

Chapter 3

World Profile of Foreseeable Strategies for the Removal of Per- and Polyfluoroalkyl Substances (PFASs) from Water



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Abstract Per- and polyfluoroalkyl substances (PFASs) have been used in wide range of industries and daily life and therefore released into the aqueous environment. Due to their unique properties, such as environmental persistence, bioaccumulation, and toxicity, PFASs have drawn increasing concern in recent years. PFASs-contaminated water has adverse effects on water microorganisms and aquatic life as well as human life. So far, tremendous efforts have been made on PFASs pollution and their treatment, yet most of the efforts have been spent on laboratory experiments. Their feasibility, cost-effectiveness, and field applicability are questionable. This review examined studies on existing while updated treatment technologies, with the goal of providing an outlook on these technologies and more importantly, proposing the most likely technique. As such, a constructed wetland-microbial fuel cell (CW-MFC) technology was recommended, which is a newly emerged technology by integrating physical, chemical, and enhanced biological processes plus the wetland plants' functions with strong eco-friendly features for the comprehensive removal of PFASs. The roles of wetland plants, substrates, and electroactive bacteria (EAB) in the removal of PFASs in the CW-MFC system were discussed with focus on highlighting the different mechanisms. It is expected that the review can strengthen our understanding of PFASs' research and thus can help select reasonable technical means of aqueous PFASs control.

Keywords Per- and polyfluoroalkyl substances · Water treatment · Degradation technologies · Construct wetlands · Microbial fuel cell

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3.1 Introduction

Per- and polyfluoroalkyl substances (PFASs) are a group of synthetic chemicals comprising more than 9000 chemicals (Cordner et al. 2021). Due to their unique hydrophobic/lipophobic physicochemical properties, they have a wide range of industrial and consumer applications such as packaging materials, waterproof material, electronics, polishes, and aqueous film-forming foams (AFFFs) (Naidu et al. 2020). PFASs have been developed for more than seven decades with the different types of fluoropolymers over 230,000 tons/year worldwide for the moment (Zhang et al. 2022). PFASs are released into the environment as a result of their widespread manufacture and application both purposefully and unintentionally. Due to persistent (P), bioaccumulative (B), and toxic (T) properties, as well as long-range transport potential (LRTP), PFASs have been detected in different environments (such as water, soil, and aerosols), which have caused great concern among the scientific community and the public. Previous studies have reported that PFASs have been linked to a slew of negative health effects in both animal and human (Naidu et al. 2020; Silva et al. 2021; Teymourian et al. 2021). Therefore, when PFASs enter the environment, they are labeled as “contaminants of emergent concerns (CECs).”

Water environment is regarded as a significant sink and source for PFASs. Many researchers have conducted a series of studies on the content level, distribution characteristics, influencing factors, and environmental behavior of PFASs in aqueous environment in recent years (Macorps et al. 2022; Pistocchi and Loos 2009; Szabo et al. 2018). In order to control and reduce the environmental risk of PFASs, some remediation technologies and methods have been tested, such as coagulation, adsorption, membrane separation, photodegradation, and hydrothermal as well as microbial degradation (Hao et al. 2022; Oyetade et al. 2018; Zhuo et al. 2020). It should be noted that these approaches have both advantages and disadvantages in foreseeable applications. When it comes to controlling or removing PFASs from water, integrated technologies are thought to be more effective than solo techniques.

Constructed wetlands (CWs) are widely applied across the world due to their cost-effectiveness and satisfactory treatment effects. CWs use plants, substrates (soil, sand, and gravel), microbial and interaction processes to remove contaminants in wastewater with minimal dependence on mechanical elements. So far, CW system has been proven effectively to removal CECs and persistent organic pollutants (POPs) in the aqueous environment (Li et al. 2014; Matamoros et al. 2008; Vymazal and Březinová 2015). In recent years, a novel technique of introducing microbial fuel cell (MFC) into CW to form CW-MFC has been studied. Compared with conventional CW, CW-MFC system is more powerful in removing several hard- or non-degradable contaminants (Wang et al. 2019a; Zhang et al. 2018). To the best of our knowledge, rare information exists on foreseeable strategies for the removal of PFASs from water in CW-MFC system.

This review covers the state of the art and understanding of treatment strategies for PFASs from water environment after briefly describing their characteristics, hazards,

and challenges. Moreover, this review focuses on the potential of employing CW-MFC system for PFASs removal. The knowledge provided in this review is hoped to improve our understanding of PFASs research and aid in the selection of suitable technological ways of aqueous PFASs control.

