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DESTROYING THE INDESTRUCTIBLE: STATE OF THE ART APPROACHES TO ENVIRONMENTAL REMEDIATION OF PER AND POLYFLUOROALKYL SUBSTANCES (PFAS)

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ABSTRACT

Per- and polyfluoroalkyl substances (PFAS) are among the most persistent and ubiquitous anthropogenic contaminants due to the exceptional strength of the carbon–fluorine bond. This comprehensive narrative review critically evaluates state-of-the-art remediation technologies for PFAS across aqueous, solid, and subsurface matrices, with emphasis on destructive approaches that achieve mineralization rather than sequestration. Drawing on peer-reviewed literature, regulatory documents, and field demonstrations (primarily 2016–2025), the review examines conventional sorption and pump-and-treat systems, advanced oxidation/reduction processes, thermal and mechanochemical destruction, biological and phytoremediation strategies, pretreatment/concentration technologies, in situ methods, and hybrid systems. Key topics addressed include analytical challenges in verifying true mineralization, byproduct toxicity, cost-effectiveness, performance against short-chain and novel PFAS replacements (e.g., GenX, PFBS), and the regulatory–technology interface at stringent MCLs such as the U.S. EPA's 4 ng L^{-1} threshold. Comparative analysis shows that while sorption technologies enable drinking-water compliance, only high-temperature thermal processes, supercritical water oxidation (SCWO), hydrothermal alkaline treatment (HALT), and optimized reductive/electrochemical systems consistently approach complete defluorination. Field-scale case studies highlight practical viability, yet economic burdens, matrix effects, and the formation of short-chain byproducts remain major limitations. The review identifies research priorities and advocates integrated treatment trains to close the widening gap between regulatory demands and technological capability.

Keywords: Carbon–Fluorine Bond; Per- and polyfluoroalkyl substances (PFAS); PFAS Pre-Treatment; Remediation Technologies; State of the Art.

1.0. Introduction

1.1. The PFAS Crisis: A Regulatory Emergency Confronting a Technology Gap

Per- and polyfluoroalkyl substances (PFAS) constitute one of the most structurally diverse and environmentally recalcitrant chemical families ever synthesized. Defined by the presence of at least one fully fluorinated methyl or methylene carbon atom (OECD/UNEP, 2021), the PFAS umbrella encompasses more than 12,000 individual compounds catalogued in databases such as the U.S. EPA's CompTox Chemicals Dashboard and the OECD Global PFAS list. PFAS derive their extraordinary persistence

from the carbon–fluorine (C–F) bond (a bond dissociation energy of approximately 544 kJ mol^{-1}, among the strongest in organic chemistry), rendering these molecules resistant to hydrolysis, photolysis, biodegradation, and thermolysis under ambient environmental conditions. This foundational stability, deliberately engineered for industrial utility, is precisely what transforms PFAS into a class of contaminants that pose an extreme challenge to environmental remediation science.

Structurally, PFAS span a vast chemical space. Legacy long-chain perfluoroalkyl acids (PFAAs) (including perfluorooctanoic acid (PFOA) and perfluoro octane sulfonate (PFOS)) are the most extensively studied subclass, characterized by fully fluorinated alkyl chains (≥C7 for carboxylic acids, ≥C6 for sulfonates) conferring extreme hydrophobicity, surfactancy, and bioaccumulation potential. Polyfluoroalkyl substances, by contrast, retain at least one non-fluorinated carbon, rendering them partially susceptible to transformation yet capable of serving as precursors to terminal perfluoroalkyl acid metabolites through environmental and biological oxidation pathways. Fluorotelomer-based compounds, perfluoroalkyl ether carboxylic acids (PFECAs), such as hexafluoropropylene oxide dimer acid (HFPO-DA, commercially known as "GenX"), perfluoroalkyl sulfonamides, and ultra-short-chain variants (C2–C3) further complicate the regulatory and analytical landscape. This structural heterogeneity (chain length, functional group identity, degree of fluorination, and branching architecture) means that no single remediation mechanism is likely to achieve broad-spectrum destruction or removal across the full PFAS universe, a challenge that defines the current technology gap.

The environmental ubiquity of PFAS is evident and global in scope. Decades of industrial manufacturing, aqueous film-forming foam (AFFF) deployment, and widespread consumer product use have distributed PFAS across all major environmental compartments. Detections have been confirmed in surface waters, groundwater, drinking water, soils, sediments, atmospheric particulates, precipitation, remote Arctic ice cores, deep-ocean water columns, and the tissues of wildlife and humans across every inhabited continent (Evich et al., 2022). The U.S. Geological Survey's 2023 national reconnaissance of drinking water estimated that approximately 45% of U.S. tap water sources contain detectable PFAS, with contamination distributed across both urban and rural systems (Smalling et al., 2023). Critically, PFAS exhibit long-range atmospheric and oceanic transport, meaning contamination is not limited to proximity to point sources. This reality renders localized remediation strategies insufficient as standalone policy responses.

