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Review

The Occurrence and Bioremediation of Emerging Polyfluorinated Compounds in Water Bodies: A Mini Review

Siphesihle Mangena Khumalo [1,2,*], Babatunde Femi Bakare [2] and Sudesh Rathilal [1]

[1] Green Engineering Research Group, Department of Chemical Engineering, Faculty of Engineering and the Built Environment, Durban University of Technology, Steve Campus, S3 L3, P.O. Box 1334, Durban 4000, South Africa
[2] Environmental Pollution and Remediation Research Group, Department of Chemical Engineering, Faculty of Engineering, Mangosuthu University of Technology, Jacobs, P.O. Box 12363, Durban 4026, South Africa
* Correspondence: khumalo.sm@outlook.com

Abstract: The occurrence and fate of polyfluorinated compounds (PFCs) in the aquatic environment resulting from anthropogenic activities has become an emerging issue of environmental chemistry. PFCs have been detected in drinking water samples, aquatic life, human tissue, and blood serum. This is attributed to their xenobiotic attributes making them environmentally persistent, bio-accumulative, and globally distributed in water receiving bodies, posing serious health problems to aquatic life and human health. This is ascribed to PFCs' peculiar physicochemical properties of being hydrophobic and oleophobic and their removal process from wastewater streams is different from any other organic pollutants. Therefore, this review summarizes the environmental occurrence and recent developments on microbial degradation of the most detected PFCs, i.e., perfluorooctanoic acid (PFOA), and perfluorooctane sulfonic acid (PFOS) in water bodies. The available literature suggests that PFOA and PFOS are susceptible to biodegradation by *Acidimicrobium* sp. strain A6, *Pseudomonas parafulva* strain YAB1, *Pseudomonas plecoglossicidia* 2.4-D, and *Pseudomonas aeruginosa* strain HJ4. Moreover, the current study presents a summary on phytoremediation of PFOA and PFOS as a sustainable green technology. Despite the extensive work undertaken on bioremediation of PFOA and PFOS by biological processes, the available literature suggests that a lot of work still needs to be carried out aimed at investigating the biodegradation pathway of PFOA and PFOS by both microbial species and plants.

Keywords: polyfluorinated compounds; perfluorooctanoic acid; perfluorooctane sulfonic acid; phytoremediation; microbial species

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1. Introduction

Polyfluorinated compounds (PFCs) are a class of anthropogenic emerging persistent organic pollutants that consist of a fully fluorinated hydrophobic alkyl chain attached to a hydrophilic chain end group. The PFCs' carbon–fluorine bond, characterized by strong polarity and strength [1], make them possess peculiar physicochemical properties such as hydrophobicity and resistance to degradation by heat and acid [2–4]. Owing to their hydrophobic and lipophobic moieties, PFCs are friction-resistant and are used as water and oil repellents [3]. Due to their peculiar chemical attributes, PFCs have cemented their application in consumer products, such as stain-resistant household products, non-sticky cooking utensils, food packaging, cosmetics, personal care products, etc., and industrial applications, such as surface coatings for textiles, metal plating, manufacturing of leather, etc., [1,3–7]. The primary purpose of using PFC-treated consumer products as well as industrial products is to improve their durability [3] whilst prolonging their exposure to humans and the environment. Hence, there has been an increase in levels of PFCs detected in aquatic environments [8–10], posing serious health problems to aquatic life and humans.

Studies [3,5,11–13] have indicated that PFCs have been detected ubiquitously in multiple environmental spheres, such as air, water bodies, and soil. Moreover, these contaminants of emerging concern have been detected in food sources, such as livestock and crustaceans [5]. Since PFCs are ubiquitous to aquatic matrices, including drinking water, humans can be exposed to them through drinking water as well as dietary intake [3,4]. Several works have been reported on human health effects as a result of exposure to PFCs. Xie et al. [13] reported on the nexus between PFCs and human thyroid dysfunction and concluded that PFCs are endocrine-disrupting compounds. Human exposure to PFCs results in thyroid malfunction and/or hormonal imbalance, particularly in pregnant women and neonates [13]. Health effects associated with PFC exposure include cardiovascular diseases, urine acid, thyroid diseases, pediatric atopy, immune toxicity, anxiety, liver damage, kidney disorder, obesity, peroxisome, reduced fertility, fecundity, etc., [3,5,10,13–15]. Because of the aforementioned human health complications, the production and regulations of PFCs have attracted public attention. According to Jian et al. [14], the 3M company, together with the United States Environmental Protection Agency (USEPA), announced the phase-out of products containing PFCs with C6 and more, aimed at eliminating long-chain PFCs which have attributes of being very persistent and bioaccumulative.

