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Source, Distribution, and Removal of PFAS in Stormwater: Challenges & Opportunity

Author
Banerjee, Swapnil

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Source, Distribution, and Removal of PFAS in Stormwater: Challenges & Opportunity

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by

Swapnil Banerjee

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ABSTRACT OF THE THESIS

Source, Distribution, and Removal of PFAS in Stormwater: Challenges & Opportunity

by

Swapnil Banerjee

Master of Science in Civil Engineering

University of California, Los Angeles, 2023

Professor Sanjay K. Mohanty, Chair

PFAS site remediation studies often overlook stormwater as a potential source of PFAS contamination. This review article looks at the transport and distribution of these anthropogenic chemicals in stormwater, to identify the species and extent of distribution of these PFAS molecules, and then brings a comparison of different adsorbents (Biochar, Granulated Activated Carbon, Ion-Exchange Resins) in stormwater biofilters as a remediation technique. We found biochar and GAC to have an extremely variable adsorption capacity (over 3 orders of magnitude) and IX Resins to have the highest PFAS adsorption. These were attributed to several factors including soil chemistry, PFAS species present, and weather conditions. However, from a cost-benefit analysis perspective IX Resins were found to be almost 9x more expensive than traditional adsorbents. Hence, either a particular type of adsorbent or a combination of sorts has been suggested, and different factors that affect the adsorption of each of these amendments has been discussed to make an informed decision.

The thesis of Swapnil Banerjee is approved.

Jennifer A. Jay

Shaily Mahendra

Sanjay K. Mohanty, Committee Chair

University of California, Los Angeles

2023

Table of Contents

List of Figures

Figure No. Page

List of Tables

Table No. Page

List of Acronyms

PFAS: Poly- and perfluoroalkyl Substances

POP: Persistent Organic Pollutant

GAC: Granular Activated Carbon

PFAA: Perfluoroalkyl Acids

PFOA: Perfluoroalkyl Octanoic Acid

PFOS: Perfluorooctane Sulfonate

PFBS: Perfluorobutane Sulfonate

PFBA: Perfluorobutanoic Acid

PFHxA: Perfluorohexanoic Acid

STS: Stormwater Treatment System

PFCA: Perfluorocarboxylate Acid

PFSA: Perfluorosulfonic Acid

EPA: Environmental Protection Agency

IX: Ion-Exchange

BMP: Best Management Practice

Acknowledgements

I want to express my sincere gratitude, starting with my committee chair, Dr. Sanjay Mohanty, whose unwavering guidance has been indispensable throughout the thesis writing journey. I'd also like to extend my heartfelt thanks to my parents and my sister, without whose unwavering support this task might have been insurmountable. Finally, I'm deeply appreciative of my friends, who believed in me and provided steadfast support through the ups and downs.

1. Introduction

Per- and polyfluoroalkyl substances (PFAS) are a class of fluorinated aliphatic compounds that has been classified as a Persistent Organic Pollutant (POP) [1] because of its adverse health effects causing liver and autoimmune conditions, enhanced risk of kidney or testicular cancer, and decrease in infant birth weights [2]. Subsurface soil and stormwater treatment systems play a crucial role in filtering surface runoff pollutants and safeguarding groundwater [3]**.** Nonetheless, there is a growing concern that emerging contaminants, such as per- and polyfluoroalkyl substances (PFAS), may not be effectively removed by these traditional systems [4] and could potentially contaminate groundwater [5]. This concern is valid as recent studies have revealed that up to 72% of drinking water treatment facilities using groundwater have been found to contain PFAS [6]**.** The concern that stormwater is a significant pathway for PFAS to enter the environment, particularly surface and groundwater, which can be used as a source of drinking water makes it critical to understand the source, distribution, and removal technology for pfas in stormwater.

PFAS regulation in stormwater can be heavily influenced by source, release, and climate. Studies have shown that stormwater, originating from contaminated sites like airports, landfills, and industries, can transport PFAS through subsurface infiltration to surface water and groundwater, further exacerbating the problem [7–9]**.** Landfill leaching and discharges have been found to be another source of PFAS release in stormwater [10,11]**.** Finally, climate can also play a role in the behavior of PFAS in stormwater. Heavy rainfall or flooding can cause PFAS to be mobilized and transported from contaminated areas, resulting in higher concentrations in stormwater runoff [12]**.** However, limited studies have examined the distribution of PFAS in stormwater and the factors that affect it.

Active treatment involves the use of a specific treatment technology that actively removes or destroys PFAS from stormwater. Some common active treatment technologies for PFAS removal include granular activated carbon (GAC), ion exchange, and membrane filtration. GAC and ion exchange are effective at removing PFAS from water by adsorbing or exchanging the contaminants onto the treatment media. Membrane filtration technologies use ultrafiltration, nanofiltration, or reverse osmosis to physically remove PFAS from the water. Active treatment methods can be highly effective at removing PFAS, but they can be costly and require ongoing maintenance [13].

Passive treatment, on the other hand, involves the use of natural or engineered systems that rely on physical, chemical, or biological processes to remove contaminants from stormwater over time. Passive treatment systems can be highly effective at removing PFAS from stormwater and can be more cost-effective than active treatment methods. Examples of passive treatment systems for PFAS removal include constructed wetlands, stormwater biofilters, and natural sorbents [14]**.**

This study focuses on understanding the fate and transport of PFAS in stormwater and identify the major PFAS species contaminating these waters. Our second goal is to compare passive treatment techniques to remediate these dominant PFAS species because of their low cost and easy availability, where we compare various adsorbents to understand what factors affect their adsorption capacity and what are some basic factors to consider making some informed decisions. Finally, a cost-benefit analysis for choosing the appropriate amendment has been provided to understand which adsorbents would be the most sustainable and economical. Previous reviews have compared adsorption capacity of different media [13–18], source of PFAS in surface water [9,19–24] or groundwater [25–28], and PFAS removal processes in subsurface [29–33]**.** However, no reviews to date have analyzed PFAS source, removal, and transport processes in stormwater system. Stormwater systems are unique because unlike wastewater treatment plant, they are subjected to transient weather conditions with intermittent flow of stormwater. These processes are known to affect the PFAS transport [34]. The current work aims to address the gap between adsorbent based remediation techniques for PFAS in stormwater systems.

2. Data collection and analysis

We examined the Web of Science database (https://www.webofknowledge.com/) in January 2023, to collect research articles using keyword combinations: “PFAS” with “adsorption”, and “amendment name”. Our goal was to quantify the distribution of PFAS in surface water and rainwater, considering the source in these waters to be surface runoff. We found 7 articles reporting PFAS concentrations in surface water over 23 sites and 8 studies reporting rainwater PFAS concentrations over 29 sites. Median concentrations were reported and found that short-chain anionic PFAS ($C \geq 8$) dominated the distribution, which allowed us to do the next part of the survey, that was to find the most effective adsorbents for removing these dominant PFAS species. Adsorbents were only considered that would not clog stormwater filter systems, so the ones considered were searched with keywords: “GAC”, “Biochar”, and “Ion Exchange Resin”. To examine adsorbent efficiency in PFAS removal, only studies reporting pseudo-second order kinetics were able to compare the true adsorption capacity of the listed amendments, and mainly focused on removal capacities of PFBA, PFBS, PFOA, and PFOS as it corresponds to the highest occurrence in surface and rainwater from our analysis. 19 articles were selected to examine adsorption data, and then compared. It must be noted that the variability caused in the analysis of adsorption data can be associated with different processing conditions and modification in certain adsorbents, for which no classification has been included in this study. RStudio (Version 2022.12.0+353) has been used for all data analysis and plotting of graphs, and all statistical analysis has been carried out using Tukey Test. The list of studies in the analysis has been provided in the supplementary material.