Regulatory pressure has escalated in response to mounting epidemiological and toxicological evidence linking PFAS exposure to thyroid dysfunction, immunosuppression, dyslipidemia, developmental toxicity, and increased risk of several carcinomas at nanogram-per-litre exposure concentrations (National Toxicology Program, 2016; Fenton et al., 2021). In April 2024, the U.S. EPA finalized the first-ever National Primary Drinking Water Regulation for six PFAS compounds under the Safe Drinking Water Act, establishing maximum contaminant levels (MCLs) of 4 ng L^{-1} for PFOA and PFOS individually, "a threshold two orders of magnitude below what conventional water

treatment infrastructure was designed to achieve" (U.S. Environmental Protection Agency, 2024). Simultaneously, the European Union's universal PFAS restriction proposal under REACH, submitted in 2023 by five Member State competent authorities, seeks to phase out the manufacture and use of approximately 10,000 PFAS across virtually all non-essential applications. This is the highest chemical restriction in EU regulatory history (ECHA, 2023). Stockholm Convention listings of PFOS (2009) and PFOA (2019) under Annex A have further internationalized the regulatory mandate for elimination (United Nations Environmental Program 2009, 2019).

The central drive for this review is the widening gap between escalating regulatory ambition and current remediation capability. Existing water treatment technologies, such as granular activated carbon (GAC), anion exchange resins (AER), and high-pressure membrane systems, concentrate or sequester PFAS rather than destroy them, thereby generating PFAS-laden waste that is merely translocated from one environmental compartment to another.

This narrative review aims to provide a comprehensive synthesis of state-of-the-art PFAS remediation technologies, emphasizing destructive approaches that achieve mineralization or near-complete defluorination while addressing practical implementation barriers. The scope encompasses both ex situ and in situ methods for aqueous, solid, and subsurface matrices; pretreatment and concentration technologies; analytical challenges in verifying destruction; byproduct formation and toxicity; cost-effectiveness; the regulatory–remediation interface; and emerging challenges posed by short-chain and novel PFAS replacements. Literature coverage prioritizes peer-reviewed studies, technical reports, and field demonstrations published between 2016 and 2025, selected for relevance, rigour, and representation of diverse matrices and scales. Unlike prior reviews that focus narrowly on specific technologies or matrices, this work integrates comparative performance metrics, real-world case studies, and forward-looking research priorities to guide practitioners, regulators, and researchers toward sustainable solutions. The review explicitly discusses novel and short-chain PFAS (e.g., GenX/HFPO-DA, PFBS, F-53B), which are proliferating as long-chain compounds are phased out, yet pose distinct mobility, sorption, and destruction challenges (Kannan et al., 2025).

1.2. Sources, Partitioning, Persistence, and Precursor Dynamics of PFAS in the Environment

PFAS contamination originates from both point and diffuse sources. Point sources dominate the documented literature, with the historical and ongoing use of aqueous film-forming foam (AFFF) at military installations, civilian airports, and firefighting training areas being one of the largest documented drivers of soil/groundwater contamination globally (Anderson et al., 2016; Leeson et al., 2021). Additional industrial point sources include fluoropolymer manufacturing facilities (e.g., Chemours Fayetteville Works), electroplating operations, textile and carpet treatment plants, and semiconductor fabrication (U.S. Environmental Protection Agency, 2024). Diffuse sources include land application of biosolids, with PFAS detected in agricultural soils at 0.1–10,000 ng/g, depending on sludge-loading history (Venkatesan & Halden, 2023; Pepper et al., 2021). Consumer product lifecycles (food packaging, non-stick cookware, personal care products) further contribute PFAS to municipal wastewater influent, where conventional WWTPs act as redistribution

hubs, transferring PFAS from aqueous to biosolid phases and releasing a portion of them in effluent (Houtz et al., 2016; ITRC, 2022).

This contamination is influenced by environmental partitioning behaviour governed by chain length, functional head group, and co-solute chemistry. Long-chain perfluoro carboxylic acids (PFCAs, C≥8) and perfluoro sulfonic acids (PFSAs, C≥6) exhibit strong sorption to soil organic carbon and mineral surfaces, with log K_oc values of 2.0–4.5 (Guelfo & Higgins, 2013). Electrostatic interactions with positively charged mineral surfaces (iron oxyhydroxides, aluminium oxides) contribute significantly to subsurface retardation, often independently of organic carbon content. This insight has reshaped transport modelling by highlighting the need to incorporate mineral-specific and pH-dependent electrostatic terms beyond simple hydrophobic partitioning (Lyu et al., 2020; Evich et al., 2025). In contrast, short-chain replacements (C4–C6, e.g., PFBS, PFBA) display lower K_oc values (generally log K_oc < 2.0), resulting in high aqueous mobility and a retardation factor approaching 1 (Guelfo & Higgins, 2013). Air-water interfacial partitioning at the vadose zone capillary fringe is now recognised as a dominant retardation mechanism for ionic PFAS, with interfacial sorption coefficients (K_ia) often exceeding those for bulk solid-phase sorption (Brusseau & Guo, 2021; Lyu et al., 2022).

