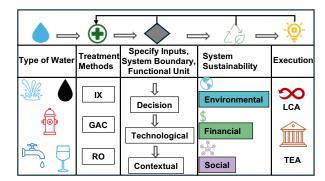
1	Balancing Act: Environmental, Social, and Economic
2	Impacts of PFAS Removal from Water
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Abstract

 Per- and polyfluoroalkyl substances (PFAS) are emerging water contaminants with significant environmental and health impacts, posing challenges in water treatment due to their degradation resistance. This study reviews 10 papers on the sustainability of technologies for PFAS removal, revealing a critical literature gap as regulations emerge. Our review shows sustainability varies across technologies and contexts. Specifically, single-use ion exchange (IX) demonstrates cost-effectiveness and environmental favorability for long-chain PFAS removal from groundwater and aqueous film-forming foam impacted water, while granular activated carbon (GAC) appears costlier due to rapid breakthroughs. These limited findings underscore the need for more comprehensive research to validate results across contexts and understand the full sustainability profile of PFAS treatment technologies, including removal and destruction. Current literature often overlooks key considerations like the ultimate fate of PFAS. To address these gaps, we propose a framework for future sustainability studies, enabling clearer technology evaluations under specific conditions. While IX shows broad applicability, treatment choice should consider water type, system boundary, functional unit, PFAS concentration, and ultimate fate of PFAS for a more holistic view of sustainability.

Graphical Abstract



1.0 Introduction.

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Poly- and perfluoroalkyl substances (PFAS) constitute a group of persistent organic pollutants characterized by strong and stable chemical bonds between carbon and fluorine atoms. 1-5 These substances exhibit remarkable resistance to environmental degradation due to their robust chemical structure, and lack of evolutionary pressure which renders them resistant to heat, alkali, and acid. 6 Consequently, PFAS have found widespread use in various industrial applications, including non-stick cookware, food packaging, 2,8,9 agueous film-forming foam, 9 plastics, 10 and semiconductor manufacturing. 11,12 Since the 1940s, 7 the discharge of PFAS from commercial and industrial sources has led to their widespread presence in water sources worldwide, with concentrations ranging from ng/L to sometimes µg/L levels.^{2,4} PFAS contamination has been detected in various water sources, including drinking water, groundwater, surface water, tap water, and industrial effluents, prompting the establishment of environmental quality standards, and guidelines in many countries. 2,13-15 Recognized as emerging contaminants, some classes of PFAS, such as perfluorooctanoic acid (PFOA), and perfluorooctane sulfonate acid (PFOS), pose significant health and environmental risks due to their carcinogenic properties. 16-21 However, the environmental persistence and bioaccumulative nature of all PFAS make them worrisome for future generations, as these chemicals do not readily degrade and can accumulate in the environment over time. To address this global challenge, numerous organizations have taken regulatory measures. For instance, the Stockholm Convention designated PFOS, and PFOA as persistent organic pollutants in 2009, and 2019, respectively. 22,23 The European Commission has outlined its chemicals strategy, which includes plans to phase out non-essential uses of PFAS within the EU.24,25 The International Chemicals Management has identified PFAS as a priority issue. In 2016, the US EPA established a drinking water health advisory level of 70 ng/L for the combined concentrations of PFOA and PFOS.²⁶ The US EPA recently established the first-ever national drinking water standard for PFAS, which will go into effect with mandatory monitoring by 2027 and compliance by 2029, with specific maximum contaminant levels for PFOA, and PFOS at 4.0 parts per trillion.²⁷ As a result of these regulations, researchers worldwide are actively seeking effective treatment techniques to remove PFAS from water sources.

The predominant focus of recent research lies in investigating the efficacy of various technologies for removing PFAS, encompassing adsorption, membrane filtration, destructive processes, and other methodologies. Among these, ion exchange (IX)^{28–31}, and granular activated carbon (GAC)^{32–34} are the most prevalent adsorption techniques employed for PFAS

removal. However, due to environmental concerns about water contamination and resource depletion, considerable attention has been directed toward the regeneration, reuse, and disposal aspects associated with IX and GAC. ^{35–39} Nanofiltration and reverse osmosis have emerged as leading membrane technologies for PFAS removal because of their high removal efficiencies. ^{40–43} However, their implementation is constrained to specific contexts due to the need for pretreatment measures, high operating cost, and energy demand. ^{44,45} Beyond these conventional approaches, a range of alternative technologies such as oxidation, ⁴⁶ activated persulfate oxidation, ⁴⁷ electrochemical advanced oxidation process, ⁴⁸ plasma, ⁴⁹ and foam fractionation ⁵⁰ have been explored for PFAS treatment from water sources. Driven by the urgency to address environmental PFAS contamination, research has primarily focused on determining the efficacy of these technologies, with little attention given to assessing their sustainability.

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Sustainability, encompassing economic, environmental, and social impacts, is a paramount consideration in PFAS treatment research.⁵¹ While meeting regulatory standards is important, demonstrating financial viability and social impacts are crucial for industry and community acceptance, respectively. For instance, the higher operational and capital expenses associated with IX, nanofiltration, and reverse osmosis in comparison to alternative technologies like GAC make these technologies less applicable in industrial settings.⁵² Sustainability facilitates the selection of treatment methods based on specific needs. The assessment of environmental impacts through life cycle impact assessment (LCA), and financial feasibility through techno-economic analysis (TEA) or life cycle cost analysis (LCCA), serve as prominent methodologies indicating sustainability.⁵³ Social impacts are usually assessed through social sustainability evaluation matrix.⁵⁴ For PFAS, most of the existing research has focused on removal efficiencies using GAC and IX technologies, resulting in limited studies that comprehensively address their economic, environmental, and social impacts. There exists a substantial research gap regarding the sustainability of these technologies, and current literature fails to provide a comprehensive understanding of the overarching concept of sustainability for PFAS treatment. Selecting the appropriate water treatment method requires considering water types, usage, regulations, costs, and final PFAS fate which is essential for informed decision-making via LCA and TEA. However, it is not clear which technologies are the most sustainable across various contexts. Therefore, a comprehensive study is needed to consolidate the literature on sustainability of PFAS treatment for effective decision-making.

