

Review

Carbon–Fluorine Bond Cleavage in PFAS Remediation: Integrating Biological and Emerging Advanced Treatment Technologies

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Abstract

Per- and polyfluoroalkyl substances (PFASs) are persistent environmental contaminants widely recognized as “forever chemicals” due to the exceptional stability of their carbon–fluorine (C–F) bonds. Their widespread occurrence in water, soil, sediments, and biota, coupled with increasing evidence of toxicity, bioaccumulation, and long-range transport, has intensified the search for sustainable remediation strategies. Conventional remediation technologies for PFASs have been widely applied but remain constrained by several technical and environmental limitations, such as incomplete mineralization, high energy requirements, secondary waste generation, and the formation of toxic transformation products. Moreover, many conventional treatment processes were not originally designed to handle the chemical stability and structural diversity of PFASs, resulting in variable removal efficiencies across different compounds. Bio-based strategies for PFAS remediation, particularly those targeting C–F bond cleavage and biological defluorination, are gaining attention due to the unique challenges posed by the chemical stability and environmental persistence of these “forever chemicals”. This review addresses the fragmented nature of PFAS remediation research by integrating biological and physicochemical strategies and critically examining mechanisms of C–F bond cleavage and defluorination. Emerging technologies, including bioelectrochemical systems, photocatalytic and electrochemical defluorination, adsorption-assisted degradation, plasma treatment, hydrothermal processes, and synthetic biology approaches, are evaluated in relation to their degradation efficiencies, defluorination capacities, and applicability in diverse environmental matrices. Particular attention is given to integrated “capture-and-destroy” systems that combine adsorption with catalytic or biological degradation to enhance remediation efficiency and reduce energy demand. PFAS treatment performance varies markedly across scalability, destruction, and cost. Key knowledge gaps and future perspectives are outlined, emphasizing the need for scalable, energy-efficient, and environmentally sustainable remediation technologies capable of achieving complete PFAS mineralization in complex environmental systems.



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1. Introduction

Per- and polyfluoroalkyl substances (PFASs) have emerged as contaminants of global concern due to their widespread production, environmental persistence, and increasing

detection across diverse environmental compartments worldwide [1–3]. They are now frequently detected in surface waters [4,5], groundwater [6], soils [7], sediments [8], and atmospheric particles [9] across both urban and remote regions. Studies have reported PFAS contamination in major river systems [10], drinking water supplies [11], agricultural soils [7], and the marine environment [12] worldwide, highlighting the pervasive nature of these substances which endanger food security [13]. Alarmingly, PFASs have also been identified in wildlife [14] and human biomonitoring studies, including blood, breast milk, and tissues [15–17], indicating their ability to bioaccumulate and biomagnify through food webs. Major PFAS contamination hotspots are often linked to historical and ongoing releases from industrial manufacturing facilities, airports and military bases where aqueous film-forming foams (AFFFs) have been heavily used, wastewater treatment plants (WWTPs) discharging PFAS-laden effluent, and landfills generating high-PFAS leachates that can migrate into surrounding environments [18–20]. This is shown in Figure 1.

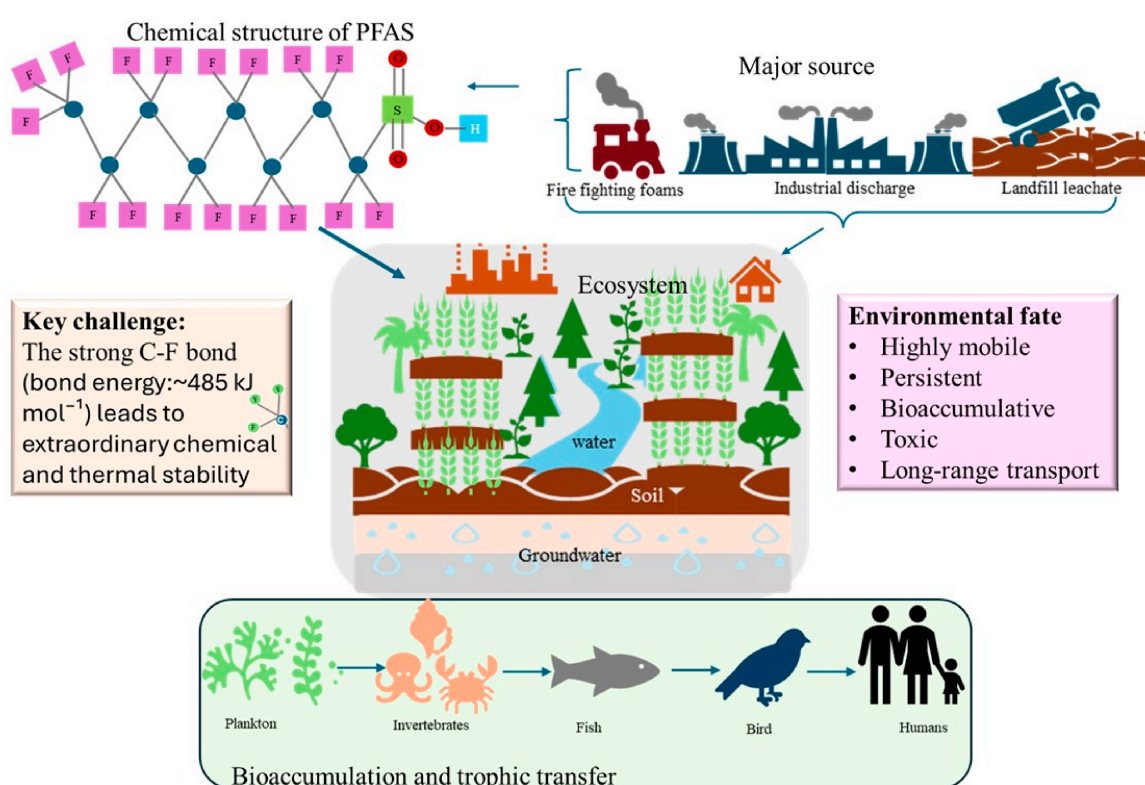


Figure 1. PFAS persistence and environmental pathways. Note: Arrows indicate contaminant transfer through the aquatic food web.

However, long-range atmospheric transport and hydrological processes have facilitated the spread of PFASs far beyond their original sources, resulting in their presence even in remote environments such as the Arctic and Antarctic regions [21–23]. Their long-term environmental persistence, contributed by C-F, has been linked to multiple toxic effects in humans and ecosystems. Exposure has been associated with liver dysfunction, immune suppression, endocrine disruption, developmental toxicity, immunotoxicity, and increased cancer risk [24–26]. Recent global monitoring efforts suggest that thousands of PFAS compounds may be present in the environment, many of which remain poorly characterized in terms of occurrence, toxicity, and environmental behaviour [27,28]. This widespread distribution, combined with their resistance to degradation and potential risks to ecosystems and human health, has led to PFASs being widely referred to as “forever chemicals” [29,30].

Conventional remediation technologies for PFASs have been widely applied but remain constrained by several technical and environmental limitations. Adsorption using materials such as activated carbon, ion exchange resins, and biochar is commonly employed to remove PFASs from contaminated water; however, this approach primarily transfers the contaminants from one phase to another rather than destroying them [31,32]. Consequently, the management and disposal of PFAS-laden adsorbents remain significant challenges, often requiring additional treatment or secure containment [33,34]. Thermal destruction methods such as incineration have been proposed to break down PFASs; however, effective mineralization typically requires extremely high temperatures and energy input, and incomplete combustion can lead to the formation of hazardous fluorinated by-products, including shorter-chain compounds and other persistent species [35,36]. Similarly, electrochemical destruction technologies have shown promise for PFAS degradation, particularly in aqueous systems, but their large-scale application is limited by high energy demands, electrode degradation, and the potential formation of toxic intermediates [37,38]. Moreover, many conventional treatment processes were not originally designed to handle the chemical stability and structural diversity of PFASs, resulting in variable removal efficiencies across different compounds [39].

Bio-based strategies for PFAS remediation, particularly those targeting C-F bond cleavage and biological defluorination, are gaining attention due to the unique challenges posed by the chemical stability and environmental persistence of these “forever chemicals” [40]. The C-F bond is one of the strongest in organic chemistry, rendering PFASs resistant to thermal, chemical, and conventional treatment methods, which contributes to their extreme environmental persistence and challenges in remediation [41]. Biological approaches, including microbial degradation and enzyme-mediated defluorination, offer a promising alternative by exploiting natural metabolic pathways capable of breaking these otherwise recalcitrant bonds, and have received increasing research attention in recent years [42]. Recent research has shown that specific bacterial strains, fungi, and engineered enzymes can initiate partial or complete defluorination of selected PFAS compounds, transforming them into less persistent or more biodegradable intermediates [43–45]. These strategies are particularly appealing because they minimize energy requirements, reduce secondary waste formation, and can potentially operate under environmentally relevant conditions, unlike high-temperature incineration or energy-intensive electrochemical methods [46]. Moreover, bio-based methods can be integrated with conventional treatments, such as adsorption or advanced oxidation, to achieve a hybrid approach that enhances removal efficiency while reducing environmental impacts [47]. As research reveals the diversity of PFAS-degrading microorganisms and the mechanisms of enzymatic defluorination, bio-based remediation is emerging as a sustainable, scalable, and environmentally compatible solution for addressing the global PFAS contamination crisis [40].

Although PFAS remediation has been extensively reviewed, most studies focus on individual treatment technologies and provide limited integration of the mechanisms governing PFAS destruction. Insufficient attention has been given to comparing remediation approaches through the common framework of C-F bond cleavage and defluorination pathways. Therefore, this review provides an integrated assessment of biological, electrochemical, photocatalytic, plasma, hydrothermal, and synthetic biology approaches, emphasizing the mechanisms of C-F bond activation and identifying opportunities for more sustainable and effective PFAS remediation.

2. Structure and Properties of PFASs and the Carbon–Fluorine (C-F) Bond

Per- and polyfluoroalkyl substances (PFASs) are a large class of synthetic fluorinated compounds characterized by carbon chains in which hydrogen atoms are partially or completely replaced by fluorine. Their unique chemical structure is dominated by the C-F bond [48–50]. This exceptional bond strength has made PFASs highly resistant to environmental degradation processes such as hydrolysis, photolysis, and biodegradation [48,51]. Structurally, PFASs consist of a hydrophobic and lipophobic fluorinated carbon tail and a hydrophilic functional head group (Figure 2), commonly carboxylate ($-\text{COO}^-$) or sulfonate ($-\text{SO}_3^-$), which imparts amphiphilic behaviour. The fluorinated tail exhibits low polarizability, high electronegativity, and strong hydrophobic characteristics, whereas the polar head group enhances aqueous solubility, surface activity, and environmental mobility [49,52–54]. PFASs can be broadly categorized into long-chain and short-chain compounds, with chain length significantly influencing their bioaccumulation potential, toxicity, and transport behaviour [55].

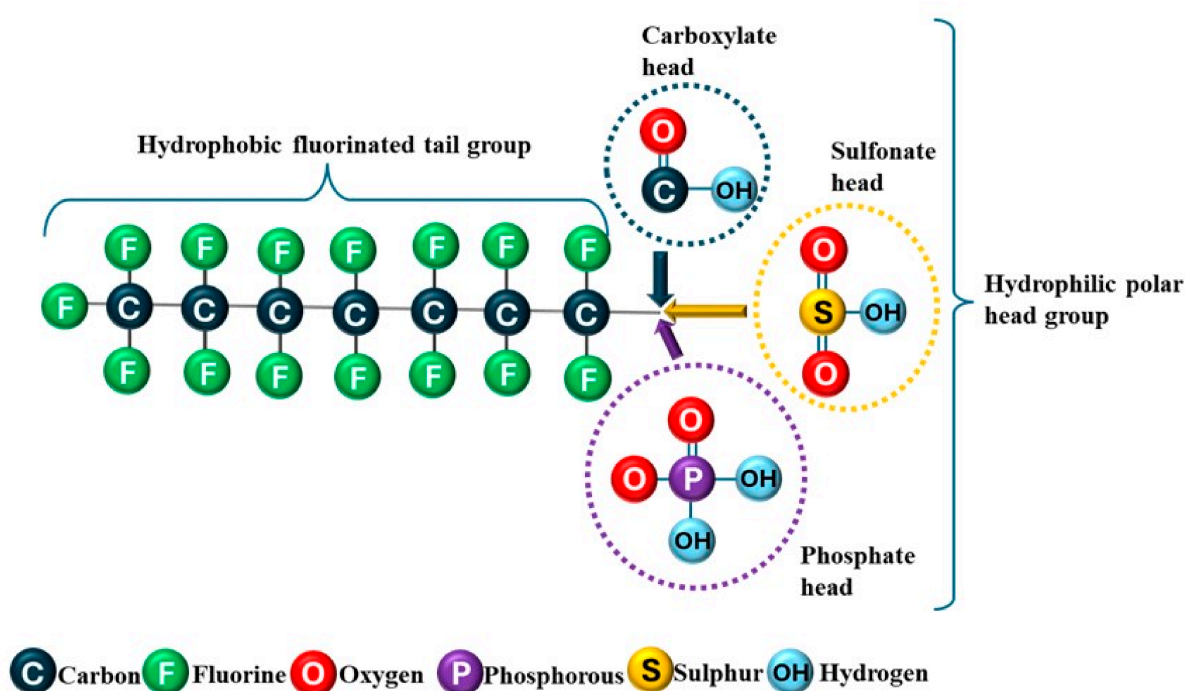


Figure 2. Structure of PFASs (adapted from Kibuye [56]).

