <p>6:2 Fluorotelomer sulfonate (FTSA)</p>	<ul style="list-style-type: none"> • Fire Fighting Foams • Mist Suppressant • Metal Plating • Ink and Toner 	[7,11]
	 <p>Perfluorooctanesulfonic acid (PFOS)</p>	<ul style="list-style-type: none"> • Cleaning Products • Metal Plating • Hydraulic Fluids • Fabric Protector 	[14,15,17]
	 <p>Perfluorooctanoic acid (PFOA)</p>	<ul style="list-style-type: none"> • Water Filtration • Non-stick Cookware • Stain-resistant Fabrics • Gore-Tex 	[13]
Polymers	 <p>Polytetrafluoroethylene (PTFE)</p>	<ul style="list-style-type: none"> • Polymer Gaskets • Chemically Resistant Components • Carpet Coatings 	[6,19,20]
Polymers	 <p>Viton A/B</p>	<ul style="list-style-type: none"> • Viton A: • General sealing • Auto/Aero Fuels • Lubricants • Viton B: Gaskets • Power-utility seals • Chemical Processing 	[10,18]
	 <p>Polychlorotrifluoroethylene (KEL-F)</p>	<ul style="list-style-type: none"> • Moisture Resistant Packaging • Cryogenic Seals and Gaskets • Lubricants for Corrosive Materials 	[19]

1.3. Thermal Degradation and Remediation for Solid Waste

The rising concern over perfluorooctanoic acid (PFOA) and perfluorooctanesulfonic acid (PFOS) has led to models being developed to understand how PFOA, PFOS, and similar compounds undergo thermal and environmental degradation [32]. Various studies have also been performed to assess the combustion of fluoropolymers and their potential to degrade in the environment to various, smaller PFAS compounds that are considered persistent organic pollutants (POPs) [2,6,33–40]. While PFOS and PFOA are non-polymers and were previously used in AFFF, fluoropolymers

also have an important use in the plastics and coatings industry. Specifically, PFAS polymers are used as binders in plastic/polymer bonded explosives (PBX) and in munition components such as gaskets, wiring sheaths, tubing, and seals, with the most prevalent materials being PTFE, vinylidene difluoride/hexafluoropropylene copolymer (Viton A, and Viton B), and polychlorotrifluoroethylene (Kel-F 800). Several studies have looked at the thermal decomposition behavior of PBX, utilizing various instruments to measure elastic and mechanical properties in order to better understand the long-term, high-temperature stability, and compatibility of the explosive mixture of 1,3,5,7-tetranitro-1,3,5,7-tetrazocane (HMX)/1,3,5-triamino-2,4,6-trinitrobenzene (TATB), and Viton A [41–43]. As shown in Table 1, PTFE has a $-CF_2-$ backbone and is terminated on both ends of the polymer with $-CF_3$.⁶ Viton A, Viton B, and Kel-F 800 have a similar structure to PTFE; however, they exhibit slight differences such as that Kel-F 800 also includes chlorine atoms in its structure.

There are currently several methods used to destroy various PFAS waste such as end-of-life fluoropolymers in various materials and include, but are not limited to, incineration, pyrolysis, and smoldering, also referred to as smoldering combustion. Incineration is a high-temperature flaming process that typically occurs in an open air space, while smoldering is a flameless combustion process that occurs on the surface of a condensed fuel [1]. While both methods can be used to treat fluorinated waste, smoldering is a self-sustaining and is, therefore, more energy efficient compared to incineration. Pyrolysis is similar to incineration in that it is a high-temperature process used for thermal decomposition, except it occurs in an inert atmosphere [44]. Critical to the effectiveness of this processes is achieving a temperature greater than 900 °C [1]. The goal of these methods pertaining to PFAS waste is to ensure that these fluoropolymers are exposed to a sufficient and sustainable temperature that ensures successful destruction of toxic PFAS compounds, as the fluoropolymers thermally degrade while still being efficient and safe. Several studies have tested the efficacy of these methods to determine if they reach temperatures high enough for a sufficient duration to allow for complete degradation [45,46]. Alternative methods to incineration such as mineralization of fluorinated compounds during thermal treatment, or a plasma based water treatment, are also being investigated as they may be safer and more efficient to treat waste [1,47,48].

Recently, there are concerns amongst communities regarding side effects from exposure at impacted locations where thermal degradation of PFAS compounds from disposal of munitions using various methods such as open burning (OB), open detonation (OD), and incineration. For example, citizens near the Holston Army Ammunition Plant in Kingsport, TN, USA; the Environmental Law Clinic at the University of California at Berkeley; and Public Employees for Environmental Responsibility (PEER) have all petitioned the Environmental Protection Agency (EPA) to prohibit burning of PFAS-contaminated munition wastes due to the risks that it poses, claiming that the current methods do not provide temperatures sufficient to destroy PFAS but rather break fluoropolymers down into smaller, toxic PFAS compounds that are released into the environment [49,50]. Research is needed, however, to determine if emissions from thermal disposal of munitions, at temperatures consistent with OB, OD, or incineration contain PFAS. The Holston Plant is one example, but PFAS compounds resulting predominantly from the use of AFFF or disposal of manufacturing waste have been identified in soils, groundwater, drinking water, animals, and humans in locations throughout the world [4,5,51,52].

Moreover, methods of the thermal degradation and waste treatment of polymers containing PFAS can be greatly expanded on to enable environmentally safe and conscientious processes. The EPA has two approved methods available to test for 29 PFAS substances in drinking water, while the Department of Defense (DoD) and Department of Energy (DoE) jointly created a quality systems manual (QSM) and data validation procedures for laboratories performing analytical testing of PFAS compounds [53–56]. While standards and guidelines have been published by the EPA on identifying and testing PFAS-impacted areas, there is still much to learn on how to reduce the footprint for PFAS.

1.4. Environmental

Over the past two decades, concern has increased over the adverse impact PFAS compounds have on human health and the environment as a probable connection between PFAS exposure and diseases such as cancer and thyroid disease [49,57–59]. Two of the most notable PFAS non-polymers that have garnered considerable attention are PFOA and PFOS due to their previous use as part of aqueous film forming foams (AFFFs) used to fight fires [3,32,60–74]. Research to determine how PFAS compounds affect health is still relatively limited; however, numerous studies have been completed on AFFF-impacted sample areas to determine the extent to which these toxic compounds have accumulated in soil, wastewater, animals, and humans [67,68,70].

The Environmental Protection Agency (EPA) is the lead organization in identifying, monitoring, and regulating PFAS throughout the United States. Most recently, the EPA released an action plan in February 2019 to address PFAS concerns [23]. Moreover, in February 2020, the EPA updated the action plan on addressing PFAS contaminant detection with various methods to include testing for 29 PFAS chemicals in drinking water via EPA Methods 533 [53] and 537.1 [54] and testing for PFAS in non-potable water via SW-846 Method 8327 [75]. They are also updating the Significant New Use Rule (SNUR) which requires PFOA manufacturers to notify the EPA at least 90 days in advance before manufacturing with these chemicals [76]. In addition to testing, the EPA is supporting government agency cleanup efforts, publishing interim guidance on the proper destruction and disposal of PFAS using high temperature incineration, and taking measures against facilities that violate current PFAS manufacturing requirements. They are also working alongside the U.S. Department of Agriculture (USDA) and Food and Drug Administration (FDA) to develop analytical methods for agricultural and food products. Lastly, to better understand human health hazards associated with PFAS, they are currently researching human health effects associated with seven common PFAS. Overall, the EPA will continue to make progress on addressing PFAS detection and remediation under guidelines established from the Safe Drinking Water Act (SDWA) [77] passed by Congress in 1974 (amended in 1986 and 1996); the Comprehensive Environmental Response, Compensation, and Liability Act (CERCLA) [78] passed in 1980 (amended in 1986); the Resource Conservation and Recovery Act (RCRA) [79] passed in 1976 (amended in 1984 and 1986); and the Toxic Substances Control Act (TSCA) [80] passed in 1976 but amended in 2016 by the Frank R. Lautenberg Chemical Safety for the 21st Century Act [81].