This review will consolidate existing research on economic, environmental, and social impacts of removing PFAS from various water sources using different water treatment techniques. The main goals of this study are to clarify the current status, future directions, and necessary actions for sustainable PFAS water treatment. The two major specific objectives are (1) to understand how and to what extent previous literature has evaluated sustainability of PFAS treatment technologies and (2) to develop a potential conceptual framework for characterizing the sustainability of PFAS treatment technologies. This review consolidated literature on evaluating the sustainability of PFAS-contaminated water treatment. From a screening of 278 articles, only 10 studies were identified as relevant to the goals of this study. Information on environmental sustainability, financial viability, and social impacts was extracted and consolidated from these articles to find out the most sustainable treatment technology for different scenarios. Next, a systematic framework was developed for future researchers to guide the selection of sustainable PFAS treatment technologies. This study will provide important insights into sustainability, offering decision-makers a valuable resource for identifying the sustainable and economically viable PFAS treatment technology from water sources.

2.0 Functional Unit to Standardize Sustainability indicators.

Studying sustainability for PFAS treatment from water requires a focus on several key components, such as the functional unit, system boundary, water type, and methodological approaches. These components are essential for assessing the environmental sustainability, social impacts, and financial feasibility of various treatment technologies (Figure 1). By standardizing sustainability indicators through these key components, researchers can develop more robust and comparable assessments, ultimately advancing the field of PFAS treatment and contributing to the broader goal of sustainable water management. Figure 1 illustrates the research focus and gaps in the current body of literature. The thickness of each line represents the number of reviewed papers, highlighting areas where research is concentrated and where further investigation is needed. This visual representation aids in identifying trends, emerging technologies, and underexplored areas, guiding future research efforts towards a more comprehensive understanding of PFAS treatment sustainability. Details on how the literature review was systematically conducted are included in the Supporting Information (SI) Section S1, and Table S1 has all the information used in the development of Figure 1.

2.1 Functional Unit and System Boundary.

Functional unit is a standardized measure used in various sustainability analyses to compare indicators across different technologies, systems, or processes. This measurement is crucial for comparisons on an equivalent basis, ensuring results are based on consistent performance metrics. In the context of sustainability of PFAS treatment, various functional units were identified across the literature, including the mass of PFAS^{55,56} and mass of sludge⁵⁷. However, Figure 1 shows a predominant emphasis on the unit volume of water. Regional and global differences in domestic water consumption, influenced by climate, infrastructure, and societal factors, highlight the challenges of establishing a standardized functional unit. The majority of examined papers opted for a functional unit based on the volume of water, with 70% of the sampled literature employing 1 m³ of treated water as the standard.^{56,58–63} Notably, one paper deviated from this trend, utilizing 800,000 gallons of groundwater per day as its functional unit.53 The chosen functional unit corresponds to the actual daily water consumption of Brighton's population, estimated at approximately 8000 people with an average daily consumption of 100 gallons per capita per day. Landfill leachate is a major source of PFAS in the environment. It is usually transported to wastewater treatment plants and treated with municipal wastewater. Therefore, this review also examines the sustainability of leachate treatment for PFAS removal. The importance of selecting a functional unit is highlighted in sustainability assessments of leachate treatment. When "1 g of PFAS removed" is used as the functional unit, the assessment reveals higher environmental and human health impacts compared to using "1 m³ of leachate treated" as the functional unit. ⁵⁶ Evaluation per mass of PFAS removed is more relevant to human and environmental health, while evaluation per volume of PFAS-impacted water treatment is more relevant to costs and engineering. This demonstrates that the choice of a functional unit significantly influences the overall sustainability outcomes of PFAS treatment technologies. However, the water treatment industry operates based on the volume of water treated, and associated costs are typically calculated per unit volume. Therefore, most of the reviewed papers have utilized water volume as the functional unit.

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A system boundary in the context of sustainability assessments defines the limits of what is included and excluded in the analysis of a system, product, or process.⁶⁴ It determines which stages, processes, flows, and impacts are considered within the scope of the evaluation.^{65,66} For PFAS treatment technologies, clearly defining these boundaries is essential to capture the comprehensive environmental, economic, and social implications associated with each technology. The persistent nature of PFAS necessitates a thorough consideration of the fate of these substances across different treatment methods, yet current literature often overlooks this

aspect. Most of the existing studies have defined their system boundaries around the removal of PFAS, with limited attention given to the ultimate destruction of these compounds (e.g., incineration^{58,59,61}). This omission is significant, as the destruction phase can entail considerable environmental, economic, and social costs that are not accounted for in current assessments. To comprehensively evaluate the sustainability impacts of PFAS treatment technologies, future research should expand system boundaries to include destruction technologies, such as advanced oxidation processes,⁶⁷ electrochemical oxidation,^{67,68} plasma,⁶⁸ and others. This broader perspective will provide a more holistic view of sustainability, enabling the accurate assessment of the true sustainability implications of PFAS treatment technologies.