The strength of the C–F bond is the bedrock of the PFAS persistence. Perfluorinated terminal compounds resist biodegradation, hydrolysis, photolysis, and oxidation under ambient environmental conditions. No known microorganism can defluorinate a fully perfluorinated carbon chain under standard environmental conditions (aerobic or anaerobic), although limited defluorination has been observed in engineered laboratory systems with enriched cultures (Wackett, 2024; Huang et al., 2025). Abiotic persistence is further reinforced by the thermodynamic stability of the fluorinated carbon backbone, which resists nucleophilic and electrophilic attack. Estimated environmental half-lives for PFOS and PFOA in soil matrices extend to decades or longer with negligible transformation rate (Interstate Technology & Regulatory Council, 2026; Evich et al., 2022).

The persistence of the C – F bond is compounded by precursor transformation. This is among the most underappreciated fate processes for PFAS. Polyfluoroalkyl precursors like fluorotelomer alcohols (FTOHs), fluorotelomer sulfonamides (FTSAs), N-substituted perfluoroalkyl sulfonamidoethanols (N-EtFOSE, N-MeFOSE), and zwitterionic/cationic PFAS present in AFFF undergo biotransformation to terminal perfluorinated acids via sequential oxidation and hydrolysis pathways. Aerobic and anaerobic microbial consortia mediate partial defluorination of the telomer (non-perfluorinated) region while leaving the perfluorinated core intact, producing PFCA homologs (e.g., 6:2 FTOH transform to → PFHxA, PFPeA and a minor amount of PFBA) (Liu & Mejia Avendano, 2013; Zhang et al., 2020). This extensive precursor reservoir means that monitoring programs targeting only terminal PFAS significantly underestimates total PFAS loading and long-term risk. Extractable organofluorine (EOF) and total oxidizable precursor (TOP) assays have therefore become essential site characterization tools; they routinely reveal 2–10× (or greater)

higher fluorine mass than targeted LC-MS/MS analysis alone (Göckener et al., 2021; Lange et al., 2024; Ateia et al., 2023).

2.0. Analytical Challenges of PFAS Monitoring in Remediation

A vital yet often overlooked aspect of PFAS remediation is the analytical challenge of accurately quantifying destruction efficiency and verifying true mineralization. Targeted liquid chromatography–tandem mass spectrometry (LC-MS/MS) methods (e.g., EPA Methods 533, 537.1, and 1633) provide high sensitivity for regulated compounds at ng L^{-1} levels but capture only a small fraction of the >12,000 known PFAS structures, missing precursors, transformation products, and nontarget species (Ateia et al., 2023; Phan et al., 2026). The Total Oxidizable Precursor (TOP) assay and Extractable Organofluorine (EOF) measurements are important for estimating total PFAS burden and routinely reveal 2–10× higher fluorine mass than targeted analyses (Ateia et al., 2023). However, TOP assay results exhibit inter-laboratory variability due to a lack of standardization, matrix interferences, and incomplete oxidation of certain precursors. Verification of mineralization requires robust fluorine mass balance (fluoride ion measurements plus residual PFAS), high-resolution mass spectrometry (HRMS) for nontargeted byproducts, and careful control of background contamination from labware and reagents. Post-degradation matrix effects (e.g., ion suppression from surfactants or salts in electrochemical or plasma systems) further complicate quantification, often requiring isotopically labelled standards and method validation with recovery matrices (Battelle, 2025).

These analytical limitations directly impact regulatory compliance assessment at the 4 ngL^{-1} MCLs and inform the need for complementary tools to confirm that "non-detect" truly implies destruction rather than transformation into mobile, unregulated short-chain byproducts.

2.1. Conventional Remediation of PFAS: Approaches and Critical Limitations

Conventional remediation strategies, while operationally established, are fundamentally limited in their capacity to achieve permanent PFAS destruction, constituting containment or transfer mechanisms rather than effective mineralization solutions. The most widely applied approaches include pump-and-treat (P&T) for hydraulic containment, sorption-based above-ground treatment using granular activated carbon (GAC) and ion-exchange resins (IEX), landfilling of contaminated solids, and high-temperature incineration.

Pump-and-treat is one of the most widely deployed hydraulic containment strategies for PFAS-impacted groundwater. The approach involves the continuous extraction of contaminated groundwater followed by above-ground treatment, typically via sorptive media. While P&T effectively controls plume migration and reduces receptor exposure, it offers no destructive capacity. In P&T, PFAS molecules are captured and concentrated but remain chemically unchanged. The technology is further limited by the "tailing and rebound" phenomenon, in which desorption from aquifer solids sustains dissolved-phase concentrations even after prolonged extraction, often extending operational timelines to decades and imposing substantial energy and cost burdens (Interstate Technology & Regulatory Council, 2022; Newell et al., 2025). For short-chain PFAS such as PFBS and PFBA (which are increasingly prevalent following the regulatory phase-out of long-chain compounds), P&T efficiency declines further due to weaker sorption to aquifer materials and greater hydraulic mobility. The extracted water still requires downstream treatment and

disposal, effectively displacing rather than resolving the contamination problem.