Hitherto, studies are still conducted aimed at understanding the environmental occurrence and fate, as well as human health complications of PFCs. This is attributed to the thousands of PFCs that have been identified and are still being manufactured and used globally [3,12]. It should be noted that, despite the wide pool of PFCs that have been detected in the environment, global environmental entities are interested in long chain perfluoroalkyl sulfonic acids ($C_nF_{2n+1}SO_3H$, $n \geq 6$) (PFSAs) and perfluoroalkyl carboxylic acids ($C_nF_{2n+1}COOH$, $n \geq 7$) (PFCAs) and their corresponding anions. The aforementioned PFCs have demonstrated to be more bioacccumulative compared to their short chain analogues reported by Rahman et al. [4] and Buck et al. [16]. Perfluorooctanoic acid (PFOA) and perfluorooctane sulfonic acid (PFOS) are conventional PFCs and the most detected perfluoroalkyl acids (PFAAs) in the environment [17,18]. The high levels of PFOA and PFOS in the environment are attributed to their xenobiotic characteristics, as reported by many researchers, which makes their complete eradication almost impossible. Therefore, the current study aims at providing an overview on the occurrence of PFCs in water bodies, as well as the latest developments in biological remediation of PFOA and PFOS, since they are characterized as xenobiotic compounds. The paper aims at contributing to the design of sustainable green PFCs bioremediation processes.

2. Occurrence of PFCs in Water Bodies

PFCs are introduced into the environment during their industrial production and application, as well as a result of leaching and their biotic or abiotic biodegradation of consumer products containing a perfluoroalkyl moiety [4,16]. Moreover, the discharge of industrial effluent containing PFCs into the environment contributes significantly to the occurrence of PFCs in both surface and ground water. Consequently, these PFCs enter waste water treatment plants (WWTPs), making them the major source of PFCs to surface waters, as reported by Rahman et al. [4]. Generally, WWTPs have demonstrated to be ineffective in the removal of PFCs from aqueous streams. This is attributed to the C–F bond becoming more stable with an increasing replacement of hydrogen by fluorine at each bond, consequently making PFCs able to withstand any degradation process (i.e., photolytic and microbial) [4]. Recently performed studies [10,14,19] have indicated that drinking water is the major exposure pathway to PFCs in communities with contaminated water. Note that the degradation of fluorotelomer alcohols and perfluoroalkyl sulfonamides lead to the formation of PFAAs, consequently resulting in their occurrence in water bodies. As such, they are classified as PFAAs precursors [4]. Moreover, the persistence of PFAAs in aquatic ecosystems is ascribed to their chemical attributes, such as simultaneous hydrophobic–hydrophilic properties and low volatility [16,20].

In their reviews of the occurrence and fate of PFCs in drinking water, Rahman et al. [4] and Jian et al. [14] reported that, apart from PFOA and PFOS, there are other frequently detected PFCs, such as perfluorooctane sulfonamide, perfluorohexane sulfonic acid, perfluorobutanoic acid, perfluorohexanoic acid, perfluoroheptanoic acid, perfluorodecanoic acid, and perfluorononanoic acid. Despite the occurrence of the aforementioned PFCs in aquatic ecosystems, PFOA and PFOS have been detected frequently, recording the highest concentration ranging from µg/L to mg/L compared to any other PFCs reported with concentration levels ranging from pg/L to ng/L. Studies [4,21] suggest that, due to the stringent regulations on PFOA and PFOS, there are other emerging fluorinated organics, such as polyfluoroalkyl phosphates, perfluorinated phosphinic acids, and perfluorinated phosphonic acids that have been detected in water bodies. However, the aforementioned phosphorus-containing fluorinated compounds are not explicitly accounted for in this current work.