3.2 Overview of PFASs in Water Environment

3.2.1 Properties

PFASs are organofluorine compounds with alkyl chain as the skeleton and hydrogen atom partially or completely replaced by fluorine atom, which can vary in chain length and linear or branched isomers (Buck et al. 2011). The general chemical formula of PFASs is $F(CF_2)_n-R$, where R is hydrophilic functional group. Recently, according to the Organization for Economic Cooperation and Development, “PFASs are defined as fluorinated substances that contain at least one fully fluorinated methyl or methylene carbon atom (without any H/Cl/Br/I atom attached to it), i.e., with a few noted exceptions, any chemical with at least a perfluorinated methyl group ($-CF_3$) or a perfluorinated methylene group ($-CF_2-$) is a PFAS” (Wang et al. 2021). All the PFASs have the structure of a C–F bond, which is extremely polar, strong, and stable, giving PFASs exceptional chemical properties such as extremely high thermal and chemical stability. Perfluoroalkyl acids (PFAAs) are the most important and basic PFASs molecule, which are divided into two groups: perfluoroalkyl sulfonic acids (PFSAs) and perfluoroalkyl carboxylic acids (PFCAs). The most frequently detected PFSAs and PFCAs in the water environment are perfluorooctanoic acid (PFOA) and perfluorooctanesulfonic acid (PFOS), respectively (Podder et al. 2021).

Regarding the carbon chain length of PFASs, PFASs can further divide into two main categories, the long-chain PFASs (the carbon number ≥ 8 with PFCAs, and ≥ 7 with PFSAs) and the short-chain PFASs (Fig. 3.1). With different chain length and hydrophilic functional group, PFASs have unique properties. As explained, for example, the short-chain PFASs, such as perfluorobutanoic acid (PFBA), are highly soluble with lower sorption tendency in soil, which leads to greater mobility and bioaccumulation than that of the long-chain PFASs in the environment (Teymourian et al. 2021; Macorps et al. 2022).

Concerns about the potential environmental issues and human hazard impact of PFASs have been investigated. Researchers have well-documented that the toxicities of PFASs involve muscle, liver, reproduction and development, neurotoxicity and potential carcinogenicity (Sunderland et al. 2019), while the arithmetic and geometric mean half-lives of serum elimination are 5.4 years for PFOS, and 3.8 years for PFOA, respectively (Olsen et al. 2007). The main exposure pathways for human may include drinking contaminated water, eating contaminated foods, and inhalation of some neutral volatile PFASs, such as fluorotelomer alcohols (FTOHs). In terms of environmental risk assessment, there has been a rise in ecological toxicity studies

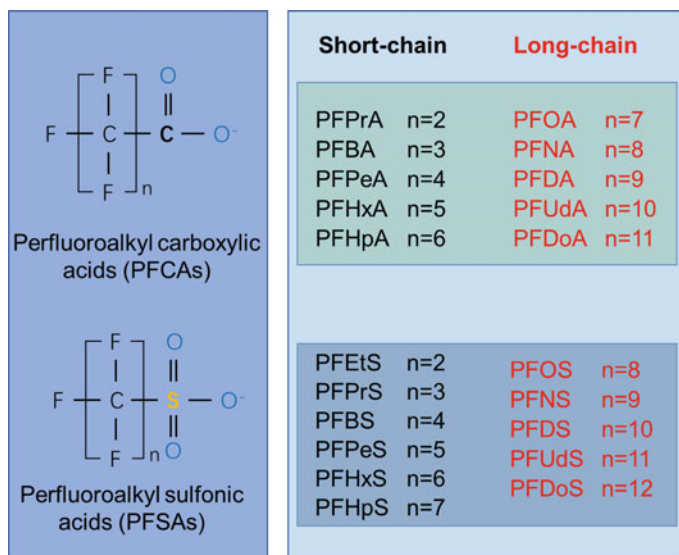


Fig. 3.1 Typical classification of PFASs

as researchers attempt to understand the impacts of PFASs on various ecosystems (Zhang et al. 2022; Cai et al. 2019). However, evaluating the toxicity of PFASs involves a number of obstacles, not the least of which being the diversity of PFASs compounds, as well as variances in their mechanisms of action in different animals. According to Sinclair et al. (2020) who reviewed the PFASs toxicity data in invertebrates, fish, and amphibians, the concentrations that induced mortality in 50% (LC₅₀) of the studied population of PFASs are unlikely to occur in the outside environment. To move forward, scientists must bridge the data gap between laboratory exposures and the actual environmental concentration.

3.2.2 Occurrence of PFASs in Water Environment

Monitoring the occurrence of PFASs in the water environment has recently become a major concern in the world. PFASs have been widely detected in wastewater, surface water, and groundwater, and even drinkable bottle water (Table 3.1).

The domestic and industrial wastewater and landfill leachate are the major sources of PFASs in water environment. As shown in Table 3.1, the high total concentrations of PFASs (0.2–6000 μg/L) were found in the wastewater from the chrome plating industry (Qu et al. 2020). Moreover, more than 800 μg/L of PFASs was detected in the effluent of electroplating wastewater treatment plant (WWTP) although this electroplating WWTP had ultrafiltration (UF) and reverse osmosis (RO) processes (Qu et al. 2020). On the other hand, landfill leachate is a significant source of high