The European Environment Agency (EEA), which has 32 European member states, has also taken measures to regulate and monitor PFAS, specifically PFOS, throughout countries within the European Union (EU) [21]. Persistent organic pollutants are a classification that encompasses different chemicals and makes classification and regulation easier; many PFAS are listed as POPs under the 2001 Stockholm Convention, with PFOA and PFOS added in 2009 [82]. PFOA and PFOS are listed to be eliminated or restricted in use, respectively. Currently, PFOS is regulated under EU Regulation No 2019/1021, with specific guidance on marketing, manufacturing, use, and waste management [83]. The Stockholm Convention continues to publish effectiveness evaluations on specific POPs, with the most recent report on PFOS from 2017 with the results stating that the production of PFOS has dropped significantly, but specifics are still unknown due to the lack of data [84]. Many countries within the European Union have set drinking water limits for PFAS [85] and are continuing research to determine the seriousness and spread of these compounds.

2. Solid Waste Degradation of Fluorinated Compounds

Degradation of fluoropolymers by incineration, biodegradation, and alternative methods used to treat PFAS solid waste throughout the environment are all relevant topics of importance discussed below.

2.1. Definition of Thermal Degradation

There are currently several methods being investigated to safely dispose of various PFAS waste, specifically end-of-life fluorinated polymers in materials [1,33,34,37,45,46,48,86]. These methods include

open burning (OB), open air detonation (OD), incineration, pyrolysis, and smoldering. Fluoropolymers have been classified as polymers of low health concern during their life span when used within specified guidelines [6]. Once they end their normal lifecycle and are disposed of, the degradation of fluorinated polymers and other non-polymer PFAS compounds must be investigated to assess all potential degradation products. It is, therefore, an important topic that the thermal degradation methods used to treat fluorinated polymer waste can properly and safely dispose of the waste. The objective of these methods pertaining to PFAS waste treatment is to ensure that these fluorinated compounds are exposed to a sufficient and sustainable temperature that guarantees successful destruction of toxic PFAS as the fluorinated compounds thermally degrade while still being efficient and safe.

According to the Department of Defense (DoD), OB is defined as an open-air combustion process by which excess, unserviceable, or obsolete munitions are destroyed to eliminate their inherent explosive hazards. Open air detonation is used for the disposal of excess, unserviceable, or obsolete munitions whereby an explosive donor charge initiates the munitions being treated [87]. OB and OD are the primary methods of disposal of munitions components including those that contain PFAS components. While OB and OD are used mainly for munitions components, other techniques exist to destroy fluorinated polymer waste in more common materials such as coatings for non-stick cookware, water proofing of fabrics, and stain repellents for carpets. Incineration is a high-temperature flaming process that occurs in open air space, while smoldering is a flameless combustion process that occurs on the surface of a condensed fuel [1]. While both methods can be used to destroy fluorinated waste, the main difference between the two is that smoldering is self-sustaining and, therefore, more energy efficient compared to incineration which requires continuous energy input [1]. Lastly, pyrolysis is similar to incineration in that it is a high-temperature process used for thermal decomposition, except it occurs in an inert atmosphere [44].

2.2. Incineration of Fluoropolymers

As discussed previously, the electronegativity and small radii of fluorine cause C-F bonds to be exceptionally strong and, therefore, require high temperatures for destruction. On 16th May 2000, the EPA and 3M, which at the time was the only manufacturer of PFOS, announced the phase out of PFOS because it was considered toxic, bioaccumulative, and persistent [88]. Research concerning potential precursors of toxic PFAS since this announcement has grown substantially as studies have been completed on commercial products treated with fluorinated polymers such as fabrics, non-stick cookware, and microwave popcorn bags [33,89]. Studies have also been completed on the incineration or combustion of common fluorinated compounds to determine degradation products [2,33–39,86,90]. Tsang et al., determined the unimolecular reaction rates to show that CF_4 molecules require 1440 °C for greater than 1 s to achieve 99.99% destruction [91]. Therefore, CF_4 molecules are a reference for the destruction removal efficiency (DRE) testing on the required temperatures, times, and turbulence during incineration [92]. Despite the ease of removing or oxidizing the polar functional groups such as decarboxylation, the resulting products of incomplete combustion (PIC) are not well understood [92]. A technique to better understand the PIC products of PFAS combustion was utilized during thermal treatment by direct- and indirect-fired thermal oxidation mitigation through a bypassing flame and packed bed reactor with $\text{Ca}(\text{OH})_2$ to form CaF_2 [47]. This approach provided a way to manage the hydrofluoric acid, carbon monoxide and carbon dioxide generated during decomposition in an elevated pH media to investigate and characterize the reaction products. A recent study by Wang et al., 2015, showed that PFAS can be created from the waste handling of fluoropolymers at the end of useful life, specifically as a result of the municipal incineration of PTFE [47]. While PTFE is inert in the environment due to its high chemical and thermal stability, Taylor et al., 2014 found that municipal waste incinerators operating with a mean gas temperature of 1000 °C over the effective length of the reactor can be used to decompose PTFE [39]. Recently in 2019, Aleksandrov et al., built upon this prior study and evaluated the potential formation of 31 PFAS compounds from the waste incineration of PTFE—one of the most common fluoropolymers used in industry [45]. This study also attempted

to validate that PTFE almost entirely transforms fluorine into hydrofluoric acid and a number of trace PFAS species ranging in concentration from 0.3 to 24 μm^3 using incineration under standard municipal waste conditions [45]. The biggest challenge from the experiment was the probability of producing false positive results. To reduce their probability of false positive results using liquid chromatography with mass spectrometry (LC-MS/MS) detection methods, the experiment was scaled up to pilot plant incineration levels, meaning the solid combustion material input was orders of magnitudes larger than in lab-based simulations. Input materials were selected to minimize external contaminants, and paired t-testing was used to identify the presence of statistical differences between blank and PTFE spiked conditions. The 31 compounds selected were based on previous literature citations, occurrence in the environment, and availability of validated methods from commercial laboratories. A constant mass flow of wood pellets was utilized while adding PTFE to the reactor to maintain a consistent thermal profile, as shown in Figure 1. To analyze PFAS concentrations, they collected flue gas samples to test for compounds of interest after they cooled from roughly 850 to 1000 °C to below 300 °C, allowing for potential condensation reactions to occur. Using thermogravimetric analysis (TGA), PTFE decomposition began around 500 °C and was completed around 650 °C. While the detection limit varied between 0.3 to 24 $\mu\text{g}/\text{Nm}^3$ depending on the compound, only 11 out of 31 compounds were able to be detected as the other 20 compounds were below the detection limit. By using paired t-testing for significance for these 11 compounds, their results showed no statistically significant evidence that the PFAS studied was created during PTFE incineration. Due to these findings, they concluded that municipal incineration is an acceptable form of waste treatment and that external environmental contamination, not PTFE incineration, was likely the reason that the 11 PFAS samples were detected [45].