It is imperative to note that the concentration levels of PFCs in water bodies can be influenced by the location of the PFC point source from the water bodies. Studies conducted on the global distribution of PFCs have shown that higher levels of PFCs, particularly PFOA and PFOS, have been detected in industrialized areas [4,20,22]. Consequently, PFCs have been detected in treated water at an average concentration of 30 ng/L [4]. However, there has been a large variability in PFC concentration levels from tap water, as shown in Table 1. Eschauzier et al. [23] reported PFOA and PFOS concentrations ranging from 0.6 to 6.6 ng/L and 0.1 to 11 ng/L, respectively, for background contaminated tap water. On the other hand, Eschauzier et al. [24] reported PFOA and PFOS concentration ranges of 22 to 519 ng/L and 3 to 22 ng/L, respectively, for highly industrialized, and/or agricultural areas. As a matter of health concern, the USEPA issued a limit for PFOA and PFOS in drinking water of 70 ng/L, both individually and combined [10,25]. Based on the reported PFOA and PFOS concentration levels present in Table 1, it is apparent that PFOA and PFOS in drinking water is below the 70 ng/L limit, at least for many regions apart from Africa. The high PFOA and PFOS concentrations in drinking water from the African region suggests that drinking water can cause imminent human health risk due to PFC contamination. Due to the occurrence of PFCs in tap water, Eschauzier et al. [23] reported that PFOA has been detected in tap water-based beverages (e.g., cola and coffee) at a concentration of 4 ng/L. The lower PFC concentration from tap water beverages is attributed to the pre-treatment of tap water prior to being used for production. Moreover, a review study by Rahman et al. [4] has indicated that high PFOA concentration levels ranging from 487–10,100 ng/L have been detected in aquatic environments near point sources.

Table 1. Concentrations of PFOA and PFOS detected in tap water.

Region	Country	PFOA, ng/L	PFOS, ng/L	Reference
Africa	Ghana	68.1–190	62.1–168.3	[26]
Oceania	Australia	0–16	0–9.7	[27]
America	Canada	3–32	2–12	[28]
	Brazil	0.35–2.82	0.58–6.70	[29]
	USA	5–30	1–57	[27]
Asia	China	115–151	0.1–11.2	[30]
	Japan	6.5–48	1.3–3.7	[29]
Europe	France	8.7–18	3–11.99	[31]
	Germany	0–6.1	0–4.7	[32]
	The Netherlands	0.6–6.6 <0.3–11	0.1–11 <0.03–0.41	[23] [33]
	Sweden	0.302–8.56	0.397–8.81	[31]
	Spain	3.8–29	2.4–140	[31]

Early studies [24] on the global distribution of PFCs in aquatic environments suggest that water-receiving bodies located in low population density areas have low levels of PFCs. This can be attributed to fewer industrial activities. It is worth noting that PFC toxicity values vary significantly from one standard to another, as indicate in Table 2. The variability in toxicity values can be attributed to the lack of federal maximum contaminant level.

Table 2. PFOA and PFOS drinking water guideline levels.

Regulatory Body	PFC	Advisory Level, ng/L	Toxicological Endpoint	Reference
U.S. EPA, 2016, Health Advisory Level	PFOA	70	Developmental	[34]
	PFOS	70	Reduced pup body weight	
Alaska DEC [1], 2016, ground water clean-up level	PFOA	400	Developmental	
	PFOS	400	Reduced pup body weight	
Maine DEP [2], 2016, Remedial action guideline	PFOA	130	Liver	
	PFOS	560	Thyroid effects	
New Jersey DEP, 2017, Maximum contaminant level	PFOA	14	Liver	
	PFOS	13	Immune response	
German Drinking Water Commission	PFOA	1500	N/A	[4]
	PFOS	600	N/A	
Drinking water inspectorate, UK	PFOA	>300	N/A	
	PFOS	>300	N/A	

[1] DEC, Department of Environmental Conservation; [2] DEP, Department of Environmental Protection.