Complementing P&T are the predominant above-ground treatment technologies for PFAS-laden water, GAC and IEX resins, both of which function exclusively through sorption. GAC removes PFAS primarily via hydrophobic interactions, supplemented by electrostatic forces, whereas single-use or regenerable anion-exchange resins exploit strong electrostatic attraction to the anionic head groups of PFAS. Both technologies can achieve substantial reductions in dissolved PFAS concentration and are endorsed in U.S. EPA drinking water treatment guidance (U.S. EPA, 2021).

However, neither GAC nor IEX destroys PFAS; they transfer contamination from the aqueous phase to a solid matrix. Spent GAC and exhausted IEX resins accumulate high PFAS loads and require either thermal regeneration or disposal as hazardous waste. Thermal reactivation of GAC at ~850°C is energy-intensive, only partially effective for PFAS mineralization, and can generate volatile fluorinated intermediates of incomplete combustion byproducts (Gagliano et al., 2023; Didenko et al., 2024). Regeneration of IEX resins produces a highly concentrated PFAS brine, a secondary waste stream with no straightforward destruction pathway, which itself requires further treatment. Short-chain PFAS exhibit significantly reduced affinity for both GAC and standard IEX media, resulting in earlier breakthrough, shorter media lifespans, and increased operational costs and waste generation (Appleman et al., 2023; Zeng et al., 2020; ITRC, 2022).

Landfill disposal of PFAS-contaminated solids is widely practised but fundamentally incompatible with PFAS destruction objectives. PFAS leach from landfill cells into leachate, with studies consistently detecting PFAS in landfill leachate at concentrations orders of magnitude above drinking water guidelines (Tolaymat et al., 2023). Modern lined landfills with leachate collection partially mitigate migration, but liner integrity degrades over decadal timescales, and collected leachate requires treatment — again cycling back to GAC/IEX without destruction. Landfilling thus constitutes deferral of the contamination problem, transferring liability to future generations while PFAS remain chemically unaltered within the waste matrix.

High-temperature incineration has historically been presumed capable of PFAS mineralization. However, emerging evidence has challenged this assumption. Complete defluorination of PFAS requires sustained temperatures exceeding 1,100°C with adequate residence time; operational incinerators frequently fail to maintain these conditions uniformly, resulting in incomplete destruction and the *de novo* formation of highly toxic short-chain fluorinated byproducts, including perfluoroalkyl carboxylic acids, tetrafluoromethane (CF_4), and hexafluoropropylene (Bjorklund et al., 2023; Weitz et al., 2024).

Beyond aqueous-phase applications, conventional soil remediation includes excavation and off-site disposal; soil washing (which separates fines and concentrates PFAS in a smaller volume for subsequent treatment); soil flushing; and in-place immobilization using amendments such as modified clays, biochar, or cementitious materials. These approaches are matrix-dependent and often transfer rather than destroy contamination, with soil washing

costs ranging from $350 to $650 per ton, depending on fines content and throughput (ITRC case studies, 2025).

2.2. In Situ Remediation Technologies

While the majority of established PFAS remediation has focused on ex situ treatment, large-scale groundwater plumes and vadose-zone contamination at AFFF release sites often render pump-and-treat impractical due to long decades of operation and high costs. In situ technologies offer promising alternatives for plume containment and source-zone treatment without extensive infrastructure.

Colloidal activated carbon (CAC) injection creates permeable reactive barriers by coating aquifer solids with sub-micron activated carbon particles. This approach rapidly immobilizes both long- and short-chain PFAS through combined hydrophobic and electrostatic sorption while preserving aquifer permeability. Field demonstrations at sites such as Grayling Army Airfield (Michigan) achieved PFAS reductions to below detection limits within one month, with sustained performance exceeding 4–7 years and no rebound observed (REGENESIS, 2023; ITRC, 2025). Cost analyses indicate that in-situ CAC is less than one-third the long-term expense of pump-and-treat while producing zero secondary waste streams.

In situ chemical oxidation (ISCO) using activated persulfate can transform precursors but often generates more mobile short-chain PFAS under alkaline conditions and shows limited direct defluorination of terminal PFAAs; acidic thermal activation improves outcomes but is logistically challenging at scale (U.S. EPA, 2025).

In situ thermal treatment (ISTT), including electrical resistance heating (ERH), thermal conduction heating (TCH), and steam-enhanced extraction (SEE), volatilizes and mobilizes PFAS for capture and above-ground destruction. These methods achieve high removal efficiencies in unsaturated soils but require energy input and vapour-phase treatment.

In situ stabilization/immobilization with amendments (e.g., organo-clays or biochar) provides long-term sequestration but does not destroy PFAS and may require performance monitoring for longevity. Collectively, in situ approaches are best suited for plume management and source control where ex situ excavation is infeasible, often serving as a bridge to more permanent destructive technologies.

2.3. Advanced Oxidation and Reduction Processes for PFAS Remediation

Advanced oxidation and reduction processes (AOPs/ARPs) are arguably the most promising class of destructive technologies for PFAS, aiming for actual mineralization or near-complete defluorination rather than mere transfer or containment. The principal approaches include electrochemical oxidation, sonochemical degradation, photocatalysis, sulfate radical-based oxidation, and direct reductive defluorination, with hybrid systems emerging as the current frontier.

