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State of the science and regulatory acceptability for PFAS residual management options: PFAS disposal or destruction options

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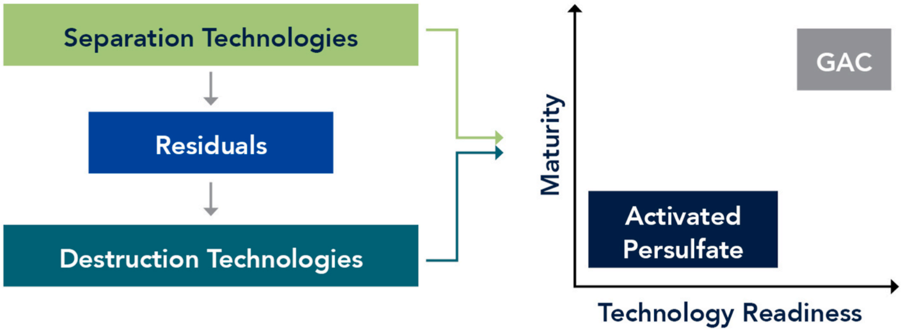
HIGHLIGHTS

- PFAS separation technologies create residuals that require further treatment.
- Regulatory compliance is key in addressing evolving PFAS regulations and guidelines.
- Managing PFAS-containing residuals necessitates a strategic and collaborative approach.
- Ongoing PFAS research is vital for informed decisions and innovative solutions.

GRAPHICAL ABSTRACT



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ABSTRACT

This systematic review covers the urgent challenges posed by per- and polyfluoroalkyl substances (PFAS) in managing residuals from municipal, industrial, and waste treatment sources. It covers regulatory considerations, treatment technologies, residual management strategies, and critical conclusions and recommendations. A rigorous methodology was employed, utilizing scientific search engines and a wide array of peer-reviewed journal articles, technical reports, and regulatory guidance, to ensure the inclusion of the most relevant and up-to-date information on PFAS management of impacted residuals. The increasing public and regulatory focus underscores the persistence and environmental impact of PFAS. Emerging technologies for removing and sequestrating PFAS from environmental media are evaluated, and innovative destruction methods for addressing the residual media and the concentrated waste streams generated from such treatment processes are reviewed. Additionally, the evolving regulatory landscape in the United States is summarized and insights into the complexities of PFAS in residual management are discussed. Overall, this systematic review serves as a vital resource to inform stakeholders, guide research, and facilitate responsible PFAS management, emphasizing the pressing

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need for effective residual management solutions amidst evolving regulations and persistent environmental threats.

1. Introduction

Public and regulatory attention to per- and polyfluoroalkyl substances (PFAS) in the environment has dramatically increased in recent years (Brennan et al., 2021). Due to the extreme persistence of PFAS in the environment, understanding their occurrence, fate, and toxicity is essential for assessing human health risks and environmental consequences (Fenton et al., 2021; Sunderland et al., 2019). Perfluoroalkyl acids (PFAAs), which include perfluorocarboxylic acids (PFCAs) and perfluorosulfonic acids (PFSAs), are the most studied PFAS. They are well-known to be extremely persistent and are also the end products of the transformation of other PFAS (Houtz et al., 2013). PFAAs, in particular the shorter-chain PFAAs (less than or equal to 7 carbons for carboxylate group and 6 for sulfonate group [Buck et al., 2011]), are quite mobile and frequently detected in water bodies, wastewater (Thompson et al., 2022), and finished drinking water (Ahrens et al., 2015; Barzen-Hanson et al., 2017; EPA., U., 2024a; Szabo et al., 2018).

Researchers began detecting PFAS in environmental samples across the United States (U.S.) around the early 2000s (Hekster et al., 2002). Several major sources or concentration points of PFAS contamination have been identified, including industrial discharges (S. Liu et al., 2021b), aqueous film forming foam (AFFF) used in fire-training areas at military bases (Moody and Field, 2000), landfills (Hepburn et al., 2019; Lang et al., 2017), and water resource recovery facilities (WRRFs) (Boulanger et al., 2005; Sinclair and Kannan, 2006).

Conventional drinking water and wastewater treatment processes are generally ineffective at removing significant PFAS from water (Appleman et al., 2014). Advanced treatment processes, such as granular activated carbon (GAC) and ion exchange (IX), have been widely adopted due to their effectiveness for treating regulated PFAS (i.e., perfluorooctanoic acid [PFOA], perfluorooctanesulfonic acid [PFOS], perfluorobutanesulfonic acid [PFBS], hexafluoropropylene oxide dimer acid [HFPO-DA], trademarked as GenX, perfluorohexanesulfonic acid [PFHxS], and perfluorononanoic acid [PFNA]) and operability. A significant drawback of these treatment technologies is that PFAS-saturated media is often disposed of through landfilling, thermal reactivation, or incineration. By removing PFAS contamination upstream, some of these PFAS separation technologies can be protective of any residuals produced downstream, such as biosolids. This approach will facilitate maintaining resource recovery programs that are currently threatened due to PFAS contamination. These potential routes of emission motivate identification of available residual management options and a comprehensive evaluation of their suitability under various operational and regulatory circumstances.

Entities such as municipal wastewater resource recovery facilities (WRRF), municipal drinking water treatment plants (WTP), and specialized waste management companies, including those managing PFAS-laden GAC or IX resins, are at the forefront of handling PFAS residuals. These organizations and facilities are critical nodes in the vast network that protects the public and the environment from PFAS contamination, bearing the brunt of mitigating its environmental impact, with WRRFs, in particular, acting as one of the primary conduits through which PFAS enters the environment. WRRFs receive PFAS from several potential sources, including residential and commercial waste, landfills, industrial sites, and sites of both historical and ongoing firefighting activities. Because conventional wastewater treatment does not significantly remove PFAS, WRRFs then discharge PFAS to the environment through effluent discharge and treated biosolids land application. PFAS occurrence has been well documented in WRRF samples (Eriksson et al., 2016; Filipovic and Berger, 2015; Kibambe et al., 2020; Kim Lazcano et al., 2020; Kurwadkar et al., 2022; Lee et al., 2014; Pernet-coudrier et al., 2008; Schaefer et al., 2023a, 2022a; Szabo et al., 2018; Tang, 2023; Tavasoli et al., 2021; Thompson et al., 2022). Annual PFAS mass in U.S. landfill leachate has been estimated to range between 563 and 6400 kg (kg) (Coffin et al., 2023).

Since PFAS has a high affinity for organic matter, these substances can accumulate in biosolids, which are rich in organic content (Kim Lazcano et al., 2020). In particular, longer chain PFAS and their precursors can accumulate in biosolids to a greater extent (Seay et al., 2023). In the United States, about 50% of biosolids are land-applied (EPA., U., 2020a), posing the risk of PFAS leaching into groundwater or being taken up by plants (Holly et al., 2024; Johnson, 2022; Pepper et al., 2021; Schaefer et al., 2022a; J. A. K. Silva et al., 2022b). Some PFAS precursors in sewage can be converted into PFAAs (terminal end products) through wastewater treatment processes such as anaerobic/aerobic oxidation primarily through biological transformation (Alukkal et al., 2024a, 2024b; Schaefer et al., 2022a). This can facilitate the release of more mobile PFAS into the environment.

Based on a comprehensive nationwide study of U.S. biosolids, the annual PFAS load in biosolids has been estimated to range from 2749 to 3450 kg (Venkatesan and Halden, 2013). DiStefano et al. (2022) reported 120 mg per kilogram (mg/kg) of PFAS in the residual spent GAC from a full-scale GAC reactivation facility during the reactivation of a load of GAC known to contain adsorbed PFAS. Such findings validate the urgent need for immediate identification of currently available residual management options and the evaluation of their suitability under various operational and regulatory circumstances.

Globally, Maximum Contaminant Levels (MCLs) for PFAS in drinking water vary significantly, with Canada adopting less stringent limits, the United States establishing intermediate thresholds, and Denmark enforcing some of the most rigorous standards. In particular for US, due to the absence of federal treatment standards for PFAS in environmental matrices except for drinking water, state regulations have emerged as the main driver for managing contaminated sites. While US-EPA has established national primary drinking water regulations (i.e., MCLs) for some PFAS and issued a Strategic Roadmap (Roadmap) for future action, inconsistencies persist across states and sectors for other non-drinking water matrices (EPA.U, 2023a, 2022, 2021a, 2021b, 2021c; ITRC, 2023a) with only recent attention being given to wastewater and biosolids (Andrews and Naidenko, 2020; Hu et al., 2016; Lang et al., 2017). As part of its Roadmap, EPA has initiated a risk assessment for PFOA and PFOS in biosolids (EPA.U, 2023b). Meanwhile, some states have enacted their own biosolids regulations, causing spikes in management costs, and in some sectors, even more significant disruption (Brown, 2022). Therefore, scientific and regulatory uncertainties complicate the quest for effective solutions. Amid growing concern over the environmental and health impacts of PFAS, the research focus has increasingly shifted toward treatment trains that couple separation technologies, such as GAC, IX, and reverse osmosis (RO), with destruction technologies. As per EPA's interim guidance, the current best management practices are as follows: (1) Class I waste injection well, (2) permitted hazardous waste landfills, or (3) thermal treatment, with emerging destructive technologies, which are still under evaluation.

While several literature reviews have summarized different PFAS treatment technologies, this is the first to specifically address PFAS removal in treatment residuals and biosolids generated in wastewater treatment plants, as well as drinking water residuals. It also examines the most appropriate destruction technologies to mineralize PFAS in these impacted residuals and the current state of PFAS regulations. With the recent release of MCLs for PFAS, utilities will begin implementing treatment technologies, which will generate PFAS-containing residuals, and this review provides timely guidance on how best to manage these residuals.

2. Methodology

A systematic literature review method was used to investigate the current state of federal and state regulations, PFAS disposal or destruction options, and PFAS management options in residuals. This methodology enabled the thorough examination of pertinent research questions and the formulation of valuable recommendations for informed decision-making. The systematic review process followed a structured five-step approach: (1) definition of the problem and selection of the subject matter, (2) identification of relevant databases, (3) selection of pertinent studies and documents, (4) rigorous evaluation of the chosen resources, including thorough assessment of their relevance, quality, and applicability, and (5) synthesis of the gathered information. The selected databases for this study encompassed peer-reviewed literature containing published methods, protocols, and research findings (Scopus, Google Scholar, and Web of Science), as well as 'grey' literature, government reports (e.g., EPA's reports) from the U.S. and international agencies (e.g., European Union (EU), Organization for Economic Cooperation and Development [OECD], and International Organization for Standardization [ISO]), as well as state reports and Interstate Technology and Regulatory Council (ITRC) documents. The keywords used to find relevant articles were "per and poly-fluoroalkyl substances disposal," "PFAS disposal," "PFAS and biosolids," "PFAS and granular activated carbon," "PFAS and ion exchange resin," "PFAS and reverse osmosis," "PFAS treatment and coagulants," "PFAS treatment and adsorbents," "PFAS and membrane filtration," "PFAS treatment and foam fractionation (FF)," "PFAS and thermal destruction," "PFAS and electrochemical treatment," "PFAS and sonochemical treatment," "PFAS and ultrasound," "PFAS and plasma," "PFAS and biodegradation," "PFAS and ultraviolet," and "PFAS and supercritical water oxidation". These searches resulted in a total of 767 peer-reviewed journal articles, technical reports, and legal documents. Upon closer examination, 528 documents were eliminated, and the remaining 239 documents were carefully evaluated, synthesized, and presented in this review (Fig. S1).