Table 3.1 Detection and investigation of PFASs in water environment

Water environment	Main compounds	PFASs analyte count	Total concentration (ng/L)	Reference
Municipal wastewater treatment plant (WWTP)	PFOS, 6:2 Cl-PFAES	4	1610–153,000	Qu et al. (2020)
Municipal WWTP	PFOA, PFOS	3–17	1030–3360	Masoner et al. (2020)
Municipal WWTP	PFOA, PFBA	21	9.3–520	Coggan et al. (2019)
Municipal WWTP	PFBA, PFHxA	27	16–1400	Lu et al. (2021)
Landfill leachate	PFHxA	21–31	19,800–48,700	Masoner et al. (2020)
Landfill leachate	PFBS	7	0.1–17,000	Harrad et al. (2019)
Industrial wastewater	6:2 Cl-PFESA, 8:2 Cl-PFESA	17	2100–4200	Liu et al. (2022)
Industry wastewater	PFOS, 6:2 Cl-PFAES	7	200–6,000,000	Qu et al. (2020)
Seawater	PFOA	21	0.98–2.64	Xiao et al. (2021)
Seawater	PFBA, PFOA	20	0.125–1015	Wang et al. (2019b)
River	PFOS, 6:2 Cl-PFAES, 6:2 FTS, PFOA	4	400–2300	Qu et al. (2020)
River	PFOA, PFOS, PFHxA	14	0.04–31.3	Cai et al. (2022)
River	PFOA, PFBA	15	11.8–281	Li et al. (2022)
River	PFOA	13	0.8–274.6	Dong et al. (2020)
Lake	PFPeA, PFHxA	16	17.7–467.0	Lee et al. (2020)
Lake	PFHxS	13	30.45–1563	An et al. (2021)
Groundwater	PFOS	3	1.9–2.4	Qu et al. (2020)
Groundwater	PFOS, PFBA	24	31.4–15,656.0	Gao et al. (2019)
Drinking Water	PFOA, PFOS	9	1.36–20.2	Jiang et al. (2021)
Drinking water	PFBA, PFHxA	6	11.8–59.7	Gao et al. (2019)
Drinking water	PFBA, PFOA	17	9.29–266.68	Chen et al. (2019a)
Bottle water	PFPrA, PFCA	32	0.17–18.87	Chow et al. (2021)

PFASs concentrations. In a study, more than 20 types of PFASs were found in landfill leachate with a total concentration of 93,100 ng/L, which was an order of magnitude greater than the influent and effluent of the WWTP receiving this landfill leachate (Masoner et al. 2020). Compared with industrial wastewater and landfill leachate, the concentrations of PFASs in domestic wastewater are generally lower. Many home products, such as fast food containers/wraps, cleaning products, and non-stick cookware, contain PFASs and release them into wastewater after usage. Coggan et al. (2019) investigated the PFASs in the influent and effluent of nineteen Australian WWTPs with varied sizes, capacities, localities, and treatment types. The results demonstrated that the range of PFASs concentrations in WWTPs influent and effluent was 9.3–520 ng/L, and there were no significant PFASs removals after WWTPs.

It is worth noting that most conventional WWTPs were not capable to remove PFASs. Thus, a considerable amount of PFASs-containing wastewater is discharged into natural water bodies, resulting in surface water and groundwater pollution. In the Asan Lake of South Korea, the 19 PFASs were detected with the total average concentration of 17.7–467.0 ng/L, the wastewater from the nearby industrial parks may be a major source of PFASs in the lake (Lee et al. 2020). Similarly, the average concentrations of PFASs in Tai Lake in China in 2018 and 2019 were 205.6 ng/L and 171.9 ng/L, respectively, while the study also suggested that the local industry was the main source of PFASs (An et al. 2021). Furthermore, the vertical transfer in soil and water bodies contaminated by PFASs leads to groundwater contamination. One study found that the total concentrations of PFAAs range from 31.4 to 15,656.0 ng/L in groundwater sample, with short-chain PFAAs being the most frequent (Gao et al. 2019). This study also found that the concentration of PFASs in groundwater decreased with increasing distance from the manufactory.

The presence of PFAS in drinking and bottled water seems inevitable due to the ubiquitous occurrence of PFASs in surface and groundwater, especially because present technologies of drinking water treatment plants (DWTPs) are incapable to completely remove PFASs. Research in Ireland found that PFOS, PFOA, PFBS, and PFHxS were all discovered in tap water from homes and offices (Harrad et al. 2019). Chen et al. (2019a) further investigated the occurrence and transport behaviors of PFASs in drinking water distribution systems (DWDS), they detected 17 kinds of PFAAs in the tap water with the average concentrations ranging from 9.29 to 266.68 ng/L. The results also found that the fate and migration of PFASs in the DWDS were influenced by their physicochemical characteristics and the sediments in the system. According to the results of PFASs in bottle water research done in the USA (Chow et al. 2021), the total PFASs concentration in 39 examined samples was 0.17–18.87 ng/L while 97% of samples were below 5 ng/L.

Overall, these studies suggest that PFASs are already widespread in a wide range of water environment. The government should take necessary and immediate action to set up relevant regulation for PFAS removal/control.