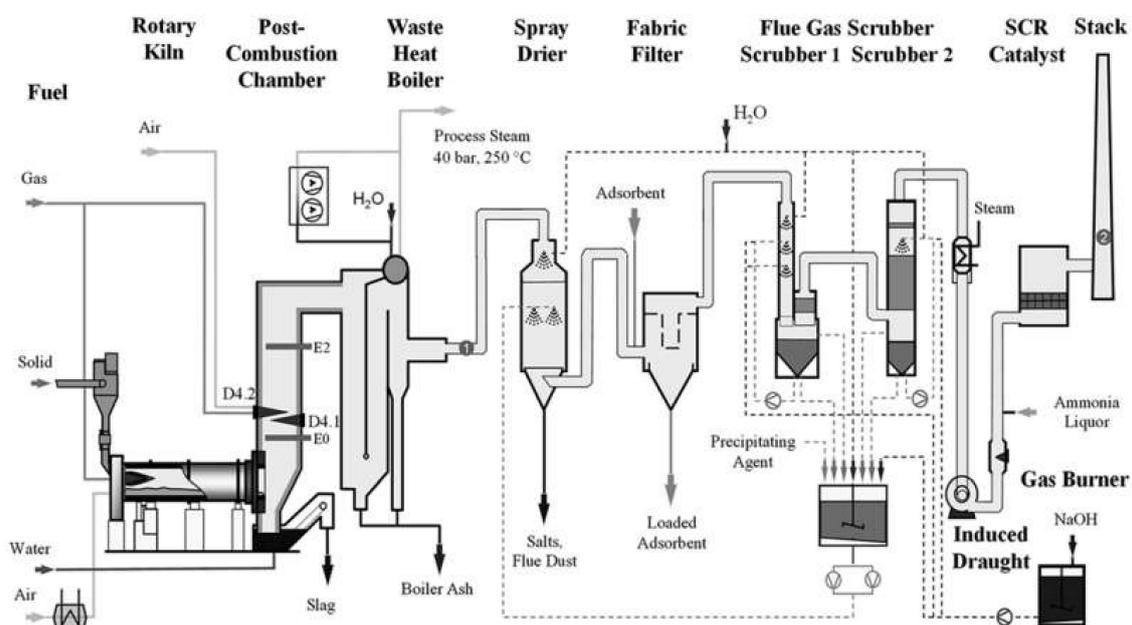


Figure 1. Schematic of the BRENDA (German acronym for “Brennkammer mit Dampfkessel”) pilot combustion facility at Karlsruhe Institute of Technology (KIT). Reproduced from Aleksandrov, K., Gehrman, H., Hauser, M., Mätzing, H., Pigeon, D., Staph, D., Wexler, M. *Chemosphere* 2019, 226, 898–906 (reference45). Reproduced with permission from Aleksandrov, K., *Chemosphere*; published by Elsevier 2019.

In terms of soil treatment, the Strategic Environmental Research and Development Program (SERDP)—the DoD’s environmental science and technology program—looked at the use of a surrogate fuel that can support smoldering and achieve temperatures above 900 °C to remediate PFAS in contaminated soils [1]. While PFAS is commonly removed from water by sorption onto carbon or

other sorbents, it is a challenge for soil contamination [1]. The smoldering of other contaminants has been shown to exceed temperatures above 900 °C, which is sufficient to treat PFAS waste; however, PFAS cannot support smoldering combustion by themselves, meaning a fuel must be used to support the smoldering process [1]. This study found that granular activated carbon (GAC) could effectively serve as a fuel and smolder at a required temperature greater than 900 °C, thus allowing ex situ and in situ treatment of PFAS contaminated materials [1]. Improvements can be made to this study by conducting more experiments to determine how much fuel is needed for successful smoldering [1]. Due to inhomogeneity in soils, smoldering techniques will not yield constant results for all areas, meaning different types of soils will require other techniques to eliminate PFAS waste [1]. Their study also found that with high PFAS combustion completion rates (not partial), HF will increase in abundance, while volatile organic fluorenes (VOF) and possibly fluorinated dioxins and furans (PFDD/F) reduce in abundance [1].

2.3. Biodegradation of Short-Chain PFAS

In addition to thermal degradation of fluorinated compounds, environmental degradation occurs in landfills where most of the solid waste is disposed [51,93–95]. Data from the EPA on the total generation and disposal of municipal solid waste (MSW) in the U.S. in 2017 found that 35% is composted and recycled, 12% is combusted for energy renewal, and slightly over 50% is discarded in landfills [96]. With over half of all U.S. waste being deposited in a landfill, the potential biodegradability of PFAS compound precursors is a relevant topic that has been investigated. In 2013, Liu and Mejia-Avendano completed a review on environmental biodegradability studies from the past decade carried out with microbial culture, activated sludge, soil, and sediment [51]. More recently in 2018, Hamid et al., completed a review focusing on the fate and transformation of PFAS in landfills specifically [95]. Allred et al., (2015) modeled how PFAS enters leachate (water that travels through waste products and contains environmental pollutants) using anaerobic bioreactors filled with municipal solid waste [94]. As commercial products treated with PFAS are often disposed of in landfills, and thus, act as a secondary point source to surface water, the study showed the abundance of PFAS in landfill leachate from six U.S. locations to have a median molar composition in the range of 0.01% to 45%. Moreover, multiple factors that contribute to PFAS concentrations in landfill leachate including abiotic leaching, pH, substrate, precursor biodegradation, and sorption were modeled as sources of PFAS in leachate by using four lab-scale anaerobic bioreactors filled with MSW operated over 273 days. This included duplicate reactors monitored under biotic and abiotic conditions to evaluate influences attributable to biological activity by using reactors to simulate methanogenic conditions that developed in landfills producing roughly 140 mL CH₄/dry g refuse. Their research connected leachate PFAS composition and MSW under time-dependent conditions relevant to landfills. To carry out the study, reactors were filled with shredded residential MSW and an anaerobic microbial inoculum to determine the roles of abiotic leaching and biological activity for 70 PFAS compounds. Reactors were periodically sampled for PFAS analysis by collecting leachate: the average total PFAS leaching was 16.7 and 2.83 nmol/kg dry refuse for biotic and abiotic leaching, respectively, indicating that biological processes were primarily responsible for leaching. The reactor gas was not analyzed for fluorinated compounds, so a future study could investigate the degree to which volatile fluorotelomers are released from MSW. In this study, biodegradation pathways were not evaluated, but from previous work, a more detailed review of biodegradation pathways can be found in the following cited publications [51,93,95,97,98].

2.4. Alternative Methods

As different methods are required to treat the presence of PFAS compounds throughout the environment, alternative methods are being investigated. One alternative method to treating PFAS in water was completed by Singh et al., in 2019 who used a novel plasma-based treatment process [48]. Using an enhanced contact plasma reactor, they were able to quantify liquid and gas phase byproducts produced during PFAS degradation by using data acquired from ultraperformance liquid

chromatograph-quadrupole time-of-flight-high resolution mass spectrometry (UPLC-QToF-HRMS) and propose a degradation mechanism for PFOA and PFOS throughout the process. From this study, as shown in Figure 2, 90% of the PFOA and PFOS was reduced in 60 and 40 min, respectively. Reduction up to 120 min was carried out to better quantify byproducts. They determined that the liquid byproducts were mainly produced by a stepwise reduction in PFOA and PFOS into linear chain PFCAs (C4 to C7), perfluoropropanoic acid (PFPA), short-chain organic acids, and elemental fluorine, and that gaseous byproducts such as cyclic perfluoroalkanes were also produced and degraded. Comparing their results to their previous study which used a higher treatment rate plasma reactor for a shorter time compared to this lower plasma treatment rate at a longer duration [99], they found that the concentrations of reactive species increased with energy input, but the composition remained the same. This meant that different concentrations of identified products were expected, but not different types of products which held true. Using a fluorine mass balance, they also noted that roughly 23% of the fluorine associated with the parent PFOA and 42% with the parent PFOS were adsorbed to the reactor walls. These findings will help contribute to their planned future work where a scaled-up version of the plasma reactor will be used to treat PFAS-contaminated investigation-derived waste (IDW) at flow rates of 3.8 to 7.6 L per minute at a U.S. Air Force base.