3. Results and discussion

3.1. Regulatory status

Exposure to some types of PFAS has been connected to adverse health effects, including cancer, immunotoxicity, dyslipidemia, neurodevelopmental effects, and metabolic effects (Sunderland et al., 2019). Due to increased public awareness and media reporting, manufacturers voluntarily started phasing out legacy PFAS, such as PFOA and PFOS, in the early 2000s and replacing them with either short-chain alternatives or nonfluorinated compounds. Studies have shown that alternatives, such as HFPO-DA (tradename GenX), a replacement for PFOA, and 6:2 chlorinated polyfluorinated ether sulfonic acid, a replacement for PFOS, are already being detected in environmental media and humans (Wang

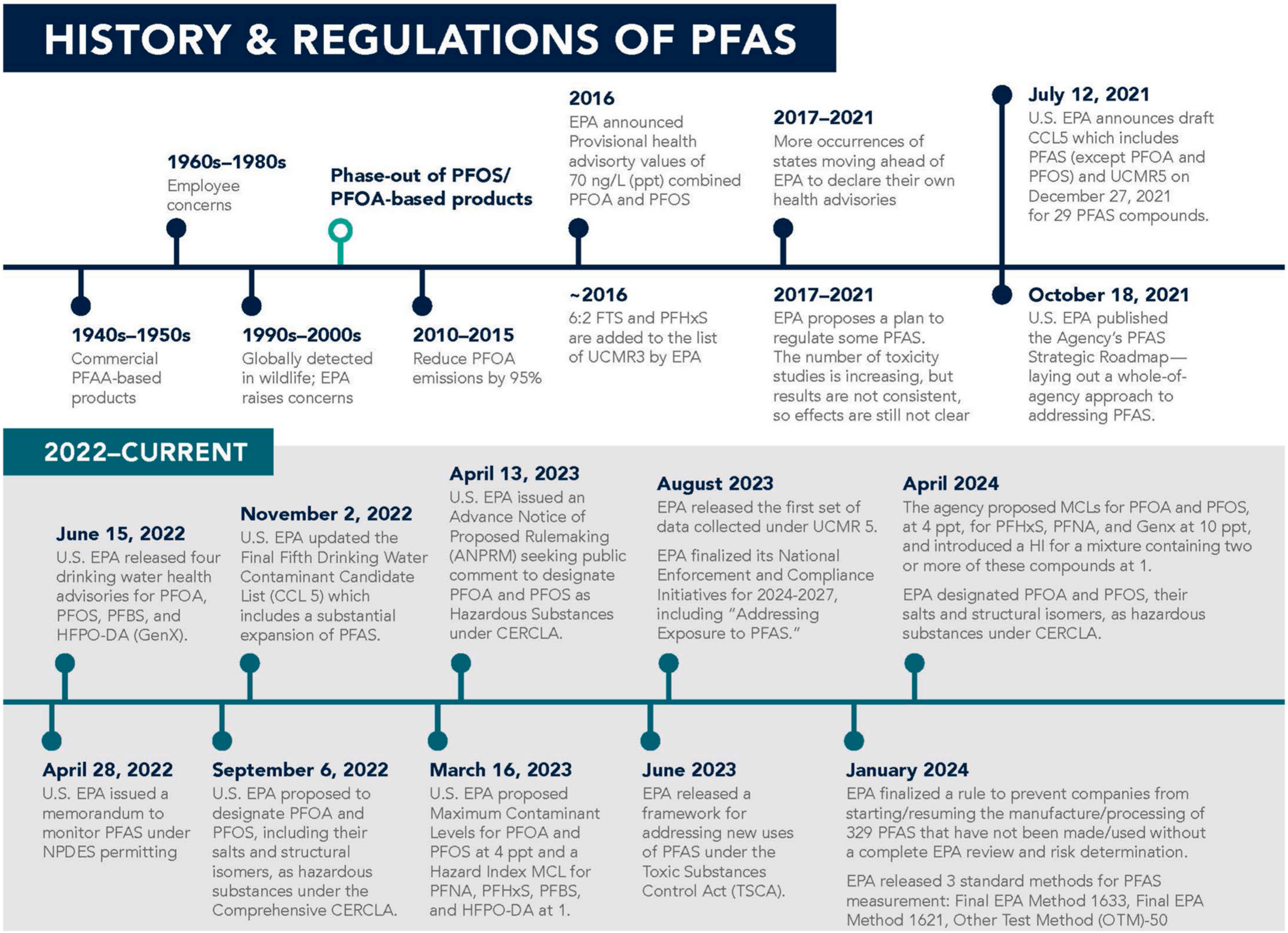

Fig. 1. Synopsis of per- and polyfluoroalkyl substances regulation history.

et al., 2019). As the regulatory landscape continues to evolve, understanding the nuances and implications of these changing guidelines is essential for stakeholders, industries, and communities to ensure compliance and protection of public health.

3.1.1. *Regulatory actions and impacts*

The PFAS regulatory landscape has been evolving in recent years (Fig. 1). In April 2024, the US EPA finalized maximum contaminant levels (MCLs) under the Safe Drinking Water Act for regulating PFAS in drinking water. The MCLs are set at 4 parts per trillion (ppt) for PFOA and PFOS, 10 ppt for PFHxS, PFNA, and GenX. In addition, EPA has established a Hazard Index (HI) MCL for a mixture containing two or more of these compounds as well as PFBS at 1 (EPA., 2024b). Around the world, MCLs for PFAS in drinking water differ widely as summarized in Table S1.

Other than drinking water, there are no federally mandated treatment standards for PFAS in other matrices. Consequently, state regulations and guidance have emerged as primary drivers for establishing guidelines at PFAS-contaminated sites. For sites owned and operated by the U.S. Department of Defense (DoD), clean-up drivers may be EPA Regional Screening Levels (RSLs) or state standards, if they exist. As reported by ITRC, 28 states have published PFAS standards or guidelines for groundwater, drinking water, surface water and biosolids, as summarized in Table S2 (ITRC, 2023a). The dataset in Table S2 reveals a general lack of consistency and agreement regarding PFAS regulations. All states must follow federal MCLs unless they have established lower limits. The federal government has initiated efforts to provide national direction and consistency in responding to PFAS contamination in the environment. This commenced with EPA's initial health advisory levels (HAL) for PFOS and PFOA in 2016, as depicted in Fig. 2. EPA also issued draft national recommended aquatic life criteria for PFOA and PFOS in freshwater, inviting public comments (ITRC, 2023a). In 2021, the EPA issued its Roadmap (EPA. U., 2021a), which included the development of effluent limitation guidelines (ELGs) for some sectors, which could impact industrial discharges to WRRFs under the National Pollutant Discharge Elimination System (NPDES). In January 2023, EPA released its Effluent Guidelines Program Plan 15, which aims to establish effluent limitation guideline (ELGs) for PFAS in some industrial discharge categories, such as landfills and textile mills, while aiming to further study PFAS at publicly owned treatment works (POTW) and the industrial dischargers into POTWs. The PFAS study period rules at POTWs required POTWs to sample and monitor PFAS levels in the influent, effluent, biosolids and industrial discharges (EPA.U, 2024c). The Roadmap also outlined plans to develop and publish a methodology for PFAS risk assessment for biosolids land application in 2024 (EPA.U, 2024b). Concomitant with the drinking water MCLs in April 2024, EPA designated PFOA and PFOS, including their salts and structural isomers, as hazardous substances under the federal Comprehensive Environmental Response, Compensation, and Liability Act (CERCLA or Superfund) (EPA.U, 2024d).

