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Distribution, transformation and remediation of poly- and per-fluoroalkyl substances (PFAS) in wastewater sources

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Distribution, transformation and remediation of poly- and per-fluoroalkyl substances (PFAS) in wastewater sources

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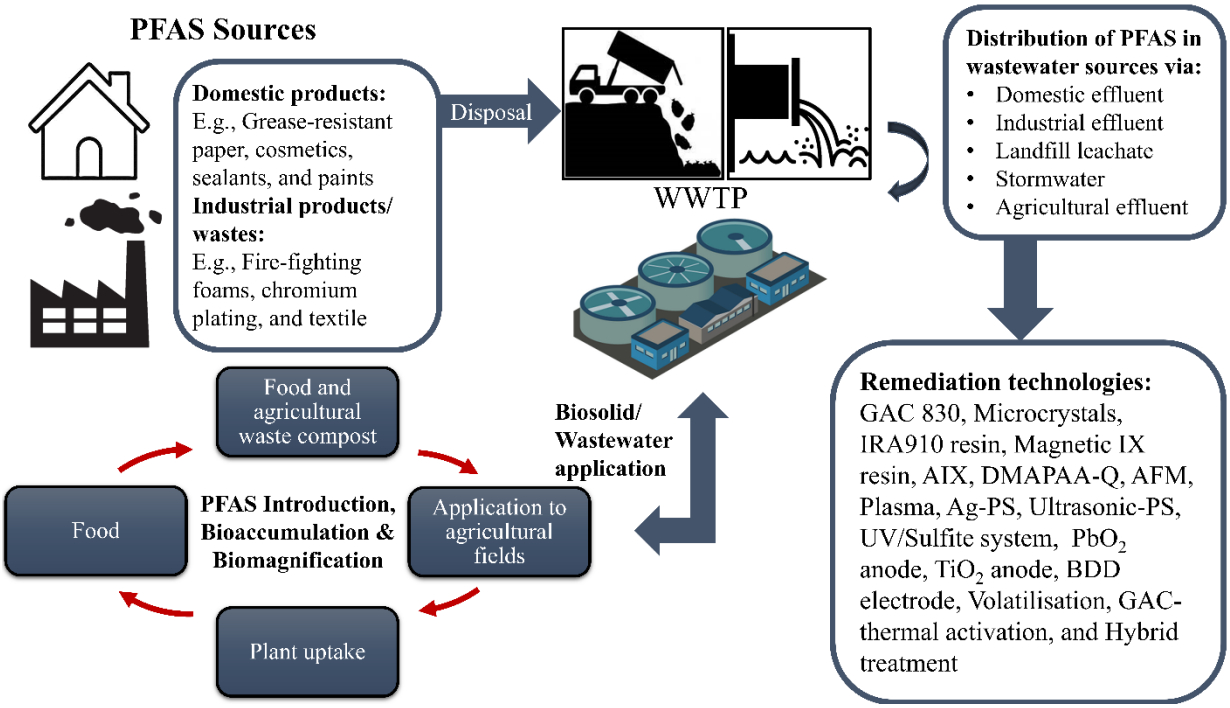
Abstract

Poly- and perfluoroalkyl substances (PFAS) are synthetic chemicals, which reach terrestrial and aquatic environments through anthropogenic activities. Major sources of PFAS in the environment include fire-fighting foams (aqueous film forming foam (AFFF)), wastewater sources, biosolids, and composts. Limited information is available about PFAS in wastewater, which is the focus of this review. PFAS wastewater sources include domestic effluents, industrial effluents, landfill leachates, stormwater, and agricultural effluents through their use in various applications. Land application of PFAS-contaminated wastewater can lead to the contamination of soil and groundwater, thereby reaching the food chain through plant uptake and consumption of potable water. Landfill leachates and industrial effluents contain the highest concentrations of PFAS, posing serious risks to surrounding waterways. Transformation of PFAS precursors can occur through abiotic and biotic processes within the treatment of wastewater, resulting in the formation of harmful PFAS compounds. Currently, there is limited data reported on novel short-chain and ultra-short chain PFAS and PFAS precursors. The strong thermal and chemical stability of PFAS and the complex nature of PFAS mixtures makes the remediation of PFAS in wastewater challenging. However, the review examines and compares current technologies which can treat and remove PFAS from wastewaters. In this review, the distribution, transformation, and remediation of PFAS and their substitutes in wastewater sources are covered.

Keywords:

PFAS, Wastewater, Effluent, Sources, Transformation, Remediation

Graphical abstract:



Abbreviations:

PFAS = Per- and polyfluoroalkyl substances; PFCA = Perfluorinated carboxylic acids; PFSA = perfluorosulfonic acid; FTS = fluorotelomer sulfonate; WWTP = Wastewater treatment plant; AFFF = Aqueous film forming foam; PAHs = Polycyclic aromatic hydrocarbons; POPs = Persistent organic pollutants; DOM = Dissolved organic matter; NF = Nanofiltration; RO = Reverse osmosis; AOT = Advanced oxidation treatment; ART = Advanced reduction treatments; MHT = Microwave hydrothermal treatment; HVED = High-voltage electric discharge; SCWT = Sub- or supercritical water treatment; AC = Activated carbon; PAC = Powdered activated carbon; GAC = Granular activated carbon; UVOx = UV-induced oxidation; PCOx = Photochemical oxidation; PSOx = Persulfate oxidation; ECOx = Electrochemical oxidation

(Insert Table 1) – abbreviations table

1 Introduction

Poly- and perfluoroalkyl substances (PFAS) are a group of manufactured fluorinated organic compounds containing one or more C atoms where F atoms replace the H substituents (Cousins et al., 2020). Polyfluoroalkyl substances are defined as aliphatic substances, where at least one fluorine atom replaces hydrogen atoms in a carbon chain (carbon-fluorine and carbon-hydrogen bonds). In comparison, perfluoroalkyl substances are aliphatic substances where fluorine replaces all H atoms in a carbon chain (carbon-fluorine bonds only) (Buck et al., 2011). There are over 3,000 synthetic PFAS chemicals, and perfluorooctane sulfonate (PFOS), perfluorooctanoic acid (PFOA), and perfluorohexane sulfonate (PFHxS) are some of the most well-known PFAS chemicals (Wang et al., 2017). PFAS are chemically stable in water and oil, and resistant to break down by heat and microorganisms.

Wastewaters are generated from several sources, including domestic effluents (municipal wastewater), industrial effluents, landfill leachates, stormwater, and agricultural effluents (Kunhikrishnan et al., 2012). Irrigation of agricultural land with wastewater, while supplying nutrients for crop production, may transfer and infuse various contaminants including PFAS and other persistent organic pollutants (POPs) (Bhanse et al., 2022), potentially toxic elements (PTEs) (Bolan et al., 2022), polycyclic aromatic hydrocarbons (PAHs) (Kumar et al., 2021), and pesticides into the food chain (Kunhikrishnan et al., 2012; Singh, 2021; Anerao et al., 2022). Most PFAS compounds are chemically and thermally stable, and thus, remediation of PFAS contamination is challenging. In the case of aqueous media such as wastewater and groundwater, remediation of PFAS can be achieved through immobilization using adsorbents and abiotic mineralization using suitable chemical agents. Biotic mineralization of PFAS has seen a limited success, because of high toxicity of these compounds to microorganisms and the high energy needed to break the strong C-F bonds in these compounds (Wackett and McMahon, 2021). Immobilization of PFAS using adsorbents, such as activated carbon, reduces the mobility and bioavailability of these compounds (Ross et al., 2018; Mahinroosta and Senevirathna, 2020; Bolan et al., 2021). A relatively successful approach is to use the remediation method of pump-and-treat with adsorbents (e.g., activated carbon) followed by incineration of the spent activated carbon saturated with PFAS or the off-site safe disposal of spent adsorbent. Other methods for *ex situ* PFAS removal include expensive high-pressure membrane treatment using nanofiltration or reverse osmosis (Kannan, 2011; Ross et al., 2018; Bolan et al., 2021). The most common abiotic PFAS mineralization remediation technologies include: (i) oxidation processes including, photolysis,

electrochemical oxidation, and photocatalysis; (ii) reduction processes involving the use of zero-valent metals/bimetals solely or supported with clay minerals; and (iii) thermal decomposition aimed at breaking C-F bonds under high temperature (Kempisty and Racz, 2021).

Overall, the goal of this paper is to analyse current literature on the sources of PFAS in wastewater, distribution of PFAS in wastewater, remediation of PFAS in wastewater, and its risk management. Figure 1 covering publications on PFAS in soil and water for the past 10 years. The analysis of the literature was performed using Science Direct, Web of Science, Google Scholar, and other sources found on the web using the following key words: 'PFAS', 'Wastewater', 'Biosolids', 'Sources', 'Point sources', 'Transformation', 'Long-chain', 'Short-chain', 'Conventional PFAS', 'Emerging PFAS', 'Domestic effluent', 'Industrial effluent', 'Landfill leachates', 'Stormwater', 'Agricultural effluents', 'Remediation', 'Mobilization', 'Immobilization', 'Adsorption', 'Filtration', 'Treatment', 'Risk management', 'Regulatory guidelines', 'Policies', 'Voluntary initiatives', and 'Monitoring'. After screening, 150 journal articles, were qualified for full-text review. The scope of this review and the article decision-making process is conveyed in SI-Figure 1.

(Insert Figure 1)

2 Sources of PFAS in wastewater and their transformation

2.1 PFAS sources

PFAS are released into the environment via domestic and industrial sources (US EPA, 2021a). Domestic products, including food packaging and commercial household materials, contain PFAS. Food packaging materials are the largest source of PFAS (Vo et al., 2020). These include popcorn bags, grease-resistant paper, fast food containers and wrappers, candy wrappers, and noodle cups (Kotthoff et al., 2015; US EPA, 2021a; Vo et al., 2020). These materials are commonly disposed of in landfills, and they subsequently pollute nearby waterways via landfill leachate. There is also a large range of PFAS-containing commercial household products, which are often discarded into the environment indirectly. These products range from cosmetics, dental floss, shampoo, water repellent chemicals used on carpets, clothing, upholstery, fabrics, cleaning products, sealants, varnishes, paints, and cooking equipment and utensils (Kotthoff et al., 2015; US EPA, 2021a). These materials are also disposed of in landfills and found in landfill leachates and domestic wastewater. Kotthoff et al.

(2015) recorded high levels of PFAS in baking paper (up to 15 μg m^{-2} PFOA), outdoor textiles (up to 19 μg m^{-2} PFOA), leather samples (up to about 200 μg kg^{-1} PFBA and 120 μg kg^{-1} PFBS), and ski waxes (up to about 2000 μg kg^{-1} PFOA). Various countries have reported significantly different concentrations and types of PFAS compounds within food packaging materials that are commonly imported.

PFAS pollution of waterways and wastewater from the mismanagement and disposal of fire-fighting foams [6:2 fluorotelomer fluorotelomer sulfonate (FTS)] is one of the most common sources of industrial PFAS pollution, particularly in the USA, Canada, and Australia (Coggan et al., 2019). Other industrial wastewater sources of PFAS that are released into the environment in high concentrations include chromium plating, textile and leather treatments, printing products, oil and paints, electronics, pesticide formulations, and other synthetic chemicals (such as hydraulic fluid in the aeronautical industry) (KEMI, 2020). The metal plating industry uses a high amount of the global PFAS production. An example of this is the hard-metal industry in Sweden which uses ~180 kg of PFOS during processing. This equates to 7.2% of the global manufacturing of PFOS (KEMI, 2020). Consequently, these PFAS compounds are found in high concentrations in industrial wastewater (shown in Table 2).

2.2 PFAS in wastewater and their transformation

Wastewater is derived from various areas and sectors including domestic and industrial effluent, stormwater, landfill leachate, and agricultural effluent. These wastewater types contain different concentrations of PFAS depending on the type of activity and associated water use. Conventional PFAS compounds (such as PFOA and PFOS) in wastewater have been covered in numerous studies, but information on the levels of PFAS precursors and short-chain PFAS in wastewater is rarely available (Göckener et al., 2021; Lenka et al., 2021). The short-chain PFAS compounds are highly polar in nature (particularly the ultra-short-chain ones), and consequently they are highly soluble and poorly retained in analytical separation columns, which make these compounds challenging to analyse in wastewater matrices (Yeung et al., 2017). Also, the unavailability of appropriate standard chemicals and poor sensitivity on conventional analytical instruments aggravate the problem of quantification of precursor PFAS compounds in wastewater (Lenka et al., 2021). Increased numbers of studies on emerging PFAS compounds in wastewater such as fluorotelomers, which are commonly used in high

amounts within various industries such as fluorinated impregnations, metal plating, oil production, and food packaging, are required (The Danish EPA, 2015).

Studies have indicated that after the ban of longer-chain PFAS compounds, shorter-chain PFAS compounds have become more prevalent (Göckener et al., 2021; Zhao et al., 2016). Göckener et al. (2021) suggest that C4-C6 perfluorinated carboxylic acids (PFCA) in suspended particulate matter in waters seem to be increasing since 2013 and can be indicative of increased use of precursors based on shorter fluorinated chains. Despite the unknown toxicity of many PFAS compounds in wastewater, Li et al. (2020) states how shorter-chain PFAS compounds can have similar or lesser toxicity than longer-chain PFAS compounds, and they are also more mobile and persistent within the environment .

PFAS transformation can occur from precursors under certain industrial, environmental, and metabolic conditions. In contrast to per-fluoroalkyl substances, poly-fluoroalkyl substances contain carbon-hydrogen, carbon-oxygen, and carbon-nitrogen bonds, which could be broken down by abiotic and biotic reactions, transforming the parent compounds to per-fluoroalkyl substances (Ji et al., 2020). Abiotic and biotic processes that cause transformation of PFAS include oxidation, photolysis, and hydrolysis. PFAS precursors can also undergo microbial transformation under anaerobic and aerobic conditions (Allred et al., 2015; Buck et al., 2011).

Unlike stormwater, landfill leachates, and agricultural wastewater, effluents derived from domestic and industrial sources are typically passed through a WWTP due to their high concentrations of contaminants. Despite the degradation of numerous contaminants in the WWTP process, PFAS cannot be removed in WWTPs by conventional treatment methods (Filipovic and Berger, 2015). After treatment, it is known that effluent PFAS levels are typically higher compared to the influent levels due to the transformation of large chain precursor PFAS compounds to smaller chain PFAS compounds such as PFOS (Coggan et al., 2019; Page et al., 2019). For instance, Coggan et al. (2019) recorded that PFAS in influent, primary effluent, secondary effluent, final effluent, and recycled water increased throughout wastewater treatment by 76, 89, 140, 140 and 120 ng $\sum$21 PFAS L^{-1}, respectively. This study showed how chemical and biological treatment in WWTPs could transform PFAS precursors into other PFAS compounds. Such PFAS precursors include FOSA, FOSAA, and FtOH, which could degrade into FOS, PFOA, and PFHxA (Zhao et al., 2013). Vo et al. (2020) compiled several other reasons by which PFAS may increase in the final effluent. One reason is that WWTP operations may integrate PFAS-containing reject water from reverse osmosis processes

with the final effluent, increasing the overall PFAS in the effluent compared to the influent. The 6:2 FTS is another compound that can undergo transformation into PFCAs. Hamid et al. (2020) reported aerobic biotransformation of 6:2 FTS within landfill leachate. PFCA formation (C4-C6) was stimulated in the leachate during nitrification resulting from the presence of ammonia monooxygenase enzyme in which the bacterium *Actinobacteria* played the key role in FTS biotransformation.

3 Distribution of PFAS in wastewater sources

3.1 Domestic effluents

PFOA, PFOS, PFHxS, PFBA, and PFHxA are the most dominant PFAS compounds found in domestic effluents. Table 2 shows the high variability of PFAS concentrations in domestic WWTP effluents, as can be seen from a low level in the study conducted by Zhou et al. (2019) at $\Sigma 15 = 38$ ng L^{-1} compared to a high concentration observed by Zhang et al. (2015) at $\Sigma 14 = 1589$ ng L^{-1}. The review by Vo et al. (2020) reported that PFAS concentrations in domestic effluents were typically >100 ng L^{-1}.

Because conventional long-chain PFAS compounds are being phased out by various countries such as the USA, EU, and Australia since the 2000s, shorter chain PFAS and PFAS precursor compounds with similar properties are inevitably replacing their role (Land et al., 2018). In the study by Coggan et al. (2019), the precursor compounds of 6:2 FTS were detected in 99% of aqueous samples at an average concentration of 7.3 ng 6:2 FTS L^{-1}. This precursor compound is typically used in AFFF and PFOS replacement chemicals used in metal plating. This suggests that harmful PFAS precursors are making their way into domestic effluents. The study by Gallen et al. (2018) recorded considerably larger concentrations of PFAS in effluents compared to influents (2 to 59 times higher), which suggested a large amount of precursors present in the wastewater.

3.2 Industrial effluents

PFAS are man-made chemicals used in various industrial and human activities. Hence, industries are the major source of PFAS in the aquatic system (Buck et al., 2011; Kim et al., 2021). PFAS can enter in the water system through fluoropolymer manufacturing, AFFF manufacturing, electrical and electronic industries, aerospace, electroplating, manufacture of

metal products, textile, oil and gas, and wood industries (Garg et al., 2020; Prevedouros et al., 2006). In addition, fabrics, coating, cosmetics, biocides, and building materials could also be a potential source of PFAS in water bodies (Janousek et al., 2019). With the exception of the results from Dauchy et al. (2017), PFOA, PFOS, PFHxS, and PFHxA were some of the most dominant PFAS compounds found in industrial effluents, as shown in Table 2. High concentrations of PFHxS (12500 ng L^{-1}), and PFOS (8410 and 1115 ng L^{-1}) were recorded in industrial effluents. Interestingly, Dauchy et al. (2017) recorded high concentrations of emerging PFAS compounds from the effluent of a fluorochemical manufacturing facility. FTS compounds (6:2 FTAB, M4, 8:2 FTOH, 6:2 FTSA, and 10:2 FTOH) were in high concentrations and were the most dominant PFAS compounds (>75%) in the industrial effluent. There is a need to perform increased studies on emerging PFAS compounds, particularly from fluorochemical manufacturing facilities, because they are likely to release vast amounts of emerging PFAS compounds into wastewater. Industrial effluents have higher concentrations of PFAS compared to domestic effluents. Variability of PFAS compounds is higher in industrial effluents than domestic effluents.