3. Source and occurrence of PFAS in stormwater and rainwater

It is challenging to attribute the source of PFAS contamination for remediation action, as both rainwater and runoff may contain PFAS. In the absence of rainwater monitoring, land use is often assumed to be the source of PFAS contamination, rendering any land-based treatment ineffective. However, if rainwater is clean, the stormwater can pick up PFAS from the land surface, which can be mitigated by changing land use. Regardless of the source attribution, polluted stormwater must be treated using Best Management Practices (BMP). The objective of this section is twofold: firstly, to compare whether rainwater may contain higher concentrations of PFAS than typically found in surface or stormwater, and if so, to identify those PFAS. Secondly, to evaluate the dominant PFAS found in surface water or stormwater and highlight the sources that could have contributed to that pollution.

3.1. Source

Source of PFAS can originate from sites including fluorochemical industrial sites [35,36]**,** firefighting training areas [8,37]**,** airports [38,39], accidental spill sites [40,41] and poorly managed landfills [5,42,43]**.** Nontraditional sources of PFAS in stormwater can also be attributed to biosolid application for land-use that transports PFAS during runoff [9,44], which are often derived from sewage sludge discharge of wastewater treatment plants [45].

PFAS in stormwater can be introduced through secondary sources, such as contaminated soil, where the release of colloids contributes to a high concentration of PFAS in surface water [46]. During a storm event, all these sources contribute to the presence of PFAS in runoff, which ultimately ends up in stormwater. Rainfall is another source that not only introduces PFAS into stormwater, but also amplifies its presence in the environment [47].

Regardless of the source, PFAS concentrations can vary based on multiple factors. Several studies [48,49] have demonstrated that proximity to a PFAS source could be an indicator of elevated concentration in neighboring areas. Climatic and temporal factors also affect PFAS concentrations, which can reintroduce PFAS into stormwater. The natural dry-wet and freeze-thaw cycle in subsurface soil can alter the leaching behavior of previously adsorbed pollutants, resulting in PFAS being released at a slow rate from PFAS-impacted subsurface soil. This diffused source of contamination persists over time [50].

3.2. Rainwater

Despite limited available data, there is an increasing body of evidence indicating the presence of PFAS in rainwater [9,12,51]. Previously, it was widely believed that Per- and Polyfluoroalkyl Substances (PFAAs) would eventually be washed off into the oceans, where they would undergo dilution over an extended period. However, it is now suggested that certain types of PFAS can become highly concentrated and transported through wet atmospheric deposition [52]. Once these PFAS reach the land, they can settle and contaminate freshwater bodies, soil, and even drinking water sources.

Although comprehensive data on PFAS concentrations in rainwater is limited, our analysis of four studies presented in Fig. 1.A. reveals a wide range of global PFAS concentrations spanning over five orders of magnitude. Interestingly, in rainwater, there is a higher percentage of long-chain PFAS (64%) compared to short-chain PFAS (36%) (Fig. 1.B.). This observation aligns with previous findings suggesting that long-chain PFAS exhibit greater hydrophobicity and lower solubility in water compared to short-chain PFAS [53]. The hydrophobic nature of long-chain PFAS can result in their adsorption onto atmospheric particles, such as dust or soot, which can then be transported by wind [54]. Consequently, these particles may remain suspended in the air and subsequently deposit onto various surfaces during rainfall events. Interestingly, the studies we referred to report extremely low levels of PFSA in rainwater. From this, we can hypothesize that PFSA are not typically present in rainwater at high concentrations. This can be attributed to their low volatility, which makes them less prone to long-distance atmospheric transport. Instead, PFSA are more likely to be deposited onto surfaces, such as soil or water, near their emission sources [55]. However, volatile precursors have been studied to undergo atmospheric oxidation and contribute to the formation of long-chain PFCA [56]. In the context of stormwater, a few studies have reported PFAS concentrations. For instance, [57] highlights the importance of utilizing proper instrumentation, such as high-resolution mass spectrometry, for routine testing. This is necessary to detect highly branched isomeric PFAS that may otherwise go undetected. [58] draws an interesting analogy to emphasize the ubiquity of PFAS and how these chemicals have surpassed our planetary boundary. These findings shed light on the complexities associated with PFAS presence in rainwater and stormwater, highlighting the need for advanced analytical techniques and the pervasive nature of PFAS contamination. [59] provides valuable insights into the contamination of PFAS in the Maltese Islands, which are geographically isolated and not connected by any borders. Despite their pristine nature, the islands exhibit high levels of PFAS contamination in surface and rainwater. This indicates that ocean currents and rainfall can serve as pathways for the introduction of PFAS into even seemingly unaffected environments. On the other hand, [52] contributes a significant dataset of PFAS concentrations in wet deposition specifically in the Midwest region. This dataset aids in assessing PFAS fluxes to rural landscapes and provides valuable information to improve models of environmental cycling of these chemicals. By better understanding the behavior and distribution of PFAS in wet deposition, we can enhance our understanding of the environmental fate and transport of these contaminants.

The origin and path of clouds are significant factors to consider when tracking the source of contaminants in precipitation [60]. This is because clouds can transport these PFAS over great distances as they move through the atmosphere. On a regional level, [61] suggests that jet streams and cloud movements play a critical role in transporting PFAS. The major mechanisms by which PFAS is transported by wet deposition are in-cloud scavenging and below cloud scavenging [59].

The concentration of PFAS in rainwater is primarily influenced by atmospheric conditions, including wind direction and speed, temperature, and humidity [61,62]**.** Furthermore, [63] suggest that the dry-wet cycle can contribute to the release of PFAS, which may subsequently become part of the cycling process and increase PFAS concentration in rainwater. However, several important questions remain unanswered when studying the transport of PFAS in rainwater. For instance, it is crucial to understand the relationship between the proximity and intensity of PFAS sources, such as industrial facilities or landfills, and the concentration of PFAS in rainwater. Additionally, further research is needed to elucidate the transport mechanisms of PFAS in rainwater based on their chain length. It is known that chain length plays a critical role in wind transport [49].

The cycling process of PFAS presents a significant challenge in our efforts to reduce global PFAS concentrations. This is evidenced by the widespread distribution of PFAAs in remote regions such as Antarctica and the Arctic, [64,65]. PFAS concentrations have been detected in ice cores and surface water in these regions, with sea-spray aerosol transport hypothesized as a potential contributing factor. [66] suggests that the atmospheric transport of volatile precursors and their eventual degradation into PFCA, especially for shorter chain C_2-C_4 plays a major role in the global distribution.