Since their large-scale industrial introduction in the mid-20th century, PFASs have been widely used in commercial and industrial applications, including firefighting foams, non-stick cookware, water- and stain-resistant textiles, food packaging, and electronics manufacturing, owing to their exceptional surfactant properties and chemical stability [48,57–59]. Their remarkable persistence is primarily attributed to the C-F bond, the defining feature of organofluorine chemistry, which possesses one of the highest bond dissociation energies among single bonds in organic compounds ($\sim 450\text{--}540\text{ kJ mol}^{-1}$) [60–62]. This exceptional bond strength renders PFASs highly resistant to thermal, chemical, and biological degradation, resulting in their long-term environmental persistence, accumulation, and global distribution [48,52,63] (Figure 3).

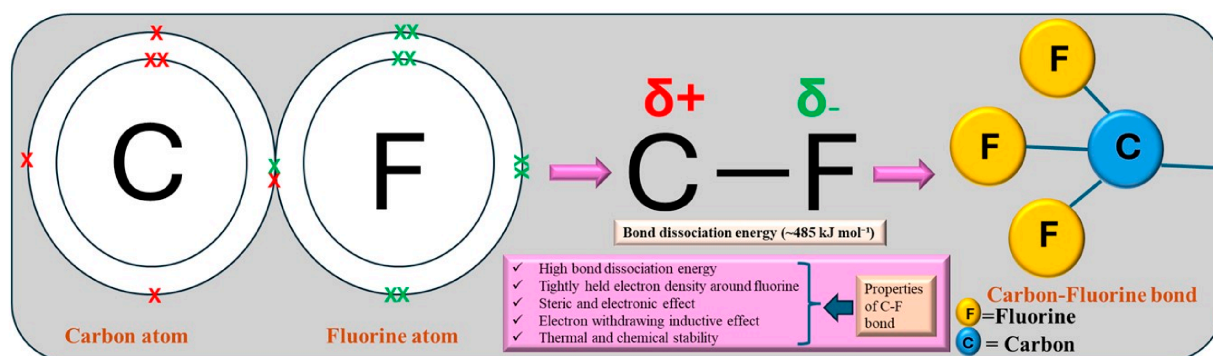


Figure 3. Carbon–fluorine dissociation bond.

The strength arises from the high electronegativity of fluorine and effective overlap of carbon and fluorine 2p orbitals, resulting in a short, highly polarized bond with partial ionic character [64]. The strong polarization of the C-F bond generates a significant dipole moment, with electron density shifted toward fluorine, and the resulting electrostatic stabilization contributes to its chemical inertness [65]. In addition to its strength, the low polarizability of the C-F bond arises from the compact and tightly held electron density around fluorine, which strongly resists distortion under external electric fields [64,65].

This significantly reduces its susceptibility to nucleophilic or electrophilic attack, thereby enhancing the chemical and biological stability of fluorinated compounds and contributing to their environmental persistence [66–68]. The C-F bond imparts pronounced hydrophobic and lipophobic properties to fluorinated molecules, particularly perfluorinated chains, owing to their low surface energy and weak intermolecular interactions with both aqueous and lipid phases, driven by the low polarizability of C-F bonds [49,69–71]. These unique physicochemical properties strongly influence the environmental partitioning, transport, and bioaccumulation behaviour of fluorinated compounds by governing their distribution across aqueous, soil, sediment, and biological compartments through structure-dependent intermolecular interactions [50,69,72–74]. They also promote accumulation at interfaces, enabling the formation of stable films and making them highly effective in industrial applications such as surfactants and surface coatings [75,76].

Steric and electronic effects play a key role in determining the reactivity of C-F-containing compounds. Increasing fluorination enhances electron-withdrawing effects and steric shielding around the carbon backbone, thereby limiting the access of nucleophiles, electrophiles, and enzymes to reactive centres and reducing overall reactivity [71,77,78]. In perfluorinated chains, the cumulative effect of multiple C-F bonds creates a highly electron-withdrawing and structurally rigid fluorinated envelope around the carbon backbone, often described as a fluorine shielding effect. This shielding, combined with strong inductive electron-withdrawing properties, stabilizes the molecular framework and renders it highly resistant to oxidative, reductive, hydrolytic, and biological degradation pathways [66,68,79]. These physicochemical properties collectively contribute to the environmental persistence of PFASs, as they resist conventional degradation pathways such as hydrolysis, photolysis, and biodegradation due to the strength and stability of C-F bonds, resulting in long environmental lifetimes [80–83].

Breaking the C-F bond typically requires extreme conditions or specialized catalytic or enzymatic systems due to its exceptional bond strength. Effective degradation pathways include reductive defluorination via electron transfer mechanisms, advanced oxidation processes, and engineered biocatalytic systems capable of overcoming the inherent stability of fluorinated compounds [84–86]. These properties support both the widespread utility

of fluorinated compounds and the significant environmental challenges associated with their persistence.

3. Conventional PFAS Remediation Technologies and Their Respective Limitations

This section discusses the conventional PFAS remediation technologies, including adsorption, incineration, advanced oxidation processes, and electrochemical processes widely applied to mitigate contamination.

Adsorption

PFAS adsorption occurs primarily through hydrophobic interactions, electrostatic attraction, pore filling, hydrogen bonding, and fluorophilic interactions between PFAS molecules and adsorbent surfaces. The fluorinated carbon tail preferentially associates with hydrophobic regions, while the negatively charged functional head groups interact with positively charged adsorption sites [49,54,87,88]. Adsorption efficiency is influenced by PFAS chain length, solution chemistry (e.g., pH, ionic strength, and natural organic matter), and adsorbent surface properties such as pore structure, surface charge, polarity, and functional groups [54,87,89]. A consistent limitation is the strong dependence on PFAS chain length. According to studies [90,91], granular activated carbon (GAC) performs well for long-chain PFASs (e.g., perfluorooctanoic acid (PFOS) and perfluorooctane sulfonate (PFOA)) but shows poor removal of short-chain compounds, which exhibit earlier advance and higher mobility. Although ion exchange resins generally outperform GAC for short-chain PFASs, their performance is still affected by competitive adsorption from co-occurring ions and natural organic matter, as highlighted in column and pilot-scale studies. Investigations by McCleaf et al. [92] and Tajdini et al. [93] further demonstrate that matrix effects such as dissolved organic carbon and sulfate can significantly reduce adsorption capacity and selectivity. In addition, adsorption performance is highly dependent on operational conditions, particularly empty bed contact time (EBCT). Short EBCTs result in rapid breakthroughs, especially for short-chain PFASs, thereby necessitating larger system designs and increasing operational costs. Under more realistic conditions, as shown in packed-column studies [94,95], these limitations become more pronounced. Both granular activated carbon and ion exchange systems exhibit strong sensitivity to hydraulic conditions, including flow variability and contact time. Reduced EBCT not only accelerates breakthrough but also compromises overall treatment efficiency, eventually requiring increased column volumes and higher capital investment. Regeneration-focused work by Sukeesan et al. [96] reveals that regeneration is often incomplete, energy-intensive, and may generate secondary PFAS-containing waste streams. Similarly, packed-column and pilot studies report fouling, reduced lifespan, and frequent media replacement, increasing operational costs [31]. Overall, these studies confirm that while adsorption is effective for PFAS capture, its limitations, which include selectivity issues, sensitivity to water chemistry, incomplete regeneration, and lack of destruction, restrict its sustainability as a standalone remediation solution.

Incineration

PFAS incineration remediation involves exposing contaminated materials to high temperatures, typically above 1000–1200 °C, under controlled combustion conditions. The intense heat cleaves the highly stable C-F bonds, resulting in thermal decomposition and eventual mineralization of PFASs to CO₂, water, and fluoride-containing end products [97–100]. Incineration efficiency for PFAS destruction depends on tightly controlled operational parameters, including high temperature, sufficient residence time, adequate oxygen supply, and effective mixing/turbulence, while emission control systems are essential to limit the formation

and release of incomplete combustion products such as volatile fluorinated species and hydrogen fluoride [97,98,100]. Studies such as Liu et al. [101] and Winchell et al. [102] show that incineration does not always achieve complete destruction of PFASs. Instead, residual PFASs or transformation products may persist in downstream waste streams, such as flue gas treatment residues and leachates, indicating incomplete mineralization under practical operating conditions. Furthermore, studies highlight the formation and release of toxic by-products and short-chain PFASs, which are often more mobile and harder to remove [103,104]. These findings suggest that thermal treatment can sometimes transform rather than fully eliminate PFASs, raising concerns about secondary contamination. Industrial-scale assessments reveal that achieving high destruction and removal efficiency requires strictly controlled high temperatures and residence times, which are energy-intensive and not always consistently maintained [105]. Consequently, operational variability can lead to fluctuating performance. Additional studies [106,107] demonstrate that PFASs can partition into ash and subsequently leach into landfill leachates. Consequently, when ash generated from PFAS incineration is disposed of in landfills, it may serve as a secondary source of PFAS release back into the environment. These findings emphasize that incineration may redistribute PFASs across environmental compartments rather than eliminate them. Summarily, these studies show that incineration is limited by incomplete destruction, by-product formation, high energy demand, and secondary contamination pathways.

Advanced oxidation processes

Advanced oxidation processes remediate PFASs by generating reactive species such as hydroxyl radicals ($\bullet\text{OH}$), sulfate radicals ($\text{SO}_4\bullet^-$), and hydrated electrons under ultraviolet (UV), photocatalytic, or electrochemical conditions, which attack the C-F backbone and initiate stepwise defluorination and partial mineralization [108–110]. Because PFASs are highly resistant, effective degradation often requires combined treatment systems or integrated reductive–oxidative pathways to achieve partial or complete mineralization into fluoride ions and smaller organic fragments, while minimizing by-product formation [100,111,112]. A report has shown that conventional advanced oxidation processes (AOPs) (e.g., hydroxyl radical-based systems) are ineffective for many PFASs, particularly fully fluorinated compounds, due to the extreme strength and stability of the C-F bond [113]. As a result, degradation is often slow, incomplete, or selective, supporting certain compounds over others. Electrochemical and electro-Fenton-based studies [114,115] demonstrate improved degradation efficiency; however, these systems require high energy input, specialized electrodes (e.g., Magnéli phase anodes), and strict operational control. This increases costs and limits large-scale applicability. Moreover, degradation often proceeds through stepwise defluorination, generating shorter-chain PFASs or intermediate products whose toxicity and persistence remain concerns. Hybrid systems, as reported by Yousefi et al. [47] and Sciscenko et al. [116], highlight efforts to enhance performance, especially for concentrated waste streams. Nevertheless, these approaches introduce additional complexity, including multi-stage operation, higher capital costs, and maintenance challenges, which hinder scalability. Despite technological advances, AOPs often result in partial PFAS degradation rather than complete mineralization, as evidenced by incomplete fluoride release, while challenges related to energy consumption, by-product formation, and scalability continue to limit their effectiveness as separate remediation technologies.

Electrochemical processes

Electrochemical remediation of PFASs occurs through the application of an external current that generates reactive species at the electrode surface, including $\bullet\text{OH}$, $\text{SO}_4\bullet^-$, and other reactive oxygen species depending on the electrolyte system, which drive PFAS

oxidation and defluorination [117–119]. Electron transfer processes initiated by hydrated or solvated electrons attack PFAS molecules, weakening and cleaving C-F bonds through stepwise reactions that progressively release fluoride ions and form partially defluorinated organic intermediates [120–122]. PFASs may also undergo direct electron reduction at the cathode via dissociative electron transfer, resulting in electron-induced C-F bond cleavage, fragmentation of the fluorinated carbon chain, and stepwise defluorination into fluoride ions and smaller organic intermediates [123]. Vershinin et al. [124] highlight that, although electrochemical systems can achieve PFAS degradation, their performance is often demonstrated under controlled laboratory conditions, limiting direct scalability to complex environmental matrices. Similarly, Saeidi et al. [125] show improved removal of otherwise recalcitrant short-chain compounds, but it requires multi-stage treatment and precise operational control, increasing system complexity and cost. A report by Guan et al. [126] emphasizes that matrix constituents (e.g., dissolved organic matter, competing ions) can significantly inhibit reaction efficiency by scavenging reactive species or interfering with electron transfer. Likewise, a report involving advanced materials [127] demonstrates high degradation efficiency but relies on expensive electrode materials and high energy input, which limit large-scale deployment. Hybrid approaches improve performance but introduce additional process complexity and potential secondary waste streams [128]. Furthermore, even with enhanced oxidation systems, degradation often proceeds via stepwise defluorination, leading to intermediate products and incomplete mineralization. Finally, challenges in selectivity and monitoring, as highlighted in Mosadegh et al. [129], emphasize difficulties in targeting diverse PFAS mixtures.