2.2 Water Type.

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Water type selection is significant in sustainability studies, guiding the choices of treatment technologies based on varying pollutant concentrations and characteristics of PFAS. PFAS concentrations range from 20 to 20,000 ng/L in groundwater, ^{69,70} 11-23 ng/L in well water,⁷¹ 0.4-40 ng/L in drinking water,^{72,73} 5.1 to 298,559 ng/L in landfill leachate,⁷⁴ 3-430 ng/L in wastewater influent, 75-82 and 6-500 ng/L in wastewater effluent 77-79,81-84 systems across USA, China, Europe, and Australia; 85,86 PFAS composition was primarily represented by PFOA, PFOS and perfluorohexane sulfonic acid (PFHxS). Groundwater has been predominantly researched for PFAS treatment, driven by its prevalence, regulatory importance, ease of study, persistence, and ecological impact (Figure 1). 53,58-60,62 Fire extinguishing water, particularly from military and industrial sites using aqueous film-forming foam (AFFF), poses significant challenges for PFAS treatment due to high PFAS concentrations. Fluorine-free alternatives have shown improved biodegradability and reduced environmental persistence compared to AFFF. 61,87 Nonetheless, some alternatives may still present similar or higher environmental impacts. 88,89 Additional research is required to fully understand the sustainability of these alternative firefighting foams. As the volume of landfilled solid waste continues to grow globally, the generation of leachate is expected to increase correspondingly.90 Landfill leachate contains a complex mix of contaminants, including PFAS. 91,92 Its high variability in PFAS concentrations necessitates robust, and adaptable treatment technologies. Studies should emphasize the requirement for systems capable of handling this variability effectively. Drinking water is a critical area for PFAS treatment due to its direct impact on public health. Recent EPA regulation for PFOA and PFOS in drinking water aims to protect communities from the adverse impacts of PFAS including cancer, and developmental impacts in children, and infants.93 Meeting the stringent levels of PFOA and PFOS concentrations requires highly effective and sustainable treatment

technologies, which are still not adequately covered in current literature. Understanding the specific challenges and opportunities of each water type allows for better tailoring of PFAS treatment technologies to achieve sustainability in environmental, social, and financial terms. This comprehensive approach ensures effective mitigation of PFAS contamination across various water sources.

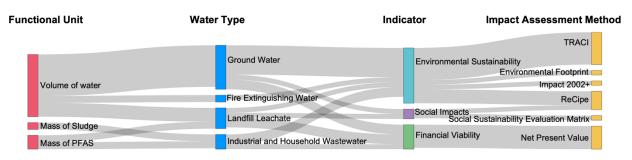


Figure 1. Literature review results for the basis of functional unit and impact assessment method by sustainability indicator (environmental sustainability, social impacts, and financial viability), and water type (ground water, fire extinguishing water, drinking water, landfill leachate, subcritical water, household wastewater). The thickness of each line corresponds to the number of studies reviewed. Full details can be found in Table S1 of the SI.

2.3 Indicator and Impact Assessment Method.

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Assessing the sustainability of treatment technologies for PFAS removal from water sources require a multifaceted approach. In this study, we reviewed existing literature to examine how treatment technologies for PFAS are evaluated in terms of sustainability. We focused on three key indicators commonly used in these studies: environmental sustainability. social impacts, and financial viability. The reviewed studies employed various methodologies, including LCA, LCCA, and TEA to quantify these indicators. Different impact assessment methods were integrated into these evaluations to provide a comprehensive analysis of the sustainability of PFAS treatment technologies. The assessment tools included TRACI, 53,56,58-60,62 ReCipe, 56,57,59 environmental footprint, 61 Impact 2002+,55 social sustainability evaluation matrix.53 and net present value 53,56,57,59,63. These tools have been utilized to assess any treatment technologies environmental sustainability, social impacts, and financial viability values. The selection of impact assessment methodology varies depending on the type of infrastructure and the research objectives. This choice can significantly influence the results due to key differences in how the environmental relevance of indicators is considered. In sustainability studies, particularly in LCA, impact assessment methodologies are generally classified into two main types: midpoint models and endpoint models. These models are used to evaluate the environmental impacts of a system, product, or process at different stages and levels of detail.

Midpoint models provide a greater level of certainty because they focus on specific impact categories, such as carbon footprint, and water footprint, before they are aggregated. In contrast, endpoint models consolidate information into a single score, making them more understandable to decision-makers by summarizing the overall damage to human health, ecosystems, and resources. Methods like TRACI, ReCiPe, and the environmental footprint can serve as either midpoint or endpoint methodologies based on their application. For example, when TRACI, ReCiPe, or environmental footprint are used to assess specific impact categories, they function as midpoint indicators. Conversely, when they are applied to evaluate comprehensive environmental damage, they align with endpoint methodologies. Integrating LCA and TEA with tools like TRACI, ReCiPe, and environmental footprint ensures a detailed analysis of environmental impacts. This integration provides a robust framework for assessing sustainability by balancing detailed environmental data with broader impact evaluations. Simultaneously, the social sustainability evaluation matrix and net present value offer valuable insights into social and financial aspects, respectively. The choice of methodology should ultimately reflect the research objectives. Selecting a method that can be supplemented with additional assessments, such as health and social impacts, aligns research goals more closely with the primary objectives of sustainability. Such a comprehensive approach not only helps identify the sustainable technologies but also facilitates understanding the trade-offs between different sustainability indicators. By balancing environmental, social, and financial considerations, this integrated methodology supports informed decision-making and promotes a holistic view of sustainability.

2.3.1 Environmental Sustainability.

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Environmental sustainability has attracted the most attention among the three sustainability indicators (Figure 1). In the majority of studies, TRACI and ReCiPe methodologies have been commonly employed to evaluate the environmental sustainability of GAC and IX technologies across various water types. ^{53,56–60,62} However, environmental footprint was utilized to assess the environmental impact of GAC in fire extinguishing water, while Impact 2002+ was employed for subcritical water decomposition technology in subcritical water. ^{55,61} LCA assesses environmental impacts using both midpoint and endpoint indicators. Ozone depletion, global warming, smog, acidification, eutrophication, carcinogenic, and non-carcinogenic effects, respiratory effects, ecotoxicity, and fossil fuel depletion are included as midpoint indicators, whereas human health, ecological quality, climate change, resources are part of the endpoint

indicators. Understanding these indicators helps to evaluate the varying impacts of GAC and IX technologies.