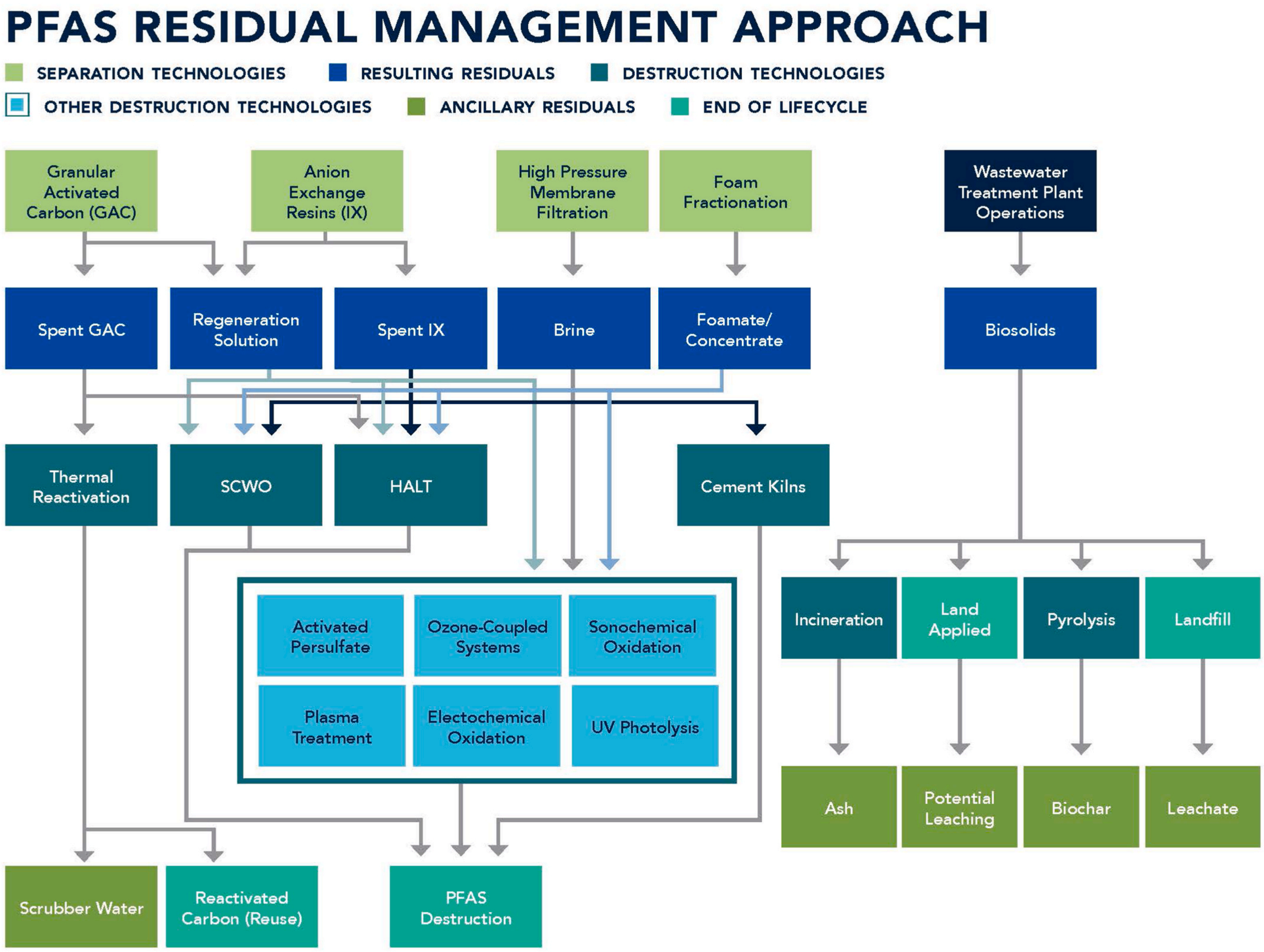

Fig. 2. Exploring PFAS Residual Management: Illustrated Approach (Note: other destruction technologies are emerging and limited to aqueous streams).

3.1.2. *Current regulatory focus*

3.1.2.1. Drinking water. So far, the regulatory focus in the USA has primarily centered on drinking water, as it represents one of the most understood and regulatable sources of PFAS exposure. UCMR3 indicated that the drinking water supplies for over 6 million US residents exceeded the EPA's 2016 HALs of 70 ppt for PFOA and PFOS (Hu et al., 2016). The method reporting limits (MRLs) in UCMR3 were too high to quantify the impacts of the federal PFAS MCLs finalized in 2024. Meanwhile, drinking water monitoring for 29 PFAS under EPA's Unregulated Contaminant Monitoring Rule 5 (UCMR5) is currently underway and expected to be completed in 2025. This dataset has lower analytical reporting limits compared to monitoring conducted under UCMR3. Among public water systems that reported data for all four quarters of quarterly sampling by April 2024, about 10% had exceedances of the federal MCLs (EPA.U, 2024a).

3.1.2.2. Wastewater. Industrial wastewater can be directly discharged to the environment after NPDES-regulated treatment or indirectly to a POTW for subsequent treatment. While PFAS remains largely unregulated across most state jurisdictions, there is an increasing focus on addressing PFAS from major source categories such as textile manufacturing facilities, municipal solid waste landfills, and metal finishing operations. For instance, the state of Colorado has implemented regulatory controls on PFAS discharges under NPDES permitting program. The EPA's guidance regarding PFAS discharges through the NPDES permitting program and pretreatment program recommends monitoring influent, effluent, and biosolids using EPA Method 1633, updating industrial user inventories for expected or suspected PFAS discharges, and employing best management practices to address PFAS discharges to POTWs. There are at least three states which have made efforts to address PFAS in NPDES permits. Massachusetts requires industrial facilities to test for PFAS if they hold a NPDES permit or discharge to POTWs which hold a permit (ITRC, 2023b). New Hampshire and New Jersey added certain PFAS to the permit application testing requirements for discharges to groundwater (ITRC, 2023b).

Municipal wastewater has also become an area of increased scrutiny, with researchers investigating PFAS occurrence, sources, fate, and transport within these systems (Lang et al., 2017; Schaefer et al., 2023b, 2022b, 2022a; Seay et al., 2023; SWRCB, 2020; Tavasoli et al., 2021; The Water Research Foundation, 2020; Thompson et al., 2022). Previous efforts have reported PFAS occurrence in domestic wastewater, likely resulting from the use of consumer products, such as cosmetics, and fabric treatments, leaching from plastic containers, laundering of PFAS-treated clothing, and, potentially, bodily excretions, with consequent discharge to sewers (EPA.U, 2023c). However, a 2021 study observed higher PFAS concentrations in municipal wastewater in the presence of industrial discharges when evaluating PFAS in influent, effluent, and residuals of WRRFs in Michigan (AECOM, 2021). Michigan is a proactive state that has enforced pretreatment of PFAS with successful source reduction (greater than 90% reduction of PFOS concentrations in treated influent) through pretreatment at the PFAS source to wastewater using GAC (AECOM, 2021).

3.1.2.3. Biosolids. Some states such as Maine, New Hampshire, Michigan, Wisconsin, Colorado and New York have issued either regulations, guidance or management strategies for PFAS in biosolids (Table S2). In April 2022, Maine banned all land application of biosolids due to PFAS concerns. In November 2022, the Environmental Council of the States (ECOS) conducted a survey involving 34 states to gather information on their policies, testing procedures, research gaps, and risk communication challenges concerning PFAS in biosolids (Sarah Grace Hughes (ECOS), 2023). Currently, no states are looking into banning landfill disposal of such materials. While there is limited information on the scope and scale of the impact of biosolids management and regulations, a 2020–2021 study surmised that in areas where PFAS regulations have been implemented, biosolids management costs have increased by 37–72% due to increased landfilling and less land application (CDM Smith, Northeast Biosolids and Residuals Association, Water Environmental Fund, and National Association of Clean Water Agencies). The overall impact on the environment and society resulting from diverting biosolids to landfill without resource recovery has not been fully recognized.

3.1.3. *Uncertainties and upcoming regulatory actions*

There are significant scientific and industrial uncertainties regarding the best solutions for PFAS solid waste disposal. Currently, EPA regulations do not classify PFAS as a hazardous waste under the Resource Conservation and Recovery Act (RCRA), although they may in the future. The industrial community's primary uncertainty centers around how to deal with PFAS-laden wastes in a cost-effective and environmentally sound manner.

To address these uncertainties and concerns, EPA is taking several actions. On February 8, 2024, EPA proposed a rule to designate nine PFAS, their salts, and their structural isomers, to its list of hazardous constituents under Appendix VIII in 40 CFR Part 261. Additionally, on April 8, 2024, the EPA released version 2 of the *"Interim Guidance on the Destruction and Disposal of Perfluoroalkyl and Polyfluoroalkyl Substances and Materials Containing Perfluoroalkyl and Polyfluoroalkyl Substances."* This document discusses the EPA's efforts to identify and evaluate emerging technologies for destroying PFAS, such as mechanochemical destruction, electrochemical oxidation, gasification and pyrolysis, and supercritical water oxidation. While the interim guidance does not provide a definitive list of accepted disposal options or methodologies, it outlines factors and data gaps to consider when disposing of PFAS-impacted materials and provides a framework to evaluate PFAS destruction and disposal technologies.

3.2. *Treatment options for managing PFAS-contaminated water and residuals*

Navigating the complex landscape of PFAS contamination necessitates a comprehensive approach to address both aqueous waste streams and solid residuals. Aqueous waste streams encompass drinking water, wastewater treatment influent or effluent, while concentrated waste streams include brines from membrane filtration, fractionate from foam fractionation, and regeneration solutions from IX resins. Solid residuals consist of spent GAC, spent IX resins, and biosolids generated from conventional WRRF operations. This diverse array of residual types requires unique treatment solutions to ensure the safe and efficient management of PFAS-contaminated waste.

Treatment strategies for PFAS management can be broadly categorized into separation and destruction technologies. Separation technologies isolate or concentrate PFAS, along with other contaminants, from the treated media into a different medium or a concentrated waste stream. These methods often leverage the distinct physicochemical properties of the contaminants, such as their chemical structures, polarities, and sizes. Regardless of the specific separation method used, PFAS-rich residuals are produced and must be managed appropriately.

In contrast, destruction technologies are designed to transform or mineralize PFAS by targeting breaking the strong carbon-fluorine bonds to yield less toxic byproducts, primarily and ideally inorganic fluoride and carbon dioxide (CO_2). These technologies leverage various physical and chemical methods, aiming to comprehensively eradicate the environmental and health risks posed by PFAS. While each of these techniques have their merits, they often falter when dealing with more complex matrices like municipal wastewater and landfill leachate (Grieco et al., 2022; Krögerström, 2021; Lutze et al., 2011; Singh et al., 2021). Separation technologies create a new waste while destruction technologies, like incineration, supercritical water oxidation, or electrochemical oxidation (EO), have the potential to create hazardous by-products (Ross et al., 2018; Wang et al., 2020; Yang, 2020). In the following sections, we explore separation technologies, an examination of the residuals generated by that particular technology, and the various destruction techniques available to manage those residuals, as illustrated in Fig. 2.

3.2.1. Separation technologies

3.2.1.1. Granular activated carbon and ion exchange resins. To date, GAC and IX resins have been widely used for PFAS removal in industrial and municipal applications (Dixit et al., 2021; Liang et al., 2022; Plumlee et al., 2022; Rodowa et al., 2020; Ross et al., 2018). GAC functions by adsorbing PFAS onto its porous surface, capitalizing on the carbon's vast surface area and affinity for organic molecules. On the other hand, IX resins employ an ionic mechanism, wherein the resin material swaps its ions for PFAS ions present in the water, facilitating their removal. While these are just two of the many separation techniques available, they demonstrate the diverse approaches to tackling the PFAS challenge. Pilot-scale and field-scale studies conducted using commercially available GACs and IX resins show near complete removal of PFAS from contaminated water for months or years before breakthrough. Breakthroughs are correlated to PFAS chain length and certain GAC performed better than others such as bituminous GACs (Liu et al., 2019; Orange County Water District, 2021; Westreich et al., 2018). The use of IX resins has proven to be more effective than GAC, particularly for removing short-chain PFAS (McCleaf et al., 2017; Woodard et al., 2017) and studies show they can remove four times the PFAS on a mass-to-mass basis than GAC (Woodard et al., 2017). Overall, the performances of GAC and IX resins for PFAS removal are competitive. Cationic and zwitterionic IX resins have also been investigated for their effectiveness in removing cationic and zwitterionic PFAS although they are typically of lesser concern in water matrices (Dixit et al., 2021, 2022). Overall, pilot and full-scale studies have documented the successful use of GAC and IX for the removal of PFAS (Belkouteb et al., 2020; Chow et al., 2022; Liu et al., 2019, 2022; Medina et al., 2022; Murray et al., 2019; Pannu and Plumlee, 2021; Rodowa et al., 2020; Woodard et al., 2017). However, selecting appropriate technology must be based on the source water composition to achieve the most cost-effective PFAS removal. Saturated media are usually disposed of at landfills until further regulations or treatment techniques are developed. However, saturated GAC can be thermally reactivated to restore its adsorptive properties.