3.2.3 Protocols of PFASs in Drinking Water

PFASs are widespread in aquatic environments and have been associated with human and animal health. They are difficult to degrade in nature. As a result, some countries and local/state governments have enacted legislation/protocol to limit the concentration of PFASs in drinking water. Table 3.2 lists the updated information.

The US Environmental Protection Agency (EPA) proposed PFASs drinking water restrictions in 2016 with maximum contamination levels (MCLs) of 70 ng/L for either PFOS, PFOA, or their total. Some states in the USA also have strict or loose regulations of PFAS in drinking water (Table 3.2). National regulations

Table 3.2 Global protocol for limiting the amount of PFASs in drinking water

Country/state	PFOA (ng/L)	PFOS (ng/L)	Year of action
US EPA (https://www.epa.gov/sdwa/drinking-water-health-advisories-pfoa-and-pfos)	70	70	2016
California (https://www.jdsupra.com/legalnews/pfas-update-state-by-state-regulation-4639985/)	5.1	6.5	2021
Minnesota (https://www.jdsupra.com/legalnews/pfas-update-state-by-state-regulation-4639985/)	–	15	2019
New York (https://www.jdsupra.com/legalnews/pfas-update-state-by-state-regulation-4639985/)	10	10	2020
Massachusetts (https://www.jdsupra.com/legalnews/pfas-update-state-by-state-regulation-4639985/)	20	20	2018
Canada (https://www.canada.ca/en/health-canada/programs/consultation-perfluorooctanoic-acid-pfoa-in-drinking-water/document.html)	200	600	2018
Denmark (https://www2.mst.dk/Udgiv/publications/2015/04/978-87-93283-01-5.pdf)	300	100	2015
Germany (https://www2.mst.dk/Udgiv/publications/2015/04/978-87-93283-01-5.pdf)	300	300	2006
Sweden (https://www2.mst.dk/Udgiv/publications/2015/04/978-87-93283-01-5.pdf)	90	90	2014
Netherlands (Kurwadkar et al. 2022)	390	200	2020
United Kingdom (https://www.dwi.gov.uk/en/private-water-supplies/pws-installations/guidance-on-the-water-supply-water-quality-regulations-2016-specific-to-pfos-perfluorooctane-sulphonate-and-pfoa-perfluorooctanoic-acid-concentrations-in-drinking-water/)	10	10	2021
Australia (https://www.pfas.gov.au/government-action/pfas-food-water)	560	70	2017
China (https://openstd.samr.gov.cn/bz/gk/gb/newGbInfo?hcno=99E9C17E3547A3C0CE2FD1FFD9F2F7BE)	80	40	2022

on PFASs in drinking water vary greatly throughout the world. The MCLs for PFOA and PFOS in Canadian drinking water are 200 and 600 ng/L, respectively (<https://www.canada.ca/en/health-canada/programs/consultation-perfluorooctanoic-acid-pfoa-in-drinking-water/document.html>), but the MCLs for these two compounds in the UK are only 10 ng/L (<https://www2.mst.dk/Udgiv/publications/2015/04/978-87-93283-01-5.pdf>). Most recently, the Chinese government announces a new “Standard for drinking water quality (GB 5749-2022),” which limits the values of PFOA and PFOS in drinking water of 80 ng/L and 40 ng/L, respectively (<https://openstd.samr.gov.cn/bz/gk/gb/newGbInfo?hcno=99E9C17E3547A3C0CE2FD1FFD9F2F7BE>). Although PFAS concentration in drinking water has been subjected to a variety of regulatory laws and standards up to this point, it is important to keep in mind that PFASs are consistently harmful to humans. It is critical to implement corresponding regulations in some developing nations and regions to limit PFASs contamination in drinking water, as well as more comprehensive national collaboration.

3.3 Treatment Technologies for PFASs in Water

As “forever chemical,” the treatment technologies for PFASs in water environment can be divided into separation technologies and destruction/degradation technologies. The main separation technologies include adsorption and membrane filtration, while thermal treatment, advanced oxidation/reduction, hydrothermal, and biological technologies are potential to the destruction of PFASs.

3.3.1 Adsorption

Adsorption is the most frequently investigated and employed treatment technology. It has been used for both ex-situ and in-situ water treatment applications. Various adsorbent materials have been tested regarding the removal performance of PFASs from water (Zhang et al. 2019a). Granular activated carbon (GAC) is considered as efficient and more economic sorbent with good performance (normally > 90%) in controlling aqueous PFASs. For example, the average removal efficiency of 15 PFASs in a full-scale DWTP with GAC as filter was from 92 to 100% (Belkouteb et al. 2020). A review of the PFASs adsorption performance of commercial carbon materials found that GAC has an adsorption capacity in the tens to hundreds of mg/g range (Pauletto and Bandosz 2022). Although powdered activated carbon (PAC) has a better adsorption capacity for PFASs than that of GAC (Zhang et al. 2019a), it has intrinsic disadvantages such as regeneration and reutilization. The impact of sorption, on the other hand, differs depending on the type of PFASs. For different PFASs, GAC/PAC has a variable adsorption capability and breakthrough time. Because the sulfonate group is more readily adsorbed onto oxide surfaces than the carboxylate

group, PFASs tend to adsorb more strongly than PFCAs with equal chain length (Du et al. 2014; Wang et al. 2012), whereas the adsorption of branched isomers is smaller than linear (McCleaf et al. 2017).