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Variations within GAC or IX technologies, such as single-use GAC adsorbents, singleuse IX resins as well as methods for reactivation and regeneration, each carry distinct environmental implications and tradeoffs. The multitude of options available for selection between these technologies significantly influences the results of LCAs. However, numerous studies suggest that from an environmental sustainability standpoint, IX generally exhibits lower environmental impacts compared to GAC. 53,58,59,61,62 IX systems have lower media usage rates (mass of adsorbent media required to treat a given volume of water) due to higher adsorption capacity and longer bed life, leading to less frequent media replacement, and fewer environmental impacts from production and disposal. For instance, single-use IX resins have lower environmental impacts in all indicators except ozone depletion compared to regenerable resins and GAC adsorbents.⁵⁹ Further research supported these findings, showing lower overall environmental impacts for IX when removing long-chained PFAS from groundwater.⁵³ Another study found that GAC generates higher average impacts, excluding ozone depletion, compared to IX-based technologies at various PFAS concentrations. 62 The high ozone depletion impacts from single-use IX resins are due to the release of ozone-depleting chemicals during their production. Despite these impacts, GAC has shown potential in specific treatment scenarios.

Although GAC is less environmentally friendly compared to IX, several studies have identified GAC as a promising treatment technology, particularly for initial high contaminant loads, low dissolved organic carbon, concentrations, longer-chained PFAS including, PFOA and PFOS. 40,53,62,94 Proper reactivation techniques can mitigate GAC's negative environmental impacts. For instance, thermally reactivated GAC has been found to have lower impacts than single-use GAC and even regenerable IX treatments. 9 Utilizing off-site thermal reactivation and reuse of spent GAC media significantly reduces impacts by decreasing the need for new adsorbent material, and operating at lower temperatures (e.g., 815°C compared to 1200°C for hazardous waste incinerators). 99,596 Regeneration techniques for IX resins play a key role in its environmental sustainability. Regenerable IX systems, while generally less impactful than single-use GAC, can have significant environmental impacts due to the infrastructure and chemical-intensive processes required for resin regeneration and regenerant recycling. The environmental impacts of IX resins regeneration could be reduced by recycling methanol and/or brine, thereby, decreasing the amount of waste sent to incineration. However, altering the composition of the regeneration solution requires careful consideration as replacing NaCl with

alternative salts can increase environmental impacts.⁵⁸ Another study showed that the regeneration and reuse of IX resins with electrochemical oxidation in groundwater significantly reduces global warming potential (GWP), especially when the energy efficiency of oxidation is between 192 kWh/m³ and 656 kWh/m³. Regeneration and reuse of IX resins coupling with electrochemical oxidation had the lowest GWP, reducing it by approximately 49% compared to IX following incineration.⁶⁰ These findings suggest that regeneration and reuse of IX resins could be one of the most environmentally friendly options for treating PFAS-contaminated groundwater.

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Several studies have focused on different water sources using various techniques for PFAS treatment. A comprehensive LCA was performed to evaluate three end-of-life options for aqueous film-forming foam containing spent fire-extinguishing waters: functional precipitation agents (PerfluorAd process). GAC, and direct incineration. ⁶¹ All three scenarios ultimately result in the incineration of PFAS. The LCA results showed that the PerfluorAd process performed best across all environmental impact categories for PFAS treatment. Another study evaluated the sorption capacity of optimized sludge-based GAC media for removing nine commonly detected PFAS from simulated wastewater.⁵⁵ Notably, it was found that reducing the ZnCl₂ impregnation ratio from 2.5 M to 1.5 M significantly decreased freshwater ecotoxicity, marine ecotoxicity, and human non-carcinogenic toxicity of the treated water by 49%.⁵⁷ The ZnCl₂ impregnation ratio refers to the ratio of ZnCl₂ used to the precursor material. Specifically, it indicates the amount of ZnCl₂ used in the chemical activation process of the sludge to produce activated carbon. Further techniques such as multistage membrane technology and subcritical water decomposition have also been investigated to find the environmental sustainability of these technologies. Multistage membrane technology for PFAS removal from landfill leachate found that offsite treatment was generally more sustainable than onsite treatment at high PFAS concentrations, highlighting the need for improved membrane, electricity, and chemical usage efficiencies for onsite treatment.⁵⁶ Exploring subcritical water decomposition of PFOS. researchers found that maintaining high temperature and pressure accounted for 99.8% of the environmental impact, suggesting that enhancing energy efficiency and catalytic effectiveness is crucial for reducing the environmental impact of subcritical water decomposition processes.⁵⁵ These findings collectively highlight the environmental trade-offs between various PFAS treatment technologies. While GAC and IX are commonly used, each technology presents unique environmental impacts. The choice of treatment should consider the specific PFAS contamination scenario and focus on optimizing the efficiency and sustainability of the selected method.

2.3.2 Financial Viability.

Financial viability is key for selecting any treatment technology. Even if a technology effectively removes PFAS from water, it won't be adopted if it's not financially sustainable. All the papers we reviewed used net present value to assess the financial viability of their chosen treatment technologies. Researchers used the USEPA's work breakdown structure model to evaluate the financial viability of IX and GAC for PFAS removal from groundwater, considering overall costs during their useful life. The life cycle inventory for each remediation system estimated lifetime costs in real dollars and on a per-unit-volume basis (\$/m³ treated). The results shows that the single-use IX system has lower capital costs compared to the regenerable IX system due to lower media usage rates, longer operational life, and fewer required components (e.g., pumps, piping). While GAC media is generally cheaper, its high media usage rates and larger vessel requirements make it less economically viable than IX treatment for PFAS. The single-use IX system costs \$0.28/m³, whereas the single-use GAC system costs \$0.45/m³, nearly twice the cost. Under the baseline scenario, single-use IX proves to be much more cost-effective than GAC. However, GAC may be cheaper when applying very stringent PFAS breakthrough criteria.