Electrochemical oxidation (EO) is currently the most scalable AOP for PFAS. Boron-doped diamond (BDD) electrodes generate hydroxyl radicals (•OH) and enable direct anodic oxidation, achieving >99% mineralization of PFOA and PFOS. Recent studies confirm that BDD electrodes deliver high removal rates (96–99%) for both short- and long-chain PFAS, outperforming alternatives such as Ti_4O_7 anodes at circumneutral pH. The key mechanism involves direct electron transfer, which

initiates sequential C–F bond cleavage via the well-established "unzipping pathway," although the energy consumption (~50–200 kWh/m³) remains a bottleneck (Mirabediny et al., 2023).

Sonochemical degradation utilizes acoustic cavitation (typically 200–1000 kHz) to generate localized hotspots (~5000 K, ~500 atm) that pyrolyze PFAS molecules at air–water bubble interfaces where amphiphilic PFAS preferentially accumulate. This process achieves defluorination efficiencies of 60–90% (and often higher in optimized systems) for PFOA within hours. However, the extreme energy intensity (~1000 kWh/m³) renders standalone sonolysis economically unfeasible at scale. Hybrid sono-electrochemical systems have demonstrated synergistic benefits, reducing energy demand by ~30% through cavitation-enhanced mass transfer combined with anodic oxidation (Awoyemi et al., 2024).

Photocatalysis, particularly UV/sulfite and UV/iodide systems, dominates reductive approaches. Sulfite generates hydrated electrons (e^-_aq, E° = −2.9 V), the most potent reductant for C–F bond cleavage, enabling near-complete defluorination (>85%) across diverse PFAS structures, including short-chain variants (Tan et al., 2025). Heterogeneous photocatalysts (Bi-based oxides, In_2O_3, g-C_3N_4) under visible light suffer from low quantum yields and catalyst fouling, although indium-based materials show exceptional performance for PFOS via ligand-to-metal charge transfer (LMCT) under UV irradiation.

Sulfate radical-based AOPs (SR-AOPs) generate highly oxidizing sulfate radicals ($SO_4^{\bullet-}$, E° = 2.5–3.1 V) through thermal, photolytic, or transition-metal activation of persulfate (PS) or peroxymonosulfate (PMS). These radicals attack PFAS primarily through electron transfer rather than hydrogen abstraction, preferentially cleaving the carboxylate or sulfonate head group and initiating stepwise chain shortening. While UV/persulfate systems commonly achieve >90% PFOA degradation, complete mineralization is challenging due to the accumulation of recalcitrant short-chain PFCAs (C2–C4), with defluorination efficiencies typically limited to 40–60% (Khalili et al., 2025). Heterogeneous photocatalytic activation (e.g., g-C_3N_4/persulfate under visible light) improves efficiency but faces scalability challenges.

In contrast, reductive defluorination pathways directly target the strong C–F bonds using hydrated electrons (e_aq^-, E° = −2.87 V) or hydrogen radicals generated via UV photolysis of sulfite/iodide, sonochemistry, or electrochemical reduction. The hydrated electron attacks the C–F bond dissociatively, releasing fluoride and forming a carbon-centred radical that enables chain degradation. UV/sulfite systems at alkaline pH achieve near-complete defluorination of both long- and short-chain PFAS (Tan et al., 2025), outperforming oxidative routes in fluoride recovery. Electrochemical reductive systems using Ti_4O_7 cathodes or indium electrodes similarly deliver high defluorination yields under controlled potentials. The principal advantage of reductive systems is their ability to degrade the entire perfluorocarbon chain rather than only the ionic head group.

Hybrid systems (sono-electrochemical, photo-electrochemical, and combined oxidative–reductive configurations) continue to advance the field by exploiting

complementary mechanisms to lower overall energy demand while progressing toward full mineralization. Nevertheless, real-world matrix complexity, natural organic matter, chloride, and competing anions pose hurdles to the transition from laboratory success to field-scale implementation.

A persistent challenge across AOPs/ARPs is incomplete mineralization, which can generate toxic fluorinated intermediates and short-chain byproducts (e.g., trifluoroacetic acid and perfluoro butanoic acid) that are more mobile and potentially equally or more bioavailable than parent compounds. These transformation products require careful monitoring via HRMS and fluorine mass balance to ensure regulatory compliance and prevent risk shifting (Björklund et al., 2023; Tan et al., 2025).

2.4. Thermal and Mechanochemical Destruction of PFAS: State-of-the-Art

Thermal and mechanochemical technologies are some of the most effective destructive approaches currently available for achieving near-complete mineralization of PFAS. These include high-temperature incineration, smoldering combustion, mechanochemical degradation, supercritical water oxidation (SCWO), and hydrothermal alkaline treatment (HALT). Each method offers distinct advantages depending on the matrix and scale, yet all involve important trade-offs in energy use, engineering requirements, and operational feasibility.