Other adsorbents, such as resins (McCleaf et al. 2017), cyclodextrin polymers (Ching et al. 2022), biochar (Sørmo et al. 2021; Xiao et al. 2017), and metal-organic frameworks (MOFs) (Pauletto and Bandosz 2022) have been tested in recent years to remove PFASs from water. It is worth noted that the properties of adsorbents and the kind of PFASs, as well as the environmental factors (i.e., solution pH, surface charge, dissolved organic matter, inorganic ions) can significantly affect the adsorption process (Oliver et al. 2019; Qi et al. 2022). When hydrophobic interaction is the primary mechanism in adsorption, short C–F chain PFASs have a more hydrophilic character as well as a smaller molecular size, resulting in a quick breakthrough time and low removal efficiency (Li et al. 2019a).

Electrocoagulation (EC) is another adsorption reaction-based technique for PFASs removal. The main mechanism of EC is adsorption of contaminants with metal hydroxide flocs which are formed by the soluble metal anode (e.g., Fe, Al, Zn) under the action of an applied electric field. At high influent concentrations (0.25 mM), a study using an air cathode in an electrocoagulation reactor demonstrated excellent PFOA removal efficiency (approximately 100%), whereas PFOS removal at low concentrations (0.1 μM) was superior than PFOA removal (Mu et al. 2021). In addition, the anode metal materials are the key to the electrocoagulation process. Liu et al. (2018) investigated the EC technique using Al-Zn electrodes for the removal of PFOA. The results achieved over 99% removal under 1 mg/L influent concentration.

Although adsorption appears to be a well-established method for PFASs removal, there are certain disadvantages to adsorption investigations to date. The majority of research has focused on the removal of a particular kind of legacy PFASs (i.e., PFOA and PFOS), despite the fact that the aquatic environment contains a variety of emerging PFASs with various functional groups and chain lengths (Oyetade et al. 2018; Zhang et al. 2019a, 2021). Furthermore, the effects of various organic matter components, inorganic ions, and microbials in water or wastewater must be investigated further. Adsorbents utilized to remove PFASs, on the other hand, should be carefully evaluated for disposal or regeneration after saturation. Information on the long-term stable adsorption of aqueous PFASs at full scale is particularly needed from an engineering standpoint.

3.3.2 Membrane Filtration

The membrane filtration, such as the reverse osmosis (RO) and nanofiltration (NF), achieves remarkable removal of aqueous PFASs (Wang et al. 2018). NF membranes have been used to remove various types of PFASs from water in several studies. They have been found to be particularly effective in removing PFASs regardless of chain length or functional group (Wang et al. 2018; Léniz-Pizarro et al. 2022;

Mastropietro et al. 2021; Xiong et al. 2021). PFOS and perfluorobutane sulfonate (PFBS) through NF (piperazine amide) membrane showed high retention rate with about 90% (Wang et al. 2018). RO is effective in the removal of mainly organic and inorganic contaminants from water and has been employed in industrial water treatment around the world. Regarding PFOS removal in water, RO membrane is better than NF (Tang et al. 2007). However, the ionic strength and solution pH significantly affected retention rate of membrane filtration process. In addition, the pretreatment processes (such as coagulation/flocculation–precipitation, adsorption) are important to avoid fouling with RO and NF to remove PFASs. Furthermore, high-pressure pumps are required to give transmembrane power to RO and NF, which subsequently hold fluid with high concentrations of PFASs. Another key question is how to manage and dispose of this hazardous liquid. The treatment train consisted of RO or NF filtration separation and degradative/destructive technologies have been recommended. This combination of several removal procedures improves removal efficiency while lowering costs and reducing hazardous by-product (Lu et al. 2020).

3.3.3 Destructive Techniques

3.3.3.1 Advanced Oxidation and Reduction

Advanced oxidation processes (AOPs) can form highly oxidizing species, like ozone (O_3), hydroxyl radicals (HO^\bullet), persulfate ($S_2O_8^{2-}$), sulfate radical anions (SO_4^\bullet), hydroperoxide (HO_2^\bullet), and superoxide (O_2^\bullet) ions, though oxidant activation with solar radiation, heat, and catalysts. However, because HO^\bullet and O_3 do not destroy the C–F bond, they are unable to fully breakdown PFOA/PFOS (Qi et al. 2022). Sulfate or/and persulfate generated can successfully breakdown PFOA under a range of circumstances, according to several lab-scale research (Wu et al. 2018; Zhang et al. 2019b). Several shorter-chain PFAAs, such as perfluoroheptanoic acid (PFHpA) and PFBA, were generated by the loss of CF_2 units from PFOA with persulfate reaction (Bai et al. 2022). Although other studies have shown that ultrasonic treatment (Lei et al. 2020), persulfate photolysis (Li et al. 2019a), photocatalysis (Jin et al. 2014; Qian et al. 2021), sonochemical (Cao et al. 2020; Rodriguez-Freire et al. 2015), and Fenton (Santos et al. 2016) can be used to destroy PFASs, these studies mostly require strict operating conditions. This suggested that, in the field application, generating highly oxidizing species to remove PFASs might be difficult due to unreasonably high costs and sluggish reaction rates.