The recent study by Kim et al. (2021) investigated the distribution of PFAS compounds within 77 different industrial plants in Korea. The study showed how wastewater samples in metal, advanced electronic, textile, and polymer industries were the greatest contributors to PFAS in wastewater (greater than 100 ng PFAS L^{-1} in 64% samples at n=9, and greater than 1000 ng PFAS L^{-1} in 35% samples at n=5). In contrast, the industries that contributed the least PFAS compounds were chemical, battery, glass, and general electronic plants. All plants had varied inputs of perfluorosulfonic acids (PFSAs), PFCAs, and FTS. For instance, PFSA was observed to be the most dominant in the advanced electronic plant samples, whereas the textile, general electronic, glass and ceramic, chemical, and battery plants were dominated by PFCAs. The polymer plants were dominated by the common FTS precursors. Changes in PFAS use patterns at industrial plants could strongly affect PFAS concentrations in wastewater (Kim et al., 2021). Hence, concentration of PFAS and effect of replaced PFAS in the industries should be investigated in future studies.

(Insert Table 2)

3.3 Landfill leachates

Table 3 shows that landfill leachates contain the highest concentrations of PFAS compounds among several other effluents. PFOA, PFOS, PFHxS, PFHxA, PFBA, PFBS, PFPeA, EtFOSAA, 6:2 FTCA, 8:2 FTCA, and 5:3 FTCA are the most dominant PFAS compounds found in landfill leachates. The compounds with the highest concentrations are PFOA with a concentration of 49246 ng L^{-1}, followed by PFBS with a concentration of 15236 ng L^{-1}. Studies that have recorded PFAS precursors and novel PFAS compounds also recorded high concentrations of EtFOSAA (290-8700 ng L^{-1}), and 5:3 FTCA (400-15000 ng L^{-1}) (Lang et al. (2017); Yan et al. (2015a)). This suggests that the extent of PFAS pollution in landfill leachates and other effluents has not yet been fully explored.

Leachate recirculation can trigger leaching, and dilution of contaminants occurs with elevated moisture levels (Huset et al., 2011). Allred et al. (2015) found short-chain PFAS when waste carpets were fed to reactors, and long- and short-chain PFAS were concentrated in bioreactors fed with clothing waste. Biodegradation intermediates, such as MeFBSAA and FTCAs, increase under methanogenic conditions (Allred et al., 2015; Xiao et al., 2012). Leached PFAS concentrations from a municipal-solid-waste-filled anaerobic reactor were ten times higher than those in a reactor that was biologically inactive (Allred et al., 2015; Gallen et al., 2016). During the methanogenic phase of a landfill, intermediates from biodegradation increased, such as n:2 and n:3 FTCAs, including Me-PFOSA. In live reactors, the 5:3 fluorotelomer carboxylate was found to be the most concentrated PFAS. The presence of PFCAs less than 8 carbons in length caused a reduction of leaching in reactors that were controlled abiotically (Allred et al., 2015). PFAS that are water-soluble, like PFAA, could be leached into the environment by landfill leachates (Yan et al., 2015b). PFAS, like FTOHs that are neutral, less water soluble, and have high vapour pressures, could be released with landfill gas (Hamid et al., 2018).

3.4 Stormwater

PFAS in stormwater are derived from non-atmospheric sources of domestic, industrial, and commercial areas, as well as domestic and industrial wastewater effluents, landfill leachates, and agricultural effluents (Page et al., 2019). Like WWTPs, conventional methods (such as constructed wetlands) to reduce levels of PFAS in stormwater are not fully efficient (Page et al., 2019). In comparison to industrial effluents and landfill leachates, PFAS

concentrations in stormwater are low (Table 3). PFOA, PFOS, PFHxS, PFHxA, and PFNA are the most dominant PFAS compounds found in stormwater. Studies exploring PFAS in stormwater are scarce, particularly for emerging and novel PFAS compounds in stormwater. Future studies are required to examine the full range of PFAS compounds found in stormwater.

In recent studies, Bai and Son (2021) and Olmsted et al. (2021) detected more PFCAs than PFSAs in US surface water and stormwater pond sediments. Additionally Bai and Son (2021) found short-chain PFAS compounds in larger concentrations than long-chain PFAS in surface water. In contrast, Olmsted et al. (2021) found an increase in long-chain PFAS compounds in sediments compared to short-chain PFAS compounds. The higher concentration of short-chain PFAS in water was due to the compounds' hydrophilic nature where short-chain PFAS compounds were more likely to desorb from sediments compared to longer-chain PFAS compounds (McCleaf et al., 2017). Moreover, short-chain PFAS compounds are becoming more prevalent due to the phasing out of long-chain PFAS compounds and, subsequently, can be recorded at higher concentrations than long-chain PFAS compounds (Land et al., 2018). Overall, in-depth studies are needed to examine the concentrations of PFAS within stormwater.

(Insert Table 3)

3.5 Agricultural effluents

Although there are multiple studies covering PFAS in domestic and industrial effluents, landfill leachates, and stormwater, information on PFAS levels in agricultural effluents is very limited. There are multiple studies and reviews that have recorded PFAS in agricultural systems, mainly in soil and crops, indicating significant PFAS risks (Bolan et al., 2021; Costello and Lee, 2020). Testing of surface and groundwater at agricultural sites is complex, expensive, and time-consuming. Sources of PFAS in agriculture include the use of organic fertilisers (e.g., composts and digestates), biosolids or sewage sludge, wastewater irrigation, pesticides, and through atmospheric deposition (Choi et al., 2019; Costello and Lee, 2020). The source, concentration level, and diversity of PFAS in agricultural effluents could be dependent on the type of organic fertilisers used, such as those derived from crop residues, biosolids, or municipal solid waste (MSW). Manures, biosolids, and composts are nutrient rich, and, hence, they are applied to soil for improving soil quality. However, the PFAS concentrations in these fertilizing products must be below a critical limit. Ghisi et al. (2019) reported that the main source of PFAS inputs in agricultural systems are from polluted water and application of

biosolids or industrial wastes. Choi et al. (2019) measured the concentration of 17 PFASs in nine municipal solid waste composts (MSW) and one backyard compost. Results revealed that the PFAA (perfluoroalkyl acids) ranged from 28.7 to 75.9 μg kg^{-1} in MSW composts, and from 2.38 to 7.60 μg kg^{-1} in the backyard compost. PFOA and PFOS were recorded in all compost samples, and most contaminants belonged to short-chain PFAA (>64%) and perfluoroalkyl carboxylic acids (PFCA) (>68%) groups (Choi et al., 2019). The evaluation of some composting plants also revealed that source and type of feedstock materials, variation of feedstock and sample collection, and maturity levels primarily influenced the type and content of PFAS compounds. A uniform distribution of PFAS compounds, especially PFCAs, PFOS PFBS, and 6:2 FTSs, was observed in various compost sources (Brändli et al., 2006). Similarly, Sungur et al. (2020) evaluated five biological waste composts applied to the soil as fertiliser in wheat and corn crops. Results revealed that the mean PFOA and PFOS content in all composts varied between 26.1-102.0 ng g^{-1} and 0.21-0.65 ng g^{-1}, respectively.

PFAS can be adsorbed to soil, however, and PFAS compounds can still be discharged from farmland into surface water and groundwater via being dissolved or through suspended particles, particularly short-chain PFAS compounds (Borthakur et al., 2021; Campos Pereira et al., 2018). A recent study by Röhler et al. (2021) examined the long-term behaviour of PFAS leaching out of agricultural fields as a result of the application of contaminated composts in Germany. The study found that long-chain and short-chain PFAS compounds remained in the soil for a long period, and the situation is likely to remain that way for decades. Short-chain PFAS remained at high levels in the field, owing to the PFAS precursors undergoing transformation. The short-chain PFAS were produced, and had accumulated throughout summer and spring, and became mobile during recharge in fall and winter (Röhler et al., 2021). In a similar study, Lindstrom et al. (2011) examined the distribution of ∑10 PFAS in biosolid-applied agricultural soil. The results showed that surrounding surface and groundwater had elevated levels of PFAS, and 22% of the samples exceeded the US EPA regulatory limit of 400 ng L^{-1}. The mobility of PFAS increased with decreased chain length (Lindstrom et al., 2011). Overall, increased number of studies are required to determine contamination of PFAS via run-off of agricultural water to surrounding waterways and their leaching into groundwater.

4 Remediation of PFAS in wastewater sources

4.1 Physical methods

4.1.1 Adsorption

Adsorption is considered as an effective method for the removal of many aquatic pollutants, including PFAS, mainly due to its environmentally friendly nature, low-cost, low-energy requirement, high efficiency, simple design, and ease of operation (Vu and Wu, 2020). Different materials, such as ion-exchange resins (Dixit et al., 2021), carbonaceous materials (e.g., granular activated carbon, biochar) (Xiao et al., 2017), natural and modified clays and clay minerals (Mukhopadhyay et al., 2021), renewable polymeric materials (e.g., chitosan, alginate) (Militao et al., 2021), and inorganic and synthesized materials [e.g., layered double hydroxides, metal organic frameworks (MOF), nanomaterials] (Huo et al., 2022; Li et al., 2021; Saleh et al., 2019) have been investigated for the adsorption of PFAS from wastewater. Similar to other organic and inorganic contaminants, PFAS removal efficacy via adsorption can be affected by several factors (e.g., medium pH, reaction temperature, competing anions, air bubbles) (Mukhopadhyay et al., 2021). Removal of PFAS through adsorption is governed by mechanisms such as electrostatic, hydrophobic, and ligand exchange interactions (Mukhopadhyay et al., 2021).

Different adsorbents have been tested for PFAS removal, and activated carbon (AC) is among the most used adsorbents. The superior adsorptive potential of AC for PFAS is due to its unique porous structure (Pauletto and Bandosz, 2022). Powdered activated carbon (PAC) exhibits greater adsorption capacity than granular activated carbon (GAC) due to the smaller pore size in PAC. The reported adsorption capacity of GAC is in the range 71.6 - 290 mg/g (for PFOS) and 41.3 - 120 mg/g (for PFOA), whereas an adsorption capacity of 560 (for PFOS) and 290 to 500 mg/g (for PFOA) is observed in the case of PAC (Zhang et al., 2019). To replace the expensive AC, several low-cost adsorbents, developed from waste materials, have also been assessed for PFAS removal. Biochar, a carbon-rich material that is prepared from residues produced by forest and agricultural wastes, could provide fiscal advantage over GAC as an adsorbent (e.g., $200~1,000/ton biochar compared to $1,500~3,000/ton GAC) (Zhang et al., 2021). Thus, biochar could be a viable and economically robust replacement for GAC when removing PFAS from wastewater.

For removing short-chain PFAS, such as PFBA and PFBS, from wastewater, anion exchange resins could be a good choice of adsorbent. More than 90% of PFAS, including short-

418 chain (PFBA and PFBS) and long-chain (PFOA and PFOS) compounds, were removed by
419 anion exchange resins (Dixit et al., 2020a), while GAC removed the short-chain compounds
420 only up to 40% (McCleaf et al., 2017). Some other short-chain compounds, such as PFHxA
421 and PFHxS, were also adsorbed (between 10% and 52%) by an alkali modified resin (IRA910)
422 (Maimaiti et al., 2018). Some synthesized adsorbents, such as polymer hydrogel and MOF,
423 were shown effective in removing a mixture of long- and short-chain PFAS from aqueous
424 media (Ateia et al., 2019) (Table 4).

425 Although adsorption is a well-established method for PFAS removal from water using
426 a variety of materials, this process has some limitations. For example, adsorption only
427 concentrates PFAS from the solution phase to the solid phase, which further necessitates
428 appropriate treatment to correctly handle the spent adsorbents. Effective destruction of
429 adsorbed PFAS, either by regeneration or safe disposal of the spent adsorbents, is necessary in
430 this case. Low removal efficiency of short-chain PFAS and mixtures of PFAS compounds and
431 their precursors is another challenge that requires immediate research attention by developing
432 novel adsorbent materials, improving removal capacity of existing materials, and enhancing
433 regeneration potential of the adsorbents.

434

435 **4.1.2 Membrane filtration**

436 Filtration is used for remediation of PFAS in wastewater. Filtration is represented as a
437 separating process of solvent from solute or rejecting solute from solvent by using a
438 semipermeable (porous) membrane (Garg et al., 2021). Filtration can be a more effective
439 method than adsorption with high PFAS removal efficiency (>99%) at high PFAS
440 concentrations (up to 1000 mg/L). The method can reject low and high molecular weight PFAS
441 compounds selectively (Franke et al., 2019; Xu et al., 2021). Filtration basically involves two
442 mechanisms: charged attraction or repulsion followed by sieving. Anionic PFAS or negatively
443 charged organic substances are effectively rejected from solvent media via electrostatic
444 repulsion of the membrane, and then the solvent or selective materials are sieved via a porous
445 membrane (Yoon et al., 2006). Separation of particulate substances is possible by a single
446 sieving operation, but not possible for organic or ionic molecules without combining the
447 rejection operation (Kumari et al., 2020). To overcome this design gap, size dependent and
448 pressure sensitive membranes have been developed, such as (i) low pressure membranes, e.g.,
449 microfiltration (100-400 nm) and ultrafiltration (~10 nm), and (ii) high pressure membranes,

e.g., reverse osmosis and nanofiltration (RO and NF, respectively) (Garg et al., 2021; Tsai et al., 2010). Low-pressure membranes enable one to separate high molecular weight (MW) PFAS (e.g., PFNA, PFOS, PFOA), but disable (or have minimum efficacy) separation of low MW PFAS (e.g., PFBS or PFHxS) from wastewater. In contrast, high-pressure membranes, specially NF, have the ability to effectively separate both low-MW and high-MW PFAS from wastewater (e.g., NF270 with PFHxS rejection ~97% and PFBS rejection ~93%) (Appleman et al., 2013).

NF and RO have been found very efficient in removing many PFAS including PFAA precursors (Mastropietro et al., 2021) Size-exclusion has been suggested as the main mechanism in this filtration process. Other mechanisms, such as electrostatic repulsion, diffusion, and cake layer filtration, have also been suggested to be involved during the process (Franke et al., 2019) As compared to RO, NF (especially the NF270) is recommended as a better option for PFAS removal due to being less expensive and having a better efficiency (Appleman et al., 2014). About 99% PFAS can be rejected using nanofiltration NF270 (Franke et al., 2019).

Reverse osmosis membranes were also used to eliminate PFOS from semiconductor wastewater, showing >99% PFOS removal efficiency (Tang et al., 2006). Boonya-atichart et al. (2016) examined the removal of PFOA by RO under different experimental conditions such as varying pressure and concentration. The efficiency of the process was also determined by comparing spiked deionized water samples and spiked groundwater samples (Boonya-atichart et al., 2016). The PFOA elimination efficacy was found to be 99.78–99.87% and 99.49–99.54% for spiked deionized water and spiked groundwater samples, respectively. The authors reported that the PFOA removal efficiency was enhanced at higher pressure. It was also noted that the total dissolved solids in the groundwater impacted the effectiveness of the NF membrane during the process.

Recent work has attempted to combine filtration with adsorption methods (i.e., AC-nanofiltration, resin-nanofiltration) to maximize rejection of PFAS with high efficacy (Franke et al., 2019). For example, a treatment train comprising the use of a NF membrane was examined for the elimination of PFHxA from industrial processed water. Subsequently, degradation of the NF concentrate using an electrochemical process was investigated (Soriano et al., 2017). The authors reported that PFHxA rejection ranged from 96.6 to 99.4% by varying the operating pressure using an NF270 membrane. The electrochemical degradation rate of PFHxA was found to be 98% followed by effective mineralization. A considerable reduction

(> 95%) of total organic carbon was reported by the authors. Similarly, NF270 and NF90 (commercial membranes) were used to assess the removal and recovery of PFOA, and a higher PFOA rejection (almost 100%) was achieved using the NF90 membrane. The trans-membrane pressure was found to increase linearly with PFOA concentration, reaching a plateau near the critical micelle concentration (Hang et al., 2015).

Membrane filtration has shown promising results for PFAS removal, but there are certain drawbacks such as membrane fouling and high cost, which hinder its large-scale application (Mastropietro et al., 2021). To overcome the drawbacks, different strategies have been proposed including novel designs, advancement in process design, and the evolution of high-functioning materials for membrane production. Moreover, the supplement of nanomaterials and/or adsorbent fillers in the polymeric membrane matrix or hybrid membranes is suggested as a feasible approach to tackle the challenges (Mastropietro et al., 2021).

4.2 Chemical and thermal treatment

4.2.1 Chemical treatments

In chemical treatment, the varying types of advanced and effective methods have been demonstrated, like redox, defluorination, plasma technology, and catalysis. During advanced oxidation or reduction treatments (AOT or ART), some highly active and nonselective free radicals ($e.g., OH^{\blacksquare}$, $CO_3^{\blacksquare -}$, $O_2^{\blacksquare -}$, $SO_4^{\blacksquare -}$), where symbol ▪ is a radical (Merino et al., 2016) or reducing nucleophiles ($e.g., SO_3^{2-}, H^-$) (Ahmed et al., 2020) are generated, which cleave the strong bonds of PFAS and convert PFAS into non-toxic smaller molecules like CO_2 and H_2O (Wanninayake, 2021). Such types of free radicals especially including $OH^{\blacksquare}$ are formed, particularly for AOT, by using some oxidizing agents like H_2O_2, UV radiation, O_3, metal (transition or noble metal), photo catalysts, potassium permanganate or persulfate, ultrasound, and electron beams (Ahmed et al., 2020; Wanninayake, 2021). Among them, some are used to develop effective degradative AOT methods for remediation of PFAS, such as UV-induced oxidation (UVOx), photochemical oxidation (PCOx), persulfate oxidation (PSOx), and electrochemical oxidation (ECOx). Interestingly, all AOT methods work on the degradation of PFAS by using a general pathway, as follows: stepwise radical formation → decarboxylation → C replacement from C-F and form HF → hydroxylation → elimination of HF → smaller molecule/shorter chain PFAS formation. These steps are reviewed by Merino et al. (2016). In UVOx, some aqueous solutions, like bicarbonate ($NaHCO_3$) or periodate ($NaIO_4$) solutions,

are used for effective decomposition of PFAS in the presence of UV radiation (Cao et al., 2010). For example, UV induced $NaHCO_3$ produces $OH^{\blacksquare}$ and $CO_3^{\blacksquare -}$ radicals, which react with PFAS and split it into CO_2, F^- and shorter chain PFAS (e.g., PFHxS) (Phan Thi et al., 2013). But, UVOx has some major limitations, including incomplete mineralization, long reaction times, and comparatively lower removal efficiency of PFOA and PFOS. Incomplete mineralization is also a limitation of the PSOx method. The PSOx method is carried out by the interaction of $SO_4^{\blacksquare -}$and $OH^{\blacksquare}$, which are derived from persulfate in the presence of light and heat. However, PSOx or the thermally activated PSOx method has some benefits over the UVOx method. For example, it provides satisfactory results with PFOA (but not PFOS) remediation, and a lower concentration of shorter chain PFAS byproducts (e.g., PFHpA, PFBA) remains after complete degradation. For example, Yin et al. (2016) observed that the degradation of PFOA was good (89.9%) at low pH (2.0) due to activation of the PSOx method. In addition, Kim et al. (2019) observed considerable decomposition of PFOS and PFOA using a hybrid radiolysis-PSOx method. PFOS was converted into PFOA by elimination of SO_3 and HF in the presence of, respectively, $SO_4^{\blacksquare -}$and $OH^{\blacksquare}$. Due to continuous interaction, the converted PFOA was again degraded until there was full conversion into F^- and CO_2. But, due to a lack of full defluorination, some intermediates were also formed like PFHpA, PFHeA, PFPeA, and PFBA.