In the subsequent sections, the recent developments on biological remediation of PFCs are summarized, despite being characterized as xenobiotic compounds.

3. Degradation and Fate of PFCs in the Environment

3.1. Bacterial Biodegradation

Conventional WWTPs use biological techniques to treat water and wastewater streams for reuse. During biological treatment of aqueous streams, biodegradable material consisting of organics in dissolved form, such as starches, alcohols, acids, aldehydes, and esters, are utilized for food by microorganisms, thus decontaminating aqueous streams. It is worth noting that microbial utilization of dissolved organics can be accompanied by oxidation, which is the deletion of hydrogen from elements of an organic molecule, or by reduction, which is the addition of hydrogen to elements of the organic molecule [35]. However, a number of previously performed studies suggests that the biodegradation pathways and fate of PFCs are unknown to a large extent. This is attributed to the general belief that the strength of the C–F bond is the limiting factor on PFC biodegradability. However, early evidence suggests that organohalogen compounds, including PFCs, are susceptible to microbial degradation, despite their high stability, as reported by Parsons et al. [36]. The biodegradation of organohalogen compounds and PFCs are relatively slow due to their kinetic stability because of the carbon–halogen bond, thus contributing significantly to their long-term environmental fate.

Hitherto, dehalogenation is the thermodynamically recognized reaction mechanism for the degradation of organohalogen compounds. Early studies [36,37] suggest that under aerobic conditions, organohalogen compounds are degraded by catabolic pathways similar to those for nonhalogenated analogues. There are a number of dehalogenation mechanisms that are reported in the literature for both aliphathic and aromatic substrates, i.e., haloacid dehalogenation, halohydrin dehalogenation, haloalkane dehalogenation, and reductive dehalogenation [36]. However, there is very little information reported on the degradation of PFCs, particularly PFOA and PFOS. This is attributed to the biodegradation mechanism

of 8:2 fluorotelomer alcohol by OH radicals, which yields PFOA and other perfluorinated carboxylic acids as products [36]. Fluorotelomer alcohols have been reported to biodegrade under aerobic and anaerobic environments [36]. In an early study conducted by Meesters and Schröder [38], it was demonstrated that both PFOA and PFOS are xenobiotic under aerobic environments. Despite the observed removal of PFOA and PFOS under anaerobic environments, no fluoride ion concentration was detected, which results in the biodegradability mechanism of PFOA and PFOS in aquatic environments remaining difficult to understand for many researchers. In the subsequent section, the biodegradation of PFOA and PFOS are explained, indicating that their xenobiotic characteristics are something of the past.

Despite the chemically challenging C–F bond cleavage, recently performed studies have demonstrated that PFCs, particularly PFOA and PFOS, can be defluorinated by strong chemical oxidants, as well as by microbial population [39–41]. Many researchers have reported limited microbial defluorination of PFCs. This is attributed to the displacement of C–F bonds, which yield a fluoride ion, F^-, that is toxic to microbial population at minute concentrations [42]. The fluoride ion, which is produced as a result of PFCs' biodegradation (i.e., defluorination), enters bacterial cells, thus poisoning them [41,42]. The poisoning of bacterial cells ceases or hinders metabolic activities, resulting in the nonbiodegradability of PFCs. Such a biological reaction mechanism explains why fluorine concentrations are not detected during biodegradation of PFCs. The revelation of the toxicity of the fluoride ion to bacterial cells explains why PFCs are characterized as xenobiotic compounds. Moreover, the biodegradation mechanism of PFCs suggests that their xenobiotic characteristics are not solely a result of the C–F bond, but it can be attributed to the toxicity of the fluoride ion which hinders microbial activities.