Several studies reported that zero-valent metals can be employed to eliminate PFASs by chemical reduction (Arvaniti et al. 2015; Chen et al. 2020; Hori et al. 2008). Hori et al. (2008) used subcritical water and zero-valent iron (ZVI) to breakdown PFASs at high pressure (20 MPa) and temperature (350 °C). The results revealed that the ZVI powder accelerated the breakdown of PFOS into F ions. Following that, other investigations revealed that PFOS and PFOA were destroyed at 380 °C, with yields of 9.2% fluorine and 21.9% sulfate, respectively. The ZVI surface area

was shown to be linked with PFASs degradation rates, indicating that the reaction is surface catalyzed. This technique, like AOPs, needs more energy and appropriate reaction circumstances. Recently, Chen et al. (2019b, 2020) discovered that hydrated electrons can quickly destroy PFOA and PFOS under UV irradiation with surfactants. They suggest that hydrated electrons generated by photo-irradiation can attack C–F bonds under ambient environments, enabling defluorination and mineralization of PFASs in actual wastewater. However, there is currently a lack of long-term and field-scale application for this technique.

Several chemical redox technologies have also been shown to reduce PFASs contamination in laboratories. There are several considerations in a full-scale application. To begin with it, the factional group (carboxylic or sulfonic) of PFASs is more susceptible to redox transition than the C–F bond, resulting in the parent compound's partial transformation (Anumol et al. 2016). It will produce various intermediates, such as short-chain PFAAs, as well as potentially more mobile or hazardous substrates. Secondly, typical co-contaminants in wastewater would have an influence on the removal of PFASs and the generation of by-products (Mu et al. 2021). Furthermore, oxidizing species are more susceptible to readily oxidized other pollutants as well as self-quenching (Lenka et al. 2021; Ross et al. 2018).

3.3.3.2 Thermal Treatment

Incineration is usually used to destroy organic compounds, especially hazardous contaminants. Due to high stability of C–F bond, the PFASs have been used to produce aqueous film-forming foam (AFFF) and non-stick cookware. Incineration process to handle PFASs, therefore, must be capable of destruction of the C–F bond. To convert PFASs into HF and non-fluorinated compounds, thermal treatment is required at high temperatures ($> 700\text{ }^{\circ}\text{C}$) for an extended length of time (Khan et al. 2020). Moreover, the greenhouse gases (GHGs), such as hexafluoroethane and tetrafluoromethane, may be formed during the incineration treatment of PFOS (Ahmed et al. 2020). High-temperature thermal treatment, on the other hand, consumes a range of fossil energy. Therefore, lowering the temperature of PFAS thermal treatment is a potential way. Wu et al. (2019) promised a novel thermal treatment technology for achieving rapid and complete destruction of PFOS via alkaline hydrothermal reaction under temperature with $200\text{--}350\text{ }^{\circ}\text{C}$ and pressures with $2\text{--}16.5\text{ MPa}$. They found that complete conversion of C–F bonds to F^{-} within 40 min for the hydrothermal reaction with 1 M NaOH . However, the intermediates and by-products may lead to secondly environmental risk. Generally, the thermal processes have yet to be proven at scale, where inefficiencies might impair performance. In addition, the incompletely destroyed of PFASs may form a potential source of air pollution.

3.3.3.3 Electrochemical Reaction

Electrochemical oxidation is the electrochemical approaches that have been proven to be effective in PFASs removal from water (Zhuo et al. 2020; Gomez-Ruiz et al. 2017). The direct oxidation of the anode or/and the indirect oxidation of active radicals are the mechanisms of electrochemical oxidation to PFASs removal (Niu et al. 2016). The removal efficiency is substantially determined by the anode material's electron transfer capacity, $\cdot\text{OH}$ -generating ability, and oxygen evolution potential (OEP) (Li et al. 2022). PFOA and PFOS may be successfully oxidized in an electrochemical manner by boron-doped diamond (BDD) anodes, with direct anodic oxidation being the predominant mechanism (Zhuo et al. 2020). However, these studies were carried out in synthetic supporting electrolyte solutions with high or low conductivity, but high initial PFASs concentrations. Nevertheless, concentrations of PFASs are relatively low and rarely exceed $1\ \mu\text{g/L}$ (Table 3.1).

Obviously, the treatment of PFASs from water using a combination of electrochemical oxidation and advanced oxidation is more effective (Wang et al. 2020a; Zhou et al. 2021). The electrode materials, on the other hand, remain a source of worry due to their high cost, limited activity, and short service life. In addition, all of the investigations relied on a reasonably pure reaction system (carried out with deionized water spiked with chosen PFASs) and were still in the theoretical/lab experimental stage.