4. Evidence of Natural PFAS Biotransformation

PFASs first undergo microbial uptake and initial biotransformation, particularly for precursor compounds such as fluorotelomers. Subsequent reactions, including oxidation, chain shortening, and enzymatic defluorination, progressively cleave C-F bonds, releasing fluoride ions [52,130,131] (Figure 4). However, biodegradation is often incomplete, frequently producing stable short-chain PFAS intermediates rather than achieving full mineralization [112,132]. This section reviews emerging evidence of microbial interactions with PFASs, including early observations of limited transformation, biodegradation of precursor compounds, and indicators of partial defluorination in environmental systems. It also compares the roles of anaerobic and aerobic microbial communities in influencing PFAS transformation pathways and degradation potential.

4.1. Early Observations of Microbial Transformation

Early observations of microbial PFAS biotransformation have significantly advanced our understanding of the environmental behaviour of these persistent contaminants by demonstrating that microbial communities can mediate the transformation of several PFAS precursors under aerobic conditions. A study has revealed that fluorotelomer sulfonates can undergo gradual microbial conversion in contaminated soils and flow-through systems, producing transformation intermediates and terminal perfluoroalkyl acids (PFAAs) [133]. Similarly, investigations into fluorotelomer alcohols highlighted the importance of microbial community composition and functional diversity in governing PFAS transformation pathways [134]. Additional evidence demonstrated that wastewater-derived microbial consortia could differentially transform PFASs depending on molecular structure, indicating that chain length and functional groups strongly influence biodegradation behaviour [74]. Furthermore, a study emphasized that microbial species exhibit varying transformation capacities [130], while another study showed that precursor compounds such as perflu-

ooctane sulfonamide (FOSA) can be microbially oxidized into more stable products including PFOS [135].

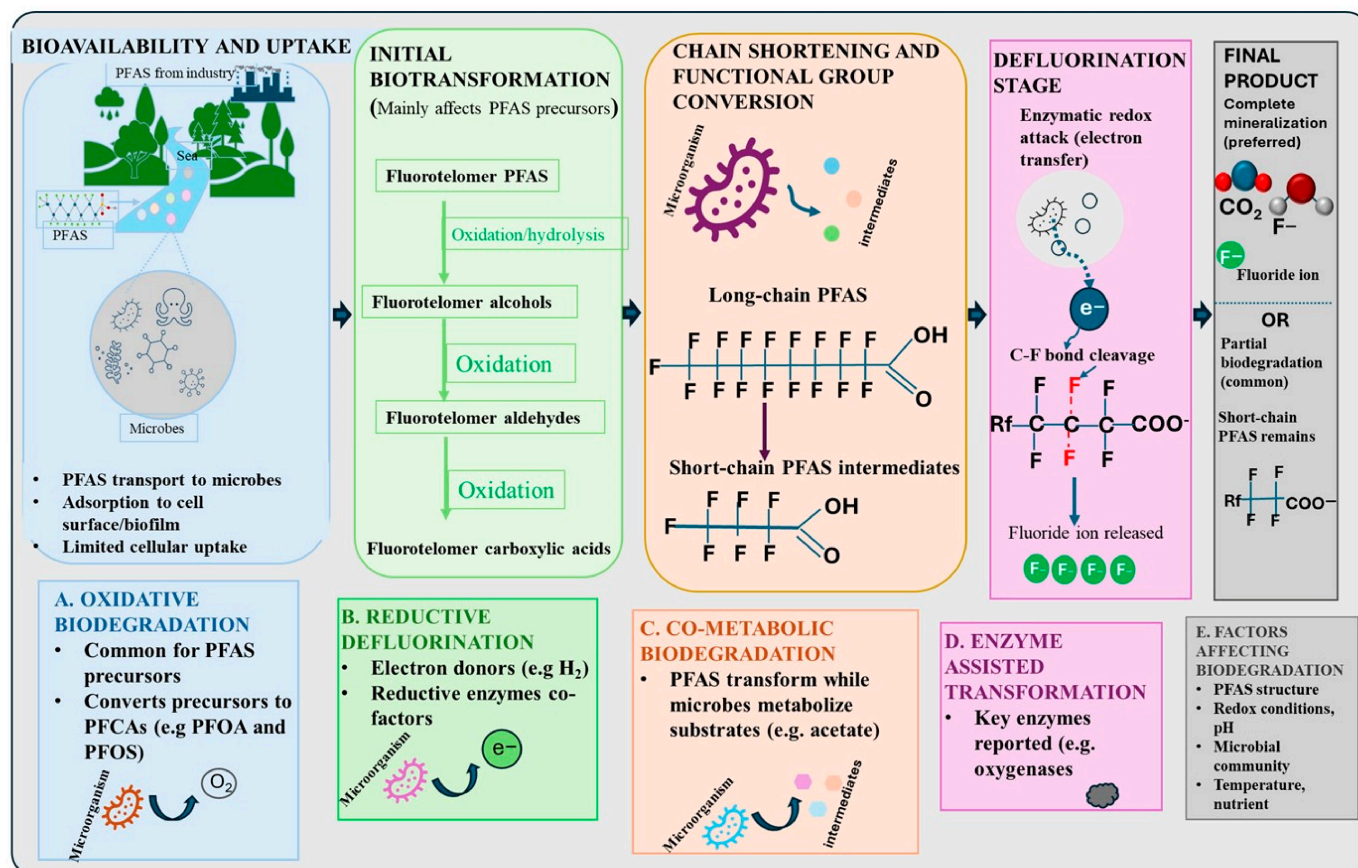


Figure 4. PFAS biodegradation pathways and mechanisms.

4.2. Transformation of PFAS Precursors

The transformation of PFAS precursors plays a significant role in the environmental fate and persistence of PFASs. Many PFASs released into the environment occur as precursor compounds, including fluorotelomer alcohols, fluorotelomer sulfonates, and other polyfluorinated chemicals, which can subsequently transform into terminal PFAAs [136]. These precursor compounds can undergo abiotic and biotic transformation processes in soils, sediments, and aquatic environments, producing persistent PFAAs as terminal products [137,138]. Microbial metabolism, oxidation reactions, and environmental photochemical processes can transform PFAS precursor compounds into more stable perfluorinated end products such as PFOA and PFOS [139,140]. Although these transformations may partially reduce the complexity of precursor molecules, they often generate highly persistent PFAAs that resist further degradation. As a result, precursor transformation can act as a secondary source of long-chain PFASs in environmental systems, prolonging contamination even after primary emissions have ceased [141]. The rate and extent of PFAS precursor transformation depend on environmental conditions such as oxygen availability, microbial community composition, temperature, and other environmental factors, as shown in Table 1. These transformations often proceed through stepwise reactions involving intermediate fluorotelomer compounds before forming terminal PFAAs (Figure 4) [142,143].

Table 1. PFAS precursor transformations.

Mother PFAS Compound (Precursor)	PFAS Class	Major Transformation Product(s)	Typical Transformation Pathway	Conditions Promoting Product Formation	Reference
FTOHs	Polyfluorinated precursor	FTALs → FTCAs → FTUCAs → PFCAs (e.g., PFOA, PFHxA, PFBA)	Sequential aerobic oxidation and chain shortening	Aerobic conditions, microbial oxidation, oxygen availability, monooxygenase activity	[144]
FTSs	Polyfluorinated precursor	FTCsAs → FTUCAs → PFCAs	Oxidative biotransformation	Aerobic microbial environments, activated sludge, soils	[145]
PAPs	Consumer product precursor	FTOHs → FTCAs → PFCAs	Hydrolysis followed by oxidation	Aerobic conditions, wastewater treatment system	[144]
diPAPs	PFAS precursor	FTOHs → FTALs → FTCAs → PFOA/shorter-chain PFCAs	Stepwise aerobic biotransformation	Activated sludge, wastewater, oxygen-rich conditions	[144]
EtFOSA	Sulfonamide precursor	EtFOSA → FOSA → FOSAA → PFOS	Oxidation and dealkylation	Aerobic microbial conditions	[146–148]
6:2 Fluorotelomer compounds	Fluorotelomer precursor	PFHxA, PFPeA, PFBA	Oxidation + chain shortening	Aerobic biodegradation	[149]
8:2 Fluorotelomer compounds	Fluorotelomer precursor	PFOA, PFHpA, PFHxA	Oxidative transformation	Aerobic conditions, biological treatment	[144]

FTOHs = fluorotelomer alcohols; FTALs = fluorotelomer aldehydes; FTCAs = fluorotelomer carboxylic acids; FTUCAs = fluorotelomer unsaturated carboxylic acids; FTSs = fluorotelomer sulfonates; PAPs = fluorotelomer mono-substituted phosphates; diPAPs = fluorotelomer di-substituted phosphates; EtFOSA = N-ethyl perfluorooctane sulfonamide; FOSAA = perfluorooctane sulfonamidoacetic acid.

The findings indicate that decreases in parent PFAS concentrations do not necessarily represent complete remediation, as persistent or potentially toxic transformation products may accumulate during treatment (Figure 5).

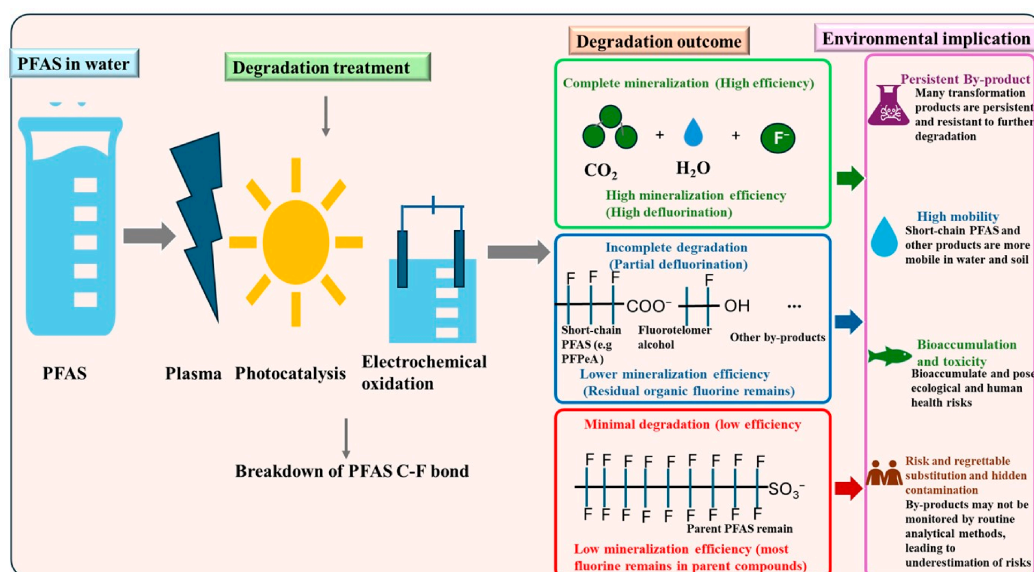


Figure 5. PFAS mineralization and by-products.