Recently, Huang and Jaffé [40] demonstrated that PFOA and PFOS are susceptible to biodegradation by *Acidimicrobium* sp. strain A6 (an autotroph) that oxidizes ammonium to nitrite while reducing ferric iron. Both PFOA and PFOS were added as carbon sources during the oxidation of ammonium to nitrite, while reducing ferric iron by *Acidimicrobium* in an anaerobic environment. It was reported that both PFOS and PFOA were metabolized recording a removal efficiency of up to 60% for both PFOS and PFOA over an incubation period of 100 days. Despite the observed biodegradation of PFOS and PFOA, it was reported that intermediate products, such as perfluorobutanoic acid, perfluoroheptanoic acid, perfluoropentanoic acid, and perfluorohexanoic acid, were detected in *Acidimicrobium* culture. The aforementioned intermediate products were detected for PFOA and PFOS initial concentrations of 100 mg/L. It is also worth mentioning that the initial concentration of 100 mg/L for the model PFCs was detrimental to the microbial population. A significant decline on *Acidimicrobium* population was observed from day 60 of incubation, as compared to an initial concentration of 0.1 mg/L. Moreover, the findings reported by Huang and Jaffé [40] suggests that PFOS is more detrimental to *Acidimicrobium* population, as compared to PFOA. Hitherto, there is no available literature reporting on PFOA and/or PFOS optimum concentration aimed at minimizing their detrimental effects on microbial population.

Ruiz-Urigüen et al. [43] investigated the biodegradation of PFOA in microbial electrolysis cells using *Acidimicrobium* sp. strain A6 as an inoculum under anaerobic conditions. It was reported that a decrease in PFOA concentration was observed after 18 days of operation, while producing electricity. For the work reported by Ruiz-Urigüen and coworkers, the biodegration of PFOA is not explicitly accounted for, on the basis that no PFOA measurements were conducted during operation but FFOA degradation intermediate products analogous to the one reported by Huang and Jaffé [40] were detected. The measurement of PFOA intermediate products is attributed to the complexities of PFOA measurements, which can be affected by sample handling and hemimicelle formation [43]. For the work reported by Ruiz-Urigüen et al. [43], 100 mg PFOA/L was the desired initial concentration; however, due to hemimicelle formation, the initial concentration measured was 47 mg PFOA/L.

Due to the recalcitrance and high stability of PFCs, particularly PFOA and PFOS, researchers have continued to test various microbial species aimed at testing their ability to biodegrade the aforementioned model contaminants. To the best of our knowledge, apart from *Acidimicrobium* sp. strain A6 [40,43] *Pseudomonas parafulva* strain YAB1 [44], *Pseudomonas aeruginosa* strain HJ4 [45], and *Pseudomonas plecoglossicida* 2.4-D [46] are the only microbial species reported to biodegrade PFOA and PFOS. Yi et al. [44] investigated the growth rate of YAB1 which was extracted from soil sediments near a PFC-producing plant. Yi et al. and co-workers conducted their investigation in an aerobic environment and PFOA was added as a carbon source for YAB1. Optimal bacteria growth rate and PFOA degradation were recorded for a PFOA concentration of 500 mg/L. Interestingly, it was reported [43] that a PFOA concentration of 1000 mg/L inhibited the growth rate of YAB1; however, the strain demonstrated considerable tolerance and growth adaptability. The findings by Yi et al. [44] suggests that higher PFOA concentrations hinder microbial activities, consequently inhibiting the bacterial proliferation. This is congruent to the work reported by Huang and Jaffé [40] and Ruiz-Urigüen et al. [43] that higher PFOA concentrations become toxic to microbial species, thus hindering microbial activities. Yi et al. [44] reported a PFOA removal efficiency of 48%, which was achieved after adding yeast to promote bacterial growth rate.