3.2.1.2. High-pressure membrane filtration. High-pressure membrane filtration emerges as a key player in PFAS management, capitalizing on the principle of size-exclusion and physicochemical interactions at the molecular level (Malaeb and Ayoub, 2011). Comprising thin layers of semi-permeable materials, NF and RO membranes are designed to permit the passage of water molecules while effectively retaining or rejecting constituents such as PFAS. Depending on the pore size and mechanism, membrane technologies can range from microfiltration and ultrafiltration to nanofiltration (NF) and RO. Particularly, NF and RO membranes, with their extremely tight pore structures, demonstrate remarkable efficacy even for short-chain PFAS such as PFHxA that are challenging for GAC and IX (Dickenson and Higgins, 2016; Ma et al., 2023; Steinle-Darling and Reinhard, 2008; Thompson et al., 2011). For example, RO systems employed in two potable water reuse plants in California removed PFAS below the method detection limits, even for short-chain compounds that were difficult to remove using other treatment techniques (Appleman et al., 2014). NF requires less energy and is less sensitive to dissolved solids than RO.

While membrane filtration offers high removal efficiencies, challenges such as fouling, which can reduce flux and overall system efficiency, require consideration. Also, the cost of disposal or destruction of PFAS-concentrated reject water from RO and NF systems must be considered (Ross et al., 2018). However, with advancements in membrane material science and system design, this technology stands as a potent tool in the comprehensive strategy for mitigating PFAS contamination in water sources. There are numerous previous studies that have successfully reported PFAS removal with membrane (Chen et al., 2023; Dirani et al., 2024; Léniz-Pizarro et al., 2023; Liu et al., 2023a,b; Ma et al., 2023; Quinnan et al., 2023; Safulko et al., 2023). For instance, Chaudhary et al. (2023) demonstrated that mixed-matrix composite nanofiltration (MMCNF) membranes, incorporating adsorption processes, led to over 99.9% removal of PFAS such as PFOA. These membranes also facilitate easier regeneration processes, improving their long-term usability. Another innovative approach, direct contact membrane distillation (DCMD), has shown promise in concentrating and removing PFAS like perfluoropentanoic acid (PFPeA), offering efficient downstream treatment or disposal (Chen et al., 2020).

Despite the effectiveness of high-pressure membrane technologies, they are found to be the most expensive technology for PFAS removal (Appleman et al., 2014). Brunswick County in North Carolina has invested $157 million in a RO plant that could treat 45 million gallons a day for PFAS (Brunswick County, 2024). Safulko et al. (2023) evaluated the rejection performance of commercially available high-pressure membranes, specifically in pilot-scale closed-circuit membrane filtration (CCMF) systems, for PFAS removal. Results indicated that tight NF and RO membranes effectively separated and concentrated PFASs during high recovery CCMF operation, with most membranes showing over 98.3% rejection, though the performance of loose NF membranes diminished at water recoveries above 90%.

3.2.1.3. Foam fractionation (FF). FF has gained significant interest for field and pilot-scale applications due to its simple underlying concept and straightforward implementation. FF is a physicochemical process based on adsorptive bubble separation technology. In FF, a gas, typically air, is introduced into a liquid solution containing the contaminants. Gas introduction creates a frothy mixture, forming bubbles that rise to the surface due to buoyancy. As the bubbles ascend through the liquid, they selectively adsorb contaminants, such as PFAS, onto the bubbles' air-water interfaces. Once the foam reaches the surface, it can be harvested. FF application for PFAS removal is gaining increasing momentum among researchers and others. PFAS removal by FF has been tested in the laboratory and at the field scale from various environmental matrices (Buckley et al., 2023, 2022; Burns et al., 2022; Dai et al., 2019; Krögerström, 2021; Malovanyy et al., 2023; McCleaf et al., 2023; Smith et al., 2023, 2022; Taseidifar, 2020; J. Wang et al., 2023a; Ziaee et al., 2021). Some researchers have tried to improve the performance of FF by adding co-surfactants (Taseidifar, 2020; Ziaee et al., 2021). Adding the co-surfactant considerably increased the volume of foam and its stability, which led to more convenient foam collection. Ziaee et al. (2021) evaluated the effect of adding a biodegradable co-surfactant 1-octanoylcysteine to remove PFOA in benchtop experiments, which resulted in improved foaming and a 73% removal of PFOA. Buckley et al. (2023) evaluated using different co-foaming agents during FF on PFAS removal (Buckley et al., 2023), where co-foaming agents with opposing charge, such as cationic surfactants performed best overall and facilitated removal of short-chain PFAS. There are on-going efforts to scale up this technology. EPOC Enviro has developed commercial-scale FF technology (surface-active FF) that can be applied ex situ and in situ for PFAS removal from water and soil. Burns et al. (2022) tested the efficacy of EPOC Enviro's system for the removal of PFAS from landfill leachate at Telge Recycling plant in Sweden. The resulting PFAS removal was >98.7% for PFOS, PFOA, and PFHxS. Whether or not a co-foaming agent is needed will depend on PFAS concentration and matrix of the media being treated. In scale-up of FF technologies where co-foaming agents may be needed, factors to consider in addition to performance include cost, toxicity, and biodegradability of the co-foaming agents.

3.2.2. Residuals management utilizing destruction technologies

3.2.2.1. Spent granular activated carbon. Full-scale PFAS treatment systems generate spent GAC which poses a unique challenge in PFAS in residuals management. DiStefano et al. (2022) conducted a stack test at a commercial GAC reactivation facility with a gas-fired multihearth Herreschoff furnace treating spent GAC with PFAS concentrations ranging from 112,943 to 119,234 ng per gram (ng/g). No targeted PFAS were detected above 1.9 ng/g in the reactivated GAC, indicating removal efficiencies of greater than 99.99%. GAC reactivation typically involves heating spent GAC at temperatures greater than 700 °C (°C) to reactivate the GAC, which then could be reused for further water treatment. Understanding the thermal stability of PFAS is critical for selecting appropriate treatment methods. PFAS thermal stabilities follow perfluorocarbons (e.g., C_xF_y structures) > perfluoroacyl fluorides (pyrolysis byproducts of PFCA salts) > PFSAs > PFCAs > perfluoropolyethers, with thermal stability decreasing with increasing chain length (Alinezhad et al., 2022; J. Wang et al., 2022b; Xiao et al., 2020). Thermal treatment processes involve complex decomposition mechanisms, including simultaneous elimination of a F and H atom (referred to as HF elimination), direct intramolecular bond cleavage, radical reactions, hydrolysis, and oxidation, with HF elimination emerging as a key initial step (Alinezhad et al., 2022; J. Wang et al., 2022b; Xiao et al., 2020). To achieve effective PFAS destruction, specific conditions must be met, with enhanced defluorination observed at higher temperatures (greater than 700 °C) and in oxidizing atmospheres (J. Wang et al., 2022b). Complete mineralization of PFAS to HF and CO_2 requires combustion temperatures above 1000 °C (Zhang et al., 2023). Therefore, many PFAS can transform into other PFAS at lower temperatures, potentially releasing fluorocarbon fragments into the environment. These products of incomplete combustion (PICs) may include greenhouse gases, substances with unknown toxicity, and compounds that can react to form new perfluorocarboxylic acids. Pilot-scale experiments using AFFF in a research combustor examined combustion conditions. Results showed that temperatures above 1090 °C achieved high removal efficiencies with minimal fluorinated PIC emissions, whereas conditions below 1000 °C yielded DEs above 99.99% but also emitted detectable concentrations of non-polar PFAS PICs (Shields et al., 2023). Thermal treatment kinetics follow first-order and Arrhenius relationships, and perfluorocarbons exhibit remarkable thermal stability even at 1000 °C, with half-lives exceeding 1 h, while PFCAs and PFSAs have sub-second half-lives above 500 °C (Wang et al., 2022a). Additives such as $Ca(OH)_2$ have demonstrated improved mineralization of PFOS at temperatures as low as 500 °C while reducing the formation of HF gas and instead forming inorganic fluorine (Abou-Khalil et al., 2024). In addition, some of the full-scale treatment systems which use wet scrubbing process produce ancillary waste streams, such as scrubber water, during the thermal reactivation process. This scrubber water, which is used to control emissions from the reactivation furnace, can become contaminated with PFAS and other byproducts of combustion. Managing scrubber water presents a significant challenge, as it requires additional treatment steps to prevent secondary contamination.

Despite these advancements, knowledge gaps remain, particularly concerning decomposition mechanisms, products, and kinetics for complex PFAS mixtures and matrices such as soils and activated carbon. Further research, including full-scale system assessments, is imperative to refine thermal treatment technologies and make them practical for real-world applications (Wang et al., 2022b; Zhang et al., 2023).

New and innovative technologies such as supercritical water oxidation (SCWO) and hydrothermal alkaline treatment (HALT) have been investigated to destroy PFAS in spent media. SCWO leverages the unique properties of water at supercritical conditions (above 374 °C and 22.1 MPa, MPa) to accelerate the oxidation of organic compounds, including PFAS. In recent years, several research studies have investigated the efficacy of SCWO in destroying PFAS. Spent GAC from full-scale PFAS treatment systems treated in a 1 ton per day SCWO system showed 75–100% destruction of PFAS (Chiang et al., 2023). Additionally, an EPA-organized PFAS innovative treatment team (PITT) highlighted SWCO as a potential solution for addressing spent GAC and IX resin treatment challenges (EPA.U, 2021d). Collectively, these studies underscore the potential of SCWO to significantly reduce PFAS concentrations. However, further research is needed on optimizing operating conditions, identifying byproducts, and treating effluent. The main challenges of SCWO include optimizing reactor design and operating conditions, as well as addressing concerns related to the formation of potentially toxic byproducts during the oxidation process.

HALT utilizes hot compressed water (300–350 °C) and strong alkali to destroy a wide range of PFAS. Continuous flow HALT reactors further enhance PFAS destruction kinetics at lower residence times compared to batch reactors (Li et al., 2022). Short residence times for effective PFAS destruction in continuous flow HALT reactors are conducive to scaling up for practical applications. The rapid kinetics suggest continuous HALT systems can be readily scaled up for treating PFAS-contaminated waters. Recent research has demonstrated the potential for regeneration of spent GAC using HALT. HALT achieved >99% destruction of PFOS and 96% defluorination of PFOS-loaded GAC, with no detectable organofluorine intermediates. HALT also degraded >96% of a spent GAC with a PFAS mixture from a field-scale pilot study (Soker et al., 2023). Notably, repeated HALT cycles did not negatively impact GAC surface area or adsorption capacity (Soker et al., 2023).These results suggest HALT enables effective on-site regeneration of spent GAC as an alternative to thermal treatment.