The financial viability of multistage membrane technology for landfill leachate treatment compared offsite and onsite scenarios for PFAS compliance. Both the onsite and offsite scenarios in the study include pretreatment processes before membrane treatment and excluded the final fate of PFAS. The primary cost driver for onsite treatment is operational expenses (45%) due to high electricity and chemical usage, while leachate transportation accounts for 95% of offsite costs. Using 1 g of PFAS treated as the functional unit, the onsite scenario's life cycle cost is 83% lower than the offsite scenario. However, treating 1 m³ of raw leachate costs \$1.96 onsite compared to \$2.50 offsite, reflecting a 21% cost reduction for onsite treatment despite its higher initial costs. 56 The net present values of a two-pass spiral-wound reverse osmosis system for additional leachate treatment in Thailand were \$577.9 million USD for a system with an evaporation pond and \$391.9 million USD for a system without one. Treatment unit costs ranged from \$1.72 to \$2.71 USD/m³ for the system with an evaporation pond and from \$1.06 to \$2.09 USD/m³ for the system without the pond depending on landfill size. 63 The two-pass spiral-wound reverse osmosis system without an evaporation pond shows lower treatment unit costs (\$1.06 - \$2.09 per m³) compared to the onsite multistage membrane technology (\$1.96 per m³). Despite higher initial costs, the overall financial viability is better for the reverse osmosis system than onsite multistage membrane technology, especially without

the evaporation pond. While each PFAS treatment technology has trade-offs, the single-use IX system and the two-pass spiral wound reverse osmosis system without an evaporation pond are more financially viable options for groundwater and landfill leachate treatment, respectively. These technologies offer lower costs per unit volume treated, and reduced operational expenses, making them more attractive for practical application.

2.3.3 Social Impacts.

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The social impacts of PFAS treatment technologies are still under-researched. All treatment technologies encompass both positive and negative social aspects. The primary social benefit is public health improvement due to PFAS removal from water sources. IX treatment effectively removes PFAS from groundwater, reducing exposure and lowering the risk of cancer and endocrine disruption. Similarly, GAC treatment significantly reduces PFAS levels in drinking water, decreasing the risk of cancer, liver damage, and thyroid disease. However, IX resin production and disposal pose social risks due to high ozone depletion potential, leading to respiratory issues, and ecological damage. Conversely, GAC emits higher levels of carcinogens and non-carcinogens compared to IX, negatively impacting air quality and human health. 53,58,59,62 Previous research highlights important distinctions between different PFAS treatment technologies, particularly in terms of health and social impacts. For example, IX has generally been found to result in higher life cycle human health impacts compared to GAC.⁶² These impacts are part of the treatment process itself and do not reflect the health benefits gained from the reduction of contaminants in the treated water. This suggests that GAC may be a more favorable option in contexts where minimizing health risks is a priority. Using the social sustainability evaluation matrix, studies have demonstrated that GAC is more socially sustainable than IX.53 The social sustainability evaluation matrix, an Excel-based tool, enables users to specify, and quantify the social aspects of a project by identifying key indicators for measurement. Its scoring system categorizes impacts into five sections: no impacts, positive, ideal, negative, and unacceptable. This comprehensive assessment framework helps highlight the social advantages of GAC over IX, reinforcing its potential benefits beyond just environmental considerations. In the context of landfill leachate treatment, the choice between onsite and offsite technologies has significant implications for human health. Research suggests that onsite multistage membrane technology may provide greater health benefits compared to offsite scenarios; however, further studies are needed to confirm these findings.⁵⁶ One of the main reasons could be the elimination of transportation risks and the ability to directly manage and optimize the treatment process. This leads to lower human health risks associated with

PFAS exposure. This finding underscores the importance of location and technology integration in optimizing the health outcomes of treatment processes. These insights collectively emphasize the need for a holistic evaluation of PFAS treatment technologies, considering health, social, and logistical factors. By understanding these nuances, decision-makers can better select and implement technologies that align with broader sustainability goals.

Table 1. Input variables (decision variables, technological parameters, and contextual parameters) for sustainable design of PFAS treatment technologies (GAC and IX). The input variables are categorized into three main sustainability indicators: environmental sustainability (blue), social impacts (purple), and financial viability (green). Note that these variables and parameters could be derived where these indicators are impacted by all; however, this work focuses on ones that will have a major impact.

Input	Treatment Technologies			Environmental Sustainability	
Variable	GAC		IX		
Decision Variables	Single Use GAC ⁵⁹	Sorbent Type ⁵⁷	Regenerable IX Resin ^{58, 59, 62}	Single Use IX Resin ⁵⁸⁻⁶⁰	Social Impacts Financial Viability
	Reactivation of GAC ^{59, 62}				Financial Viability
Technological Parameters	Media Production ⁵⁹	Incineration ^{59, 61}	Media Production ⁵⁹	Incineration ⁵⁹	
	Empty Bed Contact Time ^{53, 59}	Management of spent media ⁵⁹	Regeneration Options ^{58, 59}	Empty Bed Contact Time ⁵³	
	Bed Volume ⁵⁹	Media Usage Rate ⁵⁹	Management of spent media ⁵⁹	Media Usage Rate ⁵⁹	
	Activating Agent ⁵⁷		Bed Volume ⁵⁹		
Contextual Parameters	PFAS Length ^{57, 59}	Infrastructure ^{59, 62}	PFAS Length ⁵⁹	Infrastructure ^{59, 62}	
	Disposal option ^{59, 61}	Maintenance ⁵³	Disposal option ⁵⁹	Energy ⁵⁸	
	Energy ^{59, 61, 62}	PFAS Concentration ⁶²	PFAS Concentration ⁶²	Maintenance ⁵³	

3.0 Challenges to Sustainability.

Sustainability analysis of any treatment technologies evaluates their environmental, social, and financial impacts. Quantitative Sustainable Design (QSD) is a structured methodology used for accelerating and supporting the research, development, and deployment of technologies. Pr.98 QSD integrates concepts from sustainability science and engineering to facilitate the creation and implementation of sustainable solutions. A critical step in QSD is defining the problem space, which includes the selection of variables or parameters that influence sustainability, i.e., decision variables, technological parameters, and contextual parameters. Previous studies on the sustainability of PFAS treatment technologies primarily focus on GAC and IX. Table 1 highlights the different inputs for GAC and IX. It focuses on their effects on environmental sustainability, social impacts, and financial viability. This table will help future researchers to understand how decision variables, technological parameters, and contextual parameters affect sustainability. It will also guide the design of more sustainable PFAS treatment technologies for water sources.