3.3.3.4 Biodegradation

The most reliable and cost-effectiveness approach for wastewater treatment is bioremediation process. However, WWTPs are an important point source of PFASs. The fluorine-saturated carbon chain element structure of PFASs impeded the oxidative degradation of microorganisms (Lenka et al. 2021). Therefore, it is difficult for microorganisms to use such substances as carbon sources and energy sources.

PFASs precursors have carbon-hydrogen (C-H) and carbon-oxygen (C-O) bonds, as well as carbon-nitrogen (C-N) bonds in their molecular structure, which may convert precursor organics to PFAAs via microbial reactions (Yin et al. 2019, 2018). However, according to several studies (Huang and Jaffé 2019; Yu et al. 2020, 2022), PFASs are difficult to entirely decompose by a single biodegradation pathway.

Although a few studies reported some special microbial degradation of PFOA/PFOS, these studies were almost all long-term microbial degradation under special pure bacterial culture and sensitive to environmental changes as well as no complete mineralization (Zhang et al. 2022; Huang and Jaffé 2019; Ruiz-Urigüen et al. 2022). For example, *Acidimicrobium* sp. strain A6 is selected and employed for PFASs biodegradation with the average 77% decrease in PFOA concentration under 18 days of operation (Ruiz-Urigüen et al. 2022). Obviously, more work is highly desirable with the aim to improve the microbial biodegradation ability of PFASs.

Overall, most degradation technologies' energy costs constrain their long-term sustainability and acceptability, while the formation of hazardous by-products (e.g.,

short-chain PFAAs, bromate, and perchlorate) remains a problem. Each single treatment technique has its advantage and disadvantage (Table 3.3), so combination of different treatment techniques is often the best solution for control PFASs from water.

3.4 Potential Approach of CW-MFC

Many POPs and CECs, such as antibiotics, pharmaceutical and personal care products (PPCP), as well as insecticides, could be effectively removed in CW systems (Vymazal and Březinová 2015; Brunhoferova et al. 2021; Liu et al. 2019). Substrate serves as an adsorbent, a media for wetland plant growth, and a carrier for biofilm formation, among other components to play a key role in CW system. Various wetland substrates (GAC, zeolite, and biochar) are also used for the adsorption removal of PFASs from water (Du et al. 2014). With enough adsorbent amount and adsorption capability in the wetland substrate, both conventional pollutants and trace PFASs should be removed concurrently. There is no doubt that the use of appropriate adsorbents in the construction of CW filler beds can provide long-term effective adsorption of long-chain and/or short-chain PFASs on the field scale.

Plants are another key component of wetlands. They can uptake and accumulate PFASs in a variety of environments and applications, including laboratory experiments (Knight et al. 2021; Krippner et al. 2015; Zhang and Liang 2020; Zhou et al. 2019) and field surveys (Yamazaki et al. 2019). Because of worries about PFASs traveling via animals or crops and into humans, most previous research focused on irrigated crops in PFASs-contaminated soil or water (Zhou et al. 2019; Brown et al. 2020; Miranda et al. 2021). Several studies have found that wetland plants have potential ability for accumulating PFASs, with significant levels of PFASs found in plant tissues such as root, straw, and grains (Wang et al. 2020b; Yin et al. 2017). Short-chain PFCAs have a higher bioaccumulation potential in plant leaves than long-chain PFCAs with a low octanol/water partition coefficient (K_{ow}). For long-chain PFCAs, however, root concentration factors (RCF) rose with chain length (Lesmeister et al. 2021; Mei et al. 2021). Because of their hydrophilicity and mobility, short-chain PFASs are more likely to be uptaken and bioaccumulated in plant above-ground components. More importantly, it may be anticipated that by rationally structuring wetland substrates and plants, various components of CW could work together to remove various kinds of PFASs. Wetland plants for PFASs removal must be carefully chosen since they are a significant aspect of the biological habitat. To avoid being eaten by other organisms (animals, insects), it is advised to choose fern plants. It should be highlighted that when PFASs have been removed, disposing and regenerating the plants and substrate used in CW remain a significant challenge. The use of destructive treatment technique to achieve mineralization of PFASs is a viable option.

On the other hand, although biological approaches to the treatment of PFASs are extremely limited and not currently considered feasible, CW-MFC system provides a potential bioremediation approach. Due to unique electroactive bacteria around

Table 3.3 Advantages and disadvantages of various existing technologies for remove PFASs from water (Ji et al. 2020)