Research studies show that PFAS treatment rarely proceeds directly to complete mineralization, but instead generates a range of degradation intermediates and transfor-

mation products whose composition depends on the remediation technology and PFAS structure [150–153]. Electrochemical oxidation commonly proceeds through sequential chain shortening, producing lower-molecular-weight perfluoroalkyl carboxylic acids (PFCAs) as transient transformation products before extensive defluorination and fluoride release [154–157]. During the photocatalytic degradation of PFOA by Yang et al., sequential chain shortening is commonly observed, with transient formation of lower-molecular-weight PFCAs, including PFHpA, PFHxA, PFPeA, and PFBA. The accumulation and subsequent disappearance of these intermediates indicate progressive degradation through successive loss of $-CF_2$ groups, although complete defluorination generally occurs more slowly than parent compound degradation [158]. Plasma treatment of PFOA and PFOS generates a variety of transient fluorinated intermediates, including short-chain PFCAs (C4–C7) and perfluoroalkyl sulfonic acids (PFSAs), which are progressively degraded with continued treatment to fluoride, inorganic carbon species, and organic acids (trifluoroacetic acid (TFA), acetic acid, formic acid) [159]. In contrast, hydrothermal alkaline treatment (HALT) has demonstrated near-complete defluorination with minimal persistent organofluorine by-product formation under optimized operating conditions [160]. In contrast, biological systems predominantly transform fluorotelomer and sulfonamide precursors into intermediates such as fluorotelomer carboxylic acids (FTCAs), fluorotelomer unsaturated carboxylic acids (FTUCAs), fluorotelomer sulfonates (FTSs), FOSA, N-ethyl perfluorooctane sulfonamidoacetic acid (EtFOSAA), and perfluorooctane sulfonamide acetic acid (FOSAA), which are subsequently converted to terminal perfluoroalkyl acids (PFAAs), including PFOA or PFOS, depending on the precursor structure and microbial pathway [146,161–163].

4.3. Defluorination Indicators in Environmental Systems

Defluorination in environmental systems is commonly assessed using chemical and analytical indicators such as fluoride release and total organic fluorine measurements, which reflect the cleavage of the C-F bond [164]. One of the most direct indicators of PFAS defluorination is the release of inorganic fluoride ions (F^-), which signifies the breakdown of fluorinated compounds through C-F bond cleavage [165]. Monitoring reductions in parent PFAS concentrations, together with the formation of shorter-chain PFAAs, provides evidence of stepwise degradation and partial defluorination [166]. PFAS remediation efficiency should not be assessed solely by parent compound removal, as high removal rates do not necessarily indicate complete mineralization [151]. Complete mineralization demonstrated during HALT requires the cleavage of the strong C-F bonds, resulting in fluoride release and conversion of the PFASs to inorganic dissolved fluorides [160], whereas partial degradation often generates persistent fluorinated intermediates with uncertain environmental risks [158]. Consequently, fluoride release and fluorine mass balance provide more reliable indicators of treatment performance than PFAS removal alone [160]. The extent of mineralization varies among remediation technologies. The electrochemical oxidation study by Schaefer et al. [167] and photocatalytic/photoelectrochemical treatment study by Yang et al. [158] generally degrade PFASs through sequential chain shortening, producing transient short-chain PFAAs before extensive defluorination is achieved. In contrast, plasma and hydrothermal alkaline treatment typically achieve higher defluorination efficiencies due to rapid C-F bond cleavage [168,169], whereas biological systems primarily transform fluorinated precursors into terminal PFAAs rather than achieving complete mineralization [170]. Comprehensive assessment of treatment performance therefore requires the identification of degradation by-products using targeted liquid chromatography–tandem mass spectroscopy (LC–MS/MS), high-resolution mass spectrometry (HRMS) and combustion ion chromatography (CIC), together with complementary fluoride measure-

ments and fluorine mass balance analyses [160,171], and the elucidation of degradation pathways [172]. This is particularly important because incomplete PFAS degradation may generate transformation products, including ultra-short-chain and other fluorinated compounds, which may not be captured by routine targeted analyses, thereby potentially posing continued environmental and human health risks [173]. Therefore, the evaluation of PFAS remediation technologies should integrate parent compound removal, transformation product identification, and mineralization (or defluorination) efficiency to ensure effective contaminant destruction rather than simple chemical transformation [173]. Changes in total organic fluorine or extractable organic fluorine can further indicate transformation of organofluorine compounds into inorganic fluorine species, since analytical approaches explicitly quantify and differentiate these fluorine pools [174]. In biological systems, increased microbial activity, enzyme expression, or shifts in microbial community composition may indirectly indicate ongoing defluorination processes [175]. Additionally, mass balance approaches comparing fluorine inputs and outputs help evaluate the extent of fluorine mineralization [165,176]. However, tracking transformation products presents a major analytical challenge in detecting the biological defluorination of PFASs, as degradation generates diverse, often unknown, and low-abundance intermediates that are difficult to identify and quantify [143,172]. These transformation products exhibit extensive structural diversity, including variations in chain length, functional groups, and fluorination patterns, which, together with the limited availability of analytical standards, make them difficult to distinguish using conventional targeted analytical methods [172].

4.4. Role of Anaerobic vs. Aerobic Microbial Communities

Aerobic and anaerobic microbial communities play distinct but complementary roles in PFAS transformation in environmental systems. Under aerobic conditions, microorganisms typically drive oxidative processes, where oxygen-dependent enzymes such as monooxygenases and dioxygenases initiate the breakdown of polyfluorinated precursors, often producing more stable end products like PFAAs [139,177]. These pathways generally result in partial transformation rather than complete defluorination [143] (Figure 4). In contrast, anaerobic environments favour reductive processes, including reductive defluorination, where specialized microbes may cleave C-F bonds under electron-rich conditions [43,178]. Such pathways, though less common, are considered more promising for achieving deeper degradation of certain PFAS structures [179]. The efficiency of both aerobic and anaerobic PFAS transformation systems depends on microbial diversity, the availability of electron donors and acceptors, and prevailing geochemical conditions. In many natural settings, sequential or co-occurring aerobic and anaerobic zones can enhance overall transformation [179]. However, both systems are often limited by the strong stability of PFASs, resulting in slow transformation rates and the accumulation of persistent intermediates [143].

5. Molecular Barriers to PFAS Biodegradation

This section examines key factors governing PFAS persistence in the environment, including the exceptional thermodynamic stability of the C-F bond, the structural diversity influencing reactivity and biodegradability, and environmental bioavailability constraints. These properties explain the resistance of PFASs to conventional degradation and their long-term environmental persistence.

5.1. Thermodynamic Stability of the C-F Bond

The high electronegativity of fluorine leads to strong polarization of the C-F bond, creating a stable electronic configuration that resists cleavage under normal environmental conditions [180]. In addition, the compact size of fluorine provides steric shielding around the carbon support, reducing susceptibility to nucleophilic or electrophilic attack [181]. These factors collectively confer exceptional thermodynamic and kinetic stability, making PFASs resistant to hydrolysis, photolysis, microbial degradation, and many conventional chemical treatment processes [49]. Consequently, once released into the environment, PFASs tend to persist for long periods and can accumulate in water, soils, sediments, and biological systems [182,183]. This extraordinary stability presents a major challenge for remediation technologies and exposes the need for innovative approaches capable of effectively breaking or transforming the C-F bond.

5.2. Structural Diversity of PFASs and Biodegradability Implications

The structural complexity of PFASs presents a major challenge for biological remediation, as biodegradability varies widely among compounds and is often limited to partial transformation rather than complete mineralization [177,184]. Thus, understanding how molecular structure governs microbial susceptibility is critical for developing effective bio-based strategies for PFAS remediation. PFASs comprise a highly diverse group of thousands of synthetic chemicals characterized by varying carbon chain lengths, degrees of fluorination, and functional head groups such as carboxylates, sulfonates, phosphonates, and ether linkages [185,186]. This structural diversity strongly influences their environmental behaviour and susceptibility to biodegradation [54,131]. Long-chain PFAAs, such as PFOA and PFOS, possess fully fluorinated carbon chains that confer exceptional chemical stability and resistance to microbial attack due to the strength of the C-F bond [39,177]. In contrast, certain polyfluorinated PFASs, including fluorotelomer-based compounds and some emerging ether-linked alternatives, contain partially fluorinated structures or functional groups that may be more susceptible to microbial transformation [43,187]. Microorganisms can sometimes initiate degradation through processes such as oxidation, hydrolysis, or reductive defluorination of the non-fluorinated or less fluorinated moieties; however, these transformations often produce stable PFAAs as terminal products, which remain highly persistent in the environment [178,188].

5.3. Bioavailability Constraints in Environmental Matrices

The bioavailability of PFASs in environmental matrices is strongly influenced by physicochemical interactions with soil, sediment, and organic matter, which can limit their accessibility to microorganisms and reduce the effectiveness of biological degradation processes [55,189,190]. PFAS molecules exhibit varying degrees of hydrophobicity and ionic character depending on their chain length and functional groups, enabling them to partition between water, soil particles, and organic matter [191,192]. In soils and sediments, long-chain PFASs tend to sorb strongly to organic carbon, mineral surfaces, and metal oxides, which reduces their mobility but also restricts microbial contact with the compounds [190,193]. Electrostatic interactions, hydrophobic partitioning, and adsorption to clay minerals or iron and aluminum oxides further control their distribution within environmental matrices [191,194]. In addition, PFASs may accumulate at interfaces such as air–water boundaries, creating microenvironments where mass transfer becomes diffusion-limited, which can slow down transport and cause non-ideal breakthrough behaviour in soils and sediments [195,196]. Also, interactions with soil organic matter and minerals reduce the freely dissolved PFAS fraction, thereby limiting their biological availability for microbial uptake or enzymatic transformation [190]. Environmental factors such as

pH, ionic strength, temperature, and the presence of co-contaminants can also influence PFAS sorption–desorption dynamics and thus regulate their bioavailability [191]. Consequently, even when microorganisms capable of PFAS transformation are present, limited bioavailability in complex environmental matrices remains a major barrier to efficient biodegradation and natural attenuation [72]. Additionally, PFAS molecules can undergo micelle formation or supramolecular aggregation above a critical concentration in aqueous solutions, leading to the formation of nano-aggregates that influence their transport and fate [197]. Micelle formation can further modify the solubility, transport, and partitioning behaviour of PFASs in aqueous systems, as surfactant-like PFAS molecules aggregate into micellar structures above their critical micelle concentration [198]. Phase partitioning between water, soil, sediment, and air–water interfaces plays a key role in PFAS distribution, as their amphiphilic nature promotes accumulation at interfaces, leading to interfacial adsorption and mass-transfer limitations that contribute to non-ideal transport behaviour in porous media [193,196]. Micelle formation, driven by hydrophobic interactions, enhances PFAS degradation by improving contaminant partitioning, with more hydrophobic surfactants forming micelles at lower concentrations [199]. A study has also indicated that factors such as ionic strength, pH, and natural organic matter influence micelle stability, where loose structures at high salinity or alkaline conditions can reduce degradation efficiency [200].

6. Microbial Players in PFAS Transformation

6.1. Bacterial Taxa Associated with PFAS Transformation

Table 2 summarizes the microbial taxa reported across different environmental matrices and their associated roles in PFAS transformation, highlighting the PFAS compounds involved and the dominant biotransformation mechanisms identified. Several bacterial taxa, including members of the *Proteobacteria* (e.g., *Pseudomonas*), nitrifying bacteria and archaea, and other soil-associated groups, have been associated with the transformation of PFASs or their precursors, although overall degradation remains limited by the strong stability of C-F bonds [131,177,201]. Members of the *Pseudomonadaceae* family, particularly *Pseudomonas* spp., have been shown to metabolize certain polyfluorinated precursors under aerobic conditions, often using oxygenase-mediated pathways to partially transform the molecules [74]. Similarly, *Burkholderia* and *Rhodococcus* species have demonstrated the ability to transform fluorotelomer alcohols and sulfonates through oxidative pathways, producing shorter-chain PFAAs as intermediates [139]. Under anaerobic conditions, members of anaerobic microbial communities such as *Desulfovibrio*-related bacteria have been implicated in reductive transformations, where specialized microbes can cleave C-F bonds under electron-rich conditions, although such activity is generally slow and selective for specific PFAS structures [202]. Mixed microbial consortia from contaminated environments often exhibit synergistic capabilities, where aerobic taxa convert PFAS precursors to intermediate products that can then be further processed by other members of the community, enhancing overall transformation potential [74]. Environmental factors such as pH, redox potential, nutrient availability, and the presence of co-contaminants significantly influence the abundance and activity of microbial taxa, shaping community composition in contaminated soils and groundwater [203]. Despite these findings, the contribution of bacterial communities to complete PFAS mineralization remains limited, highlighting the need for further research into the identification, enrichment, and optimization of microbial taxa capable of more efficient PFAS transformation.

Table 2. Microbial evidence of PFAS precursor transformation across different environments.