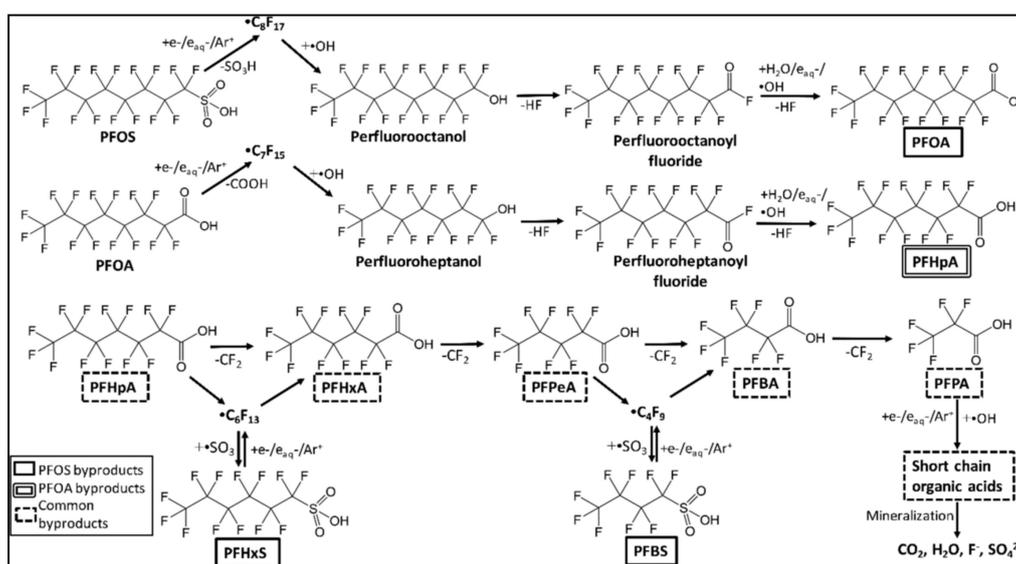


Figure 2. Proposed degradation pathway for PFOS and PFOA in plasma treatment. Plasma electrons, aqueous electrons, and argon ions are the primary species responsible for degradation of PFOS and PFOA to form perfluoroalkyl radicals. Chain propagation reactions involving the reductive and oxidative species along with subsequent hydrolysis yield short chain PFCAs. Reproduced from Singh, R.K., Fernando, S., Bygi, S.F., Multari, N., Thagard, S.M, Holsen, T.M. *Environmental Science Technology* 2019, 53, 5, 2731-2738 (reference 48). Reproduced with permission from Singh, R.K., *Environmental Science Technology*; published by American Chemical Society 2019.

3. Aqueous Film Forming Foam (AFFF)

Developed in the mid-1960s by the United States Navy and 3M (St. Paul, MN, USA), aqueous film forming foams (AFFFs) were developed to specifically combat liquid fuel fires [100]. The foams are water-based hydrocarbon surfactants that frequently contain sodium alkyl sulfate, and fluorosurfactants, such as fluorotelomers, perfluorooctanoic acid (PFOA), or perfluorooctanesulfonic acid (PFOS). Able to spread rapidly across the surface of hydrocarbon fuels and form a thin film, AFFF can prevent fuel vapor and oxygen interaction to extinguish the fire. The efficacy of AFFF led to its widespread use as a fire suppressant for airports, naval vessels, and fire departments. However, when it is discharged, the components have a propensity to runoff and contaminate surrounding environments [61,101–103]. Specifically, the PFOS within AFFF was later recognized under Annex B of the Stockholm Convention

as a toxic and bio-accumulative pollutant [76,82,104,105]. After years of firefighters training and using AFFF, runoff from the airports contaminated their local fresh water source as identified by the EPA in a study of emerging contaminants [106]. The recognition of AFFF as a toxic component has fostered a field of science focused on the detection and remediation of PFOS and many other perfluoroalkyl acid (PFAA) contaminants across the globe [62,64,67,68,70].

3.1. Early Research

An early attempt to detect and quantify AFFF contamination on military bases was conducted by Schultz et al., (2004) [62]. The study sought to detect and quantify 4:2, 6:2, and 8:2 fluorotelomer sulfonates from groundwater at 3 military bases (Naval Air Station Fallon, NV, USA; Tyndall Air Force Base, FL, USA; Wurtsmith Air Force Base, MI, USA) [62]. LC/MS-MS was utilized as a means of separation and detection of AFFF in human serum, animal tissue, and surface water samples. This method has become a common technique used in many following studies [64,67,68,70]. In addition to the detection and quantification of AFFF components, Schultz utilized fast atom bombardment mass spectrometry (FAB-MS) to also identify possible sources of the fluorotelomer sulfonate contamination observed. In terms of total fluorotelomer sulfonate concentrations, none were detected in at the Nevada site. At the Florida site, the concentrations ranged from 1100 to 14,600 µg/L, while in Michigan, it ranged from 0.60 to 182 µg/L. Schultz identified that fluoroalkylthioamido sulfonates were the main anionic fluorosurfactant in the AFFF concentrates and not fluorotelomer sulfonates as previously thought. Despite their abundance within the AFFF concentrate, fluoroalkylthioamido sulfonates were absent from groundwater samples indicating their degradation into fluorotelomer sulfonates. These results confirmed the complexity and relative lack of knowledge surrounding the biodegradation and transport of fluorotelomer sulfonates in groundwater.

3.2. Current AFFF Detection Efforts

Høisæter et al., (2019) examined the contamination at a Norwegian firefighting training facility, 15 years after the use of PFOS-based AFFF products had ceased [70]. In the soil, PFOS made up 96% of the total PFAS concentration and varied from 0.3 to 6500 µg/kg. To determine how much PFOS was still present in the source zone and if it could form a long-term source contaminating the site's groundwater, they performed unsaturated column studies with pristine soil similar to that found at the site and the same PFOS containing AFFF. Using both a high and low water infiltration rate (9.7 and 4.9 mm/day for 7 and 14 weeks, respectively) to change aqueous saturation of the soil columns, Høisæter et al., were able to model the contaminating effects of PFOS. The low infiltration rate represented 2 years of snow melt, and the high infiltration rate represented extra water added in areas of firefighting training. Utilizing LC/MS-MS, PFOS was detectable in the leachate of the higher infiltration experiment after 14 days where the concentration increased from 20 to 2200 ng/L after 49 days. Soils were also tested at various depths for PFOS detection and found that contamination reached as far as the groundwater level at 4 m below the surface. These results concluded that PFAS leaching from highly contaminated areas poses a long-term risk for contamination of local groundwater and consequently transport to neighboring surface water.