To overcome this difficulty, the PCOx methods have been used for PFAS remediation, especially for removal of PFOA and PFOS from wastewater. For maximal mineralization of the incomplete part, Xu et al. (2017) used photochemical oxidation and showed it was an effective method. According to Appleman et al. (2014), shorter chain PFHxS remained and up to ~34 % was mineralized due to accelerated oxidation and free radical formation in the presence the combined action of UV and the chemical catalyst. Generally, photochemical oxidation is considered to be an advanced stage of photolysis. Among the previously described AOT methods, photolysis is the most effective method for the degradation of PFOA and PFOS. But its single use can be ineffective in degrading carboxylic acid types of PFAS and short-chain PFAS (e.g., PFBA, PFBS). As a result, some chemical catalysts (e.g., $TiO_2, CO_3^{2-}, S_2O_8^{2-}, Fe^{3+}$) are used in combination with photolysis to form highly reactive oxidants or free radicals including $H^{\blacksquare}, OH^{\blacksquare}$ and $CO_3^{\blacksquare -}$ (Cao et al., 2010). Depending upon the catalyst, PCOx is an effective method, particularly for PFOA remediation, but it is not good for PFOS remediation. Recently, Ahmed et al. (2020) reviewed research done between 2007-2011 that considered PFOA degradation by the PCOx method. Degradation ranged between 7-

548 90%, which was not good for PFOA removal. Chen et al. (2016) introduced Pb into a UV/TiO_2
549 system, and it had an excellent degradation efficiency for PFOA (99.9%). Most recently, Sun
550 et al. (2022) demonstrated that UV/Fe^{II}NTA (nitrilotriacetic acid) based PCOx method was
551 effective in decomposing PFOS (60%) with lower activation energy (4.74 kJ/mol).

552 Despite the above-mentioned advantages, PSOx and PCOx have drawbacks. They do
553 not provide good defluorination of PFAS. They are ineffective for the removal of PFOS at
554 maximum levels. They have low degradation efficiency of short-chain PFAS (e.g., PFBS,
555 PFBA, PFHxS). They are challenging to apply for industrial purposes. Most of the drawbacks
556 are overcome by the ECOx method. It degrades PFOA with a high conversion rate. It has a
557 higher removal efficiency of PFOS compared to other AOT methods. It has complete
558 mineralization capability and is convenient and cost-effective for practical applications.
559 Therefore, it can be applied for remediation of PFAS in wastewater (Fang et al., 2019; Xu et
560 al., 2021). For example, porous Ti_4O_7 has excellent electrochemical properties as an anode.
561 Lin et al. (2018) showed that it is capable of degrading 93.1% PFOS and almost all PFOA
562 during 120-180 min. Liu et al. (2018) showed that an Al-Zn suboxide anode degraded and
563 converted PFOA (79.4%) into intermediates in a short time (60 min). Wang et al. (2020)
564 developed a Ti suboxide anode that degraded 90% PFOA and PFOS within a short time of 10-
565 30 min. Table 4 shows recent, effective studies with the ECOx method. Nevertheless, the ECOx
566 method has some limitations, like formation of toxic by-products (e.g., chlorine gas, HF,
567 organic halides, bromate, and perchlorate), a higher preparation cost of electrodes, and an
568 expensive energy cost (Trautmann et al., 2015).

569 In comparison with AOT, other chemical methods, such as ART and plasma
570 technology, do not have good performance in comparison to the ECOx method. Major
571 drawbacks include lower efficacy of reducing agents (e.g., $SO_3^{2-}, S_2O_4^{2-}, Fe^{2+}$) (Makarov,
572 2001), challenging operating conditions (e.g., requirements of high pH, temperature, and
573 reductant dose) of ART (Ahmed et al., 2020; Wanninayake, 2021), and incomplete
574 mineralization (Singh et al., 2019). For plasma technology, lower efficacy due to the
575 requirement of high voltage is its major drawback compared to AOT and ECOx. Table 4
576 compares the different methods.

577

4.2.2 Thermal treatment

Commonly known methods for thermal degradation of PFAS are incineration, the high-voltage electric discharge (HVED) process, sub- or supercritical water treatment (SCWT), and microwave hydrothermal treatment (MHT). All of these methods have a high remediation capacity of PFAS from contaminant sources. But thermal treatment has a primary limitation because it inhibits the proper treatment of PFAS in a liquid state (Phong Vo et al., 2020). Before thermal treatment, PFAS must be separated by physical treatment. However, among the above-mentioned methods, last three are used at a low scale or on a laboratory scale due to their complex maintenance and high-cost. But they have better performance compared to the first one (incineration). They require a lower temperature range, get higher removal efficiency, and generate a lower number of toxic by-products (Ahmed et al., 2020).

In general, incineration occurs at a high temperature range 600-1000˚C (Merino et al., 2016). During incineration of PFAS materials, including PFOA and PFOS, some environmentally toxic compounds, like furan and dioxins, and some greenhouse gases, such as fluorocarbons (e.g., hexafluoroethane, tetrafluoromethane), are formed (Merino et al., 2016). Despite present drawbacks, the incineration method has a wide range of use due to its being the only method for high scale application. But, during the last couple of years, these drawbacks have been minimized by investigating advanced technologies. For example, at 700˚C PFOA and PFOS degrade >99.99% to form F^- as the major by-product in the presence of an N_2atmosphere (Xiao et al., 2020). At same temperature (700˚C), another method for incineration enhanced mineralization efficiency via addition of some cationic additives, e.g., Ca^{2+}, Na^+. The recovery rate of PFOA, PFOS, and PFHxA was 74%, 90%, and 91%, respectively, which was increased from 30%, 72%, and 46% without the additives (Watanabe et al., 2018). In more advanced research, a GAC-based incineration treatment was develop that operated at a lower temperature (200˚C) (Duchesne et al., 2020). Moreover, SCWT is able to decompose the short-chain PFAS. Hori et al. (2008) showed that 94.8 % PFHxA and 83.6 % PFHxS were degraded under super- and sub-critical conditions, respectively. In comparison with SCWT, MHT treatment operates at a lower temperature (60-90˚C) for degradation of PFAS, when it is combined with persulfate (Lee et al., 2009). But the MHT system needs more development, if it is to be used in future industrial applications.

Every method has drawbacks, and any single method is not able to provide satisfactory remediation. Remediation depends on many factors, like removal efficiency (shown in Figure 2), cost effectiveness, time duration for treatment, and whether the method is environmentally

611 friendly. In the future, hybrid technologies are needed that can minimize the cost and toxic
612 effects. Figure 3 shows hybrid designs.

613 (Insert Figure 2 and 3)

614 (Insert Table 4)

615

616 5. Economy and environmental impacts of PFAS remediation technologies

617 Although, PFASs are extremely refractory to conventional water management and
618 treatment operation, there are few successful methods existing which are comprehensively
619 discussed in section 4. Nevertheless, these methods involve significant costs and high
620 environmental impacts owing to its energy intensiveness, high operational or capital costs, and
621 uses of chemicals which generated secondary pollutants (Wanninayake, 2021). Pillai (2020)
622 applied e-beam system to treat the wastewater and estimated a treatment cost ranging from
623 $0.10 (treatment at 500 kGy) to $0.80 (treatment at 3000 kGy) per gallon and a capital cost of
624 $7.7 million US dollars when e-beam system operated at 10 MeV, 560-kW, and 1500 kGy.
625 Gole et al. (2018) applied a sono-treatment method, by providing a single/dual frequencies 1
626 MHz and 500 kHz at 91L reactor and estimated the degradation of AFFFs into F^- and SO_4^{2-}.
627 The authors described that the F^- and SO_4^{2-} generated from AFFFs degradation was inversely
628 proportional to pH of the media and dilution ratio. This finding established the potency of sono-
629 chemical method for the treatment of PFAS concentrate waste streams. The energy cost of this
630 operation with 500 times dilution (at pH 4) in a 91L reactor was evaluated and found in the
631 range of $0.015 and $0.019 per liter of the media.

632 To manage the PFAS-laden waste, instead of defluorination, two approaches such as deep
633 well injection and landfill are presently adopted and considered as effective (Tow et al., 2021).
634 Even though, the per volume cost of deep well injection differs widely and has been found
635 between $0.93 m^3 and $4.58/$m^3$ for an injection stream of capacity 0.25 MGD and between
636 $0.15/$m^3$ and $0.48/$m^3$ for 2.5 MGD (Foldager, 2003). Another investigation estimated the cost
637 range between $0.54/$m^3$ and $2.65/$m^3$ (Panagopoulos et al., 2019). Furthermore, a recent
638 investigation assessed the price for third-party disposal of PFAS liquid waste as $48/$m^3$-$66/m^3
639 (DeSilva, 2019). Engineered landfill sites, which are generally used for the gradual degradation
640 and storage of solid waste, can be also used to store PFAS-laden solids from wastewater
641 treatment process such as non-regenerated PAC or GAC, spent IX resins, or solids from zero-
642 or minimal-liquid-discharge treatment operations (from an evaporation pond), nevertheless the

PFAS sequestration efficiency of landfills have not be well investigated and established (Tow et al., 2021). However, US EPA (2020) reported the estimated cost of landfilling spent GAC from PFAS removal which was $0.04/lb ($0.09/kg). More recently, a study conducted in Thailand by Kanchanapiya and Tantisattayakul (2022) and reported the treatment unit cost of landfill leachate in the ranged of 1.72–2.71 USD/m^3 for the system with an evaporation pond and ranged from 1.06-2.09 USD/m^3 for the system without an evaporation pond, depending on the size of the landfills.

Although, IX resins showed better remediation potency of PFAS contaminated wastewater than GAC-based treatment systems, still, several research gaps are persisting related to the single-use-and-dispose vs regenerate-reuse approaches of IX resins processes. However, based on life cycle analysis (LCA), production process of IX resin has been recognized as one of the crucial steps from an environmental viewpoint and attributed a majority of environmental impacts, such as global warming potential, carcinogenicity, ecotoxicity, eutrophication, and acidification, (Dixit et al., 2020b; Maul et al., 2014). Therefore, regeneration and reuse of IX resins is required in consideration with an environmental prospective, principally the environmental burden imposes by the IX resins production processes. Recent investigations revealed that regenerable organic scavenger resins can function for extended durations in comparison to PFAS-specific resins in single-use mode (Coyle et al., 2020; Dixit et al., 2020c). Nevertheless, regenerable organic scavenger resins needed more resin contact duration than its counterpart, which are single-use resins. An extended contact duration infers that a huge volume of resin is required to treat contaminated wastewater. Furthermore, management of PFAS-laden brine remains a key challenge. These days, application of IX resins for a single use is more often than carrying out regeneration owing to the contaminated brine problem. Nevertheless, for long term operation on PFAS laden wastewater sources, optimization of process parameters of IX resins regeneration and brine handling procedures is necessary. Maga et al., 2021 performed an LCA of AFFF media treatment by applying PerfluorAd®. The application of PerfluorAd® reduce PFAS concentration before its treatment with activated carbon and incineration. Results of the investigation revealed that, the application of PerfluorAd® significantly reduced the environmental impact of PFAS remediation (Maga et al., 2021).

Nzeribe et al. (2019) reported EC treatment of PFAS contaminated synthetic wastewater at the bench and pilot scale in short span of time (Nzeribe et al., 2019). Nevertheless, real environmental wastewater may demand longer treatment duration and may perceive reduced

operation performance and electrode life. Whereas, commercial boron-doped diamond (BDD) electrodes are the most preferable and widely discussed in the published work, nevertheless they are expensive (~$7,125/$m^2$) and hard to synthesize (Chaplin, 2019). Similar to BDD, other mixed metal oxide (MOO) electrodes, such as Ti_4O_7, which are cost-effective than BDD have demonstrated effectively in remediation of PFAS but have yet to be made saleable (Le et al., 2019).

6. Risk management

6.1 Regulatory guidelines, policies, and voluntary initiatives

The establishment of a rigorous risk management of PFAS compounds is extremely complex due to several parameters:

- High amounts of different PFAS compounds currently being used,
- Novel PFAS compounds and precursors currently being developed and implemented which mostly go undetected,
- Unknown toxicity of most PFAS compounds,
- Monitoring and detection of PFAS compounds are time consuming and costly,
- Difficulty in detecting and measuring PFAS compounds, particularly short- and ultra-short-chain PFAS compounds (Yeung et al., 2017),
- Expensive and highly sensitive equipment is required to analyse water samples for PFAS compounds that are typically recorded in nanograms.

Most regulatory agencies and governments worldwide have yet to establish guideline values of PFAS in wastewater effluent (Podder et al., 2021). As outlined by Podder et al. (2021), the only countries that have established guideline values are the USA, Canada, Sweden, Italy, Germany, and Australia (Table 5). However, most of these guidelines are basic, typically only as a guidance (non-enforceable) and only cover conventional PFAS compounds such as PFOS and PFOA. The US EPA is planning to set regulatory limits on industrial PFAS effluents, including nine industrial categories, and they are expected to be released in 2022 and will be ongoing (US EPA, 2021b). Currently, the US EPA has established non-regulatory lifetime health advisories of 70 ng L^{-1} limit of ∑PFOA and PFOS on drinking water (US EPA, 2016a, b). Twelve US states have also provided drinking water guidelines for PFAS and are listed in the studies by Post (2021) and Podder et al. (2021). Moreover, Germany has also set the guidelines

of ∑PFOA and PFOS at 100 ng L^{-1} (Kim et al., 2012) and Italy has set the guidelines of ∑PFOA, ∑PFOS, and ∑ other PFAS at 500, 30, and 500 ng L^{-1}, respectively (Chohan et al., 2021). In Australia, PFOS and PFOA guidelines have been set for the protection of 99% of species to 0.23 ng L^{-1} by the ANZECC water quality guideline, as mentioned in Section 2.2 (HEPA, 2020). Sweden currently covers the most PFAS compounds in drinking water. These compounds include the sum of PFOA, PFOS, PFHxS, PFHxA, PFHpA, PFBS, 6:2 FTS, PFBA, PFPeA, PFNA, and PFDA and have a standard of 90 ng L^{-1} (Chohan et al., 2021).

Currently there are policies and voluntary initiatives that are attempting to reduce the use of PFAS compounds. The Stockholm Convention is a global treaty that, since 2009, has restricted the use and production of PFOS and PFOA substances (Stockholm Convention, 2019). Although 152 countries have ratified the Stockholm Convention, numerous countries are still using other PFAS compounds as the treaty is non-enforceable. An example of use of various unregulated PFAS is in China, the major producer and consumer of PFAS, despite the fact that China ratified the Stockholm Convention (Brennan et al., 2021). Moreover, China continues to use the restricted substances of PFOA and PFOS in firefighting foams. In 2020, the European Commission published the European Chemicals Strategy, which provided a strategy to phase-out all PFAS compounds, except those that are characterised as essential for a particular application (European Commission, 2020). Likewise with the Stockholm Convention, this strategy is not currently enforceable. However, countries such as Germany, Norway, and Sweden have raised serious concerns and proposed restrictions of certain PFAS compounds, such as PFHxA, PFHxS, and the emerging compounds of HFPO-DA and GenX (Brennan et al., 2021). Shifting of the use of fluorinated fire-fighting foams to non-fluorinated foams in the aviation industry, development of a national action plan for PFAS in all countries, and dialogue between various stakeholders are also needed to minimise PFAS risk. Voluntary initiatives are another strategy that have been used by Canada to reduce PFAS released into the environment. An agreement in 2015 between PFAS manufacturers and the government has seen many companies (e.g., Arkema Canada Inc. and Asahi Glass Company LTD) eliminate the manufacture of long-chain PFAS and precursor compounds (Brennan et al., 2021; Government of Canada, 2018). Collection of PFAS analysis data from different sources, including groundwater, surface water, and drinking water, and continuous evaluation and reporting of PFAS concentrations in drinking water and surface water are required to minimize the risk to humans and aquatic organisms. Overall, efforts have been made to have adequate policies and voluntary initiatives. However, the complexity of the situation and knowledge

gaps of PFAS have caused serious barriers to properly make effective regulations, policies, and initiatives.

(Insert Table 5)

6.2 Monitoring of PFAS

Monitoring of PFAS is essential to be able to reduce the risk of high levels of PFAS compounds entering wastewater effluent (Andrews et al., 2021). As shown in Table 2 and 3, the major countries that have led the way for monitoring PFAS in wastewater are Australia, China, the USA, Canada, Korea, and some European countries, particularly in areas that are expected to have high PFAS compounds present (industrial effluent and landfill leachate). Out of the 3,000 PFAS compounds, there have only been at most 24 PFAS compounds recorded in a study, which represents only 0.8% of PFAS compounds monitored within a single study.