Environment	Taxa	PFAS Compounds Involved	Transformation Role	Reference
Agricultural soil	<i>Proteobacteria (Pseudomonas), Actinobacteria (Bacillus), Acidobacteria (Rhodococcus)</i> (enriched under PFAS stress conditions)	PFOA, PFOS, mixed PFASs	Community restructuring; stress response; indirect transformation potential via co-metabolism	[203,204]
Rhizosphere soils	<i>Proteobacteria (Pseudomonas), Actinobacteria (Streptomyces), Firmicutes (Bacillus)</i> ; reduced diversity under PFAS stress	Mixed PFASs	PFAS-driven selection of rhizosphere bacteria; altered nutrient cycling and microbial function	[205]
Activated sludge/wastewater treatment plants (WWTPs)	<i>Nitrosomonas</i> (ammonia oxidizers), <i>Nitrospira</i> , <i>Pseudomonas</i> , <i>Comamonas</i> , <i>Dechloromonas</i> ; nitrifying consortia strongly implicated	AFFF-derived PFASs, EtFOSA, sulfonamides	Co-metabolic oxidation and precursor transformation under aerobic conditions	[74,201]
Anaerobic digestion systems (biogas/bioreactors)	<i>Methanosaeta</i> , <i>Methanobacterium</i> , <i>Clostridium</i> , <i>Desulfovibrio</i>	PFOS, PFOA mixtures	Limited co-metabolic transformation; resilience of anaerobic consortia to PFAS stress	[206]
Fungal-assisted/enzyme-driven systems	Fungal taxa (<i>Phanerochaete chrysosporium</i>), white-rot fungi producing laccases and peroxidases; bacteria (<i>Pseudomonas</i> spp.) in co-metabolism systems	6:2 FTS, fluorotelomer sulfonates	Enzymatic oxidation and partial defluorination of fluorotelomers	[207,208]
Anaerobic hydrolytic transformation systems	Anaerobic bacterial consortia associated with dechlorination-linked hydrolysis (mixed fermenters + hydrolytic bacteria)	Chlorinated fluorocarboxylic acids (PFAS analogues)	Hydrolytic dechlorination enabling partial transformation pathways	[202]
Periphyton/aquatic biofilms	<i>Cyanobacteria</i> , <i>Proteobacteria (Pseudomonas-related)</i> , diatoms-associated bacteria; resistance gene carriers observed	Mixed PFAS	Stress response, resistance gene expression, biofilm-mediated retention	[209]
Soil ecosystem under long-term contamination	<i>Proteobacteria (Pseudomonas), Actinobacteria, Ascomycota fungi</i> , co-occurrence with plant-associated microbiota	Mixed PFAS + co-contaminants	Microbial network restructuring influencing functional potential (not direct degradation)	[210,211]

6.2. Fungal Contributions to PFAS Biotransformation

Fungi are increasingly recognized as important contributors to the biotransformation of PFASs, complementing bacterial activities in contaminated environments [208,212].

Certain filamentous fungi, such as species of *Phanerochaete chrysosporium*, *Trametes* spp., and others, produce extracellular oxidative enzymes, including laccases, peroxidases, and cytochrome P450s, which can attack polyfluorinated precursors and partially transform them into shorter-chain PFAAs, demonstrating fungal potential in PFAS biotransformation research [131,207,208,213] as indicated in Table 2. These enzymes are capable of generating reactive radical species or oxygenated intermediates that facilitate the cleavage of strong bonds adjacent to recalcitrant moieties, promoting subsequent oxidation or hydrolysis steps [213]. Fungi also demonstrate the ability to tolerate high concentrations of co-contaminants and harsh pH and redox conditions that may inhibit bacterial degradation, thereby extending the environmental niches for PFAS transformation [210]. Moreover, fungal hyphal networks can physically transport substrates and co-metabolites across heterogeneous soil and sediment matrices, enhancing contact between contaminants and reactive enzymes and thereby promoting co-metabolic transformation processes [214]. Although complete defluorination remains rare, fungal-mediated transformations can reduce the complexity of PFAS precursors and generate intermediates that are more accessible to bacterial consortia [139]. Environmental factors such as nutrient availability, temperature, and moisture strongly influence fungal activity and enzyme expression, making them critical determinants of transformation efficiency [211].

6.3. Microbial Community Adaptation in Contaminated Environments

Microbial communities in PFAS-contaminated environments undergo adaptive shifts that reflect both the persistence of these compounds and the selective pressures they impose [204]. Exposure to PFASs often results in changes in community composition, with the enrichment of taxa capable of tolerating chemical stress, utilizing co-contaminants, or participating in the partial transformation of PFAS precursors [205] (Table 2). These adaptations may include modifications in cell membrane structure, expression of efflux pumps, and activation of stress response pathways that enhance survival under toxic conditions [209]. In addition, microbial communities may develop co-metabolic capabilities, where enzymes originally evolved for other substrates incidentally transform PFAS compounds [206]. Over time, functional diversity within the community can increase, promoting synergistic interactions among different microbial groups, including bacteria and fungi [215]. Environmental factors such as nutrient availability, redox gradients, and contaminant mixtures further shape these adaptive responses, influencing both microbial activity and transformation potential [203]. Although complete mineralization of PFASs remains rare, adapted microbial communities can facilitate partial degradation, precursor transformation, and the formation of intermediate products [42].

6.4. Metagenomic Insights into PFAS-Responsive Microbiomes

Metagenomic approaches have provided powerful insights into the structure and functional potential of PFAS-responsive microbiomes in contaminated environments. By enabling culture-independent analysis of microbial DNA, metagenomics reveals shifts in community composition and the enrichment of genes associated with stress tolerance, xenobiotic metabolism, and co-metabolic transformation pathways in PFAS-contaminated environments [216]. In PFAS-impacted soils, sediments, and wastewater systems, metagenomic data often show increased abundance of genes encoding transporters, efflux pumps, and oxidative or reductive enzymes that may contribute to the partial transformation of PFASs or their precursors [42]. These datasets also highlight the presence of diverse metabolic networks, where different microbial taxa contribute complementary functions, supporting synergistic interactions within the community [217]. Moreover, PFASs might be biotransformed by the synergistic actions of multiple coexisting microbes rather than

by a collection of independent microorganisms, as mixed microbial consortia can enhance complex PFAS breakdown through complementary metabolic interactions [218]. Functional annotation further indicates that pathways related to carbon cycling, energy metabolism, and contaminant degradation are reorganized in response to PFAS exposure, reflecting microbial adaptation to persistent chemical stress [219]. Importantly, metagenomics can uncover previously uncharacterized enzymes and candidate genes potentially involved in defluorination or precursor conversion, offering new directions for bioremediation research [220]. Coupled with other omics techniques such as metatranscriptomics and metabolomics, these insights provide a more comprehensive understanding of how microbial communities respond, adapt, and function in PFAS-contaminated systems, ultimately informing strategies to enhance biodegradation and environmental management [221].

7. Enzymatic Pathways for C-F Bond Cleavage

7.1. Dehalogenases and Reductive Defluorination Mechanisms

Enzymes capable of degrading fluoroorganic compounds represent a growing focus in environmental biotechnology due to the persistence and chemical stability of PFASs and related compounds [222]. Known fluoroorganic-degrading enzymes include dehalogenases, monooxygenases, dioxygenases, laccases, and peroxidases, each of which acts through distinct biochemical mechanisms to cleave carbon–halogen bonds or oxidize fluorinated moieties [66,223]. Dehalogenases play a central role in the microbial transformation of halogenated organic compounds, including the rare but increasingly studied process of reductive defluorination [175]. These enzymes catalyze the cleavage of carbon–halogen bonds through hydrolytic, oxidative, or reductive mechanisms, depending on their class and the environmental conditions [224,225]. Hydrolytic dehalogenases, such as haloacid dehalogenases, typically act on short-chain halogenated substrates by catalyzing hydrolytic carbon–halogen cleavage via nucleophilic attack; however, their activity toward C-F bonds is limited due to the high bond dissociation energy [66]. In contrast, reductive dehalogenation occurs primarily under anaerobic conditions and is mediated by specialized enzymes known as reductive dehalogenases, often associated with organohalide-respiring bacteria [226]. These enzymes utilize electron donors such as hydrogen or organic acids to drive the reductive cleavage of carbon–halogen bonds, replacing halogen atoms with hydrogen in a process linked to microbial energy conservation [227]. Although reductive defluorination remains challenging due to the exceptional stability of the C-F bond, recent studies show that microbial consortia and enzyme systems can facilitate partial defluorination of PFAS precursors and short-chain fluorinated compounds [139,228,229]. Mechanistically, these processes may involve electron transfer via cofactors such as corrinoids and iron–sulfur clusters, enabling the activation of otherwise inert bonds [230,231]. Despite their limited efficiency, dehalogenases and reductive pathways represent promising avenues for biotechnological innovation, particularly when integrated with co-metabolic systems or engineered microbial consortia aimed at enhancing PFAS degradation [232].

7.2. Oxygenases and Oxidative Transformation Pathways

Oxygenases play a pivotal role in the oxidative transformation of organic contaminants by catalyzing the incorporation of oxygen into substrates, thereby enhancing their reactivity and susceptibility to further degradation [233]. These enzymes are broadly classified into monooxygenases and dioxygenases, which introduce one or two oxygen atoms, respectively, into target molecules [234]. In aerobic microbial systems, oxygenases initiate the breakdown of complex xenobiotics by hydroxylating aliphatic or aromatic structures, destabilizing chemical bonds and facilitating subsequent reactions such as cleavage, rearrangement, or mineralization [235]. In the context of PFAS precursors, oxygenases are

particularly important in transforming fluorotelomer alcohols and sulfonates into intermediate compounds, including fluorinated aldehydes and acids, which can be further oxidized into PFAAs [236,237]; however, these pathways often result in partial transformation rather than complete defluorination [237]. Oxygenase-mediated reactions are frequently supported by cofactors such as nicotinamide adenine dinucleotide (NADH) or nicotinamide adenine dinucleotide phosphate (NADPH), and involve electron transfer chains that enable the activation of molecular oxygen [238,239]. Moreover, these enzymes are often involved in co-metabolic processes, where microbes degrade fluorinated compounds incidentally while utilizing other primary substrates [177].

8. Mechanistic Constraints Governing PFAS Degradation Technologies

The principal distinction among biological and physicochemical approaches lies in how these processes are achieved, as represented in Figure 6. Biological systems rely on intracellular metabolic processes to sustain enzyme-catalyzed reactions, making them highly selective and environmentally benign. However, their effectiveness is largely restricted to PFAS precursors because the reducing power generated during microbial metabolism is generally insufficient to directly cleave the exceptionally stable C-F bond of terminal PFAAs [201,202,240]. Experimental evidence consistently shows microbial transformation of fluorotelomer- and ether-based PFAS precursors, whereas terminal PFAAs remain highly resistant to biodegradation [241]. In contrast, electrochemical, photocatalytic, plasma, and hydrothermal technologies partially overcome this limitation by supplying external energy through electrode potentials, photogenerated charge carriers, reactive radicals, or elevated temperature–pressure conditions, thereby facilitating more extensive defluorination [118,150,242,243]. The contrasting activation mechanisms also explain the different thermodynamic and kinetic constraints associated with each technology. Biological degradation is primarily limited by substrate bioavailability and microbial adaptation under environmentally relevant conditions, while enzyme specificity and co-metabolic dependence restrict efficient transformation of terminal PFAAs, resulting in preferential degradation of PFAS precursors [201,240,244]. Unlike biological systems, whose degradation efficiency is governed primarily by enzyme specificity and microbial metabolism, electrochemical and photocatalytic processes are primarily governed by electron transfer kinetics and catalyst performance, including catalyst stability and charge-carrier utilization [150,158]. Plasma technologies largely circumvent these biological constraints by generating highly reactive electrons and radical species capable of extensive PFAS mineralization, although this is accompanied by higher energy demands. Hydrothermal systems likewise achieve extensive mineralization through high-temperature and high-pressure conditions but remain constrained by specialized reactor requirements and intensive operating conditions [245,246]. Consequently, degradation efficiency is determined not only by the intrinsic stability of PFAS molecules but also by environmental conditions and molecular structure. PFAS precursors containing metabolically accessible functional groups are consistently more responsive to transformation than fully fluorinated PFAAs [133,247], while short-chain compounds generally exhibit higher aqueous motility and lower adsorption, reducing their retention and contact time within many treatment systems [248]. These mechanistic differences explain why no individual technology consistently achieves complete PFAS mineralization under environmentally relevant conditions. Instead, coupling biological transformation with physicochemical destruction offers complementary mechanisms for substrate activation and C-F bond cleavage, thereby enhancing PFAS defluorination and progressing toward more complete mineralization [45,249].