Bräunig et al., (2017), investigated the concentration of 10 perfluoroalkyl acids (PFAAs) in various environmental and biological samples including: water, soil, grass, chicken egg yolk, serums of horses, cattle, sheep, and humans to determine the extent of PFAAs leaching from a fire-fighting training area into an aquifer [68]. This study sought to expand upon the usage of both abiotic and biotic sampling introduced by Schultz et al., to not only quantify PFAA contamination but also investigate the spread of the contamination into terrestrial ecosystems [62]. Sample analysis was completed using LC/MS-MS in conjunction with isotopic dilution mass spectrometry to determine individual concentrations. PFOS in water samples ranged from 0.17 to 14 µg/L, and perfluorohexaneculphonic acid (PFHxS) between 0.07 to 6.0 µg/L. PFOS concentrations in human serum were 38 to 381 µg/L, while PFHxS ranged from 39 to 214 µg/L. Statistical significance was shown between cattle in the contamination zone

compared to outside of the zone ($p < 0.01$) in measuring PFOS and PFHxS concentrations. The highest PFOS concentration found in human serum was 30 times higher compared to the general Australian population. Their efforts concluded that insufficiently remediated sources of PFAAs further spread and contaminate terrestrial ecosystems which expose increasingly more individuals.

Houtz et al., (2016) utilized a unique approach to indirectly measure the total concentration of PFAA precursors as well as directly measure twenty specific PFAS analytes in wastewater samples from 8 wastewater treatment plants (WWTPs) that discharge into San Francisco Bay [67]. The most prevalent of these PFAS analytes include: perfluorohexanoic acid (PFHxA), perfluorooctanoic acid (PFOA), perfluorobenzoic acid (PFBA), perfluorooctanesulfonic acid (PFOS), fluorotelomer sulfonate (FtS), and perfluoropentanoic acid (PFPeA). Indirect measurement was accomplished utilizing a previously developed assay that uses hydroxyl radicals to oxidize precursors into perfluorinated carboxylates which are easily identifiable using LC/MS-MS [64,107]. Six of the WWTPs contained similar amounts of total PFASs with the following median amounts: PFHxA (24 ng/L), PFOA (23 ng/L), PFBA (19 ng/L), and PFOS (15 ng/L). The remaining two plants contained significantly higher concentrations of PFASs with one having a PFOS of 420 ng/L and another plant having PFOS of 560 ng/L, 390 ng/L 6:2 FtS, 570 ng/L PFPeA, and 500 ng/L PFHxA. These significant differences in PFAS concentrations were determined to be the result of AFFF contamination as supported by the presence of discontinued and current AFFF components in their samples. Indirectly measured PFAA precursors accounted for 33% to 63% of the total molar concentration of PFASs in samples and indicated an increase in the use of shorter-carbon PFASs as well as demonstrates the impacts of AFFF on wastewater management [104–107].

4. Electrochemical Aqueous Remediation of Fluorinated Compounds

Non-thermal degradation techniques used to remediate PFAS in water include techniques such as separation (electrocoagulation and fractionation) and electrochemical treatment (oxidation and reduction). In terms of electrochemical treatment, a promising approach is electrochemical oxidation (EO) using a boron-doped diamond (BDD) as an anode to treat PFAS-contaminated water. Previous studies have investigated the potential of this technique [108–110], among investigated materials to use as an anode, the BDD has been shown to have the highest reactivity for PFOA and PFOS degradation [110–112]. In this technique, the BDD anode potential induces oxidization that forces the transfer of electrons from oxygen atoms of the carboxylate group of PFOA or sulfonate group of PFOS to the BDD, resulting in the decomposition of PFOA and PFOS. However, a challenge associated with EO treatment using a BDD anode is that chloride in water is readily converted to perchlorate (ClO_4^-), and anodes with higher EO activity tend to produce more perchlorate. Most recently, Yang et al., (2019) used the BDD anode as a model to treat PFOA and PFOS in a chloride-bearing electrolyte and found that by adding hydrogen peroxide in a quantity greater than or equal to 50 mM H_2O_2 , the formation of perchlorate can be largely inhibited [113]. In their study, they found that a standard EO process without hydrogen peroxide can result in 40% of chloride converting into perchlorate. Using similar methods to previous studies that used a BDD anode, adding hydrogen peroxide to their strategy suppressed 88% of perchlorate formation during treatment. This happens because hydrogen peroxide quenches chlorine and hydroxyl radicals, thus inhibiting perchlorate formation. Additionally, they observed that the oxidation of PFOA and PFOS is primarily determined by direct electron transfer oxidation, regardless of the dominant radical species, meaning hydrogen peroxide would not largely inhibit PFAS degradation. This is considered a promising technique because only 4.0 L of 30% H_2O_2 can assist in the treatment of 1 m³ of groundwater at only an additional cost of USD 0.6 to 1.2/m³ with the current market price.

While the addition of hydrogen peroxide to the BDD approach is inexpensive, BDD electrodes have a high cost of approximately USD 7000/m² which limits their use for large scale application [114]. Another promising anodic material, Ti_4O_7 , has been explored for use in the removal of PFOA and PFOS from contaminated water [115–117]. Le et al., (2019) used a Magnéli phase Ti_4O_7 reactive electrochemical membrane (REM) in a single-pass, flow-through operation mode as a function of anode

potential and flow rate to investigate the extent of PFOA and PFOS degradation [112]. The criteria used to assess performance were PFOA/PFOS conversion, extent of defluorination, and energy consumption compared to other technologies. Samples were analyzed using ion chromatography and LC/MS/MS based on of EPA Method 537.1 [54]. Compared to BDD anodes, they found that Ti_4O_7 anodes show the best performance with the lowest energy consumption due to their larger surface area. In this study, 5.1 ± 0.1 kWh/m³ for PFOA and 6.7 ± 0.2 kWh/m³ for PFOS were needed to remove initial concentrations of 4.14 mg/L for PFOA and 5 mg/L for PFOS to below detection limits (86 ng/L for PFOA and 35 ng/L for PFOS). They also noted that the highest removal rates they achieved were 3415 ± 203 $\mu\text{mol}/\text{m}^2/\text{h}$ for PFOA and 2436 ± 106 $\mu\text{mol}/\text{m}^2/\text{h}$ for PFOS at a residence time of 3.8 s. These values for removal rates and energy consumption are among the lowest reported for electrochemical oxidation currently. Future work needed for REM anodes involves analyzing the success of operating multiple REMs in series in real waste streams.

Shi et al., (2019) also investigated the degradation of PFOS by using a microporous REM composed of Magnéli phase titanium suboxide [118]. To assess the performance of the REM, they ran a 2.0 μM PFOS solution with varying current density of 0.5 to 4.0 mA/cm². By comparing REM operation to a batch operation mode at an anodic potential of 3.15 V, they were able to remove $98.30\% \pm 0.51\%$ of PFOS under REM operation, a much higher efficiency than what was achieved using the batch operation mode. Their results showed that PFOS degradation formed intermediate products such as short-chain PFCAs in trace amounts and underwent rapid mineralization to form carbon dioxide and fluoride. Using density functional theory simulations, they found that PFOS degradation was initiated by direct electron transfer on the anode yielding PFOS free radicals. Then, these PFOS free radicals further reacted with hydroxyl radicals, an essential component to PFOS degradation, because PFOS free radicals had the potential to react with water and reform as PFOS. Future work for this study includes scaling the REM system to remediate larger quantities of PFOS-contaminated water.

Another technique recently applied to PFAS-contaminated water is a unique separation technique. The most important separation technique is electrocoagulation (EC) which involves the addition of charged metal hydroxides to a colloid in order to disrupt the electrostatic charges of its components, resulting in separation from the aqueous phase. EC has already been used to remove heavy metals, suspended solids, bacteria, oils, and other contaminants from water, but has been relatively unexplored in its ability to remove PFASs from water sources.