3.3. Stormwater or surface water

The concentration of PFAS in stormwater could become diluted when mixed with surface water. Thus, the concentration in surface water could be used as an indicator to examine the link between surface water and stormwater for PFAS pollution. Analyzing PFAS concentration in surface water, rainwater, and stormwater in Fig. 1.A., we observed that the first 5 PFAS species accounted for over 50% of total PFAS present, additionally 39 species of PFAS were detected. This implies that only a select few species (PFOA, PFBA, PFOS, PFBS, and PFHxA) contribute significantly to the overall PFAS concentration, and remediation strategies should be directed more towards these dominant species. Median concentration of all reported PFAS in surface water was observed to be higher than in rainwater. Street dust and vehicle traffic are observed to be also probable sources of PFAS in surface waters [59,67]. PFOS concentration in rain and surface water was higher by a magnitude of 2 and 3 respectively, which indicates that these runoff contaminants can be a source of PFAS in our drinking water infrastructure, which would be above EPA's advisory limit if not treated. Fig. 1. (B) shows the PFAS classification as Perfluoroalkyl Carboxylate Acids (PFCA) and Perfluoroalkyl Sulfonic Acids (PFSA) in surface and rainwater respectively and their chain length distribution. Contrasting trends were observed, with 93% of PFAS being short chain ($C \leq 8$) and 7% being long chain ($C > 8$) in Fig. 1. (B), inferring that short chain PFAS are more widely detected in aquatic systems. The prevalence of short chain PFAS in surface water can be attributed to their high solubility and mobility in water. Short chain PFAS exhibit greater solubility, which allows them to dissolve readily in water and remain in the aqueous phase. Moreover, their poor capacity for natural adsorption further contributes to their higher concentrations in surface water [68]. This observed trend could also be in part, due to the recent increase in use of short chain PFAS. Where in response to the persistent and bio-accumulative nature of certain PFAS chemicals [69], the European Union has included C_8-C_{14} PFAS and their sodium and ammonium salts on its candidate list of regulatory substances. Furthermore, PFOS has been added to the Stockholm Convention's Persistent Organic Pollutants (POPs) list in 2009 [70]. To comply with these regulations, manufacturers have shifted towards using alternative chemicals such as short-chain and ultra-short chain PFAS, as well as perfluoro-poly ethers, for commercial production [71,72]. Whereas no significant difference was found in the distribution of PFCA and PFSA. In conclusion, it is observed for both surface water and rain water as a source of stormwater, PFCAs are the recurring species [73].

Our analysis reveals that stormwater is rarely measured for PFAS concentration compared to surface waters because it is expensive and not required. However, identifying these dominant species in stormwater helps us to plan remediation actions.

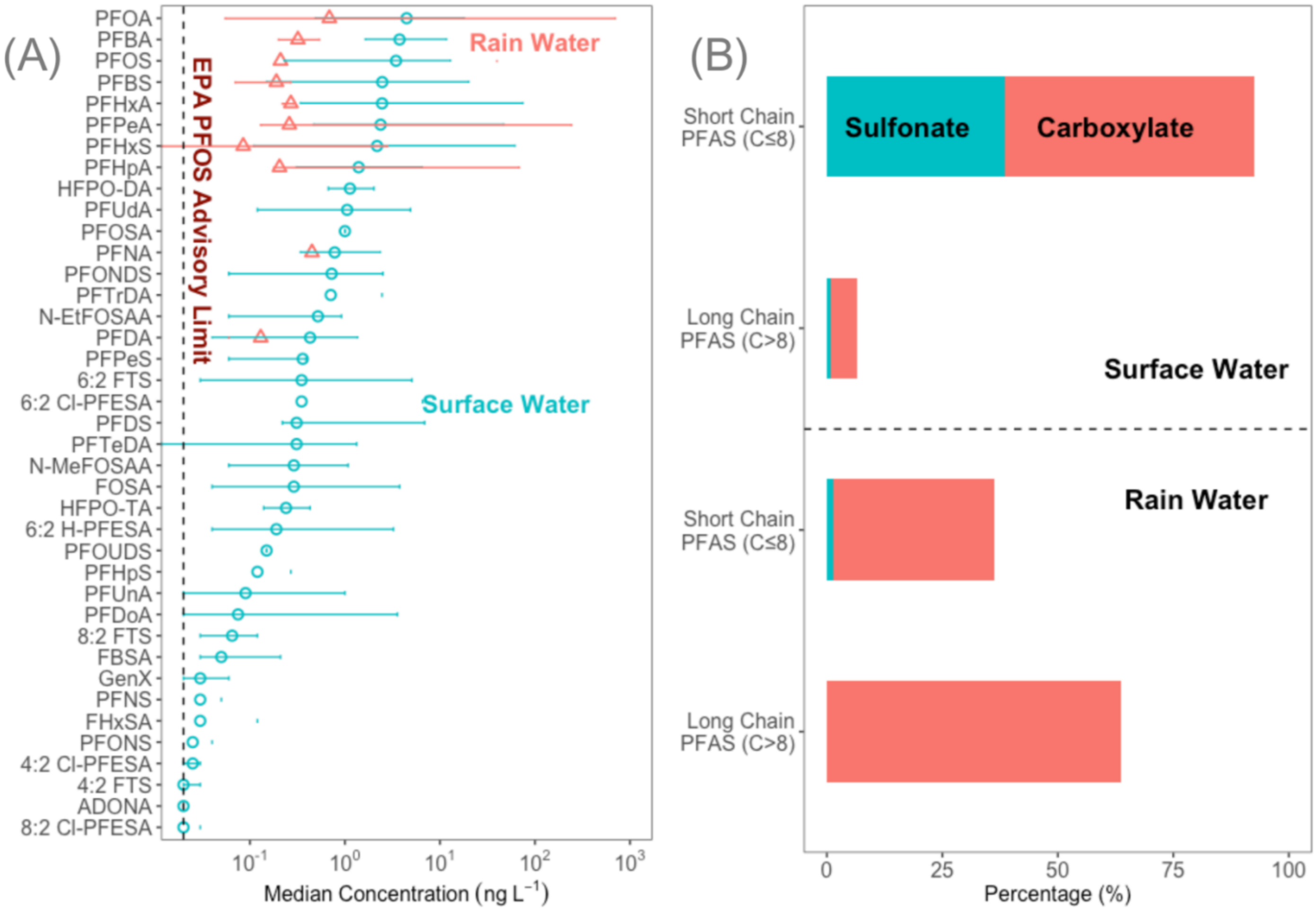

Figure 1 (A) PFAS concentration varying over 5 orders of magnitude. (B) PFAS distribution with functional group

4. PFAS transport and removal mechanism in infiltration-based stormwater treatment systems

Stormwater treatment systems (STS) are important in removing various pollutants, but they can also capture PFAS that are present in the runoff from sources like industrial sites, landfills, firefighting training areas, and agricultural areas. PFAS removal can be challenging in stormwater treatment systems. Recent studies have suggested that biofilters may effectively remove PFAS from stormwater [74–76]**,** but the effectiveness depends on the filter media used and the specific type of PFAS compound present.