3.1 Decision Variables.

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Decision variables are independent inputs that can be controlled by the designer or operator.98 For both GAC and IX technologies, an initial decision is whether to use single-use or reactivated/regenerable options. Single-use GAC has significant environmental impacts due to its high media usage rate and the need for frequent replacement and disposal. This frequent replacement can lead to higher emissions of carcinogens and non-carcinogens, negatively affecting air quality and human health in surrounding communities. ⁵⁹ In contrast, reactivating GAC media reduces these impacts by decreasing the demand for new media production; thus lowering resource consumption and waste generation. Reactivation processes also mitigate negative health impacts by reducing the frequency of GAC media replacement; thereby lowering emissions. Although the initial costs for reactivation infrastructure can be high, reactivated GAC is more cost-effective and environmentally sustainable than single-use GAC. This is particularly true at higher contaminant concentrations because reactivated GAC has lower operational costs for media replacement and disposal. 59,62 Similarly, the choice between single-use and regenerable IX also significantly affects sustainability. Both single use and regenerable IX have lower environmental impacts compared to single-use GAC, primarily due to their extended operational life and reduced media usage rate. However, the production phase of regenerable IX presents social risks, particularly high carcinogenic emissions, fossil fuel depletion, and global warming potential. These can lead to long-term health and environmental consequences. 53,58,59,62 The type of sorbent used in GAC primarily affects environmental sustainability and financial viability. Media from different carbon sources have varying adsorption capacities and lifespans, which impact the frequency of replacement, resource consumption. and waste generation. Bituminous GAC is widely recognized in research as one of the most effective media for removing PFAS. However, there is still a lack of studies addressing its environmental sustainability and financial viability. 99-102 Each decision variable can involve tradeoffs across overall sustainability of the PFAS treatment technologies. Therefore, a comprehensive evaluation is necessary to optimize treatment outcomes.

3.2 Technological Parameters.

Technological parameters, inherent to a technology's design and operations, significantly impact the sustainability of PFAS treatment systems, and are determined by the technology itself, not the designer or operator. ⁹⁸ Effective PFAS treatment technologies require careful consideration of several key technological parameters, each of which impacts environmental sustainability, social impacts, and financial viability. For both IX and GAC, media

production and incineration of spent media are the major sources of environmental impact. Media production for GAC requires activation processes and may involve mining (e.g., coal), which consumes high energy and release greenhouse gases, making this phase a major contributor to the environmental impact of GAC treatment systems. Similarly, the production of IX resins, particularly single-use resins, involves the synthesis of polymers. This process causes high environmental impacts, including ozone-depleting emissions.⁵⁹ Incineration of spent media from both GAC and IX, especially when used as single-use, is energy-intensive (above 1100°C for GAC) and results in emissions that contribute to global warming. Single-use IX systems have higher impacts from incineration compared to GAC. This is particularly applicable for human toxicity, cancer, eutrophication, and ecotoxicity due to the energy required for resin production. Incineration also involves high operational costs, increasing the financial burden of both GAC and IX treatment technologies. 59,61 Effective management of spent media can mitigate these impacts. For instance, off-site thermal reactivation of GAC media reduces the need for new media production and minimizes waste, positively impacting environmental sustainability.⁵⁹ However, the transportation and reactivation processes still contribute to environmental and social impacts. In IX systems, regenerating spent media involves handling and disposing of brine and cosolvents containing PFAS, which can cause significant environmental impacts and social risks due to the release of hazardous substances during incineration. 58,59

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Empty bed contact time and bed volume are directly proportional, as empty bed contact time is calculated by dividing the bed volume by the flow rate. While both are critical in PFAS treatment system design, their impacts on sustainability and costs are intertwined. For GAC systems, which typically require longer empty bed contact time (around 10 minutes) compared to IX systems (2-3 minutes), this means larger bed volumes, resulting in higher energy use, greater material needs, and increased costs. 53,59 In contrast, shorter empty bed contact time allow for smaller bed volumes in IX systems reduces energy and space requirements, making them more efficient environmentally and economically. 53,59 Another critical parameter affecting the sustainability of PFAS treatment technologies is media usage rate. A higher media usage rate results in frequent media replacements, increased production, and greater disposal needs, which escalate the environmental footprint and raise operational costs. High media usage rates in GAC systems lead to frequent replacements. This increases media production and disposal needs, escalating both environmental impacts and operational costs. Meanwhile, lower media usage rates in IX systems extend media life. This reduces replacement frequency, minimizes environmental impacts and lowers costs.⁵⁹ Optimizing media usage rate can balance operational efficiency and sustainability, reducing both environmental and financial impacts. The choice of activating agent in GAC significantly affects both environmental sustainability and financial viability. For example, optimizing sludge-based GAC adsorbents by reducing the activating agent (ZnCl₂) impregnation ratio can substantially lower environmental impacts and decrease production costs.⁵⁷ By understanding and optimizing these technological parameters, stakeholders can better balance the environmental sustainability, social impacts, and financial viability of PFAS treatment technologies. This comprehensive approach is essential for designing more sustainable solutions for PFAS removal from water sources.