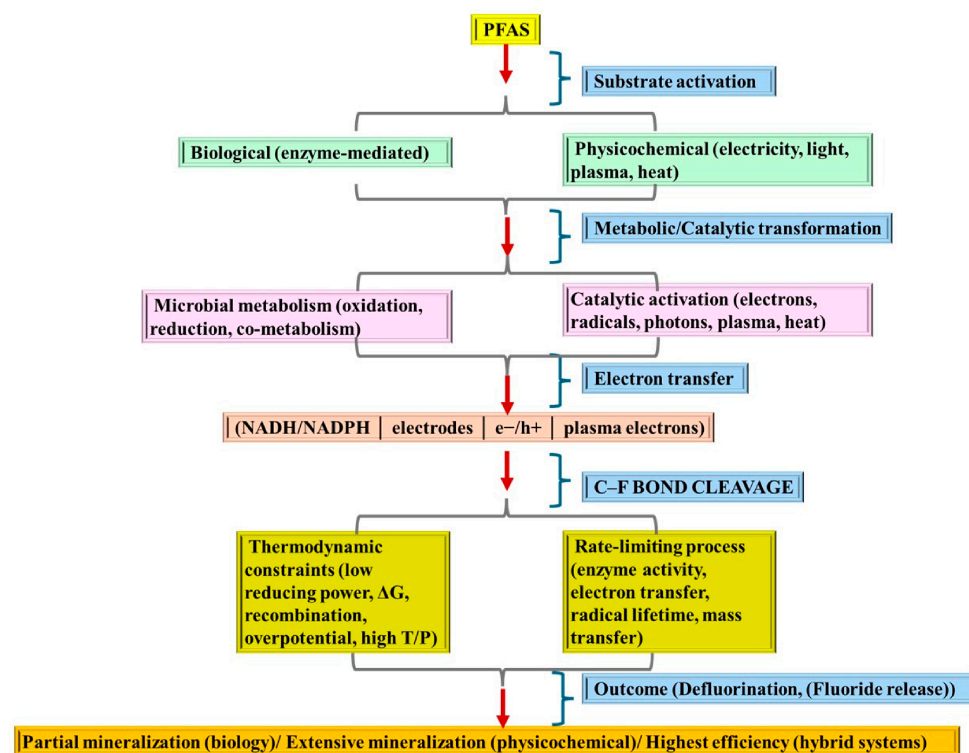


Figure 6. Mechanistic framework of PFAS degradation technologies.

9. Environmental Factors Controlling Bio-Based PFAS Degradation

Soil and sediment geochemistry play a fundamental role in governing bio-based degradation of PFASs by influencing their mobility, bioavailability, and interactions with microbial communities [250]. Key geochemical parameters such as pH, redox potential, organic matter content, and minerals (Figure 7) determine the extent to which PFAS compounds are sorbed or remain accessible for microbial transformation [72]. For instance, soils rich in organic carbon and clay minerals can strongly adsorb PFASs, limiting their availability to degrade microorganisms, whereas more porous or low-organic substrates may enhance transport and microbial contact [190,251]. This occurs because soil organic matter and clay minerals provide sorption sites that retain PFASs through hydrophobic partitioning, electrostatic attraction, and surface interactions, thereby reducing PFAS mobility and limiting their accessibility to degrading microorganisms, whereas porous low-organic soils promote PFAS transport and increase microbe–PFAS contact [190,252]. Redox conditions are particularly critical, as they regulate the activity of aerobic and anaerobic microbes involved in oxidative or reductive transformation pathways [253,254]. Additionally, the presence of competing ions and co-contaminants can influence PFAS partitioning and microbial metabolism, either inhibiting or stimulating degradation processes [42]. Sediments often act as sinks for PFAS accumulation, where stratified redox zones create microenvironments that support diverse microbial processes, including co-metabolism [219,255]. Metal ions and mineral surfaces may also catalyze abiotic reactions or facilitate electron transfer, indirectly supporting microbial degradation pathways [256].

Bio-based PFAS degradation exhibits consistent environmental control patterns across recent studies, but the magnitude of these constraints varies by system type. Waste and mixed-matrix systems show the strongest inhibition, where high PFAS loads, co-contaminants, and complex matrices suppress microbial activity and limit transformation potential [257]. In contrast, engineered systems provide improved control of pH, nutrients, and redox conditions, yet still fail to achieve complete defluorination due to intrinsic enzymatic limitations [258,259]. Soil–microbe systems offer more stable microbial com-

munities but are strongly restricted by sorption-driven reductions in PFAS bioavailability, particularly in organic-rich matrices [260]. Across all systems, precursors are more readily transformed than PFAAs, while emerging PFASs remain the least biodegradable. Compared to physicochemical technologies, biological approaches are environmentally sustainable but limited by slow kinetics and incomplete C-F bond cleavage, whereas advanced oxidation and electrochemical systems provide higher mineralization potential under broader operating conditions [118,261].

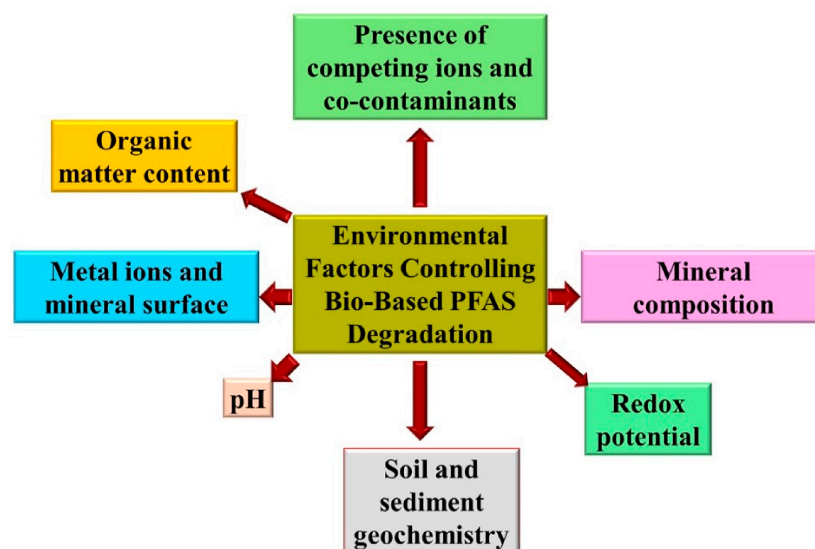


Figure 7. Factors controlling bio-based PFAS degradation.

10. Innovative Approaches for PFAS Destruction and Defluorination in Environmental Matrices

The remediation of PFASs has rapidly advanced through the development of integrated physicochemical, electrochemical, photochemical, thermal, and biological technologies targeting the highly stable C-F bond. Table 3 summarizes the major PFAS defluorination technologies according to their reported performance, technology readiness, strengths, and limitations. Detailed study-specific information, including operating conditions, defluorination efficiencies, advantages, and limitations, is provided in Supplementary Table S1 [120,127,150–152,262–323]. Among the most widely investigated PFASs are PFOA, PFOS, GenX, perfluorobutane sulfonate (PFBS), and fluorotelomer compounds, as shown in Table 3.

Beyond laboratory-scale performance, the practical implementation of PFAS remediation technologies depends on scalability, technology readiness levels, operational costs, energy requirements, reactor configurations, and long-term stability. Biological systems are attractive for in situ remediation because of their low operating costs, environmental compatibility, and simple reactor requirements, but their technological readiness for large-scale applications remains limited by slow degradation kinetics, microbial adaptation, incomplete mineralization, and variable long-term performance under fluctuating environmental conditions [177,324]. Electrochemical technologies have reached relatively higher technology readiness and have demonstrated successful pilot-scale applications, although widespread deployment remains constrained by continuous electrical demand, electrode fouling, electrode replacement costs, and mass-transfer limitations [37,325]. Photocatalytic systems operate under mild conditions and can utilize solar irradiation, reduce chemical inputs and potentially lower operational costs. However, their transition to large-scale applications remains limited by low technology readiness, challenges in scaling

photoreactors, restricted light penetration, catalyst recovery and long-term stability, and reduced treatment efficiency in complex environmental matrices [326,327]. Plasma technologies have demonstrated high PFAS destruction efficiencies and successful pilot-scale applications, indicating promising technology readiness. However, widespread implementation remains constrained by specialized reactor configurations, high electrical energy demand, capital investment, and the need for further optimization of continuous large-scale operation [159,328]. Hydrothermal treatment has demonstrated effective PFAS mineralization in contaminated groundwater, soil, and concentrated waste streams. Nevertheless, its broader implementation is constrained by the need for high-temperature, high-pressure reactor systems, substantial energy input, corrosion-resistant materials, and high capital costs, limiting current applications primarily to concentrated waste treatment [152,329].

Table 3. Recent advances in PFAS removal and defluorination technologies.

Technology Category	Representative PFAS	Highest Reported Defluorination (%)	Technology Readiness	Major Strengths	Key Challenges
Photocatalysis	PFOA, PFOS, GenX	~91%	Laboratory	Solar/visible-light driven; catalyst engineering improves C-F cleavage	UV/light dependence; catalyst stability; scale-up
Electrochemical oxidation/reduction	PFOA, PFOS, PFBS	100%	Laboratory pilot	High destruction efficiency; controllable process	Electrode cost; electricity demand; fouling
Hydrated electron-based processes	PFOA, PFOS, GenX, PFBS	100%	Laboratory	Among the highest defluorination efficiencies	UV/sulfite requirement; alkaline conditions; matrix effects
Plasma technologies	PFOA, PFOS, mixed PFAS	100%	Laboratory	Rapid degradation; minimal chemical addition	High energy demand; reactor scale-up
Hydrothermal/SCWO	Mixed PFAS	100%	Pilot	Near-complete mineralization	High temperature/pressure; capital cost
Mechanochemical/piezocatalysis	PFOA, PFOS	100%	Laboratory	Solvent-free; low chemical input	Equipment demand; scalability
Sonochemical processes	PFOA, PFOS, GenX	100%	Laboratory	Chemical-free degradation	High ultrasonic energy demand
Adsorption–destruction hybrids	PFOA, PFOS	~87%	Laboratory	Captures and destroys PFAS; reduced secondary waste	Multi-step operation; regeneration
Biological/microbial	PFOA, PFOS	30%	Laboratory	Low-energy; potential in situ remediation	Slow degradation; incomplete mineralization

SCWO = supercritical water oxidation.

Hydrated electron-based systems have shown some of the highest defluorination efficiencies in aqueous matrices. For example, the studies by Bentel et al. [120], Tenorio et al. [302], and O'Connor et al. [313] demonstrated that UV/sulfite systems achieved near-complete degradation and mineralization of PFOA, PFOS, and fluorotelomer sulfonates in water and AFFF matrices, with defluorination efficiencies often exceeding 90–99%. The major advantage of hydrated electron systems is their ability to directly cleave the C-F bond under relatively mild conditions while producing fewer persistent transformation products. Recent improvements, such as UV/sulfite/iodide systems and VUV/sulfite processes, have further enhanced electron generation and PFAS destruction, even in complex waters containing carbonate or bromide species. Hydrated electron-based PFAS treatment generally achieves high removal and strong defluorination efficiency, because hydrated electrons are extremely reducing and can directly attack C-F bonds, especially in long-chain PFASs. However, performance is structure-dependent, and complete mineralization is often difficult. Energy demand can be moderate to high, since hydrated electrons are typically generated via UV/sulfite or VUV systems requiring energy input.

Electrochemical technologies have also emerged as highly effective PFAS destruction strategies. Enschede et al. [321] used boron-doped diamond electrodes for the electrochemical oxidation of PFOA in water, achieving substantial mineralization and fluoride release. Similarly, Meng et al. [322] reported complete electrocatalytic defluorination of PFOS in an aqueous solution using abundant catalysts, highlighting the growing shift toward cost-effective electrode materials. Yin et al. [323] demonstrated the “trap-n-zap” approach using UiO-66 modified boron nitride electrodes to simultaneously adsorb and electrochemically degrade PFOA at environmentally relevant concentrations. In addition, Wang et al. [262] achieved enhanced electroreductive defluorination of unsaturated PFASs using surfactant-modified cathodes, while Liu et al. [263] utilized CeO₂ electrodes to lower the oxidation potential requirements for PFAS destruction. These electrochemical systems are advantageous because they combine adsorption, concentration, and degradation within a single treatment platform while minimizing chemical reagent consumption. Electrochemical PFAS removal generally achieves high removal efficiency, especially for long-chain PFASs, through anodic oxidation and advanced electrochemical interfaces [330]. Electrochemical PFAS degradation occurs via anodic oxidation or cathodic reduction. Anodic oxidation promotes direct electron transfer and the formation of reactive oxidizing species that induce sequential C-F bond cleavage and chain shortening [331], whereas cathodic reduction generates hydrated electrons or other reducing species that drive reductive defluorination [150]. The dominant degradation pathway depends on the electrode material and operating conditions. Its defluorination ability is comparatively strong among destructive technologies, particularly when using boron-doped diamond (BDD) or mixed-metal oxide anodes, although complete mineralization is often not achieved [155,330].