An early attempt to optimize the EC technique for the removal of PFOS/PFOA from water came from Xiao et al., (2013), who sought to examine the parameters of: flocculation time, initial contaminant concentration, concentration of dissolved organic material, coagulant dosage, and solution pH and their effects on the efficiency of removing PFOS/PFOA contaminants from water [119]. Xiao conducted a series of jar tests on prepared solutions containing deionized water and organic material from the Suwannee River, Florida, USA, that were spiked with 0.2 μM of both PFOS and PFOA. After preparing their solutions, they were coagulated using either alum or ferric chloride at various dosages and initial pHs before being filtered through a Millipore membrane. These membranes were then analyzed using LC/MS to determine the efficiency of removal of PFOS/PFOA under their respective conditions. Their testing concluded that coagulant dosage and initial solution pH were the two dominant factors affecting the efficiency of PFOS/PFOA removal as the dosage determines the surface area available for adsorption as well as pH determines the number of protonated adsorption sites. As shown in Figure 3, Xiao et al., developed an operational coagulation diagram depicting the optimal operating conditions for the removal of PFOS/PFOA [119].

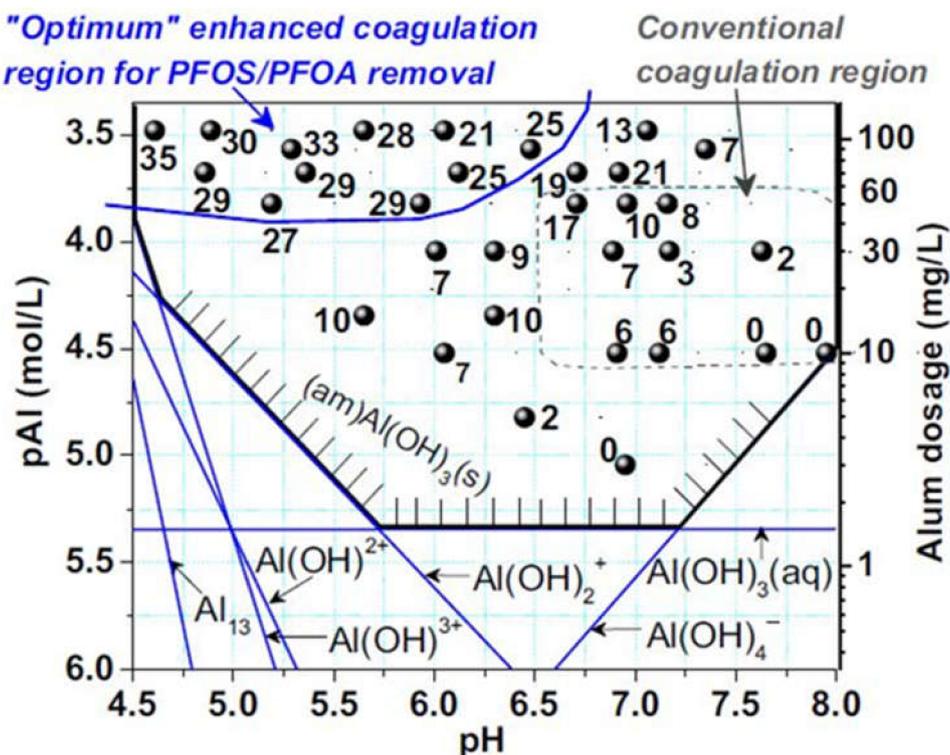


Figure 3. Removals exceeding 25% occurred in the enhanced coagulation region characterized by higher alum dosages (e.g., >60 mg/L) and (thus) lower coagulated water pH(4.5–6.5). Reproduced from Xiao, F.; Simcik, M. F.; Gulliver, J. S. *Water Research* 2013, 47 (1), 49–56 (reference 119). Reproduced with permission from Xiao, F., *Water Research*; published by Elsevier 2013.

After optimizing the EC technique to efficiently filter PFASs such as PFOS and PFOA from water, the focus became overcoming the challenge of long-term use of a single electrode leading to passivation and a drop in removal efficiency. A novel solution to this was developed by Liu et al., (2018), who modified the existing methodology by incorporating the periodically reverse electrocoagulation (PREC) technique [120]. Liu sought to determine the effectiveness, energy consumption, and application to contaminated natural groundwater using various electrode materials, applied voltages, and stirring speeds. To accomplish this, they utilized a PREC reactor with duo combinations of iron, zinc, and aluminum electrodes to treat contaminated water as well as LC/MS to determine the efficiency of removal of PFOS/PFOA and FTIR to confirm the identity of the flocs formed. Liu found that the Al-Zn electrode was the most efficient with a 99.3% conversion rate after 20 min while operating at +9.0 V and 500 rpm, and it was the least energy- and material-intensive. Under these same parameters, natural groundwater was treated using PREC achieving a removal of 79.4% in 60 min. These results combined with FTIR confirming the flocs contained PFOA demonstrated the potential for PREC to be further developed as a method of eliminating PFOA from both natural bodies of water as well as wastewater.

5. Governmental Environmental Regulatory Efforts

North American, European, and Asian governmental agencies that have identified fluorinated molecules as an emerging environmental threat each have independently enacted guidelines to help reduce the threat to both the environment and to the people that inhabit these regions.

5.1. The Environmental Protection Agency (EPA)

The Environmental Protection Agency (EPA) of the United States released a comprehensive action plan in February of 2019 on PFAS in the environment [121,122]. This action plan is designed to support the Safe Drinking Water Act (SDWA), which addresses PFAS in drinking water and outlines

measurement procedures for the amount of perfluorinated chemicals (PFCs) in water [23,77]. The EPA's current methods for drinking water detection, Method 533 and 537.1, focus on carbon chain lengths of 4 to 12 within the PFAS category, and they follow the SDWA for PFOA and PFOS [53,54]. These testing methods, which complement each other, allow for the measurement of 29 different chemicals, and the EPA is still working on obtaining more information to determine a definite limit of unsafe levels of PFAS for human health and aquatic life as well as the potential regulation of industrial wastewater [23,77]. The current Lifetime Health Advisory standard for the amount of PFOS/PFOA in drinking water is 70 ppt as recommended by the EPA [105]. In December of 2019, the Interim Recommendations for Addressing Groundwater Contaminated with PFOA and PFOS was issued using a screening level of 40 ppt to determine if further attention is warranted [23]. The preliminary remediation goal of 70 ppt was set for contaminated ground water based on the Lifetime Drinking Water Health Advisory level [23]. Currently, the EPA is in the process of designating PFOA and PFOS as hazardous substances under the Comprehensive Environmental Response, Compensation, and Liability Act (CERCLA) which would allow the government to tax manufacturers using these substances and use the money toward removing them from the environment [23,78]. Similar to method 533 and 537.1 for determining PFAS in drinking water, the EPA has developed other methods for testing non-potable groundwater, surface water, and wastewater outlined as SW-846 Method 8327 [23,75]. Guidance for destruction and disposal of PFAS using methods such as high temperature incineration is currently being developed but have not been formally established or published by the EPA [23].