3.3 Contextual Parameters.

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Contextual parameters represent non-technological values that influence the sustainability of technologies, particularly during the deployment stage. 98 These parameters are designed to capture the specific circumstances in which the technology will be implemented. The length of PFAS chains significantly impacts the environmental sustainability, social impacts, and financial viability for both GAC and IX systems. IX resins were identified as a cost-effective adsorbent for removing both long and short-chain PFAS compared to GAC adsorbents.³² Previous research indicates that IX systems have a higher adsorption capacity than GAC and are more effective at removing long-chain PFAS, like PFOA and PFOS, compared to short-chain PFAS.¹ Studies show that GAC also adsorbs long-chain PFAS better than short-chain PFAS.¹⁰³ This indicates that both IX resins and GAC adsorbents exhibit higher adsorption capacity for long-chain PFAS compared to short-chain PFAS. In another study, sludge-based GAC exhibited the lowest removal rate (35.3–57.8%) for short-chain PFAS compounds, likely due to their high solubility and low hydrophobicity. 104,105 Increasing the empty bed contact time can enhance the efficiency of GAC in removing short-chain PFAS, offering protection against the rapid breakthrough of short-chain PFAS. 32,106 GAC and IX treatments require a longer empty bed contact time for short-chain PFAS compared to long-chain PFAS.³² Both long-chain and shortchain PFAS pose significant social impacts, primarily through health risks. Compared to longchain PFAS, treating short-chain PFAS requires a higher media usage rate, which increases environmental impacts. This includes higher values for carcinogenic and non-carcinogenic effects, as well as global warming potential, indicating greater health damage compared to longchain PFAS. Consequently, the treatment of short-chain PFAS may lead to higher overall environmental, social, and financial damage.

PFAS concentration is another important parameter that effects the sustainability. Both groundwater and bottled water were examined under a range of PFOA and PFOS concentrations covering three orders of magnitude (0.7, 7.0, and 70 μ g/L) using both GAC

adsorbents and IX resins. Higher PFAS concentrations significantly impact environmental sustainability and social health issues. As PFAS levels increase, greenhouse gas emissions from GAC-based treatment rise, reaching 0.54 and 2.7 kg CO_2eq/m^3 H_2O at 7.0 and 70 $\mu g/L$ PFAS, respectively. GAC media production and reactivation processes result in greater environmental impacts compared to IX-based treatments at higher contaminant levels. Energy consumption for GAC increases substantially, contributing to impacts such as fossil fuel depletion, respiratory effects, and smog formation. At high PFAS concentrations, electricity consumption contributes significantly to climate change impacts: 15% for GAC and 63% for IX treatment. Human health impacts are also heightened, with electricity production (mainly coal-fired) responsible for over 75-90% of health and ecotoxicity impacts from both GAC and IX systems. As PFAS concentrations rise from 0.7 to 70 μ g/L, life cycle electricity consumption increases from 1.5 to 1.9 MJ/m³ for GAC and from 1.5 to 1.6 MJ/ m³ for IX, amplifying both health and environmental consequences.

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Disposal options for spent media primarily impact environmental sustainability and financial viability. For GAC systems, the disposal of activated carbon significantly impacts acidification and ozone depletion. Additionally, incineration of spent media at high temperatures (above 1100°C) results in substantial greenhouse gas emissions and other pollutants, contributing to global warming and degradation of air quality^{59,61} Similarly, for IX systems, disposal of brine and cosolvents containing PFAS can lead to hazardous emissions during incineration, impacting both environmental and social sustainability due to potential health risks. 59,60 Financially, incineration and other disposal methods incur high operational costs, increasing the overall treatment cost. Maintenance significantly impacts the financial viability of both GAC and IX systems. GAC systems face higher operational costs due to frequent media replacements, despite having lower annual operating and maintenance costs compared to IX systems.⁵³ IX systems, while having higher initial capital costs, benefit from lower media usage rates and longer operational life, reducing the need for frequent replacements. While the regeneration process adds to IX operational costs, these systems are more cost-effective over time due to reduced maintenance needs. Effective maintenance strategies are essential to optimize the financial viability of both GAC and IX treatment technologies.

Infrastructure requirements significantly affect financial viability and environmental sustainability. Treatment systems need contactors, pipes, fittings, and corrosion-resistant coatings. The infrastructure for resin treatment and regeneration is assumed to be twice as complex as that for GAC due to its added components.⁶² Despite its reusability, regenerable IX

is the second most impactful system after single-use GAC because of the additional infrastructure and chemical-intensive regeneration processes.⁵⁹ Although capital impacts are generally minimal, they are significant for single-use IX due to its low media usage rates. For reactivated GAC, the absence of high-temperature incineration shifts more impact to infrastructure requirements for regeneration. Energy consumption significantly affects the financial viability and environmental sustainability of PFAS treatment technologies. High energy use increases production costs, making technologies less efficient. GAC adsorbents production and reactivation consume substantial energy, leading to higher life cycle impacts like fossil fuel depletion and respiratory effects.⁶² Thermal reactivation of GAC adsorbents, at 815°C, is less energy-intensive than incineration at 1200°C, making it more cost-effective. ^{59,61} Regenerable IX systems also increase energy demand due to the salts used in regeneration, especially those derived from chemicals(e.g., NH₄Cl, K₂CO₃).⁵⁸ Both GAC and IX systems are sensitive to local electricity grid variations, affecting costs and environmental impacts. For example, electricity prices vary from 5.07 cents per kilowatt-hour (kWh) in Oklahoma to 35.86 cents/kWh in Hawaii, and GHG emissions from electricity range widely. 107 Switching to renewable energy sources, like wind or solar, can significantly reduce human health costs and global warming potential, making renewable energy a key target for cost and impact reduction.