3.2.2.2. Spent ion exchange (IX) resins and IX regeneration solution. IX resins used for PFAS removal are typically single-use resins, in contrast to their regenerable counterparts for other contaminants like nitrate. While a limited number of regenerable PFAS-adsorbing resins exist in the market, they require high volumes of methanol for effective PFAS extraction, thus creating a concentrated PFAS-laden methanol-based concentrate (Ellis et al., 2022). Also, until regulatory guidance is available, spent IX resins are being disposed of in landfills; however, a recent study investigating SCWO application to treat spent IX resins may provide an alternative option (Chiang et al., 2023). Using two different spent IX resins from treatment of AFFF-contaminated waters (groundwater and an Airforce base wastewater influent), SCWO effectively destroyed PFAS with 99–100% efficiency based on a rate-based elimination calculated by comparing the feed rate (rate at which PFAS enters the system) to the effluent rate (rate at which it exits the system (Chiang et al., 2023). Cement kilns incinerators or waste-to-energy incinerators are also being explored as potential options for management of spent resins (EPA.U, 2020b). These units operate at temperatures and residence times that can effectively destroy PFAS. It is important to note, however, that research gaps still persist, particularly in determining the optimal temperature and residence time requirements for destruction of various PFAS. This is especially true given the findings of a recent study where PFCAs were detected in waste-to-energy residuals which was attributed to the incomplete destruction of precursors (Björklund et al., 2024).

If regenerable IX resins become a full-scale treatment approach for PFAS in municipal waters in the future, then a solution would be needed to dispose of the PFAS-laden regenerant. Emerging treatment technologies such as electrochemical oxidation (EO), have been explored as effective means to destroy PFAS in these concentrated solutions. EO involves the transfer of electrons from PFAS to the anode facilitating efficient degradation of PFAS (e.g., Pierpaoli et al., 2021) and Liang et al. (2022) utilized EO to treat spent regenerant solution (approximately 80% methanol by volume and 2% of sodium iodide salts by weight) from a pilot-scale IX resins employed for PFAS treatment in AFFF-impacted groundwater. This study incorporated a novel resin regeneration process that enabled the recovery and reuse of the majority of the regenerant solution through distillation. However, one of the serious challenges of applying destructive technologies such as EO is generation of unwanted side products, particularly perchlorate formation. During EO, the chloride in water can get oxidized to free chlorine, chlorate (ClO_3^-), and then perchlorate (ClO_4^-). Trihalomethanes and haloacetic acids are additional byproducts generated during the disinfection process of drinking water due to interactions between free chlorine and natural organic materials (Bond et al., 2012). Therefore, some researchers have focused on the control of byproduct formation during EO processes in a real-world context and evaluated possible "Control Strategies" (Yang, 2020). While there are several strategies discussed by Yang (2020), one of the approaches that appears to be the most promising and feasible is quenching byproduct precursors. Hydrogen peroxide (H_2O_2) is suggested as an ideal quencher because it is effective, inexpensive, and widely used in site remediation projects. Overall, while electrochemical-based technologies have shown effectiveness, especially in destruction of PFOA and PFOS, further research is needed for evaluating real-world PFAS concentrations and potential toxic by-products (Liu et al., 2023a,b). Energy consumption can range from 153 to 256 kWh/m^3, depending on the level of removal (Gomez-Ruiz et al., 2017).

3.2.2.3. Nanofiltration/reverse osmosis concentrate. NF and RO processes may yield highly concentrated waste streams, necessitating additional treatment or residual management. For example, Wang et al. (2023b) applied EO to RO reject samples from a municipal WRRF and reported 59% destruction of total PFAS tested. In addition, UV, which employs ultraviolet light at 185 nm wavelength to generate hydrated electrons, has been demonstrated to reductively defluorinate a wide variety of PFAS (Bentel et al., 2019; Qanbarzadeh et al., 2023; Sahu et al., 2018). Low-pressure UV at 254 nm, which is currently used for UV disinfection and UV/AOP, can be paired with certain chemicals to produce radicals that can reductively defluorinate PFAS referred to as a UV-advanced reduction process (UV/ARP). C. J. Liu et al. (2021a) employed UV/sulfite photolysis in a field demonstration of a pilot-scale hybrid NF and UV-sulfite treatment train for PFAS remediation in AFFF-impacted groundwater at a DoD installation. UV-sulfite photolysis of the 10% reject water from NF resulted in degradation of >75% of the detected PFAS after 4 h of treatment, and >90% destruction when treatment was extended to 8 h. Factors such as initial PFAS concentration, sensitizer selection (iodide, sulfite, nitrilotriacetic acid, and indole derivatives), pH, UV type and intensity, dissolved oxygen and the presence of coexisting ions and organics were found to influence the defluorination rate (Fennell et al., 2022; Qanbarzadeh et al., 2023; Yang et al., 2020). Notably, a chain-shortening mechanism during the process produced partially defluorinated products, including short-chain analogues of PFOA (Bentel et al., 2019). UV/ARP has limitations such as selectivity towards specific PFAS like PFCAs, slower degradation of PFSAs, energy requirements for UV light sources, and the potential formation of intermediate products or incomplete degradation of PFAS. Energy requirements vary, ranging from less than 13.1 KWh/m^3 for PFCAs, 14.1 kWh/m^3 for PFOS to greater than 100 kWh/m^3 for the more recalcitrant short-chain PFSAs (C. J. Liu et al., 2021a). Limited pilot-scale studies have been conducted and further research is necessary to overcome limitations for successful full-scale application of this technology.

3.2.2.4. Foam fractionate/concentrate. Novel applications of FF present another dimension of managing PFAS-containing liquid residuals - the foam fractionate/concentrate (FFC). Wang et al. (2023b) applied EO to FFC from the final FF stage as well as the original environmental samples collected from various sources including landfill leachate samples, AFFF-impacted groundwater, municipal WRRF RO influent, RO reject samples, and electroplating wastewater. The results indicated destruction percentages ranging from 59 to 95% for FF and from 56 to 89% for the original samples pre-FF, with no consistent trend in PFAS destruction percentages between the two, suggesting that the destruction technologies were similarly effective for both FF and the original samples.

3.2.3. Potential destructive treatment options for PFAS-containing residuals

3.2.3.1. Plasma-based treatment. Plasma-based technologies have recently emerged as a promising approach for degrading persistent PFAS in water. Plasma refers to ionized gas containing highly reactive oxidative and reductive species, as well as other active components like electrons, photons, and heat. It is generated by applying a strong electrical discharge in a gas or at a gas-liquid interface. Several studies have demonstrated the ability of different plasma reactors to effectively degrade a range of PFAS including long-chain PFCAs like PFOA, PFSAs like PFOS, and fluorotelomer sulfonates (FTSs) (Singh et al., 2021; Stratton et al., 2017). Removal efficiencies >90% have been reported for long-chain PFCAs and PFSAs (Singh et al., 2021). Singh et al. (2021) reported 99.9% removal of long-chain PFAS, 10–99% removal of short-chain PFAS, and approximately 60% removal of total oxidizable precursors from landfill leachate using a plasma reactor. Furthermore, enhancements in PFAS degradation have been achieved through innovative approaches such as introducing microbubbles (Zhang et al., 2021) or adding cationic surfactants, which has improved removal of short-chain PFAS by up to 95% (Singh et al., 2020). Following PFAS destruction, products like fluoride, inorganic carbon, and by-products (e.g., smaller organic acids and cyclic perfluoroalkanes) have been identified and quantified (Singh et al., 2019a). Co-contaminants appear to have minimal effects on PFAS degradation, as reactive species directly attack PFAS accumulated at the plasma-liquid interface. This resilience to co-contaminants makes plasma promising for ROC, NFC, and FF. Required treatment times range from minutes to hours depending on the matrix and initial PFAS concentrations. Also, high solution conductivity, greater than 20 mS per centimeter (mS/cm), can reduce treatment efficiency (Lewis et al., 2020). This factor could be disadvantageous for ROC applications since ROC has elevated TDS. Among different plasma systems, gliding arc and gas discharge reactors operated in an argon or air atmosphere have resulted in high PFAS destruction efficiencies (Lewis et al., 2020; Singh et al., 2019b). Successful pilot-scale application of plasma reactors for PFAS removal have also been reported (Nau-Hix et al., 2021; Singh et al., 2019b, 2022). Plasma-based technologies can potentially be scaled up for treating large volumes of contaminated water. Energy requirements are competitive at approximately 10 to 1000 kJ per liter(s) (kJ/L), with electrical energy per order values comparable to other advanced oxidation processes (AOP) (Lewis et al., 2020; Singh et al., 2019b). In addition, an Onvector-developed plasma vortex system was demonstrated at a pilot-scale to consume 70% less energy than a plasma reactor with similar removal rates. Overall, plasma-based PFAS treatment is a promising novel solution that warrants further research and development specific to drinking and wastewater treatment, leachate and resulting residuals management.

3.2.3.2. Emerging destruction technologies. Novel methodologies for PFAS degradation, including activated persulfate and sonochemical oxidation, have demonstrated encouraging efficacy. However, their widespread implementation is hindered by inherent limitations and constrained scalability. Ozonation, an AOP where reactive oxygen species (ROS) degrade organic compounds, has negligible efficacy (Glover et al., 2018; Kumar et al., 2021; Takagi et al., 2011) for PFAA degradation given the strong C–F bonds (Franke et al., 2019; Lashuk et al., 2022). Therefore, it has been coupled with photocatalysis, activated carbon, biological filter, alkaline treatment, and UV to improve PFAS degradation efficiency in bench- and pilot-scale studies where researchers have achieved 7–97% removal of only long-chain PFAS (Dai et al., 2019; Kaiser et al., 2021; Lashuk et al., 2022; Sun et al., 2018; Thomas et al., 2020).

Persulfate ($S_2O_8^{2-}$) is a potent oxidizing agent with a redox potential of 2.1 V (Petri et al., 2011) and is effective in breaking down common groundwater contaminants such as chlorinated solvents (Chen et al., 2016; Ji et al., 2015; Lin et al., 2016). UV/persulfate is a UV/ARP in which UV photolyzes persulfate into sulfate radicals ($SO_4^{\bullet -}$) which can then react with PFAS (Hori et al., 2005; Lutze et al., 2018). Complete mineralization of PFOA through UV/persulfate is noteworthy, with end products primarily being fluoride (F^-) and CO_2 (Hori et al., 2005; Wang et al., 2010). Another innovative approach involves activating persulfate at room temperature using transitional metals like silver, proving particularly potent for PFCAs while being ineffective for PFOS (Parenky et al., 2020). Conclusively, while activated persulfate oxidation has showcased its potential in degrading PFCAs and sulfonates (Park et al., 2016), the method's efficiency tends to waver under alkaline conditions (Hori et al., 2008a, 2008b; Lee et al., 2012; Schröder and Meesters, 2005). It is worth mentioning that to the best knowledge of the authors no known studies have evaluated the effectiveness of UV/persulfate on PFAS destruction in ROC, NFC, or FF. The energy required for persulfate degradation of PFAS is on the order of 5000 kWh/m^3 (Lee et al., 2022). Despite promising results in controlled environments, scaling up these combined processes for full-scale implementation poses significant technical and cost-related challenges.