High-temperature incineration is the most established thermal technology for the destruction of PFAS. Complete mineralization requires sustained temperatures exceeding 1,100°C with residence times greater than 2 seconds in the post-combustion zone, coupled with rapid quenching to prevent reformation of fluorinated byproducts such as PFOA or perfluoro olefins (Winchell et al., 2024). Optimal conditions include excess oxygen, temperatures of at least 1,200°C for recalcitrant long-chain compounds, and robust off-gas scrubbing to capture HF and particulate fluorine. Rotary kilns and liquid injection incinerators are the preferred configurations for treating PFAS-laden solids and liquids. Failure to maintain these stringent conditions can lead to incomplete destruction and the generation of toxic byproducts, raising serious regulatory concerns.

Smoldering combustion (SC) has emerged as a promising alternative for both *in situ* and *ex situ* treatment. This self-sustaining, flameless oxidation process propagates through porous PFAS-contaminated media, achieving peak temperatures of 700–1,000°C at the smoldering front. Studies have demonstrated >99% destruction of PFOA, PFOS, and short-chain PFAS in contaminated soils and AFFF-impacted materials (Duchesne et al., 2020; Fournie et al., 2023). Its self-sustaining nature significantly reduces energy input compared with conventional incineration. However, complete mineralization is matrix-dependent, and heterogeneous temperature distribution at the field scale requires careful engineering controls.

Mechanochemical degradation (MCD) offers a non-thermal route that exploits high-energy ball milling to induce tribochemical reactions. By co-milling PFAS-contaminated solids with reagents such as CaO, Al_2O_3, or $K_2S_2O_8$, localized conditions are generated that efficiently cleave C–F bonds, yielding stable end-products such as CaF_2 and CO_2. Recent work has achieved >99.9% defluorination of PFOS and PFOA within hours using suitable co-reagents (Gobindlal et al., 2023). MCD is particularly well suited for solid-phase waste, including spent granular activated carbon. Challenges in

scalability and energy consumption at industrial throughput persist, although continuous milling systems are under active development.

Supercritical water oxidation (SCWO) operates above water's critical point (374°C, 22.1 MPa), enabling complete miscibility of organics and oxygen for rapid, homogeneous oxidation. At 450–600°C and 23–28 MPa, the process achieves near-complete mineralization of PFAS within seconds to minutes, converting C–F bonds to HF/F^-, CO_2, and H_2O. Optimized SCWO conditions have demonstrated >99.99% destruction efficiency for AFFF-contaminated streams (Rosansky et al., 2024). The technology excels at treating high-concentration liquid wastes to regulatory non-detect levels, but it is energy- and capital-intensive, with HF-induced corrosion of reactor materials presenting a major engineering challenge.

Hydrothermal alkaline treatment (HALT) combines moderate temperature and pressure with alkaline reagents (typically NaOH or KOH) in subcritical water (250–350°C, 5–20 MPa) to promote nucleophilic defluorination. Hydroxide ions attack the carbon backbone, progressively cleaving C–F bonds and generating fluoride salts, along with short-chain organics that undergo further oxidation. HALT has achieved 90–99% defluorination of complex PFAS mixtures in foam concentrates at 350°C with 1 M NaOH in under 90 minutes (Lyu et al., 2024). Operating at significantly lower temperatures than SCWO, it reduces energy demand and corrosion severity while maintaining high destruction efficiency. Ongoing optimization focuses on alkali concentration, temperature, and residence time for real-world matrices such as groundwater concentrates and still bottoms.

These technologies can achieve near-complete PFAS mineralization under optimized laboratory or pilot conditions. However, each presents distinct trade-offs in energy demand, scalability, matrix compatibility, and byproduct management that must be carefully evaluated for site-specific implementation. High-temperature incineration and related thermal methods incur substantial capital and operating costs, often exceeding those of sorption alone when off-gas scrubbing and ash disposal are included (deSilva & Deshmukh, 2025).

Field-scale validation of smoldering combustion and SCWO includes successful demonstrations on AFFF-impacted soils and concentrates, achieving >99% destruction (Duchesne et al., 2020; Rosansky et al., 2024). A notable SCWO field deployment by the U.S. Air Force at Peterson Space Force Base using the PFAS Annihilator® system verified ≥3-log reduction across multiple AFFF formulations under real-world conditions (Revive Environmental, 2025).

2.5. Biological and Phytoremediation Approaches for PFAS Remediation

Biological and phytoremediation approaches offer low-energy, eco-friendly pathways for addressing PFAS contamination. However, they currently face significant barriers to kinetic, completeness, and scalability compared to physicochemical destruction technologies.

Microbial defluorination is the holy grail of PFAS biodegradation. *Acidimicrobium* sp. strain A6 performs reductive defluorination of PFOA and PFOS under anaerobic conditions, cleaving C–F bonds via a proposed radical mechanism involving a reductive dehalogenase. This is the strongest direct evidence of microbial defluorination to

date (Huang & Jaffe, 2019; Huang et al., 2024). More recently, soil microbial consortia enriched under electron-donor-amended conditions have shown partial defluorination of short-chain PFAS (C4–C6), with measurable fluoride release (Tang et al., 2024). Cytochrome P450 enzymes and oxidoreductases have been implicated in attacking perfluoroalkyl ether carboxylic acids (PFECAs), expanding the substrate range beyond legacy compounds.