PFAS transport in biofilters depends on media depth, flow rate, and composition [77]**.** Stormwater treatment systems transport PFAS via infiltration, advection, and dispersion [78]**.** Infiltration refers to the movement of water through soil and into aquifers, carrying PFAS with it. Advection carries PFAS through the treatment system and out through discharge outlets. Intermittent flow affects transport through dispersion, where PFAS mixes in soil due to physical, chemical, and biological processes, creating a more uniform concentration that affects transport rate. Hydraulic conductivity is an important parameter that is studied to understand the infiltration of stormwater in these treatment systems, and they depend on few factors as root system of vegetation and thickness of clogging layer [79], that enables us to quantitatively measure the efficacy of the STS.

Removal mechanism is primarily governed by volatilization, sorption, and degradation. Volatilization is assumed to occur at the interface of STS and the atmosphere at the surface level, through the exchange of PFAS mass. Whereas sorption and degradation is assumed to be in the submerged zone [78].

4.1. Environmental conditions affecting transport and removal

The transport and removal of PFAS in stormwater biofilters are not only affected by design conditions but also by environmental conditions. The sorption of organic pollutants is contingent upon the intrinsic properties of PFAS, specifically their hydrophobicity and polarity. Additionally, the sorption process is affected by various physicochemical characteristics of the soil, including pH, cation exchange capacity, ionic strength, surface area, soil organic matter content, and water temperature [80]. Climate is an essential factor that can influence the biofilters' performance. During heavy rainfall events, the flow rate and volume of stormwater runoff can increase significantly, which can reduce the contact time between the water and the filter media, consequently decreasing the PFAS removal efficiency **(Mathieu et al., 2022)**. Similarly, long dry periods reduces PFAS removal due to drying of biofilters [81,82]. Water quality parameters like pH, salt, and dissolved organic carbon (DOC) also play a significant role in the transport and removal of PFAS. For example, the pH can affect the charge of the PFAS molecules and the surface charge of the filter media, altering the adsorption capacity of the filter **(Julie et al., 2023)**. A high salt concentration in the stormwater runoff can affect the PFAS adsorption by the filter media **(Song-Thao et al., 2021)**. Similarly, the presence of DOC can compete with PFAS for adsorption sites on the filter media, reducing the removal efficiency of PFAS.

4.2. Design conditions affecting the transport and removal

To effectively remove PFAS in stormwater biofilters, it is essential to consider design conditions. One significant design factor is the amount of amendment added to the filter media, which can influence the filter's ability to adsorb PFAS. While higher amendment amounts can enhance PFAS removal efficiency, they can also increase costs. The depth of the biofilter media is another crucial factor that can impact PFAS transport and removal. While a deeper media can increase PFAS removal efficiency by extending contact time, it may also increase pressure drop and lead to clogging **(Delhoménie et al., 2003)**. The flow rate of stormwater through the biofilter is also a critical design condition affecting PFAS removal efficiency. A higher flow rate can reduce contact time and decrease PFAS removal, while a lower flow rate can lead to clogging [83]. Catchment area of the STS can influence the PFAS removal capacity, with a larger system relative to the catchment area, those are the systems receiving the smallest volume per m^2 having a higher K (Hydraulic Conductivity) value [84]. Therefore, optimizing these design conditions is essential to effectively removing PFAS from stormwater runoff in biofilters.

5. Amendments for stormwater treatment

5.1. Biochar

Biochar produced at high temperatures and with high surface area has been found to have a greater capacity for PFAS adsorption from stormwater run-off. It is a cheap, natural, and environmentally friendly amendment [85]. The adsorption capacity of biochar for PFAS depends on factors such as the type of biochar, the type of PFAS, and the solution chemistry.

Our analysis shows that biochar effectively removed PFAS from stormwater irrespective of PFAS types (Fig. 2) and the removal varied from 3.5 x 10-5 - 795 $\mu M.g^{-1}$. The data shows that there are higher adsorption (95%) for longer chain (PFOA, PFOS) than short chain (PFBA, PFBS) PFAS in their respective functional groups, which can be attributed to the decreasing polarity with chain length [86–88]. However, no statistical significance was found between PFCA and PFSA with Tukey test results (p-value 0.72). However, for shorter chains PFSA sorbed more strongly than their PFCA counterpart, because of higher hydrophobicity of PFSA [89,90]. [91] observed that biochar was more effective in adsorbing short chain PFAS compared to GAC, while the opposite trend was observed for long chain PFAS.