Photocatalytic and plasma-driven systems have shown increasing promise for degrading long-chain PFASs in water and soil matrices. Duan et al. [264] employed zero-valent metal-enhanced BN/TiO₂ photocatalysts to degrade PFOA efficiently via improved electron transfer through Mott-Schottky junction regulation. Zhou et al. [265] further demonstrated singlet oxygen-mediated photocatalytic removal of PFASs using sulfur vacancy-engineered ZnIn₂S₄, while Liu et al. [266] achieved sunlight-driven C-F bond cleavage using CuInS₂/BiOCl composites. Plasma technologies also produced remarkable degradation efficiencies. X. Li et al. [267] showed efficient PFOS degradation in dielectric barrier plasma systems using Bi₂O₃ as both the catalyst and dielectric material, whereas S. Cao et al. [268] identified plasma-liquid interfacial F/OH exchange as a critical mechanism driving PFAS defluorination. Y. Zhang et al. [269] successfully remediated PFAS-contaminated soil using air-driven fluidized-bed non-thermal plasma where XGBoost

analysis identified working gas composition as the dominant factor influencing degradation efficiency. The major advantage of plasma and photocatalytic systems lies in their rapid oxidative and reductive radical generation, enabling deep mineralization without excessive chemical additives. Photocatalytic PFAS removal typically shows moderate removal efficiency, strongly dependent on catalyst type (particularly TiO₂ modifications) and light intensity, with limited defluorination under pristine TiO₂ systems that improves only under advanced photocatalysts, often resulting in partial mineralisation rather than complete degradation [332,333]; conversely, plasma-driven PFAS treatment achieves high removal efficiency through the generation of multiple reactive species, including hydrated electrons, hydroxyl radicals, and UV photons formed within non-thermal discharge environments, which collectively enable oxidative and reductive pathways responsible for PFAS bond cleavage and degradation [159]. It also exhibits a strong defluorination ability compared to most non-thermal processes, enabling partial to near-complete mineralisation under optimized conditions [334].

Thermal and mechanochemical technologies have proven particularly effective for heavily contaminated soils and concentrated PFAS waste streams. Hao et al. [152] demonstrated HALT for the destruction of PFASs in contaminated groundwater and soils, achieving extensive defluorination under elevated temperature and alkaline conditions. However, degradation in soil was slower due to soil buffering effects and the dependence on temperature. Li et al. [270] achieved efficient PFOS destruction in supercritical water oxidation (SCWO) reactors, while Pinkard et al. [271] successfully treated AFFF-impacted fire training pit water using continuous HALT. Turner et al. [272] used planetary ball milling for mechanochemical destruction of PFOS and PFOA in contaminated soils, achieving substantial degradation and fluoride release. B. Lei et al. [273] similarly demonstrated mechanochemical degradation of PFOA using heating pad waste as a reactive material. These approaches are advantageous because they can handle highly concentrated PFAS matrices and achieve near-complete mineralization, reducing long-term secondary pollution risks. Thermal PFAS treatment achieves high removal efficiency (>90%) at extreme temperatures (>700 °C) but requires high energy input and often results in incomplete mineralization, generating products of incomplete destruction and volatile fluorinated by-products [100,335–337]. Mechanochemical treatment, in contrast, enables PFAS transformation at ambient conditions via high-energy milling, offering strong defluorination potential without thermal input, but still produces solid-phase transformation intermediates [338].

Adsorption-coupled degradation systems are increasingly being developed to overcome PFAS concentration limitations. Reference [274] investigated selective PFAS adsorption using fluorous active sites, while Regmi et al. [275] fabricated nZVI/rGO composites that provided scalable remediation through sequential adsorption and oxygen-free UVC degradation, enabling sequential adsorption and photocatalytic degradation of PFASs. Ren et al. [276] combined aminated biosorbents with UV/sulfite reduction for enhanced PFOA removal from water. K.-H. Kim et al. [277] introduced a regenerable remediation platform capable of ultrafast PFAS capture and mineralization, whereas Z. Wang et al. [278] proposed an integrated adsorption–regeneration–distillation–plasma system for low-energy remediation and solvent recovery. The advantage of these hybrid systems is that adsorption enriches PFASs at reactive interfaces, significantly improving degradation kinetics and reducing energy demand. Adsorption is among the most widely applied PFAS remediation technologies, achieving high removal efficiencies, particularly for long-chain PFASs, through adsorbents such as activated carbon, ion exchange resins, and engineered biochars [275,277]. However, adsorption does not destroy PFASs or achieve defluorination;

instead, it transfers contaminants from water to a solid phase requiring regeneration or disposal [31,339].

Biological and bio-assisted approaches are also gaining attention despite the recalcitrance of PFASs. Ayers and Zhang [279] demonstrated fungal-mediated defluorination of perfluoroalkyl carboxylic acids using wood decomposer fungi, although fungi showed little evidence of defluorinating persistent legacy PFASs such as PFOA, PFOS, and GenX due to the extreme stability of long-chain C-F bonds, while Saifur et al. [74] showed that wastewater microbial consortia can biotransform structurally diverse PFASs. Huang et al. [280] further demonstrated enhanced PFBS removal through biochar–microbe synergy. These biological systems are advantageous because they offer environmentally sustainable and potentially low-cost remediation pathways, particularly when integrated with adsorption or electrochemical technologies. Biological and bio-based PFAS treatment approaches generally exhibit low-to-moderate removal efficiency, with performance strongly dependent on PFAS structure and sorbent properties. Experimental studies show that biochar and other biomass-derived sorbents can adsorb PFASs via hydrophobic and electrostatic interactions, but their efficiency is often lower and more variable than engineered adsorbents, particularly for short-chain PFASs, indicating that biosorption dominates over true biodegradation processes [340,341]. Overall, the reviewed studies collectively demonstrate that no single remediation technology universally addresses all PFAS classes or environmental matrices. However, integrated approaches combining adsorption, photocatalysis, electrochemistry, plasma treatment, hydrothermal destruction, and biological transformation are increasingly achieving high degradation and defluorination efficiencies, often exceeding 90%, while improving sustainability, selectivity, and scalability for real-world PFAS remediation.

Recent advances in PFAS removal and defluorination technologies demonstrate a clear transition from conventional separation methods toward integrated destructive processes capable of cleaving the highly stable C-F bond. The reviewed technologies (Supplementary Table S1) operate under diverse conditions, ranging from ambient biological and adsorption systems to energy-intensive hydrothermal, supercritical water oxidation, plasma, electrochemical, and mechanochemical processes. Photocatalytic technologies typically require UV, visible, vacuum, or simulated solar irradiation, and employ engineered semiconductors or heterojunction catalysts to enhance charge separation and reactive species generation [266,281,282]. Electrochemical technologies rely on controlled electrical potentials using advanced electrode materials such as BDD, Ti_4O_7 Magnéli phase anodes, CeO_2 electrodes, and modified cathodes to promote oxidative or reductive C-F bond cleavage [262,263,283,321]. In contrast, hydrothermal alkaline treatment and supercritical water oxidation require high temperatures and pressures to achieve rapid PFAS mineralization [152,270,271], whereas mechanochemical degradation utilizes high-energy ball milling to induce defluorination under solvent-free conditions [272,273]. Biological and adsorption-assisted approaches generally operate under mild conditions but primarily facilitate PFAS capture or partial transformation rather than complete destruction [279,342].

Energy demand varied substantially among the technologies. Adsorption, biological remediation, and iron-mediated reduction exhibited the lowest energy requirements because they operate under ambient conditions with minimal external energy input [274,342]. Photocatalytic and UV/sulfite-based advanced reduction processes generally required moderate energy due to continuous irradiation, although solar-driven photocatalysts, catalyst engineering, and integrated adsorption–photocatalysis strategies improved reaction efficiency and reduced overall energy demand [276,284,285]. Electrochemical technologies also demonstrated low-to-moderate or moderate energy consumption depending on electrode composition, current density, and reactor configuration, with recent studies emphasizing low-energy operation through catalyst engineering, pulsed electrolysis, and

electrosorption [285,286,322]. Conversely, plasma, ultrasonication, hydrothermal alkaline treatment, supercritical water oxidation, and mechanochemical processes remained among the most energy-intensive technologies because of their high electrical, thermal, or mechanical energy requirements [269–271,287]. Nevertheless, direct comparisons remain difficult because most studies did not report standardized energy metrics such as kWh m^{−3} treated water or kWh kg^{−1} PFAS destroyed.

The major advantages of emerging PFAS destruction technologies include improved degradation efficiency, enhanced defluorination, and an increasing emphasis on process sustainability through catalyst engineering and hybrid process integration. Several studies successfully combined adsorption with destructive treatment, including adsorption–photocatalysis, adsorption–electroreduction, adsorption–plasma, and adsorption–UV/sulfite systems, thereby concentrating dilute PFASs prior to destruction and improving treatment efficiency [276,278,288,289]. Furthermore, advanced catalyst designs, including Z-scheme heterojunctions, defect-engineered semiconductors, metal-doped photocatalysts, zero-valent metal composites, and non-precious electrocatalysts, have significantly enhanced charge separation, reactive species generation, and C-F bond activation [266,290,291,322]. Continuous-flow hydrothermal alkaline treatment and supercritical water oxidation further demonstrated the feasibility of treating real PFAS-contaminated waters, representing significant progress toward practical implementation [270,271].

Despite these advances, several limitations remain. Most technologies have only been validated at the laboratory scale using synthetic water and relatively high PFAS concentrations, with limited evaluation in complex environmental matrices containing natural organic matter and competing ions. Catalyst deactivation, electrode fouling, regeneration requirements, corrosion under harsh operating conditions, and the formation of transformation products continue to challenge practical implementation [283,292,293]. In addition, many studies report high degradation efficiencies without demonstrating complete mineralization or fluorine mass balance closure, highlighting the need for comprehensive evaluation using fluoride measurements, targeted LC-MS/MS, and high-resolution mass spectrometry [292]. Standardized economic analyses, long-term stability assessments, and energy consumption reporting are also lacking across most emerging technologies.

Based on technology readiness assessments, most catalyst-based photocatalytic, electrochemical, biological, and nanomaterial-assisted systems remain at the laboratory proof-of-concept or laboratory validation stages (TRL 2–4). Technologies incorporating continuous-flow reactors and treatment of real contaminated waters, including HALT and SCWO, demonstrate higher maturity (TRL 5–6), reflecting their progression toward pilot-scale implementation [152,270,271]. TRL values were assigned by the authors based on the NASA/European Commission TRL framework using the maturity of the technology reported in each study (laboratory validation, relevant-environment testing, pilot-scale demonstration, and commercial deployment) [343–346]. These values are interpretative assessments and were not necessarily reported by the original publications. Consequently, future PFAS remediation will likely rely on integrated treatment trains that combine efficient PFAS concentration with robust destructive technologies capable of achieving complete defluorination while minimizing energy consumption and operational costs.

11. Hybrid Bio-Physical Remediation Systems

This section highlights integrated and hybrid PFAS remediation strategies that combine biological and physicochemical processes to enhance degradation performance. It focuses on adsorption–biodegradation coupling, bioelectrochemical systems that stimulate microbial activity for PFAS transformation, and photo-biocatalytic platforms, which inte-

grate light-driven catalysis with biological processes to improve defluorination potential and treatment efficiency.

11.1. Adsorption–Biodegradation Coupling Strategies

Recent studies have increasingly demonstrated the potential of adsorption–biodegradation coupling strategies as sustainable approaches for PFAS remediation by integrating contaminant immobilization with biologically mediated transformation processes. In particular, a study highlighted how engineered biochar can serve not only as an effective adsorbent for PFBS sequestration but also as a supportive habitat for microbial communities that facilitate subsequent transformation processes [280]. Similar synergistic behaviour has been observed in hydrogel–biochar composite systems, as demonstrated in another study, where adsorption reduced contaminant mobility while simultaneously promoting microbial degradation through enhanced bioavailability and microbial attachment [347]. Although focused on microcystin, these studies provide valuable conceptual frameworks applicable to PFAS remediation. Furthermore, a study demonstrated that environmental redox conditions and electron acceptors strongly influence microbial PFAS removal efficiency, emphasizing the importance of coupling sorptive materials with optimized biogeochemical conditions [348]. Advances in bioelectrochemical systems were further illustrated by a report, which showed that electroactive microbial systems can enhance PFAS detoxification while reducing secondary environmental risks [349].