Sonochemical treatment, another type of AOP, utilizes ultrasound waves over 16 kHz (kHz) to induce acoustic cavitation, creating ROS that oxidize organic pollutants (Chowdhury and Viraraghavan, 2009; Pétrier et al., 2010). This technique has emerged as a possible tool for PFAS remediation due to its simplicity and lack of secondary pollutant generation (Cao et al., 2020; Vecitis et al., 2008, 2009). However, the effectiveness of sonochemical treatment is influenced by various factors, including temperature, atmospheric conditions, and impurities or additives (Campbell and Hoffmann, 2015; Cao et al., 2020; Nzeribe et al., 2019; Shende et al., 2023). This is attributed to cavitation and mass transfer processes occurring at the bubble-water interface (Cao et al., 2020). Despite its potential, there are challenges like scalability and addressing variances in real-world environmental conditions. Field trials have identified barriers, with certain compounds inhibiting degradation efficiency (Collings et al., 2010). Kulkarni et al. (2022) reported the energy required to degrade PFAS by an order of magnitude ranged from 940 to 2500 kWh/m^3 for combined PFOA + PFOS, which is higher than other destruction technologies. However, further exploration is essential before concluding the practicality or not of implementing ultrasound technology at the pilot- and field-scale for treating influents and effluents in wastewater with elevated PFAS concentrations. Given its substantial energy requirements, this technology is particularly well-suited for addressing wastewater scenarios characterized by relatively small volumes and significantly high PFAS concentrations such as return flows. To maximize its effectiveness and minimize operational costs, there is potential for integrating ultrasound treatment with complementary processes such as physical separation (e.g., FF) and other AOPs. The energy requirement for sonochemical oxidation is on the order of 5000 kWh/m^3 (Lee et al., 2022). Given all these limitations, such destruction technologies have minimal application in degrading or removing PFAS from concentrated wastes. Another newer technology being developed include an Electric field-assisted nanofiltration for PFOA removal with exceptional flux, selectivity, and destruction demonstrates an electric field-assisted nanofiltration system, which achieves 97% PFOA rejection and high water flux with low energy consumption by placing a nanofiltration membrane between reactive electrodes, offering a practical solution for PFAS removal and degradation from water (Ji et al., 2023). Overall, the high energy demands and sensitivity to environmental conditions can hinder its feasibility for widespread use in larger-scale applications.

In addition to residuals from treatment technologies specific for PFAS removal in WWTPs, Biosolids are also considered as one of the main residuals from wastewater treatment plants as illustrated in Fig. 2 and discussed in next section.

3.2.4. Biosolids

PFAS are typically not effectively degraded through traditional wastewater treatment processes and tend to accumulate in various locations, including biosolids. During the treatment of influent wastewater, organic matter is biologically degraded, while solids are separated from the liquid phase. This results in the formation of sludge, which undergoes further processing in many cases to produce biosolids. These biosolids are typically rich in organic matter and nutrients, making them a valuable resource for land application as a soil conditioner and fertilizer.

In recent years, biosolids, traditionally considered a valuable resource in wastewater treatment, face evolving challenges in their management due to PFAS contamination concerns. Traditionally, most biosolids are land applied as a fertilizer, disposed of in landfills, or incinerated. However, concerns over industrially impacted biosolids with high levels of PFAS contamination have impacted the feasibility of land application and landfilling for some utilities. Although many of the treatment technologies presented previously can greatly reduce PFAS concentrations in biosolids when applied for industrial wastewater streams upstream of wastewater treatment plants, the main challenge remains regarding municipal wastewater. Treatment technologies applicable to PFAS management in biosolids will be explored here.

3.2.4.1. Incineration. There is an increasing interest in understanding PFAS fate during incineration of biosolids. Current research suggests that PFAS destruction occurs to some extent during incineration processes. For example, a recent study conducted at an undisclosed municipal WRRF operating a fluidized bed sewage sludge incinerator (SSI) with typical operator conditions of 830 °C and a residence time of 2 s. The study showed approximately 51% PFAS destruction for a total of 32 PFAS including PFCAs, PFSAs, FTSs, sulfonamides, and GenX (Seay et al., 2023). Generally, temperatures above 1,000 °C are required to completely destroy PFAS but traditional biosolids incinerators only operate up to 850 °C. There are also concerns that low temperature destruction might release intermediate fluorinated compounds such as fluorocarbon and hydrofluorocarbon gases, some of which can contribute to ozone depletion in the atmosphere. Although achieving temperatures above 1,000 °C is possible, there are a limited number of facilities that can reach these temperatures necessary to destroy PFAS. Winchell et al. (2021) focused on PFAS fate in incineration systems, particularly SSIs. PFAS levels in products from pollution control measures during incinerators such as flue gas, or ash can be undetectable. However, scrubber water from SSIs has been found to contain high concentrations of PFAS. This scrubber water is usually collected in plant return systems, further complicating the overall management of PFAS residuals in thermal treatment systems. For example, S. Liu et al. (2021b) found that leachate from municipal solid waste (MSW) incineration plants contained high levels of PFAS (mostly shorter-chain), with substantial annual discharges (up to 384 kg from one plant in China), while PFAS (mostly PFOS) levels were relatively lower in fly ash (mean 16.4 ng/g) and bottom ash (14.6 ng/g), indicating effective destruction of PFAS during high-temperature incineration. Fournie et al. (2023) reported complete 'removal' of PFAS (PFAS 4C–8C) following smoldering in sewage sludge. However, in most cases, loss was due to volatilization with the exception of some degradation at higher temperatures and with CaO additions. For example, in laboratory-scale, PFOS and PFOA were effectively eliminated from the sludge, but their high concentrations in the gaseous emissions (comprising 79–94% of the total PFAS by mass) indicated volatilization without undergoing degradation (Fournie et al., 2023). Addition of CaO reduced 97–99% of the emissions and minimal PFAS was found in the ash, thus assumed to be mineralized. Further full-scale studies are required to confirm the efficiency of PFAS destruction during incineration and to investigate the potential generation of other compounds of concern. While laboratory-scale research has shown near complete PFAS decomposition of 99.99% at representative temperatures, it has also identified the presence of fluorinated by-products (Winchell et al., 2021). Factors like residence time and turbulence, in addition to temperature, play critical roles in destruction in combustion systems. Full-scale SSIs, which provide longer residence times may promote PFAS destruction.

During incineration, several complexities are anticipated. For instance, although advanced analytical methods have been developed that surpass the EPA's list of 40 targeted compounds in standard methods such as Method 1633 (EPA.U, 2024e), there remains a significant gap in air emissions testing. Air emissions undergo sampling and analysis using Other Test Methods (OTMs). Introduced in 2021 (USEPA, 2021), OTM-45 represents the inaugural PFAS air emissions test method for stationary source air emissions. This method can assess 50 semi-volatile and non-volatile compounds in both gaseous and particulate-bound media. Currently, OTM-45 is unable to detect non-polar PFAS; however, modifications are being made to the method to accommodate non-polar species. Furthermore, the development of OTM-50 is in progress for non-polar volatile PFAS compounds using whole air canisters, and OTM-55 is under development for non-polar, semi-volatile, and non-volatile PFAS compounds, encompassing fluorotelomer alcohols (FTOHs) and products of incomplete combustion or destruction (PICs or PIDs). However, challenges persist with these methods, as only a limited number of commercial laboratories are equipped to perform the analyses, resulting in a scarcity of reported data.

The EPA has yet to establish a risk assessment or air emission standards that would clarify the required destruction percentage, acceptable trace emissions, or required flue stack monitoring and air pollution controls. It is crucial to comprehensively evaluate the fate of PFAS in full-scale SSIs, understand their presence in wastewater solids, identify significant compounds in stack emissions and pollution control residues, and develop strategies to minimize PFAS emissions and their associated risks.

3.2.4.2. Pyrolysis. Recent case studies have demonstrated the potential of thermal treatment in achieving high levels of PFAS destruction in biosolids. For example, a pilot study with a commercial pyrolysis system coupled to a thermal oxidizer demonstrated PFAS removal efficiencies ranging from 81.3 to >99.9% (Thoma et al., 2022). However, during biosolids' pyrolysis, shorter-chain PFAS, volatile organofluorine compounds, CO_2, HF, and solid residues like coke were generated (Zhang et al., 2023). It is difficult to quantify such byproducts without proper analytical methods, but based on the limited studies that have determined the emission rates during thermal treatment, minor emission rates for products of incomplete destruction (PICs) are expected. Minnesota Pollution Control Agency (2023) suggests pyrolysis followed by thermal oxidation as a potential technique for PFAS destruction in biosolids.

3.2.4.3. Hydrothermal liquefaction. A recent study demonstrated the PFAS destruction in wastewater sludge using hydrothermal liquefaction under temperatures ranging from 260 to 350 °C for a duration of up to 90 min and pressures ranging from 4 to 16.5 MPa showed removals of >99% PFCA, 34% PFSAs such as PFOS and 67% 8:2 FTS (Yu et al., 2020). Although this is a promising technology for recovering energy in the form of liquid biofuel, there are concerns regarding the partitioning of PFAS into the liquid biofuel, and the management of liquid and solid phase byproducts.

3.2.4.4. Thermal hydrolysis. Thermal hydrolysis (TH) offers several advantages for the digestion of biosolids, including improved dewaterability and pathogen reduction (Garg et al., 2023). However, it has minimal impact on PFAS concentrations. Research by Lazcano and colleagues (Kim Lazcano et al., 2019) found that THP had no significant effect on PFAS concentrations, in contrast to heating and composting methods which increased PFAS concentrations primarily by promoting biotransformation. This is not unexpected given the temperature utilized for TH being below 1000 °C, which is insufficient to achieve the effective destruction of PFAS.