The growing interest in the investigation of various microbial species to degrade PFCs has demonstrated that PFOS can be degraded by *Pseudomonas aerginosa* strain HJ4 under an aerobic environment [45]. PFOS was added as a carbon source at an initial concentration ranging from 1400 µg/L to 1800 µg/L for the growth of strain HJ4. Kwon et al. [45] reported an overall PFOS biodegradation of 67%, which was recorded after adding glucose to enhance the growth rate of strain HJ4 for PFOS biodegradation. However, according to Kwon et al. [45], no defluorination was observed. Fluoride ion concentration remained constant during biodegradation of PFOS. On the other hand, Chetverikov et al. [46] reported 75% PFOS degradation as a carbon source for the growth of *Pseudomonas plecoglossicida* 2.4-D. A significant growth rate on the 2.4-D strain was observed after adding NaCl which was maintained at less than 5% to avoid any microbial inhibition. Chetverikov and co-workers reported a fluoride ion concentration of up to 150 mg/L in the culture medium to account for the fact that defluorination indeed took place. Table 3 presents a summary of studies conducted to date aimed at microbial degradation of PFOA and PFOS at the laboratory scale.

Table 3. Microbial degradation of PFOA and PFOS.

Pollutant	Microbial Species	Matrix Conditions	Concentration (mg/L)	System Efficacy (%)	Incubation Period (Day)	Reference
PFOA	*Acidimicrobium* sp. strain A6	Anaerobic	0.1–100	<60	100	[40]
			47	77	18	[43]
	Pseudomonas parafulva strain YAB1	Aerobic	500	48	5	[44]
PFOS	*Acidimicrobium* sp. strain A6	Anaerobic	0.1–100	<60	100	[40]
	Pseudomonas aerginosa strain HJ4	Aerobic	1.4–1.8	67	2	[45]
	Pseudomonas plecoglossicida 2.4-D	Aerobic	1000	75	6	[46]

3.2. Microbial Degradation Pathway of PFOA and PFOS

Despite the experimentally illustrated biodegradation of PFOA and PFOS by microbial species [40,43–45], there is no available literature confirming a precise biodegradation mechanism of PFOA and PFOS. In the few studies that have reported on PFC biodegradation,

particularly PFOA and PFOS, the mechanism is based on the detection of intermediate metabolites. According to Zhang et al. [47], this is attributed to PFOA and PFOS having a low redox potential of about −450 mV. Typical microbial oxidizable growth substrates demonstrate redox potentials greater than −450 mV [39,47]; therefore, at a low redox potential, the transfer of an electron from a substrate for PFC reduction will not be thermodynamically attractive for microbial species. However, as indicated in the previous sections, studies have demonstrated that selected bacteria can degrade PFOA and PFOS under anaerobic [40] and aerobic [43–45] environments. Besides the redox potential, Wackett [41] reported that, in order for a microbe to degrade a PFC, it needs to transport the fluorinated compound into the cell, harbor a recently evolved enzyme to catalyze the C–F bond cleavage, detect the poisonous fluoride ion generated, and protect itself against the fluoride with a fluoride–proton antiporter. However, the disadvantage in C–F bond reduction is that it can be selected against an evolution on the basis that it provides no benefit to the microbial species but drains cellular energy [39]. Despite the shortcomings of C–F bond reduction, selected bacteria which have been exposed to environments with fluoride concentrations exceeding 1 ppm have developed systems to protect themselves against the toxic fluoride ion. Last et al. [48] reported that bacteria can resist the toxicity of environmental fluoride by expelling the fluoride ion from its cytoplasm by the aid of a proton-coupled fluoride ion antiporter, *E. casseliflavus* being the common antiporter. The microbe attributes necessary to biodegrade PFCs suggest that bacteria that are found near PFC-producing subjects or in PFC-contaminated environments can develop antiporters and survive the toxic fluoride ion. It is worth noting that in the available literature reporting on PFOA and PFOS degradation by novel microbial species, all reported bacteria were harvested from PFCs contaminated sites, which explains why the reported bacteria can biodegrade PFOA and PFOS.