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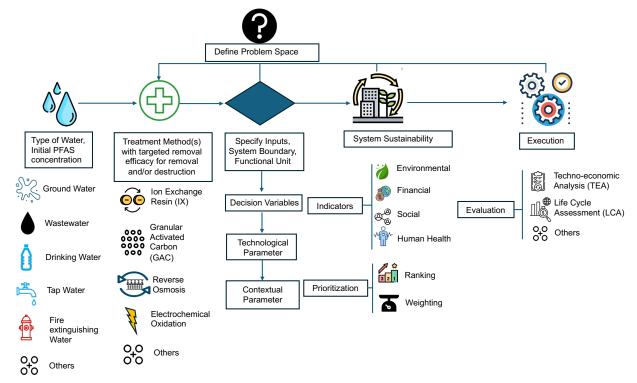


Figure 2. Developed framework for understanding the sustainable PFAS treatment in water. This framework outlines the sustainable treatment of PFAS from water, encompassing steps such as defining the problem space, selecting water types, and treatment methods, characterizing system sustainability

with various indicators (e.g., environmental, financial, social, and human health), prioritizing these indicators via ranking, weighting, and executing the treatment impacts using evaluation methods like LCA, TEA, or LCCA.

4. Charting Pathways for Sustainable Treatment of PFAS.

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The new US EPA regulation requires public water systems to complete initial monitoring of six PFAS, including PFOA, PFOS, PFHxS, perfluorononanoic acid (PFNA), hexafluoropropylene oxide dimer acid (HFPO-DA), and mixtures containing at least two or more of PFHxS, PFNA, HFPO-DA, and perfluorobutane sulfonic acid (PFBS) by 2027. 108 If the concentration of these PFAS exceeds the maximum contaminant levels (MCL), solutions to reduce their concentrations must be implemented by 2029. The EPA estimates that the new regulation will reduce PFAS exposure for about 100 million Americans, leading to fewer cases of cancer, liver disease, and birth complications. This will result in annual health benefits of approximately \$1.5 billion, covering reduced medical costs and lost income. 109 The actual benefits are likely higher, considering unquantified health effects like developmental and cardiovascular issues. While, according to AWWA, the national cost for treating long-chain PFAS (PFOA, PFOS, PFHxS, PFHpA, and PFNA) at a 4 ppt level is approximately \$60 billion in the United States. This underscores the need for sustainable and cost-effective PFAS treatment technologies that effectively remove these contaminants from water. 110 As current situation demands sustainable treatment technologies to eliminate persistent PFAS from water sources. comprehensive sustainability studies are required. These studies must encompass existing technologies and explore new solutions under various scenarios, including different water types, indicators of sustainability, prioritization and weighting of the indicators. To navigate the extensive opportunity space for sustainable PFAS treatment, it is mandatory to integrate sustainability analyses throughout the research, development, and deployment phases of technologies. Our review of existing literature on PFAS water treatment technologies has led to the development of a proposed framework for understanding sustainable PFAS treatment (Figure 2). The initial step in the sustainable treatment of PFAS involves identifying the type of water requiring treatment, e.g., groundwater, wastewater, drinking water, tap water, or fire extinguishing water. Each water type presents unique challenges and necessitates different treatment approaches. Based on the water type, appropriate treatment methods, such as IX, GAC, RO, or other technologies, are selected. Next, inputs are specified, including decision variables (e.g., single-use or regenerable media), technological parameters (e.g., media production, bed volume), and contextual parameters (e.g., PFAS length, concentration). Sustainability is assessed across three primary indicators: environmental sustainability, financial

viability, and social impacts. Tools such as TEA, LCA, and others are employed to evaluate these indicators. Models like MIVES integrate results from environmental, financial, and social assessments.⁵³ The prioritization of indicators through ranking and weighting guides the final sustainability assessment. For example, the MIVES model was used to integrate results from environmental, financial, and social assessments for GAC and IX under different weighting scenarios. The scenarios included equal weighting of all indicators (33% each), higher weighting on social impacts (60%), and higher weighting on financial viability (50%). The results suggested that IX treatment is generally more sustainable than GAC treatment.⁵³

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By following this framework (Figure 2), we can identify the most sustainable treatment technology for specific scenarios. If the results do not meet environmental guidelines or financial constraints, we can revisit any point in the proposed framework and choose alternative options. This framework is iterative, allowing for adjustments in treatment methods and re-prioritization of indicators. This iterative approach ensures the selection of sustainable and effective PFAS treatment method for particular water and contaminant types. Each treatment technology involves trade-offs. This framework supports the sustainable treatment of PFAS by guiding the evaluation of both established and emerging technologies. For established technologies like GAC and IX, historical data and proven methods are used to specify inputs, assess sustainability, and optimize execution. For emerging technologies, it accommodates flexible parameters, experimental data, and iterative testing to refine and validate new treatment methods. This approach ensures balanced consideration of environmental, social, and financial impacts for all PFAS treatment technologies. The US EPA's three R's (Research, Restrict, and Remediate) are key directives in tackling PFAS, the "forever chemicals." Our study finds that research on sustainability of PFAS treatment technologies is still in its developmental stages and requires further investigation across various scenarios. Alternative technologies, including reverse osmosis and nanotechnology for PFAS removal, as well as destruction technologies like advanced oxidation processes, electrochemical oxidation, and plasma, require thorough sustainability assessments to ensure their effectiveness and feasibility for comprehensive PFAS treatment. Additionally, existing sustainability studies on treatments like GAC and IX are limited, as they rarely examine all three sustainability parameters: environmental sustainability, social impacts, and financial viability. This review underscores the uncertainties in sustainability indicators and the trade-offs within different treatment systems. Overall, this study highlights the potential to reveal the sustainability implications of PFAS removal technologies in specific contexts. This enables stakeholders to make informed decisions about the multidimensional

648	effects of these technologies, ultimately contributing to society's transition towards greater
649	sustainability.
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652	University.

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