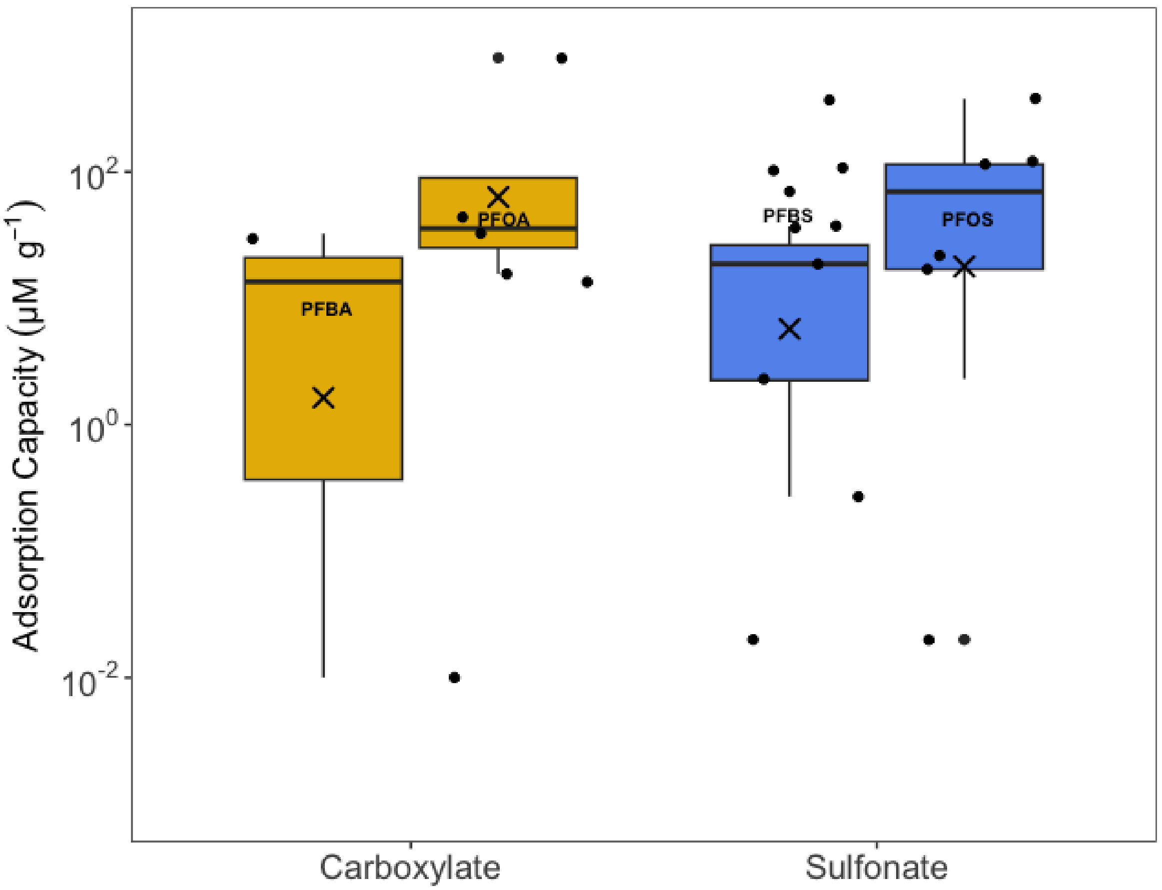

Figure 2 Adsorption capacity of Biochar based on short chain PFAS as PFBA (C4) and longer chain PFAS as PFOS(C8) and classified in Perfluorinated Carboxylic acids (PFCA) and Perfluorinated Alkyl Sulfonates (PFSA)

The adsorption of PFAS onto biochar is generally governed by both electrostatic and hydrophobic interactions [92,93]. The mechanism of PFAS adsorption onto biochar is thought to be due to the surface chemistry of biochar, which contains functional groups such as carboxylic acids and phenols that can interact with PFAS molecules [94,95]. The high surface area and porosity of biochar also provide a large surface area for adsorption [96]. Researchers have found that sorption of organic pollutants increases with an increase in the C-content, as number of strong sorption sites increases [97].

In practical applications, it has been observed that the adsorption capacities of biochar for PFAS exhibit a significant degree of variability, spanning over two orders of magnitude. Additionally, comparative studies [17,91] have indicated that biochar may exhibit relatively lower PFAS adsorption capabilities when compared to alternative adsorbents.

Based on our comprehensive literature survey, we have identified several key factors that contribute to the considerable variability observed in biochar's adsorption of PFAS. These factors include the type of feedstock material used, the pyrolysis temperature applied during biochar production, and the resultant surface area of the biochar. The observed variability in adsorption capacities spans a wide range, from 3.5 x 10^{-5} µmol/g to 795 µmol/g. Generally, biochar derived from feedstocks with higher lignin and cellulose content exhibits larger pores and a greater surface area, which, in turn, enhances its capability for adsorbing PFAS [97]. We were unable to locate any specific study that directly reported the impact of ash content on the adsorption of PFAS. However, it was observed that feedstocks with high organic matter content tended to yield biochar with relatively low adsorption capacity. This outcome can be attributed to the abundance of organic matter present, which has the potential to bind to the pores of the biochar and impede its adsorption capacity by causing pore clogging [97]**.** Raising the pyrolysis temperature can produce biochar with a greater capacity for PFAS adsorption by enhancing surface area and more micropores, which offer a larger surface area for adsorption. Further, the water and volatile organic components in the feedstock are also more completely removed at higher temperatures, which can increase the stability and carbonaceous content of the final biochar product [98] and enhance PFAS removal performance. However, extremely high pyrolysis temperatures (>900° C) could limit PFAS sorption by destroying the micropores porosity and eventually the surface area [99,100]. Moreover, it should be noted that extremely high pyrolysis temperatures can lead to the degradation of organic functional groups within the biochar. These functional groups play a significant role in contributing to the adsorption capacity of the biochar. Therefore, the degradation of these functional groups at excessively high temperatures can lead to a reduction in the biochar's overall adsorption capacity [95]. The pH level of the solution also plays a notable role in the sorption of PFAS onto biochar. It has been generally observed that a decrease in pH corresponds to an increase in sorption, particularly when the adsorption process is not primarily governed by hydrophobic interactions. In such cases, a lower pH environment promotes greater PFAS sorption onto the biochar surface [91,93]**.** This can be explained with the electrostatic interaction which is enhanced at low pH resulting in higher sorption onto biochar. It has been generally demonstrated that dissolved organic carbon (DOC) has a negative impact on PFAS adsorption. This effect can be attributed to two potential mechanisms. Firstly, the presence of DOC can competitively sorb onto the biochar surface, limiting the available sorption sites for PFAS. Secondly, DOC may act as a blocking agent, physically obstructing the sorption sites on the biochar surface, thereby hindering the sorption of PFAS molecules [101,102].

Biochar has the potential to immobilize PFAS in soil, but the degree of immobilization can vary significantly depending on factors such as the type of PFAS, the characteristics of the sorbent material, and the chemical properties of the soil [103]. Based on our findings, it can be concluded that the impact of biochar on PFAS adsorption exhibits a high degree of variability. However, identifying the appropriate biochar type can prove to be extremely beneficial for remediation efforts at PFAS-impacted sites.

Future studies can be directed towards investigating modified biochar, as we see in some cases where lab grade and commercial biochar outperforms GAC in PFAS adsorption efficiency [17].

5.2. Granular activated carbon

Granular Activated Carbon (GAC) is a highly porous material made from carbon rich organic resources is found to be highly effective in treating certain PFAS species [104,105].

Due to its high surface area, porosity, ability to fix surface charge based on manufacturing process, and long-term durability if regularly maintained makes GAC an extremely effective filter media. It can be used in several ways when used as a stormwater biofilter media for removal of PFAS [77]**.** GAC can be used as an infiltration media, where the stormwater runoff first passes through a layer of GAC before infiltrating the soil for groundwater recharge [106]**.** It can also be used as a filtration media where the runoff is passed through layers of GAC and sand to remove PFAS and suspended particles respectively. Next application comes as an amendment to the soil media, to enhance the immobilization of PFAS as well as providing physical filtration [107].

We evaluated 22 studies and found relevant data in 8 studies to determine the adsorption capacity of long and short chain PFAS, that showed varying adsorption capacities.