3.2.4.5. Supercritical water oxidation. While this SCWO technique has shown promising results for destroying PFAS in liquid streams (Jama et al., 2020), no known studies have evaluated the effectiveness of SCWO for PFAS destruction in biosolids. However, there is an active SERDP-ESTCP project (SERDP-ER22-3384, 2022), which looks at bench-scale demonstration of PFAS destruction in solids including soils using SCWO. The primary aim of this proof-of-concept endeavor is to explore the technical viability of employing SCWO to eliminate PFAS from solid matrices, particularly in the context of a soil/sludge slurry. To the best knowledge of the authors, there is no full scale application of SCWO for PFAS removal in biosolids (Munson et al., 2023).

3.2.4.6. Sonochemical oxidation/ultrasound. Lab-scale studies have shown up to 90% PFOS destruction in biosolids using frequencies ranging from 400 to 1000 KHz and when treated for more than 4 h (James Wood et al., 2020). This technology has been identified by EPA's PITT as a potential non-combustion destruction method for PFAS in biosolids (EPA.U, 2021d).

3.3. PFAS in residuals: persistence and management

It is essential to exercise caution when considering the utilization of treatment technologies individually or in combination and to carefully assess the pre-treatment requirements. As advised by the Minnesota Pollution Control Agency (2023), pre-treatment costs can sometimes outweigh the expenses associated with PFAS removal and destruction. Factors such as site-specific objectives, conditions, and limitations can significantly influence options for and potential efficacy of source control or PFAS treatment, necessitate site-specific evaluations. Minnesota Pollution Control Agency (2023) has explored several PFAS management alternatives, providing insights into their estimated capital and operational costs. Table 1 provides an overview of the treatment technologies discussed in this study, along with details about successful pilot studies, advantages and disadvantages, energy consumption, and commercially available vendors for these technologies. Fig. 3 represents a graphic depicting the applicability of treatment technologies.

Furthermore, it is imperative to acknowledge that managing PFAS-laden residuals from treatment technologies is not without its challenges. For instance, in certain states like New York, the disposal of investigation-derived waste (IDW) mandates out-of-state transportation, leading to elevated disposal costs. Furthermore, a growing number of site owners are apprehensive about sending waste to landfills due to potential liability concerns stemming from PFAS-contaminated leachate. As highlighted by Grieco et al. (2022), the disposal landscape for PFAS-contaminated solids, IDW, and single-use sorptive media, such as GAC and IX resins, has become increasingly complex and expensive in recent years due to mounting regulatory scrutiny.

3.4. Cost

The establishment of MCLs for six PFAS has sparked growing conversations within the water industry about the potential cost of removing PFAS from the natural environment. Based on the current rate of release, estimates suggest the annual expense could range anywhere from 20 to 7000 trillion USD (Ling, 2024). Notably, the cost of implementing PFAS residual management strategies is a critical factor to consider, intertwined with energy consumption. Costs play a pivotal role in the feasibility and scalability of PFAS residuals management techniques. These cost variations, along with energy consumption details, are comprehensively summarized in Table 1, offering a clear comparison

Table 1
Comparative analysis of PFAS treatment methods.

Technologies	Treatment	Pilot Test/Full-Scale System	Pros	Cons	Energy	Commercial Vendors
Separation Technologies	GAC/IX Resins	(Belkouteb et al., 2020; Chow et al., 2022; Liu et al., 2019, 2022; Medina et al., 2022; Murray et al., 2019; Rodowa et al., 2020; Woodard et al., 2017; Pannu and Plumlee, 2021)	Effective for removing a wide range of PFAS; suitable for large-scale WRRFs	Regeneration and disposal of spent media can be costly; may generate secondary waste, not effective for shorter-chain PFAS	0.01 kWh/m^3 ((Yadav et al., 2022)	Calgon Carbon, ECT2
	FF	(Burns et al., 2022; Krögerström, 2021; Smith et al., 2022)	Low energy consumption, potential for scalability, easily can integrated into existing infrastructure in water and WRRFs	Limited removal for shorter- chain PFAS, the foamate should be treated for destruction	3–7 kWh/m^3 (Yadav et al., 2022)	EPOC, Allonia, Altra-Sanexen, Evocra
	RO	(Safulko et al., 2023; Appleman et al., 2014)	High removal efficiency for PFAS; can be applied at water treatment plants	High energy consumption; concentrated waste stream disposal	RO: 0.4 kWh/m^3 (Yadav et al., 2022) NF: 0.528 kWh/m^3 (Das and Ronen, 2022)	DuPont, Toray, Xylem
Destructive Technologies for Concentrated Waste Streams/ Residuals	Plasma	(Nau-Hix et al., 2021; Singh et al., 2019b, 2022)	Efficient PFAS destruction; requires no chemical additions and produces no residual waste; total organic compound and co-contaminants do not affect the process; mobile and scalable; versatile in bench and continuous systems	High energy consumption; specialized equipment and expertise required; limited removal of short-chain PFAS; excessive foam formation caused by argon bubbling	2-6 kWh/m^3 PFOA: 28 (Nzeribe et al., 2019)	Clarkson University/GSI Solutions, Purafide, Onvector
	UV/sulfite	(C. J. Liu et al., 2021a)	Effective for certain PFAS; relatively lower energy consumption compared to some alternatives	Limited applicability to specific PFAS; scalability challenges for large treatment volumes	<13.1 to >100 kWh/m^3 (C. J. Liu et al., 2021a)	Xylem
	Supercritical Water Oxidation	(Battelle, 2021; Jama et al., 2020)	High PFAS destruction efficiency; potential for treatment at wastewater facilities	High energy and equipment costs; limited commercial availability	Aqueous: 250 kWh/m^3 Solids: <2500 kwh/ton (Denmark, 2021)	374 Water's AirSCWO™, Revive Environment,
	UV/Persulfate	Coupled with ozonation (Franke et al., 2019)	Effective for PFAS destruction; versatile in various water matrices	May require activation mechanisms; byproducts may require further treatment	PFOA: 5000 kWh/m^3 (Lutze et al., 2018) TFA: 55 kWh/m^3 (Lutze et al., 2018)	LENNTECH (ozone coupled with iron-oxide based catalysis and persulfate)
	EO	Liang et al. (2022)	Efficient PFAS degradation; potential for treatment at water treatment plants.	High energy consumption; electrode fouling challenges; byproducts may require further treatment	29 PFAS in WRRFs: 153–256 kWh/m^3 (Gomez-Ruiz et al., 2017) PFOS: 99–136 kWh/m^3 (Schaefer et al., 2018)	Aclarity, Batelle, Axine, OVIVO
	Sonochemical Oxidation	Kulkarni et al. (2022)	Effective PFAS degradation	High energy consumption; limited pilot/field scale studies; longer treatment times	PFOA: 5000 kWh/m^3 (Nzeribe et al., 2019) 940–25,000 kWh/m^3 Kulkarni et al. (2022)	PCT Systems Inc.
	Alkaline Hydrothermal	(SERDP-ER21-7591, 2021)	Potential for PFAS destruction; adaptable to various matrices	Limited commercial availability; scalability challenges.	110–317 kWh/m^3 127 kWh/m^3 (Aquagga, 2023; Pinkard et al., 2023; SERDP-ER18-1501, 2018)	Aquagga, Inc
	Thermal Treatment	(DiStefano et al., 2022; Shields et al., 2023; Winchell et al., 2021)	High destruction efficiency for PFAS; Effective for a broad range of contaminants; Well-established technology	High energy consumption. High initial capital and operational costs. Limited scalability for large WRRFs due to size constraints	55 kWh per wet ton (Thoma et al., 2022)	Clean Harbors, Calgon
Other	Zeolites	NA	Good PFAS adsorption capacity; potential for regeneration	Regeneration processes can be energy-intensive; limited scalability for large treatment volumes	–	Calgon Carbon
	Coagulants	NA	Can be used in conjunction with other treatment methods; relatively low cost	May require careful optimization for specific PFAS; effectiveness can vary.	–	PerfluorAd®
	CAC/Deep Well Injection	(McGregor, 2023) Niarchos et al. (2023)	Effective for deep well injection; potential for PFAS immobilization in geological formations	May require substantial energy for deep well injection; site-specific feasibility.	–	Calgon Carbon, Regenesis

(*continued on next page*)

Table 1 (*continued*)

Technologies	Treatment	Pilot Test/Full-Scale System	Pros	Cons	Energy	Commercial Vendors
	Surface Modified Clay Adsorbent	(Das et al., 2013; Grieco et al., 2021; Hwang and Grieco, 2021; Pannu and Plumlee, 2021)	High PFAS adsorption capacity; potential for regeneration	Regeneration processes can be energy-intensive; limited scalability.	–	Fluoro-Sorb® matCARE™
	Cyclodextrin Polymer	NA	Selective for certain PFAS; potential for innovative applications	Limited effectiveness for a broad range of PFAS; scalability challenges.	Carboxycellulose nanofibers: 1.49 kWh/g (Li et al., 2021)	CycloPure
	Biodegradation	NA	Environmentally friendly and sustainable; low operational and maintenance costs; applicable to a wide range of PFAS	Lack of sufficient research supporting the biodegradation and mineralization of PFAS with bacteria	NA	NA

Notes: kwh/ton = Kilowatt hour per ton NA = Not applicable.