Monitoring of PFAS is essential for tracking down point-source pollution occurring intentionally or unintentionally. These point sources typically dispose of large amounts of PFAS in effluents. Therefore, higher volumes of monitoring of various PFAS compounds are required to help with tracking the point source of the PFAS pollution, particularly in the industrial context where improper PFAS waste disposal typically goes unnoticed (Andrews et al., 2021). The case studies examined by Andrews et al. (2021) convey how large scale monitoring resulted in the identification of a large number of PFAS sites in Michigan and California. These studies have provided a deeper understanding of the main polluter of PFAS in wastewater effluent (Michigan Department of Environment, 2019). The Michigan government launched a program to identify and eliminate point sources of PFAS. The state's PFAS water quality guidelines are >12 ng PFOS L^{-1}; >420 ng PFOS L^{-1}. Thirty-one out of 95 WWTP sites had exceeded the guidelines. Based on these results, and the testing of other potential sites, landfill leachates represented the largest source of PFAS being discharged with 49 out of 56 landfills recording PFAS levels over the guidelines (ranging from 13-5000 ng L^{-1}). Ninety-five percent of WWTPs that received landfill leached PFOS that exceeded the guidelines. Forty-seven out of 320 metal plating facilities that had effluent discharged into the WWTP exceeded the PFOS limit of which 33 out of 50 of these were chrome plating. These facilities had high levels of PFOS (20-240,000 ng L^{-1}). Four out of 17 chemical manufacturers had effluent exceeding the PFOS guidelines. One of the sites had an extremely high reading of 4,600,000 ng L^{-1} and showed how chemical manufacturers can be one of the biggest PFAS

polluters in wastewater effluent. Similarly, testing in California monitored the inputs of PFAS in wastewaters. However, the study Andrews et al. (2021) is still mostly underway, because only 9 out of 217 chrome plating facilities have been tested. The testing includes chrome plating facilities (n=9), landfills (n=189), municipal wastewater plants (n=135), and airport sites (n=21). Currently for these sites, the median value of 25 PFAS at airports, landfills, municipal wastewater plants, and airports were 54 ng L^{-1}, 59 ng L^{-1}, 95 ng L^{-1} and 6569 ng L^{-1}. However, despite the higher concentration at the airport sites, the other sites of main concern are more frequent and widespread (Andrews et al., 2021). Overall, these case studies are consistent with the findings in Section 3, where landfill leachates and industrial effluents result in the greatest inputs of PFAS into wastewater. The case studies also show how, by monitoring of effluents, large amounts of PFAS from point sources can be identified and eliminated.

7. Conclusions

Wastewater sources are one of the most important point sources of PFAS reaching surface and groundwaters due to disposal of various domestic and industrial PFAS-containing products. The biological and chemical treatments within WWTPs can transform PFAS precursors. Abiotic and biotic processes that cause transformation of PFAS include oxidation, photolysis, and hydrolysis. Landfill leachates and industrial effluents contain the highest concentrations of PFAS levels and are released into waterways. The occurrence, distribution, and transformation of PFAS in landfill sources are dependent on a number of factors that include the nature of waste disposed, age of the landfill site, the type of landfill (i.e., engineered or conventional), and climatic conditions (i.e., temperature and rainfall). The fate of accumulated PFAS inside the wastewater sources is regulated by a combination of biological (biodegradation) and abiotic (desorption) processes.

After the ban of longer-chain PFAS compounds, shorter-chain PFAS compounds have replaced these compounds and hence, have become more prevalent in wastewater sources. Shorter-chain PFAS can be more mobile and persistent within soils and wastewaters. Moreover, harmful PFAS precursors are making their way into domestic effluents. Treatment of long-chain PFAS compounds are more effective compared to shorter-chain compounds and are often poorly removed. Anion exchange resins and SCWT are more efficient at the removal of short-chain PFAS compared to other treatments. While various technologies have been tested for treating

803 firefighting foam-derived PFAS in groundwater, our knowledge is limited on the treatability
804 of PFAS in wastewater sources.

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806 **References**

807 Ahmed, M.B., Alam, M.M., Zhou, J.L., Xu, B., Johir, M.A.H., Karmakar, A.K., Rahman,
808 M.S., Hossen, J., Hasan, A.T.M.K., Moni, M.A., 2020. Advanced treatment
809 technologies efficacies and mechanism of per- and poly-fluoroalkyl substances
810 removal from water. Process Saf. Environ. Prot. 136, 1-14.
811 https://doi.org/10.1016/j.psep.2020.01.005

812 Allred, B.M., Lang, J.R., Barlaz, M.A., Field, J.A., 2015. Physical and biological release of
813 poly-and perfluoroalkyl substances (PFASs) from municipal solid waste in anaerobic
814 model landfill reactors. Environ. Sci. Technol. 49, 7648-7656.
815 https://doi.org/10.1021/acs.est.5b01040

816 Andrews, D.Q., Hayes, J., Stoiber, T., Brewer, B., Campbell, C., Naidenko, O.V., 2021.
817 Identification of point source dischargers of per-and polyfluoroalkyl substances in the
818 United States. AWWA Water Science 3, e1252. https://doi.org/10.1002/aws2.1252

819 Anerao, P., Kaware, R., kumar Khedikar, A., Kumar, M., Singh, L., 2022. Phytoremediation
820 of persistent organic pollutants: Concept challenges and perspectives. In
821 Phytoremediation Technology for the Removal of Heavy Metals and Other
822 Contaminants from Soil and Water (pp. 375-404). Elsevier.
823 https://doi.org/10.1016/B978-0-323-85763-5.00018-0.

824 Appleman, T.D., Dickenson, E.R.V., Bellona, C., Higgins, C.P., 2013. Nanofiltration and
825 granular activated carbon treatment of perfluoroalkyl acids. J. Hazard. Mater. 260,
826 740-746. https://doi.org/10.1016/j.jhazmat.2013.06.033

827 Appleman, T.D., Higgins, C.P., Quiñones, O., Vanderford, B.J., Kolstad, C., Zeigler-Holady,
828 J.C., Dickenson, E.R.V., 2014. Treatment of poly- and perfluoroalkyl substances in
829 U.S. full-scale water treatment systems. Water Res. 51, 246-255.
830 https://doi.org/10.1016/j.watres.2013.10.067

831 Ateia, M., Arifuzzaman, M., Pellizzeri, S., Attia, M.F., Tharayil, N., Anker, J.N., Karanfil, T.,
832 2019. Cationic polymer for selective removal of GenX and short-chain PFAS from
833 surface waters and wastewaters at ng/L levels. Water Res. 163, 114874.
834 https://doi.org/10.1016/j.watres.2019.114874

835 Ateia, M., Attia, M.F., Maroli, A., Tharayil, N., Alexis, F., Whitehead, D.C., Karanfil, T.,
836 2018. Rapid removal of poly-and perfluorinated alkyl substances by poly
837 (ethylenimine)-functionalized cellulose microcrystals at environmentally relevant
838 conditions. Environ. Sci. Technol. Lett. 5, 764-769.
839 https://doi.org/10.1021/acs.estlett.8b00556

840 Bai, X., Son, Y., 2021. Perfluoroalkyl substances (PFAS) in surface water and sediments
841 from two urban watersheds in Nevada, USA. Sci. Total Environ. 751, 141622.
842 https://doi.org/10.1016/j.scitotenv.2020.141622

843 Benskin, J.P., Li, B., Ikonomou, M.G., Grace, J.R., Li, L.Y., 2012. Per-and polyfluoroalkyl
844 substances in landfill leachate: patterns, time trends, and sources. Environ. Sci.
845 Technol. 46, 11532-11540. https://doi.org/10.1021/es302471n

Bentel, M.J., Liu, Z., Yu, Y., Gao, J., Men, Y., Liu, J., 2020. Enhanced degradation of perfluorocarboxylic acids (PFCAs) by UV/sulfite treatment: Reaction mechanisms and system efficiencies at pH 12. Environ. Sci. Technol. Lett. 7, 351-357. https://doi.org/10.1021/acs.estlett.0c00236

Bhanse, P., Kumar, M., Singh, L., Awasthi, M.K., Qureshi., 2022. Role of plant growth-promoting rhizobacteria in boosting the phytoremediation of stressed soils: Opportunities, challenges, and prospects. Chemosphere. 134954. https://doi.org/10.1016/j.chemosphere.2022.134954

Bolan, N., Kumar, M., Singh, E., Kumar, A., Singh, L., Kumar, S., Keerthanan, S., Hoang, S.A., El–Naggar, A., Vithanage, M., Sarkar, B., 2022. Antimony contamination and its risk management in complex environmental settings: A review. Environ. Inter. 158, 106908. https://doi.org/10.1016/j.envint.2021.106908.

Bolan, N., Sarkar, B., Vithanage, M., Singh, G., Tsang, D.C., Mukhopadhyay, R., Ramadass, K., Vinu, A., Sun, Y., Ramanayaka, S., 2021. Distribution, behaviour, bioavailability and remediation of poly-and per-fluoroalkyl substances (PFAS) in solid biowastes and biowaste-treated soil. Environ. Int. 155, 106600. https://doi.org/10.1016/j.envint.2021.106600

Boonya-atichart, A., Boontanon, S.K., Boontanon, N., 2016. Removal of perfluorooctanoic acid (PFOA) in groundwater by nanofiltration membrane. Water Sci. Technol. 74, 2627-2633. http://doi.org/10.2166/wst.2016.434

Borthakur, A., Wang, M., He, M., Ascencio, K., Blotevogel, J., Adamson, D.T., Mahendra, S., Mohanty, S.K., 2021. Perfluoroalkyl acids on suspended particles: Significant transport pathways in surface runoff, surface waters, and subsurface soils. J. Hazard. Mater. 417, 126159. https://doi.org/10.1016/j.jhazmat.2021.126159

Bossi, R., Strand, J., Sortkjær, O., Larsen, M.M., 2008. Perfluoroalkyl compounds in Danish wastewater treatment plants and aquatic environments. Environ. Int. 34, 443-450. https://doi.org/10.1016/j.envint.2007.10.002

Brennan, N.M., Evans, A.T., Fritz, M.K., Peak, S.A., von Holst, H.E., 2021. Trends in the regulation of per-and polyfluoroalkyl substances (PFAS): A scoping review. Int. J. Env. Res. Public Health 18, 10900. https://doi.org/10.3390/ijerph182010900

Buck, R.C., Franklin, J., Berger, U., Conder, J.M., Cousins, I.T., De Voogt, P., Jensen, A.A., Kannan, K., Mabury, S.A., van Leeuwen, S.P., 2011. Perfluoroalkyl and polyfluoroalkyl substances in the environment: terminology, classification, and origins. Integr. Environ. Assess. Manag. 7, 513-541. https://doi.org/10.1002/ieam.258

Campos Pereira, H., Ullberg, M., Kleja, D.B., Gustafsson, J.P., Ahrens, L., 2018. Sorption of perfluoroalkyl substances (PFASs) to an organic soil horizon – Effect of cation composition and pH. Chemosphere 207, 183-191. https://doi.org/10.1016/j.chemosphere.2018.05.012

Cao, M.H., Wang, B.B., Yu, H.S., Wang, L.L., Yuan, S.H., Chen, J., 2010. Photochemical decomposition of perfluorooctanoic acid in aqueous periodate with VUV and UV light irradiation. J. Hazard. Mater. 179, 1143-1146. https://doi.org/10.1016/j.jhazmat.2010.02.030

CARE, 2019. GROUNDWATER INFORMATION SHEET: Perfluorooctanoic Acid (PFOA) & Perfluorooctanesulfonic Acid (PFOS). State Water Resources Control Board Division of Water Quality GAMA Program.

CELA, 2019. THE THREAT OF PFAS: THE FOREVER CHEMICALS, Canadian Environmental Law Association. A call for Canadian Citizen Action to protect theGreat Lakes – St. Lawrence River Ecosystem.

Chaplin, B.P., 2019. The prospect of electrochemical technologies advancing worldwide water treatment. Acc. Chem. Res. 52, 596-604.

Chen, M.-J., Lo, S.-L., Lee, Y.-C., Kuo, J., Wu, C.-H., 2016. Decomposition of perfluorooctanoic acid by ultraviolet light irradiation with Pb-modified titanium dioxide. J. Hazard. Mater. 303, 111-118. https://doi.org/10.1016/j.jhazmat.2015.10.011

Chohan, A., Petaway, H., Rivera-Diaz, V., Day, A., Colaianni, O., Keramati, M., 2021. Per and polyfluoroalkyl substances scientific literature review: water exposure, impact on human health, and implications for regulatory reform. Rev. Environ. Health 36, 235-259. https://doi.org/10.1515/reveh-2020-0049

Choi, Y.J., Kim Lazcano, R., Yousefi, P., Trim, H., Lee, L.S., 2019. Perfluoroalkyl acid characterization in U.S. municipal organic solid waste composts. Environ. Sci. Technol. Lett. 6, 372-377. https://doi.org/10.1021/acs.estlett.9b00280

Clara, M., Scharf, S., Weiss, S., Gans, O., Scheffknecht, C., 2008. Emissions of perfluorinated alkylated substances (PFAS) from point sources—identification of relevant branches. Water Sci. Technol. 58, 59-66. https://doi.org/10.2166/wst.2008.641

Coggan, T.L., Moodie, D., Kolobaric, A., Szabo, D., Shimeta, J., Crosbie, N.D., Lee, E., Fernandes, M., Clarke, B.O., 2019. An investigation into per- and polyfluoroalkyl substances (PFAS) in nineteen Australian wastewater treatment plants (WWTPs). Heliyon 5, e02316. https://doi.org/10.1016/j.heliyon.2019.e02316

Cordner, A., De La Rosa, V.Y., Schaider, L.A., Rudel, R.A., Richter, L., Brown, P., 2019. Guideline levels for PFOA and PFOS in drinking water: the role of scientific uncertainty, risk assessment decisions, and social factors. J. Expo Sci. Environ. Epidemiol. 29, 157–171. https://doi.org/10.1038/s41370-018-0099-9.

Costello, M.C.S., Lee, L.S., 2020. Sources, fate, and plant uptake in agricultural systems of per- and polyfluoroalkyl substances. Curr. Pollut. Rep., 1-21. https://doi.org/10.1007/s40726-020-00168-y

Cousins, I.T., DeWitt, J.C., Glüge, J., Goldenman, G., Herzke, D., Lohmann, R., Miller, M., Ng, C.A., Scheringer, M., Vierke, L., 2020. Strategies for grouping per-and polyfluoroalkyl substances (PFAS) to protect human and environmental health. Environ. Sci.: Process. Impacts. 22, 1444-1460. https://doi.org/10.1039/d0em00147c

Coyle, C., Ghosh, R., Leeson, A., Thompson, T., 2020. US department of defence-funded research on treatment of per- and polyfluoroalkyl substance-laden materials. Environ. Toxicol. Chem. https://doi.org/10.1002/etc.4836.

Crownover, E., Oberle, D., Kluger, M., Heron, G., 2019. Perfluoroalkyl and polyfluoroalkyl substances thermal desorption evaluation. Remediation 29, 77-81. https://doi.org/10.1002/rem.21623

Dauchy, X., Boiteux, V., Bach, C., Colin, A., Hemard, J., Rosin, C., Munoz, J.-F., 2017. Mass flows and fate of per- and polyfluoroalkyl substances (PFASs) in the wastewater treatment plant of a fluorochemical manufacturing facility. Sci. Total Environ. 576, 549-558. https://doi.org/10.1016/j.scitotenv.2016.10.130

936 DeSilva, V. (2019). PFAS in landfill leachate. http://www.
937 mowastecoalition.org/resources/Documents/2019conference/MWCC2019-
938 deSilva(SCS)-PFAS-July16,2019.pdf

939 Dixit, F., Barbeau, B., Mostafavi, S.G., Mohseni, M., 2020a. Removal of legacy PFAS and
940 other fluorotelomers: Optimized regeneration strategies in DOM-rich waters. Water
941 Res. 183, 116098. https://doi.org/10.1016/j.watres.2020.116098

942 Dixit, F., Barbeau, B., Mostafavi, S.G., Mohseni, M., 2020b. Efficient removal of GenX
943 (HFPO-DA) and other perfluorinated ether acids from drinking and recycled waters
944 using anion exchange resins. J. Hazard Mater. 384, 121261.
945 https://doi.org/10.1016/j.jhazmat.2019.121261.

946 Dixit, F., Barbeau, B., Mostafavi, S.G., Mohseni, M., 2020c. PFAS and DOM removal using
947 an organic scavenger and PFAS-specific resin: trade-off between regeneration and
948 faster kinetics. Sci. Total Environ. 142107.
949 https://doi.org/10.1016/j.scitotenv.2020.142107.

950 Dixit, F., Dutta, R., Barbeau, B., Berube, P., Mohseni, M., 2021. PFAS removal by ion
951 exchange resins: A review. Chemosphere 272, 129777.
952 https://doi.org/10.1016/j.chemosphere.2021.129777

953 Duchesne, A.L., Brown, J.K., Patch, D.J., Major, D., Weber, K.P., Gerhard, J.I., 2020.
954 Remediation of PFAS-contaminated soil and granular activated carbon by smoldering
955 combustion. Environ. Sci. Technol. 54, 12631-12640.
956 https://doi.org/10.1021/acs.est.0c03058

957 European Commission. 2020. Communication from the Commission to the European
958 Parliament, The Council, The European Economic and Social Committee and the
959 Committee of the Regions Chemicals Strategy for Sustainability Towards a Toxic-
960 Free Environment. https://eur-lex.europa.eu/legal-
961 content/EN/TXT/?uri=COM%3A2020%3A667%3AFIN. (Accessed 10/10/2021).

962 Fang, C., Sobhani, Z., Niu, J., Naidu, R., 2019. Removal of PFAS from aqueous solution
963 using PbO2 from lead-acid battery. Chemosphere 219, 36-44.
964 https://doi.org/10.1016/j.chemosphere.2018.11.206

965 Filipovic, M., Berger, U., 2015. Are perfluoroalkyl acids in waste water treatment plant
966 effluents the result of primary emissions from the technosphere or of environmental
967 recirculation? Chemosphere 129, 74-80.
968 https://doi.org/10.1016/j.chemosphere.2014.07.082

969 Foldager, R. A., 2003. Economics of desalination concentrate disposal methods in inland
970 regions: Deep-well injection, evaporation ponds, and salinity gradient solar ponds.
971 New Mexico State University.

972 Franke, V., McCleaf, P., Lindegren, K., Ahrens, L., 2019. Efficient removal of per- and
973 polyfluoroalkyl substances (PFASs) in drinking water treatment: nanofiltration
974 combined with active carbon or anion exchange. Environ. Sci. Water Res. Technol. 5,
975 1836-1843. https://doi.org/10.1039/C9EW00286C

976 Franke, V., Ullberg, M., McCleaf, P., Wålinder, M., Köhler, S.J., Ahrens, L., 2021. The Price
977 of really clean water: combining nanofiltration with granular activated carbon and
978 anion exchange resins for the removal of per- and polyfluoralkyl substances (PFASs)
979 in drinking water production. ACS ES&T Water. 1, 782-795.
980 https://doi.org/10.1021/acsestwater.0c00141

Gagliano, E., Sgroi, M., Falciglia, P.P., Vagliasindi, F.G.A., Roccaro, P., 2020. Removal of poly- and perfluoroalkyl substances (PFAS) from water by adsorption: role of PFAS chain length, effect of organic matter and challenges in adsorbent regeneration. Water Res. 171, 115381 https://doi.org/10.1016/j.watres.2019.115381.