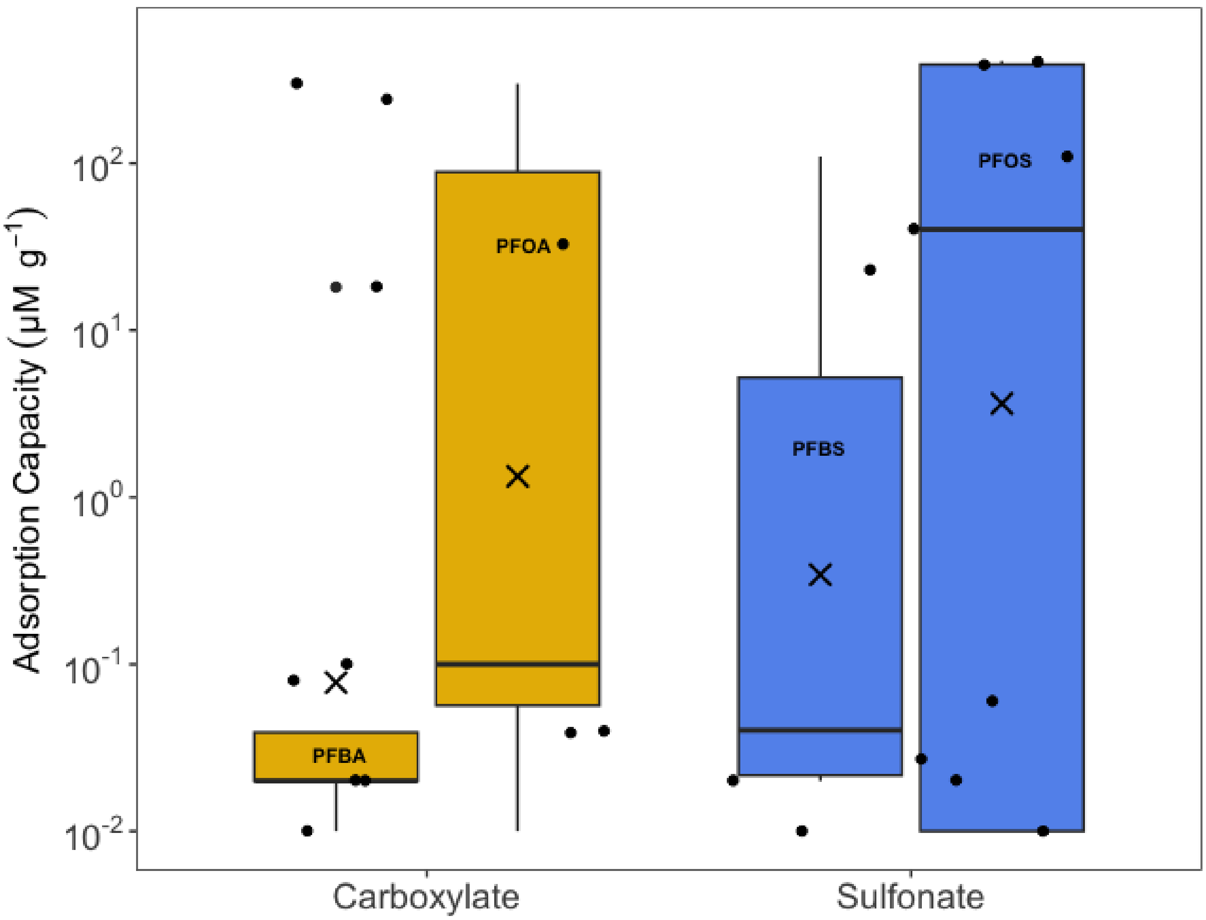

Figure 3 Adsorption capacity of GAC based on short chain PFAS as PFBA (C4) and longer chain PFAS as PFOS(C8) and classified in Perfluorinated Carboxylic acids (PFCA) and Perfluorinated Alkyl Sulfonates (PFSA)

Fig. 3. illustrates the adsorption capacity of short and long chain PFCA and PFSAs. We notice that GAC exhibits poor performance in the removal of short chain PFAS. This can be attributed to the fact that shorter-chain PFAS compounds are generally more hydrophilic, leading to reduced adsorption capacity when interacting with hydrophobic adsorbents such as GAC [68]. Even though we observe a high degree of removal in some cases of PFOA and PFOS, these were mainly attributed to extremely high sorption time, of at least 168 hours [108] or the GAC had being modified to a regenerable magnetic AC [109]. A p-value of 0.42 between the PFCA and PFSA indicates that there is no statistical significance in the removal capacities in these

functional groups. Based on our survey, we have observed a wide range of PFOS adsorption capacities, spanning from 0.01 µmol/g to 390 µmol/g. This significant variability is hypothesized to be influenced by the manufacturing processes employed for GAC production. Furthermore, the adsorption of PFAS onto GAC is affected by various intrinsic factors. pH levels, ionic strength, and specific properties of the GAC material itself all play important roles in determining the extent of PFAS adsorption.

Most studies [91,108,109] reported a negative relation between increase in pH and sorption capacity. This is particularly due to pk_a of certain PFAS and their increase in electrostatic interaction with GAC with a decrease in pH. [110] reports that ionic strength comes into effect, and is enhanced in systems with competing ions, that facilitate aggregation via divalent cation bridging effect and charge neutralization. Predominantly, the ionic strength effect keeps decreasing with an increase in PFAS chain length as the adsorption mechanism is favored by hydrophobic interactions in case long chain PFAS [111]**.** One of the most important characteristics for determining the adsorption of PFAS onto GAC is the pore distribution. [112,113] report that the presence of meso- and macro- pores, help in the adsorption of long chain PFAS. Presence of organic matter had varying results on the adsorption of PFAS, hence no conclusive inference could be drawn. However, [114] suggests that performance of bituminous based GAC had a reduced influence by varying DOC content. These factors indicate that, even though GAC can be useful at decreasing the concentration of PFOA and PFOS in water, it is quite unreliable for eliminating a wide range of PFAS.

It is crucial to address the knowledge gaps pertaining to the use of GAC as a potential adsorbent, particularly considering its relatively low adsorption capacity for short-chain PFAS. Future studies can focus on exploring modification methods aimed at enhancing the adsorption of these PFAS species, given their significant prevalence in stormwater (Fig. 1.B.).

5.3. Ion exchange resin

Ion exchange resins are solid, porous, insoluble substances that are frequently employed in water and wastewater treatment procedures to exchange or remove certain ions from a solution. IX resins can be utilized in the wastewater treatment process to eliminate certain pollutants or trace contaminants including various PFAS from drinking water sources and effluent streams.

Due to the negative charge of anionic PFAS, they may be selectively adsorbed onto the resin, as it has been observed that anion exchange resins are very effective in eliminating PFAS compounds. The utilization of ion exchange (IX) in filter systems offers a notable advantage in the form of the regeneration process. Particularly in the realm of stormwater biofilters, where the dependability of treating runoff assumes significant importance, the ability to regenerate the resins proves highly advantageous [115].

The mechanism of PFAS removal from water using ion exchange resins involves a dual mechanism with selective adsorption of PFAS onto the anion exchange resin, mostly governed by hydrophobic and electrostatic interactions [116] and exchange of ion, because of both these mechanism working at play IX resins have an increased PFAS removal [117,118]**.** Aggregation may play an important role in the adsorption of long chain PFAS like PFOA and PFOS at active sites that are favored by hydrophobic interactions [119]. Positively charged functional groups found in anion exchange resins enable the attraction and exchange of negatively charged ions. The adsorption process is highly selective, meaning that certain types of PFAS compounds will be preferentially adsorbed over others, depending on the resin chemistry [120].

Looking at data from 6 studies, we observed the adsorption capacity of AE resins and found very effective removal of PFAS.