The primary methods used to detect PFASs in drinking water are Method 533 and 537.1, and SW-846 Method 8327 is used to test PFASs in groundwater, surface water, and wastewater [53,54,75]. All three of these methods utilize liquid chromatography/tandem mass spectrometry, but the differences in extraction provide the differences in compounds detected [53,54,75]. US EPA Method 533 was published in November of 2019, and consists of a solid phase extraction (SPE) (LC-MS/MS) while utilizing the Multiple Reaction Monitoring (MRM) mode of the MS/MS to enhance selectivity [53]. Method 533 focuses on short chain PFASs, those with 4 to 12 carbon chain length, and can measure 25 chemicals, 11 of which cannot be detected with Method 537.1 [123]. Methods 533 and 537.1 are designed to complement each other, with the two combined being able to detect 29 different PFASs [123]. US EPA Method 537.1 is an update to the original method, 537, which was published in 2008 and revised in 2015 [124]. Method 537.1 was published in November of 2018 and similarly uses SPE and LC-MS/MS as in Method 533 [54]. Method 537.1 detects a total of 18 PFASs with 4 that cannot be detected with Method 533 [54,123]. Like Methods 533 and 537.1, Method 8327 uses LC-MS/MS but instead is designed to measure PFAS in groundwater, surface water, and wastewater as opposed to drinking water [75]. This method was published in June of 2019 and requires the MS to be in MRM mode [75].

5.2. International Regulatory Efforts

The Stockholm Convention seeks to restrict or eliminate different chemicals that are determined to be POPs. Within the list of POPs, there are classifications for risk levels with Annex A substances being the most harmful, instructing parties to eliminate the production and use of the substances under Annex A [82]. Currently PFOA, its salts, and PFOA-related compounds are listed under Annex A for being persistent, bioaccumulative, and toxic to living organisms and the environment [82]. The Stockholm Convention cites a link between kidney cancer, testicular cancer, thyroid disease, and high cholesterol and PFOA for its inclusion on the highest threat level list [82]. The second, less harmful classification of POPs is Annex B, which calls for the restriction of production and use of the substances [82]. PFOS, its salts, and perfluorooctane sulfonyl fluoride (PFOSF) are currently listed under Annex B for substantial bioaccumulating and biomagnifying properties [82]. In 2017, the Stockholm Convention published effectiveness evaluations for select chemicals on the POPs list, including one for PFOS, its salts, and PFOSF [84]. After being placed on the POPs list in 2009, there was an increase in the number of countries that emplaced legal and administrative measures to control PFOS production, import, export,

and use [84]. Ultimately, it was not possible to determine a comprehensive global assessment of the production and use of PFOS because there is still limited information and data on the topic [84]. There are different acceptable use exceptions for each POP, with the most recent update being in 2019 when the Stockholm Convention agreed upon new acceptable uses including insecticide and firefighting foams [85]. Member states have until December of 2020 to implement these changes in their national laws [125].

5.3. European Union Regulatory Efforts

Within the European Union (EU), the Registration, Evaluation, Authorization, and Restriction of Chemicals (REACH) is a regulation of the EU that provides guidelines to better protect the environment and human health from the dangers of different chemicals [126,127]. The regulatory organization was implemented in June 2007, and requires manufacturers to register and report the chemicals used in their products. Through the REACH regulation, the EU has the authority to ban any substance that is determined to have unmanageable risk [126]. EU Regulation No. 1907/2006 first established the REACH program, and outlines reporting procedures, safety sheets, and appeal processes for different chemicals listed under different classifications [128]. The list of substances of very high concern (SVHC) includes PFOA (added June 2013), or PFHxS, (added July 2017), and Perfluorobutane sulfonate (PFBS), (added January 2020) [129]. Under EU Regulation No. 440/2008, there are specific testing measures for determining the amount of SVHC in products and manufacturing designed to ensure that manufactures are complying with the guidelines [128]. Similarly, EU Regulation No. 340/2008 outlines fees for non-compliance with REACH standards [128]. PFOS, although not listed as an SVHC, is still a restricted compound under REACH [21]. Some of the most recent legislation from the EU concerning POPs was passed in 2019. EU Regulation No. 2019.1021 was adopted in June 2019 and outlines further restriction and exceptions for manufacturing with PFOS as an addition to the preexisting EU Directive 2006/112/EC from 2006 [130,131]. The standing legislation from 2006 was based on the Organization for Economic Cooperation and Development (OECD) hazard assessment on PFOS carried out in July of 2002 that deemed the compound to be “persistent, bioaccumulative, and toxic to mammalian species” [130]. As a result of this assessment, the EU restricted substances or preparations with a PFOS concentration equal to or higher than 0.005% by mass, semi-finished products with a PFOS concentration equal to or higher than 0.1% by weight, and textiles or other coated materials with the amount of PFOS equal to or higher than 1 $\mu\text{g}/\text{m}^2$ of coated material [130]. Exemptions from the restrictions were granted to photographic coatings on films, paper, or printing plates, and mist suppressants for hard chromium plating and wetting agents for controlled electroplating and hydraulic fluids for aviation; fire-fighting foams placed on the market before December 2006 could be used until June 2011 [130]. The most recent legislation, passed in June of 2019, added additional restrictions to PFOS stockpiles and waste management in addition to allowing more exemptions including the selling of PFOS to labs for research purposes [131]. Member countries must report stockpiles of more than 50 kg PFOS and sets a waste limit of 50 mg/kg PFOS [131]. If the PFOS in waste exceeds this limit, it must be disposed of in such a way that the PFOS is “destroyed or irreversibly transformed so that the remaining waste releases do not exhibit the characteristics of POPs” citing the only known way to transform PFOS is through degradation [131].

Across the EU, there is no consistent limit for the amount of PFOS and PFOA in drinking water, although many Member States have set their own limits [21,132–135]. Sweden has the most recent drinking water limitations, with recommendations on the management of both drinking water and fish contamination being made in 2014 [135]. The recommendation from the Swedish National Food Agency was the sum of 11 PFAS including perfluorobutanesulfonic acid (PFBS), PFHxS, PFOS, 6:2 fluorotelomersulfonate (6:2 FTS), pentafluorobenzoic acid (PFBA), and PFOA, which should not exceed 90 ng/L in drinking water [135]. The original recommendation was updated in 2016, and although there are currently no legally binding limits for PFAS in drinking water, the Swedish National Food Administration SLVFS legislation 2001: 30 § 7 states that drinking water should not contain

substances in levels high enough that they could pose a risk to human health [135]. There is a proposed 530 ng/L limit of PFOS in drinking water in Norway, and the advised maximum acceptable amount of PFOS in drinking water is 300 ng/L [133,136]. The United Kingdom Health Protection Agency established the 300 ng/L risk limit of PFOS in drinking water, and also established a 10 µg/L risk limit for PFOA in drinking water [133]. This report, originally published in August of 2007, also outlines different tiers with maximum concentrations [133]. Within the lowest tier (Tier 1) is when the concentration is above 300 ng/L; there should be increased monitoring and consultation of local health professionals [133]. This also applies to the highest tier where concentrations of PFOS are above 90 µg/L and people should contact local health professionals immediately as well as refrain from drinking the water for 7 days [133]. The Drinking Water Commission of the German Ministry of Health established a value for safe lifelong exposure of PFOS and PFOA to be 300 ng/L in drinking water [132]. Some countries, such as Italy, have established legally binding limits to PFAS after detecting extremely high levels [134]. The EU established a project called Perfluorinated Organic Compounds in the European Environment (PERFORCE) which had the goal of sampling and measuring PFCs across Europe, primarily in surface water, sewage treatment plants, and sediments [137]. This study showed that the highest concentrations of PFHpA, PFOA, and perfluorononanoic acid (PFNA) across all cities in Europe were found in the Po River in Italy [137]. This triggered an investigation by the Italian government into other sources of water in Italy in the early 2010s, with the highest concentration of PFAS being 1,475 ng/L [134]. The government installed activated carbon filters and was able to lower the concentration to 386 ng/L [134]. Threshold levels were established in January of 2014 in Italy with a Ministerial Decree to be 500 ng/L for PFOA, 30 ng/L for PFOS, and 500 ng/L for all other PFASs in drinking water [134]. Norway has established regulations for the amount of PFOS and PFOA in soil based on a study on the reproductive effects of these chemicals on earthworms [138,139]. The Netherlands has established similar risk limits for both soil and groundwater, but these limits are based on human exposure through consuming vegetables or animals that were in contact with these substances [140]. The limit for PFOS in soil is 2.3 µg/kg, and the risk limit for PFOS in groundwater is 23 mg/L; the Dutch National Institute for Public Health and the Environment stated that there were not enough data to derive risk limits for vegetables, milk, or meat [140]. A summary of United States and European regulations regarding PFOS/PFOA limits is given in Table 2.