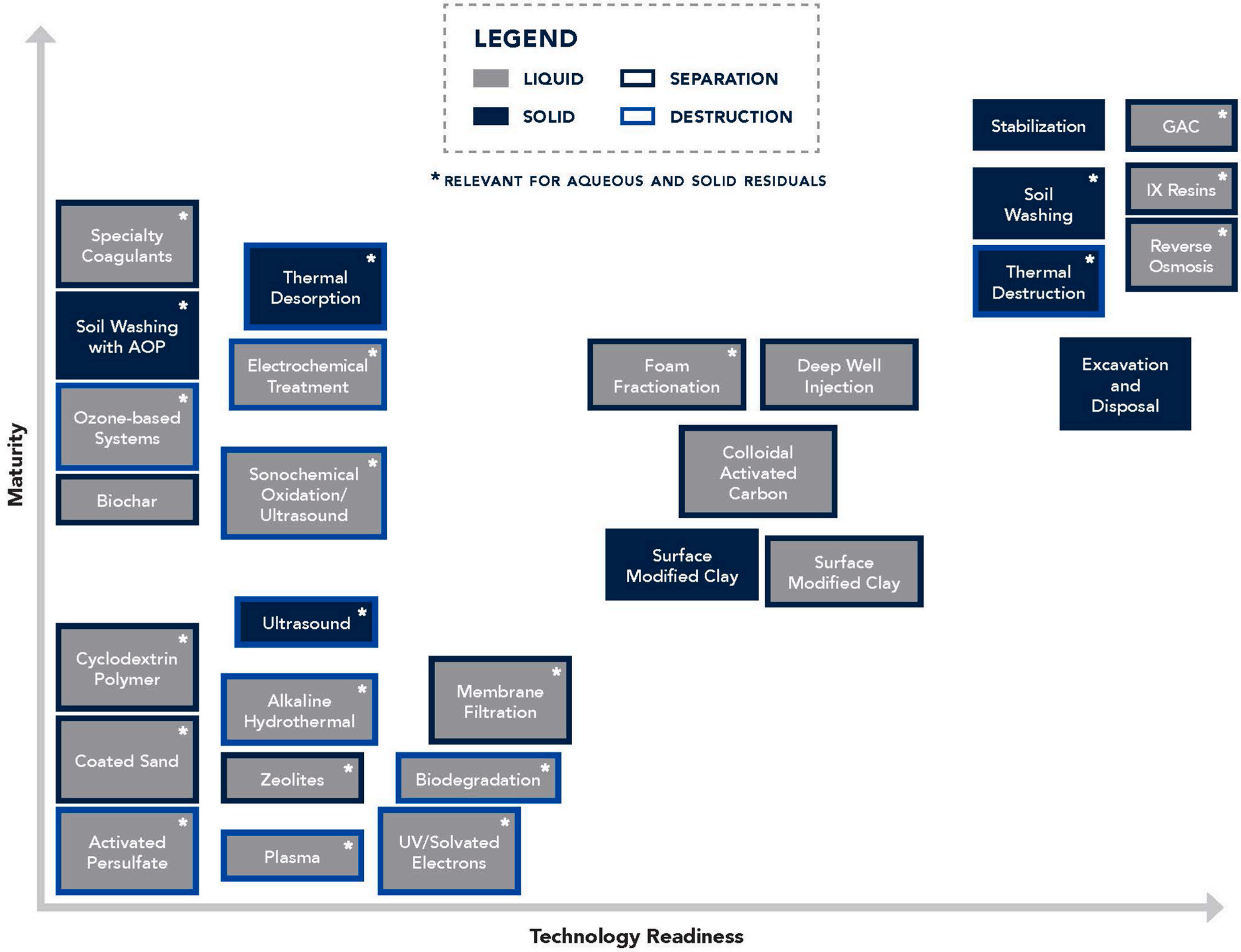

Fig. 3. Progress on applicability of PFAS removal techniques.

of the economic and environmental aspects of each technique. It is imperative to bear in mind that when making decisions regarding PFAS residuals management, not only should the individual technique costs be considered, but also the potential benefits of employing treatment trains, where separation technologies are coupled with destruction methods. The cost-effectiveness of such integrated approaches can significantly impact the overall strategy for addressing PFAS residuals. Therefore, a thorough evaluation of costs coupled with energy considerations, are paramount for making informed decisions in the quest for effective PFAS management.

4. Conclusions and recommendations for effective PFAS residuals management

4.1. Regulatory landscape and evolving science

The regulatory landscape and evolving science surrounding PFAS is undergoing significant changes in response to growing concerns about their adverse health effects and widespread environmental contamination. While voluntary phase-outs of legacy PFAS by manufacturers have been initiated, the emergence of replacement PFAS with unknown toxicity underscores the need for comprehensive regulation and management. The EPA has made notable strides in addressing PFAS concerns, issuing a national primary drinking water regulation for specific PFAS. These regulatory efforts primarily focus on drinking water due to its relatively straightforward risk assessment. However, studies indicate the majority of human PFAS exposure actually occurs through diet. The industrial and municipal wastewater sectors are also under scrutiny, with a growing emphasis on monitoring, treatment, and regulation of PFAS in these areas. Biosolids management is subject to state-specific regulations, with several states implementing PFAS concentration limits and monitoring requirements. However, the environmental, social, and financial impacts of these regulations on biosolids management costs, as demonstrated by increased landfilling and loss of resource recovery, highlights the complexities associated with PFAS regulation. Despite these efforts, uncertainties remain. Upcoming regulatory actions or future triannual updates to the EPA's PFAS destruction and disposal guidance may one day provide clarity on disposal best practices for this pressing environmental and public health issue. Therefore, policymakers should focus on establishing national-level guidelines for PFAS disposal and destruction. Additionally, policymakers can play a crucial role in funding research into alternative materials to PFAS and supporting innovative treatment technologies through incentive programs for industry. This collaborative approach will help to ensure that PFAS regulations are adaptable to new scientific discoveries.

4.2. PFAS separation technologies

Separation technologies play a pivotal role in addressing the challenges associated with these persistent contaminants. These techniques offer several advantages, including the ability to selectively target and concentrate PFAS from various environmental matrices. GAC and IX resins, for instance, have shown remarkable potential in achieving near-complete PFAS removal from contaminated water. Cyclodextrin polymers, with their unique molecular structure, have emerged as potent tools for efficient and selective PFAS removal. Additionally, high-pressure membrane filtration, encompassing NF and RO, provides a robust solution for PFAS removal from aqueous solutions. Their high removal efficiencies are notable, albeit with the challenge of fouling that requires careful consideration. Separation technologies can be a valuable tool for PFAS management for residuals produced downstream. For instance, employing these technologies at PFAS point sources in the sewershed upstream of a wastewater treatment plant can be an effective management strategy for protection of the wastewater treatment plant's biosolids produced. However, it is crucial to address one significant drawback associated with these separation technologies—the generation of PFAS-rich waste streams. Finally, industry stakeholders should prioritize adopting separation technologies at point sources to minimize PFAS entry into water treatment facilities. Concurrently, researchers must focus on developing energy-efficient and scalable separation technologies that can handle diverse PFAS structures and reduce the volume of PFAS-rich waste streams for subsequent destruction.

4.3. PFAS destruction technologies

Destruction technologies often represent innovative approaches focused on the comprehensive eradication of PFAS, targeting their C–F bonds, which are known for their remarkable stability. The aim here is not merely to isolate these persistent compounds but to completely transform them into non-toxic byproducts, primarily inorganic fluoride and CO_2. These methods leverage various physical and chemical mechanisms, presenting promising avenues for mitigating the environmental and health risks posed by PFAS contamination. Among these technologies, plasma-based systems, UV/ARP, SCWO, electrochemical treatments, sonochemical oxidation, HALT, and thermal treatment stand out as potential solutions. They each offer distinct advantages and capabilities, yet they also come with their specific challenges that necessitate careful consideration. Generally, these technologies usually eliminate the resource-recovery opportunities of the residuals when employed. As we explore these methods in greater detail, it is essential to recognize the varying energy consumption requirements associated with each technology. Additionally, byproduct formation is a significant concern that warrants meticulous examination. Furthermore, the scalability of these destruction technologies remains a critical factor in practical implementation. In essence, the journey towards effective PFAS management encompasses a rich tapestry of technologies, with each thread holding a promise for addressing this persistent challenge. Nevertheless, it is essential to navigate all the complexities to ensure a sustainable and comprehensive approach to PFAS remediation. Researchers should focus on enhancing the scalability of destruction technologies, particularly exploring hybrid approaches that combine destruction with separation technologies for optimal results. Industry stakeholders should work towards pilot-scale implementation of destruction technologies like electrochemical oxidation and SCWO to better understand their feasibility for large-scale PFAS management. Additionally, both policymakers and industry need to collaborate on funding initiatives for scaling up these destruction technologies to ensure widespread adoption.

4.4. Challenges and considerations

The management of PFAS-containing residuals presents a multifaceted challenge that demands tailored solutions across various forms of contamination. From advanced treatment methods for liquid concentrates to emerging approaches in biosolids and spent media management, addressing PFAS residuals requires a dynamic and adaptable strategy. However, it is essential to acknowledge the growing complexities in PFAS waste disposal, marked by increasing regulations and concerns surrounding liability. Despite these hurdles, ongoing research and innovative technologies offer hope in the quest for effective PFAS residuals management. As we navigate this evolving landscape, a comprehensive understanding of these strategies empowers us to make informed decisions in our collective effort to combat the persistent environmental threat posed by PFAS contamination. Policymakers should ensure clear guidelines for PFAS waste disposal that minimize liabilities for industries while upholding environmental safety. Researchers must continue to explore innovative technologies that not only remove PFAS but also address safe byproduct management. Industry must focus on compliance with new regulations and proactive strategies to reduce the volume of PFAS-laden residuals in landfills or incinerators by adopting best available technologies for destruction.

4.5. Strategic future pathways

In conclusion, addressing the challenges associated with PFAS-containing residuals management requires a strategic and collaborative approach. Regulatory compliance and advocacy efforts are crucial in navigating the evolving landscape of PFAS regulations and guidelines. Regular monitoring and comprehensive risk assessments are essential to identify contamination risks and potential liabilities. Integrating advanced treatment technologies that combine separation and destruction methods can efficiently remove and mitigate PFAS contaminants while minimizing waste generation. Waste minimization strategies should be explored and resource recovery opportunities should be investigated. Active involvement in research projects and staying up-to-date with the latest advancements in PFAS management are pivotal for informed decision-making and innovative solutions. Environmental responsibility should be a top priority, emphasizing stakeholder engagement and thorough environmental impact assessments. Capacity building, innovation, and pilot projects can drive progress in PFAS management, while collaboration and information exchange among industry stakeholders are vital for collective success. Continuous improvement through performance metrics and long-term planning for sustainability will further enhance PFAS residuals management efforts. By embracing these recommendations, actively participating in research endeavors, and staying engaged in the field, utilities can navigate the complexities of PFAS contamination and contribute to a sustainable and responsible approach to PFAS residuals management. For policymakers, it is essential to continue refining regulations to keep up with evolving science, providing industry incentives for adopting PFAS-free alternatives, and funding research. Industry should prioritize pilot projects to test emerging PFAS separation and destruction technologies, and researchers must explore advanced methods for reducing PFAS waste streams. Collaboratively, all stakeholders should establish performance metrics to assess the long-term effectiveness and sustainability of PFAS treatment strategies.

CRediT authorship contribution statement

Mahsa Modiri: Writing – review & editing, Writing – original draft, Visualization, Validation, Supervision, Resources, Project administration, Methodology, Investigation, Funding acquisition, Formal analysis, Data curation, Conceptualization. **Pavankumar Challa Sasi:** Writing – review & editing, Writing – original draft, Visualization, Methodology, Investigation, Data curation, Conceptualization. **Kyle A. Thompson:** Writing – review & editing, Validation. **Linda S Lee:** Writing – review & editing, Validation, Methodology, Formal analysis. **Katie Marjanovic:** Writing – review & editing. **Graeme Hystad:** Writing – review & editing, Conceptualization. **Kamruzzaman Khan:** Writing – review & editing. **John Norton:** Writing – review & editing.

Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: Mahsa Modiri reports financial support was provided by Water Research Foundation. If there are other authors, they declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.chemosphere.2024.143726.

Data availability

No data was used for the research described in the article.

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