Gallen, C., Drage, D., Eaglesham, G., Grant, S., Bowman, M., Mueller, J., 2017. Australia-wide assessment of perfluoroalkyl substances (PFASs) in landfill leachates. J. Hazard. Mater. 331, 132-141. https://doi.org/10.1016/j.jhazmat.2017.02.006

Gallen, C., Drage, D., Kaserzon, S., Baduel, C., Gallen, M., Banks, A., Broomhall, S., Mueller, J., 2016. Occurrence and distribution of brominated flame retardants and perfluoroalkyl substances in Australian landfill leachate and biosolids. J. Hazard. Mater. 312, 55-64. https://doi.org/10.1016/j.jhazmat.2016.03.031

Gallen, C., Eaglesham, G., Drage, D., Nguyen, T.H., Mueller, J.F., 2018. A mass estimate of perfluoroalkyl substance (PFAS) release from Australian wastewater treatment plants. Chemosphere 208, 975-983. https://doi.org/10.1016/j.chemosphere.2018.06.024

Garg, S., Kumar, P., Mishra, V., Guijt, R., Singh, P., Dumée, L.F., Sharma, R.S., 2020. A review on the sources, occurrence and health risks of per-/poly-fluoroalkyl substances (PFAS) arising from the manufacture and disposal of electric and electronic products. J. Water Process. Eng. 38, 101683. https://doi.org/10.1016/j.jwpe.2020.101683

Garg, S., Wang, J., Kumar, P., Mishra, V., Arafat, H., Sharma, R.S., Dumée, L.F., 2021. Remediation of water from per-/poly-fluoroalkyl substances (PFAS) – Challenges and perspectives. J. Environ. Chem. Eng. 9, 105784. https://doi.org/10.1016/j.jece.2021.105784

Ghisi, R., Vamerali, T., Manzetti, S., 2019. Accumulation of perfluorinated alkyl substances (PFAS) in agricultural plants: A review. Environmental Research 169, 326-341. https://doi.org/10.1016/j.envres.2018.10.023

Glover, C.M., Quiñones, O., Dickenson, E.R.V., 2018. Removal of perfluoroalkyl and polyfluoroalkyl substances in potable reuse systems. Water Res. 144, 454-461. https://doi.org/10.1016/j.watres.2018.07.018

Göckener, B., Fliedner, A., Rüdel, H., Fettig, I., Koschorreck, J., 2021. Exploring unknown per- and polyfluoroalkyl substances in the German environment – The total oxidizable precursor assay as helpful tool in research and regulation. Sci. Total Environ. 782, 146825. https://doi.org/10.1016/j.scitotenv.2021.146825

Gole, V. L., Sierra-Alvarez, R., Peng, H., Giesy, J. P., Deymier, P., Keswani, M., 2018. Sono-chemical treatment of per- and polyfluoroalkyl compounds in aqueous film-forming foams by use of a large-scale multi-transducer dual-frequency based acoustic reactor. Ultrason Sonochem. 213–222. https://doi.org/10.1016/j.ultsonch.2018.02.014

Government of Canada. 2018. Perfluorocarboxylic Acids and Their Precursors: Environmental Performance Agreement Overview. https://www.canada.ca/en/environment-climate-change/services/environmental-performance-agreements/list/perfluorocarboxylic-acids-overview.html. (Accessed 15/10/2021).

Hamid, H., Li, L.Y., Grace, J.R., 2018. Review of the fate and transformation of per-and polyfluoroalkyl substances (PFASs) in landfills. Environ. Pollut. 235, 74-84. https://doi.org/10.1016/j.envpol.2017.12.030

1025 Hamid, H., Li, L.Y., Grace, J.R., 2020. Formation of perfluorocarboxylic acids from 6:2
1026 fluorotelomer sulfonate (6:2 FTS) in landfill leachate: Role of microbial communities.
1027 Environ. Pollut. 259, 113835. https://doi.org/10.1016/j.envpol.2019.113835

1028 Hang, X., Chen, X., Luo, J., Cao, W., Wan, Y., 2015. Removal and recovery of
1029 perfluorooctanoate from wastewater by nanofiltration. Sep. Purif. Technol. 145, 120-
1030 129. https://doi.org/10.1016/j.seppur.2015.03.013

1031 HEPA. 2020. PFAS National Environmental Management Plan Version 2.0. National
1032 Chemicals Working Group of the Heads of EPAs Australia and New Zealand.
1033 (Accessed 26/11/2021).

1034 Hori, H., Nagaoka, Y., Murayama, M., Kutsuna, S., 2008. Efficient decomposition of
1035 perfluorocarboxylic acids and alternative fluorochemical surfactants in hot water.
1036 Environ. Sci. Technol. 42, 7438-7443. https://doi.org/10.1021/es800832p

1037 Houtz, E.F., Sutton, R., Park, J.-S., Sedlak, M., 2016. Poly- and perfluoroalkyl substances in
1038 wastewater: Significance of unknown precursors, manufacturing shifts, and likely
1039 AFFF impacts. Water Res. 95, 142-149. https://doi.org/10.1016/j.watres.2016.02.055

1040 Huo, J., Min, X., Dong, Q., Xu, S., Wang, Y., 2022. Comparison of Zn–Al and Mg–Al
1041 layered double hydroxides for adsorption of perfluorooctanoic acid. Chemosphere
1042 287, 132297. https://doi.org/10.1016/j.chemosphere.2021.132297

1043 Huset, C.A., Barlaz, M.A., Barofsky, D.F., Field, J.A., 2011. Quantitative determination of
1044 fluorochemicals in municipal landfill leachates. Chemosphere 82, 1380-1386.
1045 https://doi.org/10.1016/j.chemosphere.2010.11.072

1046 Inyang, M., Dickenson, E.R.V., 2017. The use of carbon adsorbents for the removal of
1047 perfluoroalkyl acids from potable reuse systems. Chemosphere 184, 168-175.
1048 https://doi.org/10.1016/j.chemosphere.2017.05.161

1049 Janousek, R.M., Lebertz, S., Knepper, T.P., 2019. Previously unidentified sources of
1050 perfluoroalkyl and polyfluoroalkyl substances from building materials and industrial
1051 fabrics. Environmental Science: Processes & Impacts 21, 1936-1945.
1052 https://doi.org/10.1039/C9EM00091G

1053 Ji, B., Kang, P., Wei, T., Zhao, Y., 2020. Challenges of aqueous per- and polyfluoroalkyl
1054 substances (PFASs) and their foreseeable removal strategies. Chemosphere 250,
1055 126316. https://doi.org/10.1016/j.chemosphere.2020.126316

1056 Jin, T., Peydayesh, M., Joerss, H., Zhou, J., Bolisetty, S., Mezzenga, R., 2021. Amyloid
1057 fibril-based membranes for PFAS removal from water. Environ. Sci. Water Res.
1058 Technol. https://doi.org/10.1039/D1EW00373A

1059 Kanchanapiya, P., Tantisattayakul, T., 2022. Analysis of the additional cost of addressing
1060 per-and polyfluoroalkyl substance contamination from landfill leachate by reverse
1061 osmosis membranes in Thailand. J. Water Process. Eng. 45, 102520.

1062 KEMI. 2020. Report 7/15: Occurrence and use of highly fluorinated substances and
1063 alternatives. https://www.kemi.se/en/publications/reports/2015/report-7-15-
1064 occurrence-and-use-of-highly-fluorinated-substances-and-alternatives. (Accessed
1065 23/11/2021).

1066 Kempisty, D.M., Racz, L., 2021. Forever chemicals: Environmental, economic, and social
1067 equity concerns with PFAS in the environment. CRC Press.

1068 Kim, K.Y., Ndabambi, M., Choi, S., Oh, J.-E., 2021. Legacy and novel perfluoroalkyl and
1069 polyfluoroalkyl substances in industrial wastewater and the receiving river water:
1070 Temporal changes in relative abundances of regulated compounds and alternatives.
1071 Water Res. 191, 116830. https://doi.org/10.1016/j.watres.2021.116830

1072 Kim, M.H., Wang, N., McDonald, T., Chu, K.H., 2012. Biodefluorination and
1073 biotransformation of fluorotelomer alcohols by two alkane-degrading Pseudomonas
1074 strains. Biotechnol. Bioeng. 109, 3041–3048. https://doi.org/10.1002/bit.24561.

1075 Kim, T.-H., Lee, S.-H., Kim, H.Y., Doudrick, K., Yu, S., Kim, S.D., 2019. Decomposition of
1076 perfluorooctane sulfonate (PFOS) using a hybrid process with electron beam and
1077 chemical oxidants. Chem. Eng. J. 361, 1363-1370.
1078 https://doi.org/10.1016/j.cej.2018.10.195

1079 Kotthoff, M., Müller, J., Jürling, H., Schlummer, M., Fiedler, D., 2015. Perfluoroalkyl and
1080 polyfluoroalkyl substances in consumer products. Environ. Sci. Pollut. Res. 22,
1081 14546-14559. https://doi.org/10.1007/s11356-015-4202-7

1082 Kumar, M., Bolan, N.S., Hoang, S.A., Sawarkar, A.D., Jasemizad, T., Gao, B., Keerthanan,
1083 S., Padhye, L.P., Singh, L., Kumar, S., Vithanage, M., 2021. Remediation of soils and
1084 sediments polluted with polycyclic aromatic hydrocarbons: To immobilize, mobilize,
1085 or degrade? J. Hazard. Mater. 420, 126534.
1086 https://doi.org/10.1016/j.jhazmat.2021.126534.

1087 Kumari, P., Bahadur, N., Dumée, L.F., 2020. Photo-catalytic membrane reactors for the
1088 remediation of persistent organic pollutants – A review. Sep. Purif. Technol. 230,
1089 115878. https://doi.org/10.1016/j.seppur.2019.115878

1090 Kunacheva, C., Tanaka, S., Fujii, S., Boontanon, S.K., Musirat, C., Wongwattana, T.,
1091 Shivakoti, B.R., 2011. Mass flows of perfluorinated compounds (PFCs) in central
1092 wastewater treatment plants of industrial zones in Thailand. Chemosphere 83, 737-
1093 744. https://doi.org/10.1016/j.chemosphere.2011.02.059

1094 Kunhikrishnan, A., Bolan, N.S., Müller, K., Laurenson, S., Naidu, R., Kim, W.-I., 2012. The
1095 influence of wastewater irrigation on the transformation and bioavailability of heavy
1096 metal (loid) s in soil. Adv. Agron. 115, 215-297. https://doi.org/10.1016/B978-0-12-
1097 394276-0.00005-6

1098 Land, M., de Wit, C.A., Bignert, A., Cousins, I.T., Herzke, D., Johansson, J.H., Martin, J.W.,
1099 2018. What is the effect of phasing out long-chain per- and polyfluoroalkyl
1100 substances on the concentrations of perfluoroalkyl acids and their precursors in the
1101 environment? A systematic review. Environ Evid. 7, 4.
1102 https://doi.org/10.1186/s13750-017-0114-y

1103 Lang, J.R., Allred, B.M., Field, J.A., Levis, J.W., Barlaz, M.A., 2017. National estimate of
1104 per-and polyfluoroalkyl substance (PFAS) release to US municipal landfill leachate.
1105 Environ. Sci. Technol. 51, 2197-2205. https://doi.org/10.1021/acs.est.6b05005

1106 Le, T.X.H., Haflich, H., Shah, A.D., Chaplin, B.P., 2019. Energy-efficient electrochemical
1107 oxidation of perfluoroalkyl substances using a Ti4O7 reactive electrochemical
1108 membrane anode. Environ. Sci. Technol. Lett. 6, 504-510.

1109 Lee, Y.-C., Lo, S.-L., Chiueh, P.-T., Chang, D.-G., 2009. Efficient decomposition of
1110 perfluorocarboxylic acids in aqueous solution using microwave-induced persulfate.
1111 Water Res. 43, 2811-2816. https://doi.org/10.1016/j.watres.2009.03.052

1112 Lei, Y.-J., Tian, Y., Sobhani, Z., Naidu, R., Fang, C., 2020. Synergistic degradation of PFAS
1113 in water and soil by dual-frequency ultrasonic activated persulfate. Chem. Eng. J. 388,
1114 124215. https://doi.org/10.1016/j.cej.2020.124215

1115 Lenka, S.P., Kah, M., Padhye, L.P., 2021. A review of the occurrence, transformation, and
1116 removal of poly-and perfluoroalkyl substances (PFAS) in wastewater treatment
1117 plants. Water Res., 117187. https://doi.org/10.1016/j.watres.2021.117187

1118 Li, F., Duan, J., Tian, S., Ji, H., Zhu, Y., Wei, Z., Zhao, D., 2020. Short-chain per-and
1119 polyfluoroalkyl substances in aquatic systems: Occurrence, impacts and treatment.
1120 Chem. Eng. J. 380, 122506. https://doi.org/10.1016/j.cej.2019.122506

1121 Li, R., Alomari, S., Stanton, R., Wasson, M.C., Islamoglu, T., Farha, O.K., Holsen, T.M.,
1122 Thagard, S.M., Trivedi, D.J., Wriedt, M., 2021. Efficient removal of per- and
1123 polyfluoroalkyl substances from water with zirconium-based metal–organic
1124 frameworks. Chem. Mater. 33, 3276-3285. https://doi.org/10.1016/j.jece.2021.105271

1125 Lin, H., Niu, J., Liang, S., Wang, C., Wang, Y., Jin, F., Luo, Q., Huang, Q., 2018.
1126 Development of macroporous Magnéli phase Ti4O7 ceramic materials: As an
1127 efficient anode for mineralization of poly- and perfluoroalkyl substances. Chem. Eng.
1128 J. 354, 1058-1067. https://doi.org/10.1016/j.cej.2018.07.210

1129 Lindstrom, A.B., Strynar, M.J., Delinsky, A.D., Nakayama, S.F., McMillan, L., Libelo, E.L.,
1130 Neill, M., Thomas, L., 2011. Application of WWTP biosolids and resulting
1131 perfluorinated compound contamination of surface and well water in Decatur,
1132 Alabama, USA. Environ. Sci. Technol. 45, 8015-8021.
1133 https://doi.org/10.1021/es1039425

1134 Liu, Y., Hu, X.-M., Zhao, Y., Wang, J., Lu, M.-X., Peng, F.-H., Bao, J., 2018. Removal of
1135 perfluorooctanoic acid in simulated and natural waters with different electrode
1136 materials by electrocoagulation. Chemosphere 201, 303-309.
1137 https://doi.org/10.1016/j.chemosphere.2018.02.129

1138 Lorenzo, M., Campo, J., Morales Suárez-Varela, M., Picó, Y., 2019. Occurrence, distribution
1139 and behavior of emerging persistent organic pollutants (POPs) in a Mediterranean
1140 wetland protected area. Sci. Total Environ. 646, 1009-1020.
1141 https://doi.org/10.1016/j.scitotenv.2018.07.304

1142 Maga, D., Aryan, V., Bruzzano, S., 2021. Environmental assessment of various end-of-life
1143 pathways for treating per- and polyfluoroalkyl substances in spent fire-extinguishing
1144 waters. Environ. Toxicol. Chem. 40, 947–957. https://doi.org/10.1002/etc.4803.

1145 Maimaiti, A., Deng, S., Meng, P., Wang, W., Wang, B., Huang, J., Wang, Y., Yu, G., 2018.
1146 Competitive adsorption of perfluoroalkyl substances on anion exchange resins in
1147 simulated AFFF-impacted groundwater. Chem. Eng. J. 348, 494-502.
1148 https://doi.org/10.1016/j.cej.2018.05.006

1149 Makarov, S., 2001. Recent trends in chemistry of sulfur-containing reductants.

1150 Mastropietro, T., Bruno, R., Pardo, E., Armentano, D., 2021. Reverse osmosis and
1151 nanofiltration membranes for highly efficient PFASs removal: overview, challenges
1152 and future perspectives. Dalton Trans. https://doi.org/10.1039/D1DT00360G

1153 Maul, G.A., Kim, Y., Amini, A., Zhang, Q., Boyer, T.H., 2014. Efficiency and life cycle
1154 environmental impacts of ion-exchange regeneration using sodium, potassium,
1155 chloride, and bicarbonate salts. Chem. Eng. J. 254, 198-209.
1156 https://doi.org/10.1016/j.cej.2014.05.086

McCleaf, P., Englund, S., Östlund, A., Lindegren, K., Wiberg, K., Ahrens, L., 2017. Removal efficiency of multiple poly- and perfluoroalkyl substances (PFASs) in drinking water using granular activated carbon (GAC) and anion exchange (AE) column tests. Water Res. 120, 77-87. https://doi.org/10.1016/j.watres.2017.04.057

Merino, N., Qu, Y., Deeb, R.A., Hawley, E.L., Hoffmann, M.R., Mahendra, S., 2016. Degradation and removal methods for perfluoroalkyl and polyfluoroalkyl substances in water. Environ. Eng. Sci. 33, 615-649. https://doi.org/10.1089/ees.2016.0233

Michigan Department of Environment. 2019. Michigan Department ofEnvironment, Great Lakes, and Energy. 2018 PFAS sampling ofdrinking water supplies in Michigan. Project 60570309. . https://www.michigan.gov/documents/pfasresponse/2018_PFAS_Sampling_of_Drinking_Water_Supplies_in_Michigan_663543_7.pdf. (Accessed).

Militao, I.M., Roddick, F.A., Bergamasco, R., Fan, L., 2021. Removing PFAS from aquatic systems using natural and renewable material-based adsorbents: A review. J. Environ. Chem. Eng. 9, 105271. https://doi.org/10.1016/j.jece.2021.105271

Mukhopadhyay, R., Sarkar, B., Palansooriya, K.N., Dar, J.Y., Bolan, N.S., Parikh, S.J., Sonne, C., Ok, Y.S., 2021. Natural and engineered clays and clay minerals for the removal of poly- and perfluoroalkyl substances from water: State-of-the-art and future perspectives. Adv. Colloid Interface Sci. 297, 102537. https://doi.org/10.1016/j.cis.2021.102537

Murakami, M., Shinohara, H., Takada, H., 2009. Evaluation of wastewater and street runoff as sources of perfluorinated surfactants (PFSs). Chemosphere 74, 487-493. https://doi.org/10.1016/j.chemosphere.2008.10.018

Nguyen, V.T., Reinhard, M., Karina, G.Y.-H., 2011. Occurrence and source characterization of perfluorochemicals in an urban watershed. Chemosphere 82, 1277-1285. https://doi.org/10.1016/j.chemosphere.2010.12.030

Nzeribe, B.N., Crimi, M., Mededovic Thagard, S., Holsen, T.M., 2019. Physico-chemical processes for the treatment of per-and polyfluoroalkyl substances (PFAS): a review. Crit Rev Environ Sci Technol. 49, 866-915.