Table 2. Summary of United States and European Union regulations governing PFOS/PFOA limits for given medium.

Country and Agency	Pfos Limit	Pfoa Limit	Medium	References
US EPA	70 ppt	70 ppt	Drinking Water	[123]
UK Drinking Water Inspectorate (Tier 1)	300 ng/L	300 ng/L	Drinking Water	[134]
UK Drinking Water Inspectorate (Tier 2)	10,000 ng/L	1,000 ng/L	Drinking Water	[134]
UK Drinking Water Inspectorate (Tier 3)	90,000 ng/L	9,000 ng/L	Drinking Water	[134]
Swedish National Food Agency	90 ng/L	90 ng/L	Drinking Water	[136]
Norwegian Pollution Control Authority	530 ng/L	N/A	Drinking Water	[134]
German Drinking Water Commission	300 ng/L	300 ng/L	Drinking Water	[132]
Italian National Institute of Health	30 ng/L	500 ng/L	Drinking Water	[134]
Dutch National Institute for Public Health and the Environment	530 ng/L	N/A	Drinking Water	[137]
Norwegian Pollution Control Authority	0.1 mg/kg	0.16 mg/kg	Soil	[140]
Dutch national Institute for Public Health and the Environment	2.3 µg/kg	N/A	Soil	[137]
Dutch National Institute for Public Health and the Environment	23 mg/L	N/A	Ground Water	[137]

5.4. Environmental Regulatory Efforts in Asia

Moreover, there is significantly less governmental regulation of PFAS in Asia compared to that in Europe or the United States [141]. In countries such as Bangladesh, Egypt, India, Indonesia, and Malaysia, PFAS are essentially unregulated and not monitored [141]. Japan has taken measures to

regulate PFOA, PFHxA, PFHxS, and PFOS in manufacturing with no maximum levels for any PFAS set for tap water in the country [141,142]. Additionally, there are limitations under the Chemical Substances Control Law for reporting the PFAS listed above to the Ministry of Economy, Trade, and Industry if manufacturers import or manufacture over 1 ton [141,142]. In the early 2000s, as 3M announced that they would halt production of PFOS-based products, production of PFAS products shifted to other countries, especially China [143]. This caused a large increase in manufacturing of PFAS in China before the government began regulating it [143]. Currently, both the Chinese and South Korean governments have taken measures to ban PFOS and its salts with some limited exceptions in accordance with the Stockholm Convention [144,145]. A summary of United States, European, and Asian regulations regarding PFOS/PFOA limits is given in Table 3.

Table 3. Summary of international regulations by country that govern production and import of PFAS.

	Country and Regulation	Path Taken	Category of PFAS Addressed	Life Cycle State(s) Addressed	Method of Approach	Ref
United States	2010/15 PFOA Stewardship Program	Encourage industry phaseout	Long-chain perfluorocarboxylic acids and related substances	All	Regulatory	[122]
	Significant New Use Rule (40 CFR §721.9582)	Manage, manufacture, import, and processing	Perfluoroalkyl sulfonates (PFASs)	Chemical manufacture and import; processing of chemicals	Regulatory	[122]
	Significant New Use Rule (40 CFR §721.10536)	Manage manufacture, import, and processing	Perfluoroalkyl carboxylate chemicals	Chemical manufacture and import; processing of chemicals	Regulatory	[122]
	Proposed Significant New Use Rule (SNUR) to designate import of perfluoroalkyl carboxylate chemicals (Proposed rule: 80 FR 2885)	Manage manufacture, import, and processing	Perfluoroalkyl carboxylate chemicals	Chemical manufacture and import; processing of chemicals	Regulatory	[122]
European Union	PFOA and APFO harmonized classification under Regulation. (EC) No. 1272/2008 (2013)	Manage the manufacture, sale, import and export	PFOA and APFO	Chemical manufacture, product (mixtures and articles) manufacture and use	Regulatory	[127]
	PFOA and APFO covered by Annex XVII entries 28-30 of REACH	Manage the manufacture, sale, import and export	PFOA and APFO	Mixtures for consumer use	Regulatory	[127]
	Proposal for restriction of PFOA, its salts and PFOA-related substances under REACH (2014)	Manage the manufacture, sale, import and export	PFOA, its salts and PFOA-related substances	Chemical manufacture, product (mixtures and articles) manufacture and use	Regulatory	[127]
	Several PFASs listed on draft Community Rolling Action Plan (CoRAP) for substance evaluation withing the next years	Evaluation	Several (especially alternatives to PFOA- and PFOA-related substances)	Whole life cycle	Assessment	[127]
China	Monitoring of PFASs production and emissions	Continuous monitoring	PFOS and PFOA	N/A	Monitoring	[144]
	Ban of production, transportation, application, imports and exports of PFOS, its salts and PFOSE, except for specific exemptions and acceptable use	Ban	PFOS, its salts and PFOSE	Manufacture, use, sale, and import	Regulatory	[144]

Table 3. Cont.

Country and Regulation	Path Taken	Category of PFAS Addressed	Life Cycle State(s) Addressed	Method of Approach	Ref
Restriction of the production of PFOS and PFOA and support of R&D for alternatives to these substances	Restriction as part of the National Development and Reform Commission, which issues an Industrial Reconstructing Guide Directory	PFOS and PFOA	-	-	[144]
Substances listed under the High Pollution, High Environmental risk Product Catalogue	Political target to establish financial and environmental protection priorities	High temperature melting membrane fluorine resin coating/PFOA	-	Policy	[144]
Japan PFOS and its salts listed in Class I Specified Chemical Substances under the Chemical Substances Control Law (CSCL)	Manage the manufacture, import, export and use	PFOS and its salts	Manufacture, import, export, use	Regulatory	[142]
South Korea Under the Persistent Organic Pollutions Control Act, PFOS, its salts, PFOS-F are designated as restricted	Restriction on manufacture, import, export, use	PFOS, its salts and PFOS-F	Manufacture, import, export, use	Regulatory	[145]

6. Conclusions

Since its inception and widespread industrial and commercial uses, fluorinated compounds have become essential for many everyday applications and are not considered to be a health hazard during their typical life cycle. At end-of-life, these same compounds that are dependent upon conditions can become a health hazard that needs to be easily detected and removed from the environment. As presented throughout this review, much effort has been devoted to the detection and remediation in both solid and aqueous media to ensure that contamination is limited to safe levels. Regulatory requirements are often developed independently within their region or country. These independent non-uniform regulations could potentially present environmental challenges across international borders. This review has focused on current state and advancements in classification, solid and aqueous waste remediation, and environmental regulatory guidelines to eliminate the threat of contamination from fluorinated compounds. Looking forward, continued advancements in detection across a wider range of fluorinated compounds is imperative and large-scale implementation of remediation techniques for solid and aqueous waste remain a technical challenge.

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