Technology	Application media	Scale	Advantages	Disadvantages
Adsorption	Wastewater Surface water Drinking water	Full-scale	<ul style="list-style-type: none"> • Easy to operate • Tailor-made of adsorbents • Good removal of several long-chain PFASs 	<ul style="list-style-type: none"> • Poor in clearing short-chain PFASs • Unable to remove trace level of PFASs • Difficulty regeneration • Impact with other pollutants
Membrane filtration	Wastewater Surface water Drinking water	Field pilot	<ul style="list-style-type: none"> • Good performance • Wide range of application scenarios 	<ul style="list-style-type: none"> • High-operating costs • Based on the PFASs' molecular weight • High concentrations of residues
Advanced oxidation & reduction	Wastewater Surface water	Field pilot	<ul style="list-style-type: none"> • Able to mineralize PFASs • Quicker reaction capability 	<ul style="list-style-type: none"> • Require extreme operating conditions, centralized equipment, and massive chemical • Poor in treating some PFASs • Forming intermediate product • Impact with other pollutants
Thermal treatment	Wastewater	Lab-scale	<ul style="list-style-type: none"> • Able to mineralize PFASs 	<ul style="list-style-type: none"> • Required at high temperatures • May form a potential source of air pollution
Electrochemical	Wastewater Surface water Drinking water	Field pilot	<ul style="list-style-type: none"> • Able to mineralize PFASs • Could be used in combination with other treatment methods 	<ul style="list-style-type: none"> • High-energy • High-operating costs • Poor in treating some PFASs • Forming intermediate product
Bioremediation	Surface water	Lab-scale	<ul style="list-style-type: none"> • Cost-efficiency • Green solution 	<ul style="list-style-type: none"> • Few evidences could completely degrade PFASs • Requires specific environmental

anode chamber, CW-MFC has been investigated in recent years to remove a variety of biorefractory organic contaminants. CW-MFC could effectively treat wastewater with high antibiotic and PPCPs (Wang et al. 2020c; Zhang et al. 2017a, b). Biodegradation, substrate adsorption, and plant uptake were found to have distinct roles in removing pollutants in these studies, notably biodegradation, and plant uptake. It has been demonstrated that a bioelectrochemical system not only creates a battery circuit that can promote microbial metabolism but also selectively enriches electrochemically active microbial communities for refractory organics (Li et al. 2019b; Wen et al. 2022; Xu et al. 2019).

The accumulation of PFASs in the substrate with long hydraulic retention time, and bioelectrochemical stimulation in CW-MFC offers a favored condition for incubation and/or domestication PFASs-degrading microorganisms. Moreover, there are few studies, showing that the special microbial can degrade of PFOA in lab experiment (Sect. 3.3.4). It seems that there is a condition and pathway for PFOA biodegradation or biotransformation due to the abundant microbial population and suitable microenvironment in CW-MFC system. However, further studies are highly desirable.

Based on the above clues, using CW-MFC to removal PFASs in water was proposed by Ji et al. (2020) (Fig. 3.2). Subsequently, related experiments were carried out, while the results showed that when the concentrations of PFOA and PFOS in the influent were $6.46 \pm 0.52 \mu\text{g/L}$ and $9.34 \pm 0.87 \mu\text{g/L}$, respectively, both closed-circuit and open-circuit operations of the CW-MFC systems demonstrated over 96% removal performance of PFASs (Ji 2022). These results indicate that the CW-MFC system can effectively remove PFASs from wastewater.

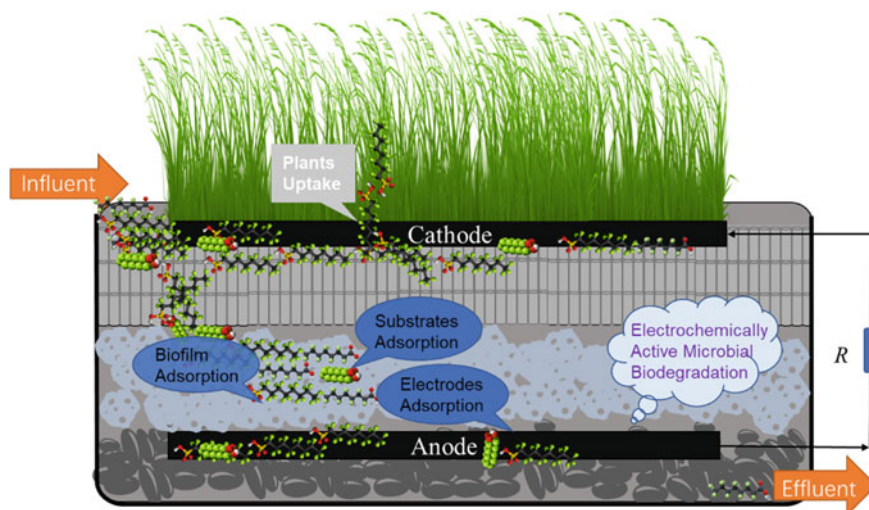


Fig. 3.2 Principle of PFASs removal in proposed CW-MFC system

3.5 Conclusion

The current review provides a brief overview of the occurrence and properties of PFASs in water environment, with an emphasis on problems and feasible removal solutions based on published literature. Some physical, chemical, and limited biodegradation methods have been documented to degrade or transfer PFASs to some extent. However, these methods are insufficiently thorough and not environmentally friendly. Given the fact that PFASs are very stable compounds with distinct physicochemical features, the integration approach or a combined system may be a viable option for PFASs control. CW-MFC system could provide an integrated and environmentally friendly while sustainable way to remove PFASs from water through substrate adsorption, plant uptake, and enhanced biodegradation.

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