Olmsted, J.L., Ahmadireskety, A., Da Silva, B.F., Robey, N., Aristizabal-Henao, J.J., Bonzongo, J.-C.J., Bowden, J.A., 2021. Using regulatory classifications to assess the impact of different land use types on per-and polyfluoroalkyl substance concentrations in stormwater pond sediments. J. Environ. Eng. 147, 06021005. https://doi.org/10.1061/(ASCE)EE.1943-7870.0001906

Page, D., Vanderzalm, J., Kumar, A., Cheng, K.Y., Kaksonen, A.H., Simpson, S., 2019. Risks of perfluoroalkyl and polyfluoroalkyl substances (PFAS) for sustainable water recycling via aquifers. Water 11, 1737. https://doi.org/10.3390/w11081737

Palma, D., Papagiannaki, D., Lai, M., Binetti, R., Sleiman, M., Minella, M., Richard, C., 2021. PFAS degradation in ultrapure and groundwater using non-thermal plasma. Molecules 26, 924-936. https://doi.org/10.3390/molecules26040924

Panagopoulos, A., Haralambous, K. J., Loizidou, M., 2019. Desalination brine disposal methods and treatment technologies – A review. Science of the Total Environment. 693, 133545. https://doi.org/10.1016/j.scitotenv.2019.07.351.

Parenky Akshay, C., de Souza Naomi, G., Nguyen Hiep, H., Jeon, J., Choi, H., 2020. Decomposition of carboxylic PFAS by persulfate activated by silver under ambient

conditions. J. Environ. Eng. 146, 06020003. https://doi.org/10.1061/(ASCE)EE.1943-7870.0001808

Park, M., Daniels, K.D., Wu, S., Ziska, A.D., Snyder, S.A., 2020. Magnetic ion-exchange (MIEX) resin for perfluorinated alkylsubstance (PFAS) removal in groundwater: Roles of atomic charges for adsorption. Water Res. 181, 115897. https://doi.org/10.1016/j.watres.2020.115897

Pauletto, P.S., Bandosz, T.J., 2022. Activated carbon versus metal-organic frameworks: A review of their PFAS adsorption performance. J. Hazard. Mater. 425, 127810. https://doi.org/10.1016/j.jhazmat.2021.127810

Phan Thi, L.-A., Do, H.-T., Lee, Y.-C., Lo, S.-L., 2013. Photochemical decomposition of perfluorooctanoic acids in aqueous carbonate solution with UV irradiation. Chem. Eng. J. 221, 258-263. https://doi.org/10.1016/j.cej.2013.01.084

Phong Vo, H.N., Ngo, H.H., Guo, W., Hong Nguyen, T.M., Li, J., Liang, H., Deng, L., Chen, Z., Hang Nguyen, T.A., 2020. Poly-and perfluoroalkyl substances in water and wastewater: A comprehensive review from sources to remediation. J. Water Process. Eng. 36, 101393. https://doi.org/10.1016/j.jwpe.2020.101393

Pillai, S. D., 2020. Ex situ remediation of investigation-derived wastes containing PFAS by electron beam technology distribution. Strategic Environmental Research and Development Program (SERDP).

Podder, A., Sadmani, A.H.M.A., Reinhart, D., Chang, N.-B., Goel, R., 2021. Per and poly-fluoroalkyl substances (PFAS) as a contaminant of emerging concern in surface water: A transboundary review of their occurrences and toxicity effects. J. Hazard. Mater. 419, 126361. https://doi.org/10.1016/j.jhazmat.2021.126361

Post, G.B., 2021. Recent US state and federal drinking water guidelines for per-and polyfluoroalkyl substances. Environ. Toxicol. Chem. 40, 550-563. https://doi.org/10.1002/etc.4863

Prevedouros, K., Cousins, I.T., Buck, R.C., Korzeniowski, S.H., 2006. Sources, fate and transport of perfluorocarboxylates. Environ. Sci. Technol. 40, 32-44. https://doi.org/10.1021/es0512475

Röhler, K., Haluska, A.A., Susset, B., Liu, B., Grathwohl, P., 2021. Long-term behavior of PFAS in contaminated agricultural soils in Germany. J. Contam. Hydrol. 241, 103812. https://doi.org/10.1016/j.jconhyd.2021.103812

Saleh, N.B., Khalid, A., Tian, Y., Ayres, C., Sabaraya, I.V., Pietari, J., Hanigan, D., Chowdhury, I., Apul, O.G., 2019. Removal of poly- and per-fluoroalkyl substances from aqueous systems by nano-enabled water treatment strategies. Environ. Sci.: Water Res. Technol. 5, 198-208. https://doi.org/10.1039/C8EW00621K

Singh, A., 2021. A review of wastewater irrigation: Environmental implications. Resources, Conservation and Recycling 168, 105454. https://doi.org/10.1016/j.resconrec.2021.105454

Singh, R.K., Fernando, S., Baygi, S.F., Multari, N., Thagard, S.M., Holsen, T.M., 2019. Breakdown products from perfluorinated alkyl substances (PFAS) degradation in a plasma-based water treatment process. Environ. Sci. Technol. 53, 2731-2738. https://doi.org/10.1021/acs.est.8b07031

Solo-Gabriele, H.M., Jones, A.S., Lindstrom, A.B., Lang, J.R., 2020. Waste type, incineration, and aeration are associated with per- and polyfluoroalkyl levels in landfill leachates. Waste Management 107, 191-200. https://doi.org/10.1016/j.wasman.2020.03.034

Soriano, Á., Gorri, D., Urtiaga, A., 2017. Efficient treatment of perfluorohexanoic acid by nanofiltration followed by electrochemical degradation of the NF concentrate. Water Res. 112, 147-156. https://doi.org/10.1016/j.watres.2017.01.043

Stockholm Convention. 2019. At COP-4 in 2009, the Conference of the Parties decided to list perfluorooctane sulfonic acid (PFOS), its salts and perfluorooctane sulfonyl fluoride (PFOSF) in Annex B to the Stockholm Convention (decision SC-4/17). http://chm.pops.int/Implementation/IndustrialPOPs/PFOS/Overview/tabid/5221/Default.aspx. (Accessed 18/11/2021).

Sun, Z., Zhang, C., Jiang, J., Wen, J., Zhou, Q., Hoffmann, M.R., 2022. UV/FeIINTA as a novel photoreductive system for the degradation of perfluorooctane sulfonate (PFOS) via a photoinduced intramolecular electron transfer mechanism. Chem. Eng. J. 427, 130923. https://doi.org/10.1016/j.cej.2021.130923

Sungur, Ş., Çevik, B., Köroğlu, M., 2020. Determination of perfluorooctanoic acid (PFOA) and perfluorooctane sulfonic acid (PFOS) contents of compost amended soils and plants grown in these soils. Int. J. Environ. Anal. Chem., 1-9. https://doi.org/10.1080/03067319.2020.1745200

Tang, C.Y., Fu, Q.S., Robertson, A.P., Criddle, C.S., Leckie, J.O., 2006. Use of reverse osmosis membranes to remove perfluorooctane sulfonate (PFOS) from semiconductor wastewater. Environ. Sci. Technol. 40, 7343-7349. https://doi.org/10.1021/es060831q

The Danish EPA. 2015. Short-chain Polyfluoroalkyl Substances (PFAS). https://www2.mst.dk/Udgiv/publications/2015/05/978-87-93352-15-5.pdf. (Accessed 10/12/2021).

Tow, E.W., Ersan, M.S., Kum, S., Lee, T., Speth, T.F., Owen, C., Bellona, C., Nadagouda, M.N., Mikelonis, A.M., Westerhoff, P., Mysore, C., 2021. Managing and treating per- and polyfluoroalkyl substances (PFAS) in membrane concentrates. AWWA water science. 3, e1233.

Trautmann, A.M., Schell, H., Schmidt, K.R., Mangold, K.M., Tiehm, A., 2015. Electrochemical degradation of perfluoroalkyl and polyfluoroalkyl substances (PFASs) in groundwater. Water Sci. Technol. 71, 1569-1575. https://doi.org/10.2166/wst.2015.143

Tsai, Y.-T., Yu-Chen Lin, A., Weng, Y.-H., Li, K.-C., 2010. Treatment of perfluorinated chemicals by electro-microfiltration. Environ. Sci. Technol. 44, 7914-7920. https://doi.org/10.1021/es101964y

US EPA. 2016a. Drinking water health advisory for perfluorooctane sulfonate (PFOS). EPA 822-R-16-004. Washington, DC. (Accessed 12/12/2021).

US EPA. 2016b. Drinking water health advisory for perfluorooctanoic acid (PFOA). EPA 822-R-16-005. Washington, DC. Washington, DC. (Accessed 12/12/2021).

US EPA. 2020. Interim guidance on the destruction and disposal of perfluoroalkyl and polyfluoroalkyl substances and materials containing perfluoroalkyl and polyfluoroalkyl substances (EPAHQ-OLEM-2020-0527-0002).

US EPA. 2021a. Our Current Understanding of the Human Health and Environmental Risks of PFAS. https://www.epa.gov/pfas/our-current-understanding-human-health-and-environmental-risks-pfas. (Accessed 23/11/2021).

US EPA. 2021b. PFAS Strategic Roadmap: EPA's Commitments to Action 2021-2024. (Accessed 23/12/2021).

Uwayezu, J.N., Carabante, I., Lejon, T., van Hees, P., Karlsson, P., Hollman, P., Kumpiene, J., 2021. Electrochemical degradation of per- and poly-fluoroalkyl substances using boron-doped diamond electrodes. J. Environ. Manage. 290, 112573. https://doi.org/10.1016/j.jenvman.2021.112573

Vo, H.N.P., Ngo, H.H., Guo, W., Nguyen, T.M.H., Li, J., Liang, H., Deng, L., Chen, Z., Nguyen, T.A.H., 2020. Poly-and perfluoroalkyl substances in water and wastewater: A comprehensive review from sources to remediation. J. Water Process. Eng. 36, 101393. https://doi.org/10.1016/j.jwpe.2020.101393

Vu, C.T., Wu, T., 2020. Recent progress in adsorptive removal of per-and poly-fluoroalkyl substances (PFAS) from water/wastewater. Crit. Rev. Environ. Sci. Technol. 52, 1-40. https://doi.org/10.1080/10643389.2020.1816125

Wackett, L.P., McMahon, K., 2021. Why is the biodegradation of polyfluorinated compounds so rare? mSphere 6, e00721-00721. https://doi.org/10.1128/mSphere.00721-21

Wang, Y., Pierce, R.D., Shi, H., Li, C., Huang, Q., 2020. Electrochemical degradation of perfluoroalkyl acids by titanium suboxide anodes. Environ. Sci. Water Res. Technol. 6, 144-152. https://doi.org/10.1039/C9EW00759H

Wang, Z., DeWitt, J.C., Higgins, C.P., Cousins, I.T., 2017. A never-ending story of per-and polyfluoroalkyl substances (PFASs)? ACS Publications.

Wanninayake, D.M., 2021. Comparison of currently available PFAS remediation technologies in water: A review. J. Environ. Manage. 283, 111977. https://doi.org/10.1016/j.jenvman.2021.111977

Watanabe, N., Takata, M., Takemine, S., Yamamoto, K., 2018. Thermal mineralization behavior of PFOA, PFHxA, and PFOS during reactivation of granular activated carbon (GAC) in nitrogen atmosphere. Environ. Sci. Pollut. Res. 25, 7200-7205. https://doi.org/10.1007/s11356-015-5353-2

Xiao, F., Davidsavor, K.J., Park, S., Nakayama, M., Phillips, B.R., 2012. Batch and column study: sorption of perfluorinated surfactants from water and cosolvent systems by Amberlite XAD resins. J. Colloid Interface Sci. 368, 505-511. https://doi.org/10.1016/j.jcis.2011.11.011

Xiao, F., Sasi, P.C., Yao, B., Kubátová, A., Golovko, S.A., Golovko, M.Y., Soli, D., 2020. Thermal Stability and Decomposition of Perfluoroalkyl Substances on Spent Granular Activated Carbon. Environ. Sci. Technol. Lett. 7, 343-350. https://doi.org/10.1021/acs.estlett.0c00114

Xiao, F., Simcik, M., Gulliver, J., 2011. Perfluoroalkyl acids in urban stormwater runoff: Influence of land use. Water Res. 46. https://doi.org/10.1016/j.watres.2011.11.029

Xiao, X., Ulrich, B.A., Chen, B., Higgins, C.P., 2017. Sorption of poly-and perfluoroalkyl substances (PFASs) relevant to aqueous film-forming foam (AFFF)-impacted groundwater by biochars and activated carbon. Environ. Sci. Technol. 51, 6342-6351. https://doi.org/10.1021/acs.est.7b00970

1333 Xu, B., Ahmed, M.B., Zhou, J.L., Altaee, A., Wu, M., Xu, G., 2017. Photocatalytic removal
1334 of perfluoroalkyl substances from water and wastewater: Mechanism, kinetics and
1335 controlling factors. Chemosphere 189, 717-729.
1336 https://doi.org/10.1016/j.chemosphere.2017.09.110

1337 Xu, B., Liu, S., Zhou, J.L., Zheng, C., Weifeng, J., Chen, B., Zhang, T., Qiu, W., 2021. PFAS
1338 and their substitutes in groundwater: Occurrence, transformation and remediation. J.
1339 Hazard. Mater. 412, 125159. https://doi.org/10.1016/j.jhazmat.2021.125159

1340 Yan, C., Zhang, W., Zhang, Z., Liu, Y., Deng, C., Nie, N., 2015a. Assessment of water
1341 quality and identification of polluted risky regions based on field observations & GIS
1342 in the honghe river watershed, China. PloS one 10, e0119130.
1343 https://doi.org/10.1371/journal.pone.0119130

1344 Yan, H., Cousins, I.T., Zhang, C., Zhou, Q., 2015b. Perfluoroalkyl acids in municipal landfill
1345 leachates from China: Occurrence, fate during leachate treatment and potential impact
1346 on groundwater. Sci. Total Environ. 524, 23-31.
1347 https://doi.org/10.1016/j.scitotenv.2015.03.111

1348 Yeung, L.W., Stadey, C., Mabury, S.A., 2017. Simultaneous analysis of perfluoroalkyl and
1349 polyfluoroalkyl substances including ultrashort-chain C2 and C3 compounds in rain
1350 and river water samples by ultra performance convergence chromatography. J.
1351 Chromatogr. A 1522, 78-85. https://doi.org/10.1016/j.chroma.2017.09.049

1352 Yin, P., Hu, Z., Song, X., Liu, J., Lin, N., 2016. Activated persulfate oxidation of
1353 perfluorooctanoic acid (PFOA) in groundwater under acidic conditions. Int. J. Env.
1354 Res. Public Health 13, 602. https://doi.org/10.3390/ijerph13060602

1355 Yoon, Y., Westerhoff, P., Snyder, S.A., Wert, E.C., 2006. Nanofiltration and ultrafiltration of
1356 endocrine disrupting compounds, pharmaceuticals and personal care products. J.
1357 Membr. Sci. 270, 88-100. https://doi.org/10.1016/j.memsci.2005.06.045

1358 Zhang, C., Yan, H., Li, F., Zhou, Q., 2015. Occurrence and fate of perfluorinated acids in two
1359 wastewater treatment plants in Shanghai, China. Environ. Sci. Pollut. Res. 22, 1804-
1360 1811. https://doi.org/10.1007/s11356-013-2044-8

1361 Zhang, D., He, Q., Wang, M., Zhang, W., Liang, Y., 2021. Sorption of perfluoroalkylated
1362 substances (PFASs) onto granular activated carbon and biochar. Environ. Technol. 42,
1363 1798-1809. https://doi.org/10.1080/09593330.2019.1680744

1364 Zhang, D.Q., Zhang, W.L., Liang, Y.N., 2019. Adsorption of perfluoroalkyl and
1365 polyfluoroalkyl substances (PFASs) from aqueous solution - A review. Sci. Total
1366 Environ. 694, 133606. https://doi.org/10.1016/j.scitotenv.2019.133606

1367 Zhao, L., McCausland, P.K., Folsom, P.W., Wolstenholme, B.W., Sun, H., Wang, N., Buck,
1368 R.C., 2013. 6:2 Fluorotelomer alcohol aerobic biotransformation in activated sludge
1369 from two domestic wastewater treatment plants. Chemosphere 92, 464-470.
1370 https://doi.org/10.1016/j.chemosphere.2013.02.032

1371 Zhao, P., Xia, X., Dong, J., Xia, N., Jiang, X., Li, Y., Zhu, Y., 2016. Short- and long-chain
1372 perfluoroalkyl substances in the water, suspended particulate matter, and surface
1373 sediment of a turbid river. Sci. Total Environ. 568, 57-65.
1374 https://doi.org/10.1016/j.scitotenv.2016.05.221

1375 Zhou, Y., Meng, J., Zhang, M., Chen, S., He, B., Zhao, H., Li, Q., Zhang, S., Wang, T., 2019.
1376 Which type of pollutants need to be controlled with priority in wastewater treatment

1377 plants: Traditional or emerging pollutants? Environ. Int. 131, 104982.
1378 https://doi.org/10.1016/j.envint.2019.104982

1379 Zushi, Y., Masunaga, S., 2009. First-flush loads of perfluorinated compounds in stormwater
1380 runoff from Hayabuchi River basin, Japan served by separated sewerage system.
1381 Chemosphere 76, 833-840. https://doi.org/10.1016/j.chemosphere.2009.04.004