11.2. Bioelectrochemical Systems for PFAS Transformation

Bioelectrochemical systems have emerged as promising technologies for PFAS transformation by integrating microbial metabolism, electrochemical stimulation, and conductive material interfaces to enhance contaminant removal and defluorination processes. Pilot-scale microbial fuel cells have demonstrated the capacity to simultaneously reduce PFASs and other contaminants of emerging concern while generating bioelectricity, highlighting the multifunctional potential of bioelectrochemical treatment systems in water reuse applications [350]. Similarly, a report showed that electroactive microbial communities could maintain stable electricity generation despite continuous exposure to PFOA, indicating microbial resilience and adaptation under PFAS stress conditions [351]. The integration of electrical stimulation into engineered treatment systems has also shown significant benefits for PFAS removal. For example, a study demonstrated that the application of a direct current enhanced redox reactions and improved PFAS removal efficiency within wetland environments [352]. Likewise, a report revealed that electrochemical assistance could alleviate PFAS-induced microbial inhibition by influencing extracellular polymeric substance interactions and reconstructing microbial metabolic pathways, thereby improving reactor performance and microbial stability [353]. Furthermore, a study emphasized the critical role of conductive material–microbe interactions in facilitating electron transfer and promoting deeper anaerobic PFAS defluorination [45].

11.3. Photo-Biocatalytic Degradation Platforms

Photo-biocatalytic degradation platforms are emerging as innovative approaches for the transformation of persistent contaminants by integrating photocatalytic activation with enzymatic catalysis to achieve selective and energy-efficient degradation pathways. Although not directly focused on PFASs, one study demonstrated the potential of coupling photocatalysis with enzymatic reactions to enhance the breakdown and detoxification of highly halogenated organic pollutants [354]. In this system, photocatalytic processes generated reactive species that promoted enzymatic activity and accelerated contaminant transformation, illustrating how combined photo-bio systems can overcome the recalcitrance typically associated with halogenated compounds. Similarly, a report highlighted

the capability of light-assisted enzymatic catalysis to drive oxidative reactions under mild conditions through efficient electron transfer mechanisms [355]. The study demonstrated how photoactivated enzymatic systems can facilitate difficult oxidation reactions while maintaining catalytic specificity and reduced energy demand.

12. Bio-Inspired Catalysis and Synthetic Biology Approaches

12.1. Engineering Microbes for PFAS Degradation

Engineering microbes for PFAS degradation represents a promising frontier in environmental biotechnology, aimed at overcoming the inherent chemical stability and persistence of these compounds [232]. Naturally occurring microorganisms often exhibit limited capacity to transform long-chain PFASs, and complete mineralization is rare [42]. To address limited natural microbial activity, synthetic biology and metabolic engineering approaches are employed to enhance microbial capabilities, either by optimizing native pathways or introducing heterologous enzymes such as dehalogenases, oxygenases, and reductases [232]. These engineered strains can be designed to express multiple catalytic systems simultaneously, enabling sequential or co-metabolic degradation of PFAS precursors and their intermediates [356]. Strategies include increasing enzyme expression levels, improving substrate specificity, and integrating catalytic capacities to facilitate oxidative or reductive transformations [357]. Additionally, microbial consortia can be rationally designed, combining complementary aerobic and anaerobic microbes to achieve more comprehensive degradation than single strains [358]. Advances in gene-editing tools such as Clustered Regularly Interspaced Short Palindromic Repeats (CRISPR)-Cas systems allow precise manipulation of microbial genomes to enhance stress tolerance, cofactor availability, and metabolic flux toward contaminant transformation pathways [359]. These engineered strains can be designed to express multiple catalytic systems simultaneously, enabling sequential or co-metabolic degradation of PFAS precursors and their intermediates, an approach grounded in advances in synthetic biology and metabolic pathway engineering [360].

12.2. Synthetic Microbial Consortia for PFAS Transformation

Recent studies on synthetic microbial consortia have demonstrated growing potential for the biological transformation and remediation of PFASs, particularly through the integration of microbial ecology, synthetic biology, and data-driven approaches. Research showed that cooperative microbial interactions within mixed consortia can improve PFAS removal through combined biosorption and biodegradation mechanisms [361], highlighting the importance of synergistic metabolism in overcoming PFAS persistence. Similarly, a study demonstrated that diverse microbial assemblages possess varying capacities to transform PFASs depending on molecular structure and environmental conditions [74]. Advances in anaerobic systems were further illustrated, revealing that different microorganisms contribute distinct metabolic roles during ether-PFAS defluorination and transformation pathways [43]. Synthetic biology has also emerged as a promising frontier, as demonstrated in a study where cyanobacterial systems were proposed as engineered chassis for targeted PFAS remediation [362]. The incorporation of computational tools is further transforming the field; studies emphasized the use of machine learning and predictive modelling to design optimized microbial consortia with enhanced degradation efficiency and stability [363,364].

13. Research Gaps and Future Perspectives for PFAS Remediation

Despite the significant progress in PFAS remediation technologies, several critical research gaps continue to limit their large-scale and sustainable application. Most current

remediation studies are performed under controlled laboratory conditions using synthetic water matrices and relatively high PFAS concentrations, which do not adequately represent the complexity of real environmental systems such as groundwater, sediments, wastewater, landfill leachates, and AFFF-impacted soils. The presence of competing ions, dissolved organic matter, carbonate species, and co-contaminants often suppresses degradation efficiency and hydrated electron availability, thereby reducing practical performance. Furthermore, many studies focus primarily on legacy PFASs such as PFOA and PFOS, while emerging short-chain PFASs, ether-PFASs (e.g., GenX and ADONA), fluorotelomer compounds, and PFAS precursors remain insufficiently investigated despite their increasing environmental occurrence and mobility.

Another major limitation is the poor understanding of degradation pathways, intermediate transformation products, and long-term toxicity. Although high degradation and defluorination efficiencies have been reported for UV/sulfite [120,302,313], electrochemical [124,321], plasma [267], photocatalytic [264], and hydrothermal systems [271], many processes generate partially fluorinated intermediates whose environmental persistence and toxicity are not fully characterized.

There is also a scarcity of reports needed to improve the selectivity, regeneration capacity, and environmental compatibility of adsorbent materials. While activated carbon, ion exchange resins, and biochar [31,32] demonstrate strong PFAS adsorption, many suffer from saturation, secondary waste generation, and limited efficiency toward ultra-short-chain PFASs. Hybrid systems integrating adsorption with photocatalysis, electrochemical reduction, biodegradation, or plasma treatment offer promising opportunities for simultaneous enrichment and destruction of PFASs, but these integrated systems are still in the early stages of development.

Current biological remediation strategies remain constrained by the limited number of experimentally verified PFAS-defluorinating microorganisms and the incomplete understanding of the enzymatic mechanisms responsible for C-F bond cleavage [365]. Current biological PFAS degradation is limited by the small number of known defluorinating microorganisms and the restricted substrate range of their associated enzymatic pathways [222,365]. Although several C-F bond-cleaving enzymes have been identified, their catalytic efficiency and substrate specificity toward environmentally relevant PFASs remain limited [222]. In addition, despite encouraging laboratory-scale results, significant knowledge gaps remain regarding the long-term performance, scalability, and operational stability of biological PFAS treatment systems under realistic environmental conditions [177]. Many PFAS remediation studies continue to evaluate treatment performance primarily based on the disappearance of parent compounds, with limited characterization of transformation products and insufficient assessment of fluorine mass balance and mineralization, potentially leading to an overestimation of treatment effectiveness [160,173]. Finally, although numerous PFAS remediation technologies have demonstrated promising laboratory-scale performance, relatively little attention has been given to evaluating their environmental sustainability and economic viability for large-scale implementation [177]. These limitations highlight that current PFAS remediation technologies remain constrained by limited validation under environmentally relevant conditions. Consequently, the following future perspectives should be considered to advance PFAS remediation technologies:

1. Future studies should evaluate remediation technologies in real environmental matrices rather than relying predominantly on synthetic laboratory systems. The influence of competing ions, dissolved organic matter, carbonate species, and co-contaminants on treatment performance should be systematically investigated to improve field applicability. Greater attention should be directed toward the remediation of emerging PFASs, including short- and ultra-short-chain PFASs, ether-PFASs (e.g., GenX

and ADONA), fluorotelomer compounds, and precursor substances, which remain considerably less studied than legacy PFASs.

2. Future research should also prioritize the development of next-generation adsorbent materials with enhanced selectivity for both legacy and emerging PFASs, particularly ultra-short-chain compounds, while improving regeneration efficiency and long-term stability. Rational material design through surface functionalization and the incorporation of advanced porous materials, including engineered biochar, metal–organic frameworks, and covalent organic frameworks, may enhance adsorption capacity and reduce secondary waste generation.
3. Further investigation is needed to optimize adsorption–destruction treatment trains that combine adsorptive concentration with subsequent PFAS degradation, thereby improving overall treatment efficiency and minimizing the environmental burden associated with consumed adsorbents. Priority should therefore be given to the discovery of novel defluorinase enzymes and microorganisms capable of degrading a wider range of PFASs, particularly short- and ultra-short-chain compounds.
4. Priority should be given to overcoming the limitations that currently restrict the practical application of biological PFAS remediation. Furthermore, advances in synthetic biology and protein engineering, supported by artificial intelligence for enzyme design and prediction, may enable the development of biocatalysts with enhanced catalytic efficiency and broader substrate specificity.
5. Advances in synthetic biology and protein engineering, supported by artificial intelligence for enzyme design and prediction, may improve catalytic efficiency and broaden substrate specificity.
6. Greater emphasis should be placed on continuous-flow and pilot-scale reactor studies to evaluate long-term performance under realistic environmental conditions.
7. Research should incorporate transformation product identification, fluorine mass balance, and mineralization efficiency alongside parent compound removal to provide a more comprehensive assessment of treatment performance.
8. Life-cycle assessment and techno-economic analysis should be integrated into technology development to determine the environmental sustainability and economic feasibility of large-scale implementation.

14. Conclusions

This review integrates biological and emerging PFAS remediation technologies within a unified framework centred on C-F bond cleavage. Unlike previous studies that treat approaches separately, it emphasizes mechanistic defluorination pathways across biological and physicochemical systems. The synthesis highlights key limitations in achieving complete PFAS destruction and identifies opportunities for developing more efficient and sustainable hybrid remediation strategies. PFAS contamination remains a critical environmental challenge due to their extreme chemical stability and resistance to conventional treatment processes. The strength of the C-F bond limits natural degradation, resulting in long-term persistence and bioaccumulation in water, soil, and biota. Consequently, effective remediation must prioritize both degradation and complete defluorination to avoid the formation of persistent intermediates. Emerging technologies such as electrochemical oxidation, advanced reduction processes, plasma treatment, and catalytic systems show potential for breaking C-F bonds and achieving mineralization. Comparative evaluation revealed that adsorption offers the greatest scalability and lowest by-product formation but negligible defluorination, whereas thermal treatment achieves the highest PFAS destruction and defluorination efficiencies at the expense of substantial energy demand and cost. Electrochemical, hydrated electron, and plasma-based technologies provide strong destructive

performance with varying scalability constraints, while photocatalytic, mechanochemical, and biological/bio-based approaches represent promising alternatives that balance treatment efficiency, energy requirements, and environmental sustainability but generally exhibit lower mineralisation rates or limited large-scale implementation. However, scalability and efficiency remain limiting factors, necessitating the continued development of integrated, cost-effective defluorination strategies.

Supplementary Materials: The following supporting information can be downloaded at: <https://www.mdpi.com/article/10.3390/separations13070206/s1>, Table S1: Detailed Comparison of PFAS Defluorination Technologies.

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Abbreviations

The following abbreviations are used in this manuscript:

AOP	Advanced oxidation process
BDD	Boron-doped diamond
AFFFs	Aqueous film-forming foams
C-F	Carbon–fluorine
CIC	Combustion ion chromatography
CRISPR	Clustered regularly interspaced short palindromic repeats
EBCT	Empty bed contact time
EtFOSA	N-ethyl perfluorooctane sulfonamidoacetic acid
FOSA	Perfluorooctane sulfonamide
FOSAA	Perfluorooctane sulfonamide acetic acid
FTCA	Fluorotelomer carboxylic acid
FTS	Fluorotelomer sulfonate
FTUCA	Fluorotelomer unsaturated carboxylic acid
GAC	Granular activated carbon
HALT	Hydrothermal alkaline treatment
HRMS	High-resolution mass spectrometry
HTL	Hydrothermal liquefaction
LC-MS/MS	Liquid chromatography–tandem mass spectroscopy
NADH	Nicotinamide adenine dinucleotide
NADPH	Nicotinamide adenine dinucleotide phosphate
nZVI	Nanoscale zero-valent iron
PFAAs	Perfluoroalkyl acids
PFAS	Per- and polyfluoroalkyl substance
PFOA	Perfluorooctanoic acid
PFOS	Perfluorooctane sulfonate
PFSA	Perfluoroalkyl sulfonic acid
SCWO	Supercritical water oxidation
TFA	Trifluoroacetic acid
UV	Ultraviolet
WWTPs	Wastewater treatment plants

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