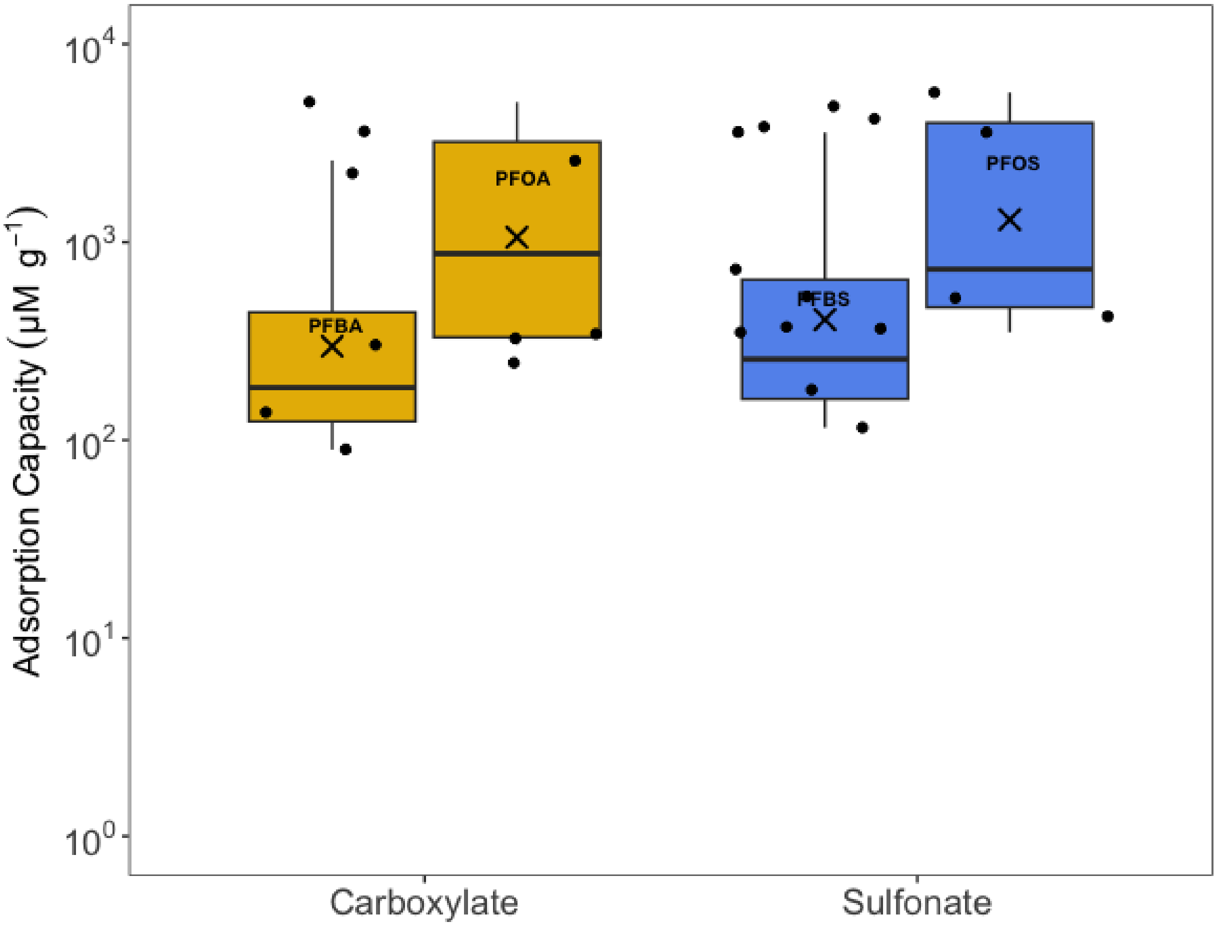

Figure 4 Adsorption capacity of Ion Exchange Resin based on short chain PFAS as PFBA (C4) and longer chain PFAS as PFOS(C8) and classified in Perfluorinated Carboxylic acids (PFCA) and Perfluorinated Alkyl Sulfonates (PFSA)

The sorption capacity of ion exchange resin in the adsorption PFAS is depicted in Fig. 4. Upon initial examination, it is evident that the adsorption capacity of the IX resin is notably superior to that of any other adsorbents studied. One of the main takeaways from this data is the extent of removal of short chain PFAS, that other carbon-based adsorbents are unable to do. No statistical significance was observed between the removal of PFCA and PFSA with a p-value of 0.56 between the two groups and no significant difference was found with the functional group associated with the resin. However, IX resins showed the least variability when compared with other adsorbents. The utilization of quaternary ammonium functional groups as the preferred choice of resin functional group for PFAS adsorption has been consistently observed across various studies. Several significant reasons can be hypothesized to support this choice. Firstly, quaternary ammonium functional groups possess a positive charge, enabling them to form strong interactions with the negatively charged PFAS molecules. Furthermore, these functional groups exhibit high stability, allowing them to withstand harsh conditions including elevated temperatures, extreme pH levels, and exposure to organic solvents [121]. This allows for repeated use and regeneration of the resin [118]**,** resulting in lower costs and environmental impact, which makes it a sustainable choice for incorporating IX resins in green infrastructure for treating surface runoff. In addition, quaternary ammonium functional groups exhibit significant hydrophobicity, implying a low affinity for water and a high affinity for non-polar compounds [122]**.** This allows for efficient adsorption of the highly hydrophobic PFAS molecules. Finally, quaternary ammonium functional groups being a strong exchange group have a low tendency to leach out from the resin, resulting in lower potential for secondary contamination of the water [123]**.** This is important for the removal of PFAS, which are persistent and bio-accumulative in the environment [124–126]. [127] shows how resin matrix has significant effect on the sorption of PFAS. Resins with a hydrophilic matrix showed higher sorption kinetics for long chain PFOS. The high removal capacity for IX resins can be attributed to several factors. Firstly, anion exchange resins are highly selective for anionic PFAS compounds and can specifically attract and exchange these ions resulting in high removal efficiency [128]**.** Secondly, Ion exchange resins have a high adsorption capacity for PFAS compounds, which allows them to adsorb a significant amount of PFAS from water before reaching saturation [15,17]**.** This enables for enhanced efficient and cost-effective treatment, as the resin can be used for a longer period before requiring regeneration. Hydrophobicity plays a crucial role in determining the sorption of PFAS onto resins. [119] highlights that highly hydrophobic resins performed much better than low or no hydrophobic resins, which may depend on the species of PFAS present. The decreased solubility of highly hydrophobic PFAS compounds in water can result in the formation of PFAS aggregates or clusters. These clusters can adversely affect the effectiveness of adsorption by resin-based systems. The presence of PFAS aggregates can hinder the access of the resin to individual PFAS molecules, reducing their availability for sorption onto the resin surface. As a result, the adsorption capacity of the resin may be impacted, potentially leading to decreased efficiency in removing PFAS contaminants from water.