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Figures

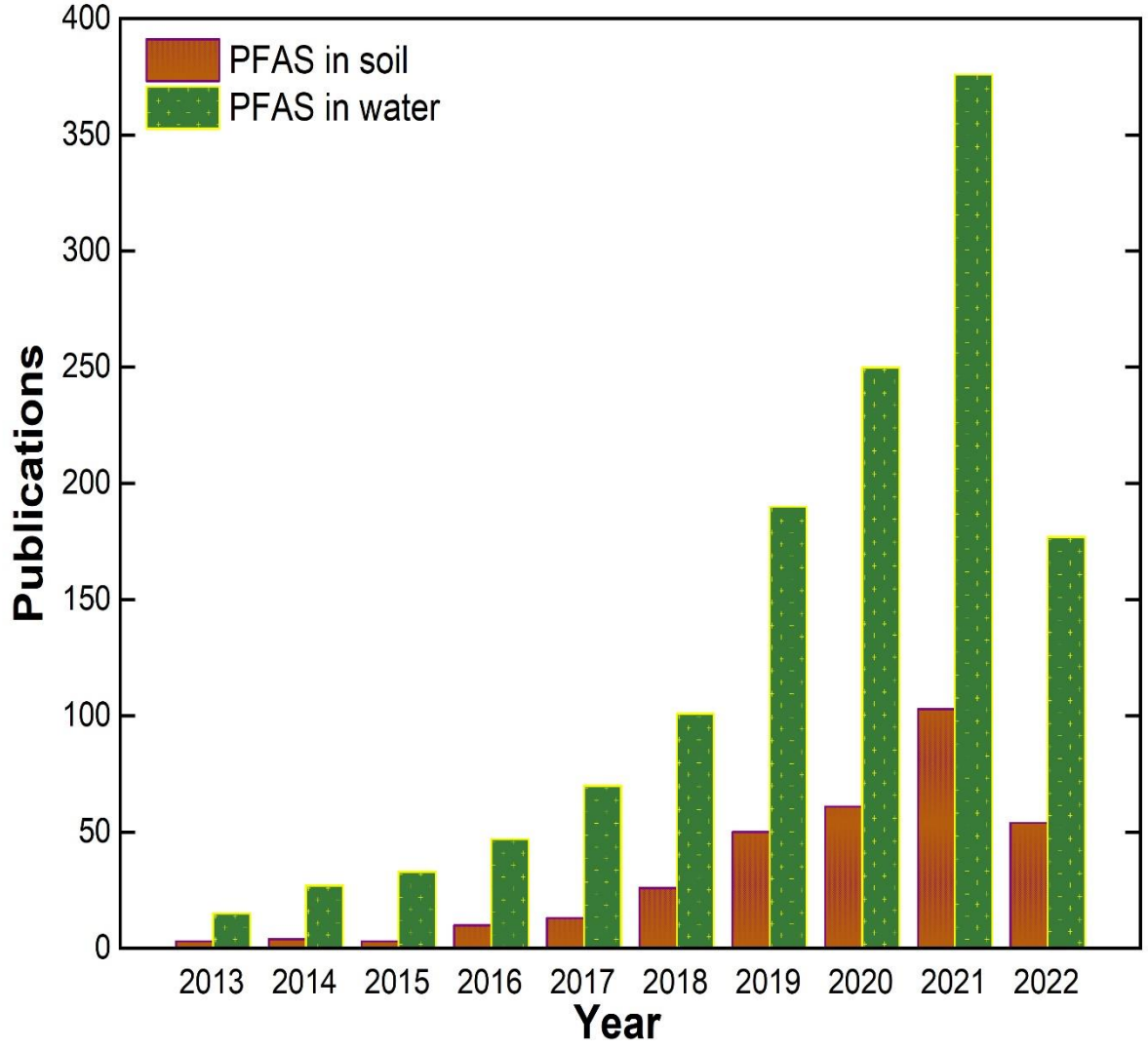


Figure 1: Publications on PFAS in soil and water for the past 10 years.

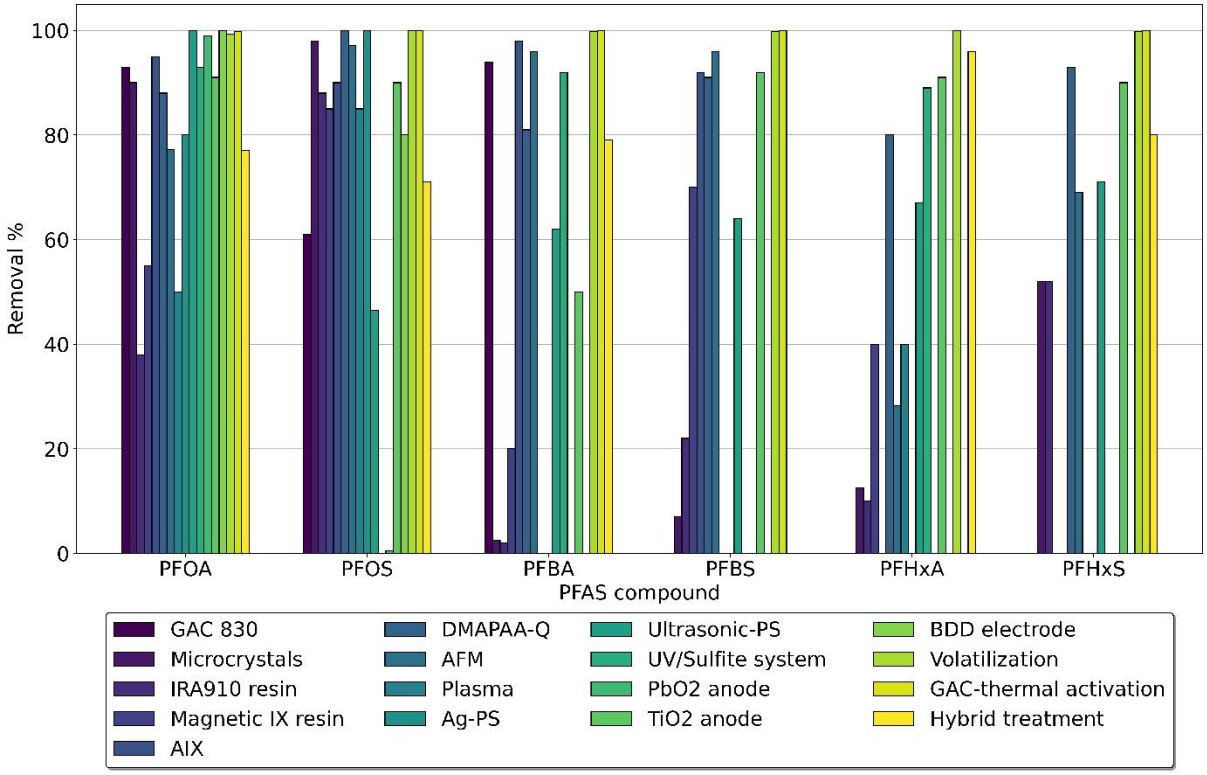


Figure 2: Performance of different materials and technologies for PFASs removal from wastewater

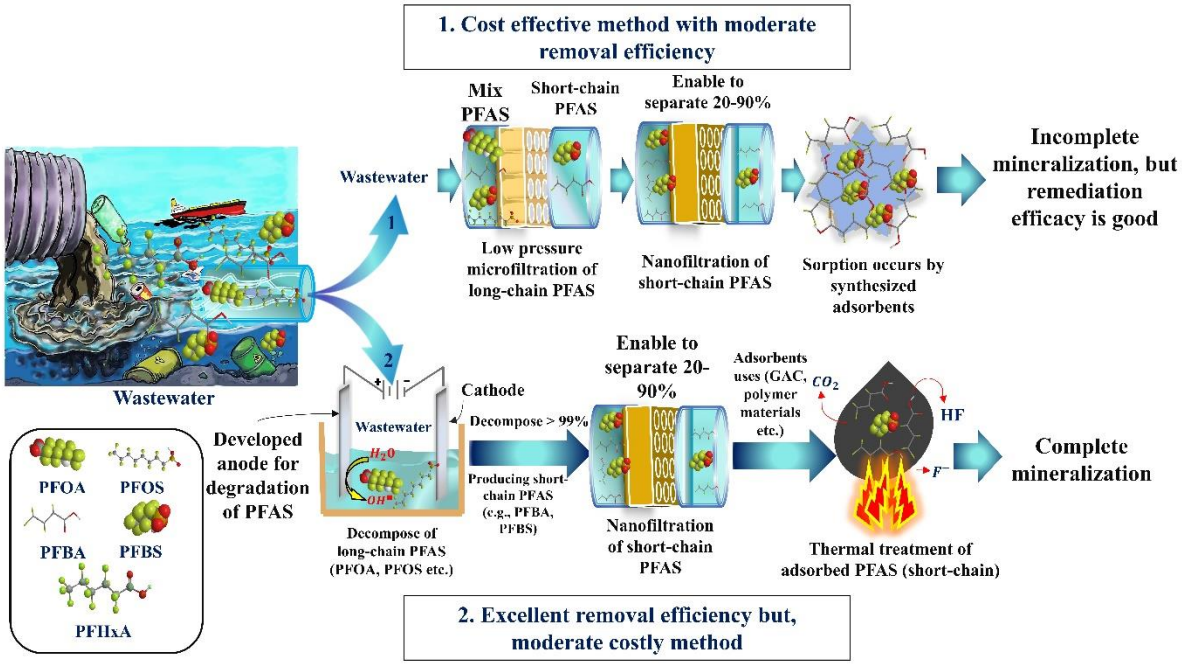


Figure 3: Integrated hybrid process and comparative analysis of different removal processes for PFASs removal from wastewater

Tables

Table 1: List of PFAS abbreviations used within the review

Class	Abbreviation	Chemical Name	Formula
PFCA	PFPrA	Pentafluoropropionic acid	C_2F_5COOH
	PFBA	Perfluorobutanoic acid	C_3F_7COOH
	PFPeA	Perfluoropentanoic acid	C_4F_9COOH
	PFHxA	Perfluorohexanoic acid	$C_5F_{11}COOH$
	PFHpA	Perfluoroheptanoic acid	$C_6F_{13}COOH$
	PFOA	Perfluorooctanoic acid	$C_7F_{15}COOH$
	PFNA	Perfluorononanoic acid	$C_8F_{17}COOH$
	PFDA	Perfluorodecanoic acid	$C_9F_{19}COOH$
	PFUnDA	Perfluoroundecanoic acid	$C_{10}F_{21}COOH$
	PFDoDa	Perfluorododecane acid	$C_{11}F_{23}COOH$
	PFTrDA	Perfluorotridecanoic acid	$C_{12}F_{25}COOH$
	PFTeDA	Perfluorotetradecanoic acid	$C_{13}F_{27}COOH$
	PFODA	Perfluorooctadecanoic acid	$C_{17}F_{35}COOH$
PFSA	PFBS	Perfluorobutane sulfonic acid	$C_4F_9SO_3H$
	PFPeS	Perfluoropentane sulfonic acid	$C_5F_{11}SO_3H$
	PFHxS	Perfluorohexane sulfonic acid	$C_6F_{13}SO_3H$
	PFHpS	Perfluoroheptane sulfonic acid	$C_7F_{15}SO_3H$

	PFOS	Perfluorooctane sulfonic acid	$C_8F_{17}SO_3H$
	PFNS	Perfluorononane sulfonic acid	$C_9F_{19}SO_3H$
	PFDS	Perfluorodecane sulfonic acid	$C_{10}F_{21}SO_3H$
	PFUnDS	Perfluoroundecane sulfonic acid	$C_{11}F_{23}SO_3H$
	PFDoDS	Perfluorododecane sulfonic acid	$C_{12}F_{25}SO_3H$
	PFTrDS	Perfluorotridecane sulfonic acid	$C_{13}F_{27}SO_3H$
	PFTeDS	Perfluorotetradecane sulfonic acid	$C_{14}F_{29}SO_3H$
Other	PFOSA	Perfluoroctylsulfonamide	$C_8H_2F_{17}NO_2S$
	FOSA	Perfluorooctane sulfonamide acid	$C_8F_{17}SO_3NH_3$
	FOSAA	Perfluorooctane sulfonamideoacetate acid	$C_{10}H_4F_{17}NO_4S$
	MeFOSAA	2-(N-Methylperfluorooctanesulfonamido)acetic acid	$C_{11}H_6F_{17}NO_4S$
	EtFOSAA	N-ethylperfluorooctane sulfonamidoacetic acid	$C_{12}H_8F_{17}NO_4S$
	MeFBSA	N-(Methyl)nonafluorobutanesulfonamide	$C_5H_4F_9NO_2S$
	MeFBSAA	2-(N-Methylperfluorobutanesulfonamido)acetic acid	$C_7H_6F_9NO_4S$
	4:2 FTSA	4:2 Fluorotelomer saturated sulfonic acid	$C_6H_5F_9O_3S$
	6:2 FTSA	6:2 Fluorotelomer saturated sulfonic acid	$C_8H_5F_{13}O_3S$
	8:2 FTSA	8:2 Fluorotelomer saturated sulfonic acid	$C_{10}H_5F_{17}O_3S$
	6:2 FTCA	6:2 Fluorotelomer saturated carboxylic acid	$C_{17}H_{21}F_{13}N_2O_3S$
	8:2 FTCA	8:2 Fluorotelomer saturated carboxylic acid	$C_{10}H_3F_{17}O_2$

5:3 FTCA	5:3 Fluorotelomer saturated carboxylic acid	$C_8H_5F_{11}O_2$
7:3 FTCA	7:3 Fluorotelomer saturated carboxylic acid	$C_{10}H_5F_{15}O_2$
10:2 FTCA	10:2 Fluorotelomer saturated carboxylic acid	$C_{12}H_3F_{21}O_2$
6:2 FTUCA	6:2 Fluorotelomer unsaturated carboxylic acid	$\underline{C_8H_2F_{12}O_2}$
8:2 FTUCA	8:2 Fluorotelomer unsaturated carboxylic acid	$\underline{C_{10}H_2F_{16}O_2}$
10:2 FTUCA	10:2 Fluorotelomer unsaturated carboxylic acid	$C_{12}H_3F_{20}O_2$
6:2 FTOH	6:2 Fluorotelomer alcohol	$C_8H_5F_{13}O$
8:2 FTOH	8:2 Fluorotelomer alcohol	$C_{10}H_5F_{17}O$
10:2 FTOH	10:2 Fluorotelomer alcohol	$C_{12}H_5F_{21}O$
12:2 FTOH	12:2 Fluorotelomer alcohol	$C_{14}H_5F_{25}O$
6:2 FTI	6:2 Fluorotelomer iodide	$C_8H_4F_{13}OI$
6:2 FTMAC	6:2 Fluorotelomer methacrylate	$\underline{C_{12}H_9F_{13}O_2}$
8:2 FTAC	8:2 fluorotelomer acrylate	$C_{13}H_7F_{17}O_2$
8:2 FTMAC	8:2 Fluorotelomer methacrylate	$\underline{C_{12}H_9F_{13}O_2}$
6:2 FTAB	6:2 fluorotelomer sulfonamide alkylbetaine	$\underline{C_{15}H_{20}F_{13}N_2O_4S^+}$
M4	6:2 Fluorotelomer sulfonamide propyl N,N dimethylamine	$\underline{C_{13}H_{18}F_{13}N_2O_2S^+}$

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Table 2: Concentrations of PFAS (ng L^{-1}) in domestic and industrial effluent recorded. Values recorded as mean or range. (See List of Abbreviations for associated compound names)

	(Gallen et al., 2018)	**(Coggan et al., 2019)**	**(Houtz et al., 2016)**	**(Zhou et al., 2019)**	**(Zhang et al., 2015)**	**(Lorenzo et al., 2019)**	**(Clara et al., 2008)**	**(Kunachev a et al., 2011)**	**(Dauchy et al., 2017)**	**(Bossi et al., 2008)**	**(Kim et al., 2021)**
Effluent type	Domestic	Domestic	Domestic	Domestic	Domestic	Domestic/ Industrial	Industrial	Industrial	Industrial	Industrial	Industrial
Country	Australia	Australia	US	China	China	Spain	-	Thailand	France	Denmark	Korea
No. of sites	14	19	6	6	2	10	9	1	1	4	77
PFPrA	-	-	-	-	13.5-15.3	-	-	-	-	-	-
PFBA	-	13	16	20.24	27.6-93.5	4.4	-	-	2-50.7	-	317
PFPeA	-	8.3	-	-	<LOQ-754	1.3	-	7.9	0.9-3.6	-	110
PFHxA	18	21	26	4.02	15.8-68.7	3.2	ND-180	1	5.1-45.3	-	167
PFHpA	3.6	6.1	4.4	0.75	7.4-11	20.4	ND-55	1.8	1-2.7	-	29.6
PFOA	22	19	21	6.07	252-429	65	1.4-76	16.9	<2-5.9	<2-115.4	58.7
PFNA	1.3	0.92	8.4	0.26	2.9-5.54	9.6	ND-320	353.2	<2	<0.8-76	14.5

PFDA	3.0	1.3	3.5	0.14	3.4-2.32	2.8	ND-96	1.8	<2-3.7	<1.6-35.7	8.9
PFUnDA	0.49	<LOQ	-	0.03	1.1-11.7	2.7	ND-70	157.6	<2	<2.2-18.8	<LOQ
PFDoDa	0.11	0.47	-	0.06	<LOQ	1.6	ND-4.2	ND	<2	-	35
PFTrDA	-	<LOQ	-	0.03	-	4.8	-	-	-	-	-
PFTeDA	-	0.27	-	0.04	ND-<LOQ	2.2	-	-	-	-	-
PFODA	-	-	-	-	-	ND	-	-	-	-	-
PFBS	-	4	2.7	2.17	70.4-702	16.7	-	-	-	-	344
PFPeS	-	1.9	-	-	-	-	-	-	-	-	343
PFHxS	20	13	4.8	0.24	ND-42.7	18	-	50.4	-	<0.2-19.8	12500
PFHpS	-	0.86	-	-	-	2.5	-	-	-	-	5
PFOS	25	15	13	1.86	44.6-84.5	35	ND-8410	552.8	-	<1.5-1115	103
PFNS	-	-	-	-	-	-	-	-	-	-	-
PFDS	-	0.21	-	ND	-	ND	ND	-	-	-	-
PFUnDS	-	-	-	-	-	-	-	-	-	-	-
PFDoDS	-	-	-	-	-	-	-	-	-	-	-

PFTrDS	-	-	-	-	-	-	-	-	-	-	-
PFTeDS	-	-	-	-	-	-	-	-	-	-	-
PFOSA	-	-	-	-	-	-	-	-	-	-	-
FOSA	-	-	-	-	-	-	-	-	-	-	-
FOSAA	-	-	-	-	-	-	-	-	-	-	-
MeFOSAA	-	-	-	-	-	-	-	-	-	-	-
EtFOSAA	-	-	-	-	-	-	-	-	-	-	-
MeFBSA	-	-	-	-	-	-	-	-	-	-	-
MeFBSAA	-	-	-	-	-	-	-	-	-	-	-
4:2 FTSA	-	-	-	-	-	-	-	-	<0.8-14.0	-	-
6:2 FTSA	-	7.3	-	-	-	-	-	-	443-4413	-	<LOQ
8:2 FTSA	-	0.53	-	-	-	-	-	-	21.9-110	-	9.3
6:2 FTCA	-	-	-	-	-	-	-	-	2.2-3.0	-	-
8:2 FTCA	-	-	-	-	-	-	-	-	-	-	-
5:3 FTCA	-	-	-	-	-	-	-	-	-	-	-

7:3 FTCA	-	-	-	-	-	-	-	-	-	-	-
10:2 FTCA	-	-	-	-	-	-	-	-	-	-	-
6:2 FTUCA	-	-	-	-	-	-	-	-	-	-	-
8:2 FTUCA	-	-	-	-	-	-	-	-	-	-	-
10:2 FTUCA	-	-	-	-	-	-	-	-	-	-	-
6:2 FTOH	-	-	-	-	-	-	-	-	10.9-110	-	-
8:2 FTOH	-	-	-	-	-	-	-	-	12.5-4265	-	-
10:2 FTOH	-	-	-	-	-	-	-	-	14.1-1820	-	-
12:2 FTOH	-	-	-	-	-	-	-	-	0.1-1.1	-	-
6:2 FTI	-	-	-	-	-	-	-	-	0.7-3.5	-	-
6:2 FTMAC	-	-	-	-	-	-	-	-	1.3-2.6	-	-
8:2 FTAC	-	-	-	-	-	-	-	-	<10-136	-	-
8:2 FTMAC	-	-	-	-	-	-	-	-	0.2-<10	-	-
6:2 FTAB	-	-	-	-	-	-	-	-	3987-45462	-	-

M4	-	-	-	-	-	-	-	-	957-5748	-	-
ΣPFAS	Σ9=94	Σ21=110	Σ9=100	Σ15=38	Σ14=438.7 -1589	Σ16=192	Σ10=22-617	Σ10=1143	Σ23=5459-62200	Σ15=5180	Σ16=5180

LOQ = Values below the quantification limit; ND = Not detected; ΣPFAS = Sum of PFAS; No. of sites = number of sites or WWTPs recorded.