The degradation of PFOA and PFOS by novel bacteria, as reported in the literature [40,43–45], can be attributed to certain enzymes cleaving the C–F bond through either oxidation by inserting oxygen atoms or reduction by adding extra electrons to the bond [47]. The dictation of fluoride ions during microbial degradation of PFCs indicates that microbes can break the C–F bond under control environments, which can be either aerobic or anaerobic. Yu et al. [49] reported that during microbial degradation of PFCs, C–F bond cleavage occurs at the sp^2 and sp^3 C–F bonds. However, Yu and co-workers reported that, despite the tertiary sp^3 C–F bond having the lowest bond dissociation energy for the model PFCs (i.e., perfluoro (4-methylpent-2-enoic acid) and 4,5,5,5-tetrafluoro-4-(trifluoromethyl)-2-pentenoic acid), the sp^2 C–F bond was more microbially active. The production of fluoride ions [40] and intermediate products [43–45] during the biodegradation of PFOA and PFOS is an indication that the C–F bond was cleaved, despite having a bond dissociation energy of up to 130 kcal/mol [47]. There is no available literature reporting on the specific pathways for PFOA and PFOS biodegradation. It is worth noting that the extensively reported biodegradation of fluorotelomer alcohols and other PFCs, apart from PFOA and PFOS, is limited to the removal of non-fluorinated moieties rather than C–F bond cleavage.

Factors Affecting PFOA and PFOS Biodegradation

It is apparent that the microbial biodegradation of PFOA and PFOS can be affected by certain factors, such as the type of microbial species, initial concentration of model PFCs, and co-substrates. To date, *Acidimicrobium* sp. strain A6, *Pseudomonas parafulva* strain YAB1, *Pseudomonas aeruginosa* strain HJ4, and *Pseudomonas plecoglossicida* 2.4-D are the only reported microbial species in the literature that are capable of PFOA and PFOS biodegradation. This suggests that the type of microbial species plays a significant role in the biodegradation process of PFCs, which are characterized as xenobiotic during their bio-remediation process. Moreover, the available literature on the biodegradation of PFOS and PFOA have demonstrated that higher PFC concentrations, ranging between 100 mg/L and 1000 mg/L, can inhibit microbial growth rate depending on the microbe strain, thus compromising PFC degradation. Moreover, selected microbes use PFOA and PFOS as

carbon sources and simultaneously perform bio-defluorination [40,47]. The presence of co-substrates can enhance microbial growth rate due to microbial preference [40,43,45].

3.3. Other Biological Remediation Processes of PFOA and PFOS

Previously conducted studies have reported on a number of effective remedial technologies of PFCs from aquatic environments, such as photocatalysis and electrochemical oxidation [50–53], plasma technology [54,55], and adsorption by granular-activated carbon [15]. However, the aforementioned treatment technologies are energy-intensive, consequently resulting in high operational costs [56]. Hence, this section of the paper focuses on PFOA and PFOS remediation by phytoremediation, characterized as an environmentally green and cost-effective process.

Phytoremediation of PFOA and PFOS

The removal of organic pollutants by plants is known as phytoremediation. The mechanism of the phytoremediation process consists of (1) phytofiltration, which is essentially the uptake of pollutants by the plant; (2) phytovolatilization, which is the conversion of pollutants to volatile form; and (3) phytodegradation, which involves the participation of root exudes and microbial population [56]. Early studies [57,58] have demonstrated that plants can absorb PFCs, where long-chain PFCs are likely to be accumulated in roots, and short chains are more likely to be accumulated in buds, fruits, and crops [15]. Gobelius et al. [57] investigated the uptake of PFCs by various plant species, and it was reported that high concentrations of short-chain PFCs were detected in leaves, with PFOA and PFOS being detected in roots. It is worth mentioning that Gobelius et al. [57] did not report on PFOA and PFOS phytovolatilization and phytodegradation. The findings suggest that only phytofiltration was observed for PFOA and PFOS phytoremediation mechanisms.