Despite these advances, the inherent recalcitrance of the C–F bond imposes critical limitations on biodegradation. Defluorination rates are extremely slow, with half-lives of months even under optimized laboratory conditions, rendering them impractical at the field scale. Most pathways yield incomplete mineralization, producing shorter chain perfluorinated intermediates that are equally or more mobile and bioavailable. Additional constraints include toxicity to microbial communities at environmentally relevant PFAS concentrations, the lack of well-characterized catabolic genes (which hinders effective bioaugmentation), and matrix interference, where soil organic matter and competing electron acceptors suppress defluorinating activity.

Phytoremediation primarily drives PFAS removal through phytoextraction, involving root uptake and translocation to shoots. High-transpiration species such as *Festuca arundinacea*, *Helianthus annuus*, and *Lolium perenne* exhibit measurable shoot bioconcentration factors (BCF > 1) for short-chain PFAS (PFBA, PFHxA), while long-chain compounds (PFOS, PFOA) are largely sequestered in roots (Gobelius et al., 2017 in Adu et al., 2023). Stimulation of the rhizosphere microbiome may provide indirect co-metabolic benefits. Nevertheless, phytoremediation is mainly a containment or extraction strategy rather than true mineralization. Harvested biomass constitutes PFAS-laden waste that requires high-temperature incineration (>1,100°C). Low BCF values for long-chain PFAS, combined with the decadal timeframes needed for meaningful mass removal, severely limit its practicality at many contaminated sites.

Bio-electrochemical systems (BES), including microbial electrolysis cells (MECs) and microbial fuel cells (MFCs), are one of the most promising hybrid biological approaches. Cathode-driven reductive defluorination under applied potentials (−0.8 to −1.2 V vs. SHE), combined with electroactive biofilms (Geobacter, Shewanella spp.), enhances electron transfer to PFAS molecules. Defluorination efficiencies of 60–80% for PFOA have been reported in two-chamber MECs within 30-day operation periods. Simultaneous anodic oxidation can generate reactive oxygen species that attack fluorinated intermediates. However, these systems still face limitations, including energy input requirements, electrode fouling, reduced performance in complex real-world matrices, and unresolved challenges in scale-up economics and long-term stability. A comparative summary of PFAS remediation technologies and their limitations is shown in Table 1.0.

3.0. PFAS Pretreatment and Concentration Technologies.

Destruction technologies such as electrochemical oxidation, sonochemical degradation, and supercritical water oxidation require concentrated PFAS feeds for efficient operation. Dilute matrices such as groundwater and landfill leachate therefore necessitate robust preconcentration steps. This preconcentration stage is the principal engineering bottleneck in most full-scale PFAS treatment trains.

Ion exchange resins (IEX) dominate current practice, with both single-use and regenerable high-capacity anion exchange resins widely used. Single-use resins (e.g., Purolite A600E, Lanxess Lewatit series) exploit combined hydrophobic and electrostatic mechanisms to achieve>99% PFAS removal even at ng/L influent concentrations. Regenerable resins (e.g., Purolite A592E, Amberlite HPR9200) allow elution of brine with methanol/NaCl or NaCl/NaOH mixtures, producing a concentrated PFAS eluate suitable for downstream destruction. Short-chain PFAS (C4–C6, such as PFBS and PFHxA) is problematic, as conventional anion exchange resins exhibit markedly reduced affinity for these lower-molecular-weight species. Recent demonstrations confirm that regenerable IEX integrated with advanced reduction processes (e.g., UV/sulfite) can form a technically viable closed-loop system for both capture and mineralization (Liu et al., 2025). Resin fouling by natural organic matter (NOM) and competing anions (sulfate, nitrate) remains a key operational challenge that typically requires targeted upstream pretreatment.

Foam fractionation (also known as air sparging concentration) takes advantage of the amphiphilic surfactant properties of PFAS, which preferentially partition to air–water interfaces. Air is sparged through contaminated water to generate a PFAS-enriched foam that is subsequently collapsed and collected as a concentrated liquid fraction. Concentration factors of 100–10,000× have been reported, depending on chain length and initial matrix (Meng et al., 2023; Burns et al., 2024). Long-chain PFAS (PFOS, PFOA) concentrate efficiently, while short-chain analogues perform less effectively due to weaker surface activity. The technology is particularly attractive for AFFF-impacted groundwater and landfill leachate, where PFAS concentrations range from µg/L to mg/L. Pilot- and full-scale demonstrations have established foam fractionation as a low-energy, first-stage concentrator upstream of IEX or direct destruction, substantially reducing downstream resin loading and overall operational costs.

Pressure-driven membrane processes, including nanofiltration (NF) and reverse osmosis (RO), achieve high PFAS rejection (NF: 90–99%; RO: >99%) primarily through size exclusion and charge repulsion. RO is essentially non-selective, concentrating all dissolved solids alongside PFAS and producing a small-volume, high-TDS concentrate that complicates downstream destruction. NF offers greater selectivity; tight NF membranes (e.g., NF270, NF90, DK series) effectively reject long-chain PFAS while allowing monovalent ions to pass, thereby simplifying brine chemistry. However, short-chain PFAS rejection by conventional NF is poor (<70% for PFBA and PFHxA), a well-documented limitation (Ma et al., 2024). Membrane fouling, concentration polarization, and management of the concentrate stream are persistent operational hurdles. Consequently, NF/RO are best deployed as a polishing or concentration step within integrated treatment trains rather than as a standalone solution.