Table 3: Concentrations of PFAS (ng L^{-1}) in landfill leachates and stormwater recorded. Values recorded as mean or range. (See List of Abbreviations for associated compound names)

	(Lang et al., 2017)	**(Gallen et al., 2017)**	**(Yan et al., 2015)**	**(Solo-Gabriele et al., 2020)**	**(Benskin et al., 2012)**	**(Huset et al., 2011)**	**(Bai and Son, 2021)**	**(Zushi and Masunaga, 2009)**	**(Murakami et al., 2009)**	**(Nguyen et al., 2011)**	**(Xiao et al., 2011)**
Effluent type	Landfill leachates	Landfill leachates	Landfill leachates	Landfill leachates	Landfill leachates	Landfill leachates	Stormwater	Stormwater	Stormwater	Stormwater	Stormwater
Country	US	Australia	China	US	Canada	US	US	Japan	Japan	Singapore	US
No. of sites	18	27	5	5	2	4	2	1	3	5	4
PFPrA	-	-	5872	-	-	-	-	-	-	-	-
PFBA	300-1000	-	3518	1164	70-660	325	1.3	-	-	-	-
PFPeA	200-1000	-	2494	891	570-1300	-	29.05	-	-	-	-
PFHxA	500-2000	1700	1430	2276	650-2500	340	49.35	7.4	-	-	-
PFHpA	100-1000	430	1297	1600	240-690	210	14.5	6.4	-	<0.7-10.5	ND-6.8
PFOA	100-1000	690	49246	1640	210-1500	663	17.1	28.4	17-174	7.2-38.2	3.5-30.6

PFNA	5-100	49	146	93	15-450	20	ND	84.9	4.7-70	0.9-78.3	ND-10.7
PFDA	3-100	18	4	83	10-1100	7	ND	5	1.8-77	0.7-28.2	ND-10.6
PFUnDA	-	3.1	-	-	<3-120	2	-	6.7	NA-45	0.2-3.1	ND-2.9
PFDoDa	-	1.2	-	-	<1.4-16	0.2	ND	-	-	ND-1	-
PFTrDA	-	-	-	-	-	0	ND	-	-	-	-
PFTeDA	-	-	-	-	<1.5-5.1	1.8	-	-	-	-	-
PFODA	-	-	-	-	-	-	-	-	-	-	-
PFBS	10-500	-	15236	1694	28-190	247	11.45	3	-	-	-
PFPeS	-	-	-	-	-	-	1.5	1.5	-	-	-
PFHxS	20-500	1200	131	1684	85-570	177	8.8	15.8	-	1.2-73	-
PFHpS	-	-	-	-	-	-	ND	-	-	-	-
PFOS	-	310	2716	598	80-4400	59	7.55	44.1	2.8-50	4.5-139	<LOQ-156
PFNS	-	-	-	-	-	-	-	-	-	-	-
PFDS	-	-	-	-	<1.8-63	0.4	5.8	-	-	-	-
PFUnDS	-	-	-	-	-	-	-	-	-	-	-

PFDoDS	-	-	-	-	-	-	-	-	-	-	-
PFTrDS	-	-	-	-	-	-	-	-	-	-	-
PFTeDS	-	-	-	-	-	-	-	-	-	-	-
PFOSA	-	-	-	-	-	-	-	-	-	-	-
FOSA	-	-	-	-	3.4-220	0	-	-	-	ND-3.2	-
FOSAA	-	-	-	-	22-990	0.2	-	-	-	ND-1.8	-
MeFOSAA	-	-	-	-	180-5000	77	-	-	-	0.3-4	-
EtFOSAA	-	-	-	-	290-8700	140	-	-	-	0.3-14.7	-
MeFBSA	-	-	-	-	-	0.5	-	-	-	-	-
MeFBSAA	-	-	-	-	-	92	-	-	-	-	-
4:2 FTSA	-	-	-	-	-	-	-	-	-	-	-
6:2 FTSA	5-300	-	-	-	-	93	-	-	-	<0.4-35.9	-
8:2 FTSA	0.3-100	-	-	-	-	1.8	-	-	-	-	-
6:2 FTCA	100-3000	-	-	-	<5.7-280	-	-	-	-	-	-
8:2 FTCA	10-400	-	-	-	<8.6-5200	-	-	-	-	-	-

5:3 FTCA	400-15000	-	-	1231	-	-	-	-	-	-	-
7:3 FTCA	30-900	-	-	-	-	-	-	-	-	-	-
10:2 FTCA	-	-	-	-	36-775	-	-	-	-	-	-
6:2 FTUCA	-	-	-	-	7.1-64	-	-	-	-	-	-
8:2 FTUCA	-	-	-	-	49-2100	0.7	-	-	-	-	-
10:2 FTUCA	-	-	-	-	7.3-430	-	-	-	-	-	-
6:2 FTOH	-	-	-	-	-	-	-	-	-	-	-
8:2 FTOH	-	-	-	-	-	-	-	-	-	-	-
10:2 FTOH	-	-	-	-	-	-	-	-	-	-	-
12:2 FTOH	-	-	-	-	-	-	-	-	-	-	-
6:2 FTI	-	-	-	-	-	-	-	-	-	-	-
ΣPFAS	Σ19=2000-29000	Σ9=4401	Σ11=82076	Σ11=11791	Σ24=2546-36207	Σ24=2738	Σ16=190	Σ10=203	Σ5=3.3-387.6	Σ13=29.4-643.9	Σ6=3.5-217

LOQ = Values below the quantification limit; ND = Not detected; ΣPFAS = Sum of PFAS; No. of sites = number of sites or WWTPs recorded.

1412 *Table 4: Removal efficacy with advantages and disadvantages of different remediation of PFAS during 2017-2021*

Method	Materials	Removal efficacy of PFAS (% or, ngL^{-1})						Advantages	Disadvantages	Ref.
		PFOA	PFOS	PFBA	PFBS	PFHxA	PFHxS			
Sorption	GAC	~93%	~61%	~94%				➢ Better for short-chain PFBA removal ➢ more sorption capability.	➢ low performance for short-chain PFAS. ➢ Longer operation time and pores block	(Inyang and Dickenson, 2017)
	Poly(ethylenimine)-functionalized cellulose microcrystals	~90%	~98%	~2.5 %	~7%	~12.5%	~52%	➢ Cost effective ➢ Natural based adsorbent ➢ potential for long-chain PFAS	➢ Not good for short-chain PFAS removal	(Ateia et al., 2018)
	*IRA910 resin	~38%	~88%	~2%	~22%	~10%	~52%	➢ Enable to competitive sorption ➢ Effective sorption media for long-chain PFAS removal.	➢ Decrease removal efficacy in presence of inorganic ions.	(Maimaiti et al., 2018)
	Magnetic ion-exchange (IX) resin	~55%	~85%	~20%	~70%	~40%		➢ Cost effective and potent for short chain PFAS removal. ➢ Have higher stability, fast recover of sorption capability	➢ Not effective for long chain of PFAS. ➢ Regeneration media are not friendly	(Park et al., 2020)
	Anionic IX resin (AIX)	~95%	~90%	~98%	~92%			➢ >99% PFAS removal ➢ Resin regeneration efficiency too good with >10% NaCl. ➢ Overall cost effective	➢ need backwash	(Dixit et al., 2020)

	DMAPAA-Q* synthesized polymer hydrogel	~88%	~100 %	~81%	~91%	~80%	~93%	➢ Excellent adsorption capability of short- and long-chain PFAS ➢ Fast removal kinetics (1-2 h) ➢ Not affected by pH range. ➢ Excellent regeneration	➢ Need to effective design instead of lab scale.	(Ateia et al., 2019)
Filtration	Nanofiltration with AIX	24 $\pm$ 3.3^a	142 $\pm$ 24^a	17 $\pm$ 4.7^a	44 $\pm$ 9.7^a	37 $\pm$ 9.2^a	306 $\pm$ 24^a	➢ Capable to remove 11 PFASs. ➢ 98% removal performance ➢ Safe and acceptable method	➢ Challenging in process monitoring and maintaining.	(Franke et al., 2021)
	β-lactoglobulin amyloid fibril membrane	~50%^{b}	~100%	-	~100%	~10%^{b}	~99%^{b}	➢ Good for long and short chain of PFAS removal. ➢ Environmentally friendly and high sustainability	➢ complex mechanism.	(Jin et al., 2021)
Chemical redox	Non-thermal plasma generator	50%	85%			40%		➢ activate oxidation, electric field, free radical & ozone ➢ Energy saving treatment.	➢ Not good for short-chain PFAS ➢ byproducts form	(Palma et al., 2021)
	Transition Metal (Ag^+)– Activated PS*	~80%	~100 %					➢ Decomposition occurs within a comparable short time.	➢ Complex system.	(Parenky Akshay et al., 2020)

	Ultrasonic activated PS*	100 ± 1.2%	46.5 ± 1.0%			62-71%		➢ Synergistic effects of degradation. ➢ Easy operating.	➢ byproducts form ➢ Need development for field application	(Lei et al., 2020)
	UV/sulfite reductive	93 ± 1.6%		92 ± 2.8%		89 ± 2.4%		➢ Materialization performance too good.		(Bentel et al., 2020)
Electrochemical	PbO_2 activated anode	>99%	0.5%					➢ High current density to enhance degrade capability. ➢ Cost effective	➢ High operation time ➢ Complex process ➢ Ion & HF gas leaked	(Fang et al., 2019)
	Titanium suboxide anode	>90%	>90%	50%	>90%	>90%	>90%	➢ High fluoride release ratio ➢ Complete mineralization. ➢ Degrading all PFAS (10 min)	➢ Need to more development ➢ Costly system.	(Wang et al., 2020)
	B-doped diamond electrode	~100%	~80%					➢ High current density ➢ Produce minimal amount of short-chain byproduct	➢ Needed extra treatment time	(Uwayezu et al., 2021)
Thermal treatment	Volatilization of PFAS at 350˚C	99.22 %	99.92 %	99.82 %	99.80 %	99.96%	99.84 %	➢ Excellent degrading capability ➢ At 400˚C all PFAS are degraded almost 100%	➢ Not cost effective ➢ Toxic byproducts	(Crownover et al., 2019)
	GAC*-thermal reactivation	70-99.81 %	98.67 - 100%	94.31 - 99.98 %	97.51 - 99.97 %		98.30-99.94 %	➢ Mineralization capability of PFAS is high ➢ Degradation occurs within a short time.	➢ toxic byproducts. ➢ high temperature	(Xiao et al., 2020)

Hybrid treatment	*[RO-UV/AOP + O_3-BAC-GAC].	>77 ± 14%	>71%	>79 ± 10%	>96 ± 6%	>80%	➢ Cover removal of almost all PFAS materials.	➢ System management too challenging	(Glover et al., 2018)

*DMAPAA-Q: Poly (N-[3-(dimethyl amino)propyl] acrylamide, methyl chloride quaternary); *GAC: Granular activated carbon; *[RO-UV/AOP + O_3-BAC-GAC]: [Reverse osmosis-UV/advanced oxidation process + ozone-biological activated carbon filtration-granular activated carbon adsorption]; *IRA910: Strong base anion-exchange resin; *PS: persulfate; Filtration^a: Captured concentration (ngL^{-1}) of PFAS from feedwater during two stages stepwise nanofiltration by two Filmtec-NF90 and NF270 membrane respectively; Adsorption^b: Removal efficiency (%) of PFAS from NF concentrate at 20×10^3 BV during treating by Purofine PFA694 resin.

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1415 *Table 5: PFAS compounds and the regulatory guidelines in selected countries/states*

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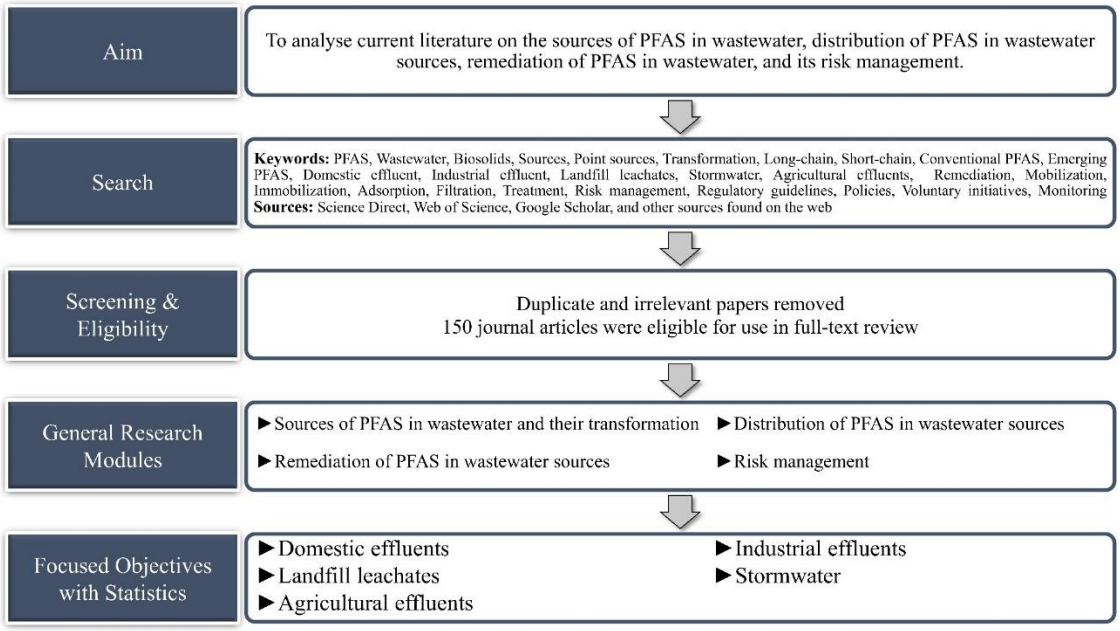
Country/state	**Compound**	**Standard**	**Advisory/regulatory body**	**Type of water**	**References**
Australia	PFOA, ∑PFOS, ∑PFHxS	10 μg L^{-1}, 2 μg L^{-1}	National Health and Medical Research Council	Recreational water	(Coggan et al.,2019)
Australia	PFOS, PFOA	0.23 ng L^{-1}, 1900 ng L^{-1}	National Environmental Management Plan	Fresh water and marine water	(Coggan et al.,2019)
Germany	∑PFOA, ∑PFOS	100 ng L^{-1}	German Ministry of Health	Drinking water	(Kim et al., 2012)
Sweden	∑PFOA, ∑PFOS, ∑PFHxS, ∑PFHxA, ∑PFHpA, ∑PFBS, ∑PFBA, ∑PFPeA, ∑PFNA, ∑PFDA	90 ng L^{-1}	National Food Agency	Drinking water	(Chohan et al., 2021)

Italy	∑PFAS, ∑PFOA, ∑PFOS,	500 ng L^{-1}, 30 ng L^{-1}, 500 ng L^{-1}	Italian National Health Service	Drinking water	(Chohan et al., 2021)
Canada	PFOA, PFOS	200 ng L^{-1}, 600 ng L^{-1}	Health Canada	Drinking water	(CELA, 2019)
USA	∑PFOA, ∑PFOS	70 ng L^{-1}	US EPA	Drinking water	(Cordner et al., 2019)
USA/Minnesota	PFOA, PFOS	35 ng L^{-1}, 27 ng L^{-1}	Minnesota Department of Health	Drinking water	(Cordner et al., 2019)
USA/Vermont	∑PFOA, ∑PFOS, ∑PFHxS, ∑PFHpA, ∑PFNA	20 ng L^{-1}	Department of Environmental Conservation	Drinking water	Cordner et al., 2019)
USA/ New Jersey	PFOA, PFOS	14 ng L^{-1}, 13 ng L^{-1}	New Jersey Department of Environmental Protection	Drinking water	Cordner et al., 2019)

USA/California	PFOA, PFOS	14 ng L^{-1}, 13 ng L^{-1}	California State Water Resources Control Board	Drinking water	(CARE, 2019)
USA/New Hampshire	PFOA, PFOS, PFHxS, PFNA, PFOA	12 ng L^{-1}, 15 ng L^{-1}, 18 ng L^{-1}, 11 ng L^{-1}	New Hampshire Department of Environmental Services	Drinking water	(Gagliano et al. 2020)
USA/Wisconsin	PFOA, PFOS	2 ng L^{-1}	Wisconsin Department of Health	Drinking water	(Gagliano et al. 2020)

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Supplementary Material



SI-Figure 1. Flowchart of the journal article decision-making process and scope of the review