Colomer-Vidal et al. [58] investigated the sorption of PFCs in the water-sediment-plant system along the Dongzhulong and Xiaoqing rivers by floating and rooted plant species. The average PFC concentration in water was reported to be 84,000 ng/L and 2300 ng/L in sediments, respectively, with PFOA accounting for up to 97% in both water and sediment. It was reported that PFOA demonstrated high sorption affinity with floating plant species absorbing a higher concentration of PFCs as compared to rooted plants. The high uptake capacity by floating plants is attributed to large proliferation rates. Higher PFOA concentrations were detected in the roots of plant species for both floating and rooted plant species, suggesting that phytofiltration took place. The PFCs distribution analysis within the model plant species for the work reported by Colomer-Vidal et al. [58] demonstrated that short-chain PFCs were concentrated more in the shoots than in the roots, congruent to the work reported by Gobelius et al. [57]. Short-chain PFC uptake is attributed to the water potential gradient resulting from plant transpiration, consequently promoting the upward uptake of PFCs through the xylem [58]. On the other hand, the accumulation of long-chain PFCs in plant roots can be attributed to the proteinphilic-linked sorption. Previously undertaken studies on the phytoremediation of PFOA and PFOS suggest that long-chain PFCs are bound strongly to the roots' surface due to a positive correlation with protein content as opposed to short-chain PFCs [58,59].

The phytoremediation of PFOA and PFOS is strongly dependent on plant species. Wen et al. [59] investigated the role of proteins and lipids in the accumulation of PFOA and PFOS by different plant species, i.e., alfalfa, lettuce, maize, mung bean, radish, ryegrass, and soyabeans. It was reported that the concentration of PFOA and PFOS in the roots of the aforementioned model plant species did not follow the same trend, with ryegrass, maize, and radish roots recording the lowest PFOA and PFOS concentrations. The observed behavior was attributed to the low protein content in the roots of ryegrass, maize, and radish, as reported by Wen et al. [59]. Moreover, it was reported that the binding of PFOS to roots was stronger than that of PFOA; therefore, the transfer potential of PFOA from roots to shoots is more favorable compared to PFOS [59]. The observed behavior could

be attributed to the relatively large molecular structure and lipophilicity of PFOS when compared to PFOA.

Based on the available literature, it is evident that PFCs can be eradicated from the environment by phytoremediation. However, the detection of PFCs in plant leaves suggests that nonedible plant species should be considered for phytoremediation of PFCs to avoid human exposure to PFCs. Moreover, the reported literature on phytoremediation focuses on the translocation of PFCs in plant species. There is no available literature reporting on the degradation of PFCs in plant tissues.

4. Conclusions and Future Perspectives

Recently undertaken studies [40,43–45] have demonstrated that PFCs, particularly PFOA and PFOS, are susceptible to degradation by certain microbial species. The metabolism mechanism of PFOA and PFOS follows a peculiar path relative to the degradation of fluorotelomer alcohols and organohalogens. The available literature [40,43–45] suggests that certain microbial species (i.e., *Acidimicrobium* sp. strain A6, *Pseudomonas parafulva* strain YAB1, *Pseudomonas plecoglossicidia* 2.4-D, and *Pseudomonas aeruginosa* strain HJ4), which can develop antiporters to protect themselves from the toxic fluoride ions, can biodegrade PFOA and PFOS. Despite the novel developments on PFOA and PFOS degradation by microbial species, further investigation needs to be carried out aimed at optimising PFOA and PFOS microbial degradation. Based on the reported literature, it is apparent that certain bacteria can use PFOA and PFOS as carbon sources; however, high concentrations of the aforementioned compounds can hinder the microbial growth rate, thus compromising the treatment efficiency. Such observations warrant further investigation aimed at optimizing the microbial growth rate under high PFOA and PFOS concentration levels. Moreover, further investigation needs to be performed on the co-substrate dose for different microbial species in enhancing microbial growth rate. The available literature suggests that there is lack of cellular benefits from PFC biodegradation; hence, microbial species require a co-substrate to facilitate their growth for effective PFC removal.

Moreover, the current study has demonstrated that it is apparent that phytoremediation of PFOA and PFOS is one of the promising sustainable green technologies which can be utilized in eradicating PFCs in water bodies. Further investigation needs to be conducted on the biotransformation of PFOA and PFOS in plant tissues. Currently, there is very little information reported on the degradation of PFOA and PFOS in plant tissues. In conclusion, the reported PFOA and PFOS removal by microbial species and phytoremediation is associated with a high retention time, which makes it difficult for PFOA and PFOS remediation in receiving water bodies. Genetically engineered microbes need to be considered in order to eradicate the occurrence of PFCs in water bodies.

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