In summary, no single preconcentration technology is universally sufficient. Emerging best practice converges on modular treatment trains, typically foam fractionation for bulk volume reduction, followed by IEX for polishing and further

concentration, and ultimately destruction. NF/RO find niche applications where regulatory drivers mandate near-zero effluent PFAS levels. Short-chain and novel PFAS (e.g., GenX, PFBS) continue to challenge capture efficiency across foam fractionation, IEX, and membranes due to weaker surface activity and lower affinity, necessitating ongoing innovation in resin chemistry and hybrid designs (Kannan et al., 2025).

Comparative Summary of PFAS Remediation Technologies

Table 1.0. Comparative Technology Matrix

Technology	**Primary Matrix**	**Defluorination Efficiency (%)**	**Energy Use**	**Scalability**	**Approximate Cost**	**Limitation**
GAC/IEX (sorption)	Water	0 (sequestration)	Low	Full-scale	Capital $0.75 – 8.89 / gal; O&M $0.06 – 0.28 / gal	No destruction; short chain breakthrough
Electrochemical Oxidation	Water	96 - 99	50 – 200 kWh/m^3	Pilot to full	High (energy + electrodes)	Energy-intensive; matrix effects
Sonochemical	Water	60 – 90	~1000 kWh/m^3	Lab/pilot	Very high	Not Economical
UV/Sulfite	Water	>85	Moderate	Pilot	Moderate	pH-sensitive; lampfouling
SR-AOPs	Water	40 - 60	Variable	Pilot	Moderate	Incomplete mineralization
High-Temp Incineration	Solid/ Liquids	>99 (at >1,100^oC)	High	Full-scale	High (energy + Scrubbing)	Byproduct Formation risk
Smoldering Combustion	Soil	>99	Low (self-sustaining)	Pilot/ field	Moderate	Matrix dependent temperature
Mechanochemical	Solids	>99.9	Moderate (milling)	Lab to pilot	Moderate	Scalability challenges
SCWO	Concen-trates	>99.99	High (450-600^oC)	Field demo	Volume dependent	Capital intensive
HALT	Concen-trates	90 - 99	Moderate (250-350^oC)	Pilot	Lower than SCWO	Needed alkali Optimization

Biological/ phyto	Soil/ water	Low	Very low	Lab/field	Low	Kinetic limitation
In Situ CAC (PlumeStop)	Ground water	0 (immobili zation)	Non	Full scale	High	Long term monitoring
In Situ Thermal	Soil/ Vadose	High(volatilizati on + capture)	High	Pilot	High	Energy and infrastructure

Note: Costs are approximate and site-specific; values drawn from recent U.S. treatment plant data and case studies (deSilva &Deshmukh, 2025; ITRC, 2025).

3.1. Regulatory–Remediation Interface

Regulatory mandates have advanced, yet technological capability lags. The U.S. EPA's 2024 National Primary Drinking Water Regulation sets MCLs of 4 ng L^{-1} for PFOA and PFOS (and hazard indices for mixtures including HFPO-DA and PFBS), requiring treatment trains that achieve consistent non-detect levels (U.S. Environmental Protection Agency, 2024). Sorption technologies (GAC/IEX) can meet these thresholds in drinking water but generate concentrated waste requiring destruction. Destructive technologies (SCWO, HALT, electrochemical oxidation) excel at concentrate mineralization yet struggle with dilute matrices without pretreatment. No single technology currently achieves broad-spectrum compliance for entire PFAS mixtures at the field scale while addressing precursors and short-chain replacements. "Non-detect" interpretation varies across analytical methods (targeted LC-MS/MS vs TOP/EOF), complicating verification. Practitioners must therefore deploy integrated trains (concentration followed by Destruction) with robust analytical validation to comply with the regulations and protect public health.

4.0. Conclusion

This review synthesizes the current state of PFAS remediation science, highlighting that while destructive technologies have advanced significantly, no single approach fully resolves the multifaceted challenges of persistence, matrix complexity, short-chain PFAS, and regulatory stringency. Sorption and concentration technologies are essential enablers, but true risk reduction demands mineralization via thermal (SCWO, HALT, smoldering), electrochemical, or hybrid reductive systems, supported by robust pretreatment and analytics.

Key research priorities include: (1) development of low-energy hybrid systems to reduce costs and improve scalability; (2) field validation of in situ immobilization and thermal methods for large plumes; (3) advanced nontargeted HRMS and fluorine mass-balance protocols to verify mineralization and track byproducts; (4) targeted innovation for short-chain and ether-based PFAS; and (5) life-cycle cost analyses integrating destruction of secondary wastes.
Looking forward, integrated treatment trains that combine foam fractionation or regenerable IEX with SCWO/HALT destruction and in situ CAC for plume control offer the most promising pathway to regulatory compliance and environmental restoration.

Continued collaboration among researchers, engineers, regulators, and practitioners is essential to close the technology gap and protect human and ecological health from these indestructible contaminants.

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