One of the key factors that makes ion exchange resins highly advantageous is their regenerability. Once saturated with PFAS compounds, IX resins can be effectively regenerated [115]**,** enabling their reuse and minimizing waste generation. This regenerative capability not only ensures a more sustainable and cost-effective approach but also contributes to environmental conservation by reducing the overall disposal of used resins. In addition, as per life cycle analysis (LCA) studies, the production process of IX resin has been identified as a crucial step with significant environmental impacts. It has been found to contribute to various environmental factors, including global warming potential, carcinogenicity, ecotoxicity, eutrophication, and acidification [26]. Regeneration is typically achieved using a concentrated salt solution or other strong anion, which displaces the PFAS molecules from the resin and releases them into the regeneration solution [116]. These make AE resins much more suitable, both functionally and economically for treating PFAS from stormwater runoff. Future studies can look at advanced adsorbent regeneration methods, as traditional techniques require great amount of organic solvents, which becomes a challenge from both, an economic and environmental perspective [17].

5.4. Feasibility and application of the amendments

Comparing all amendments, Fig. (5) highlights ion-exchange resins to have the highest adsorption capacity. However, practical application necessitates considering a multitude of factors that impact not only PFAS removal but also the development of an economical and reliable system.

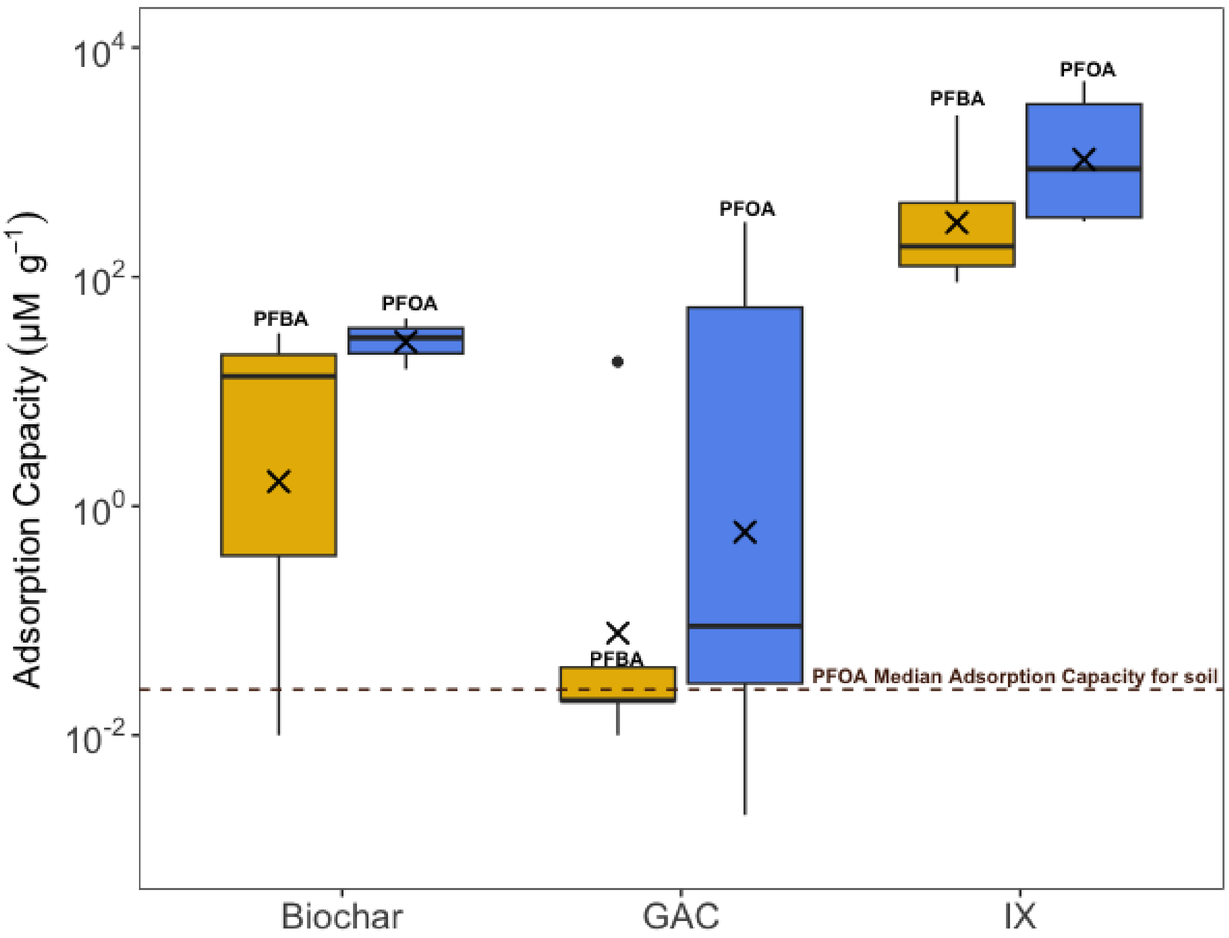

Figure 5 Adsorption capacity of PFCA (PFOA and PFBA) by different adsorbents- Biochar, GAC, and Ion-Exchange Resin. The brown dashed line represents the median PFOA adsorption capacity for soil without any amendments.

Stormwater Treatment Systems (STS) should be designed keeping several factors in consideration. Use of adsorbents in the treatment system depends on adsorption capacity, variability, leaching behavior, material cost, availability, and reusability. While designing BMP for STS, it is important to understand the soil chemistry and presence of co-existing ions, as they can highly influence in the adsorption of not only PFAS but other pollutants of interest [129].

Considering the poor removal of PFAS by soil (Fig. 5.), we propose amending the soil, while establishing green infrastructure such as stormwater biofilters with either one or a combination of adsorbents specific to the site. For example, if a site has a contamination of a relatively higher concentration of shorter chain PFAS, it would be cognizant to consider using IX-Resin despite the cost, as other adsorbents governed by hydrophobic interaction would fail to remove short chain PFAS. Comparing different adsorbents, we can see that biochar and GAC has a wide variability for its adsorption capacity (Fig. 5.). Even though, they have sufficient removal capability for long chain PFAS, they fail to remediate short chain PFAS. Hence, it is crucial to consider the target PFAS before considering these amendments for the STS.

Desorption of PFAS from the adsorbents is another factor that must be considered, as they can serve as secondary sources of contamination. There are two scenarios in which leaching should be considered for adsorbents: during service to prevent degradation of runoff quality and after disposal in landfills. Adsorbents with low leaching potential should be preferred for practical use [129]**.** Long-term studies to determine leaching potential are rarely used due to lengthy experiment durations. Hence, evaluating chemical leaching during disposal is crucial to prevent secondary pollution. Leaching tests [130] can rapidly assess chemical leaching in different situations. Once adsorption and leaching evaluations are passed, the adsorbents can be recommended for use in stormwater treatment.

When selecting amendments, it is important to consider the proximity of local sources as it can affect the delivery and shipping costs. To avoid prohibitive costs, it is recommended to opt for more cost-effective options that can still deliver the desired level of treatment for the targeted PFAS.

However, to our knowledge there is no guidance to use of amendments in STS for PFAS treatment, hence we have tried to summarize some critical information (Table 1.) to help BMP designers make a more informed decision for remediation.

Table 1. Cost, availability, and feasibility of different adsorbents for PFAS removal in Stormwater treatment systems.

Adsorbent	PFAS adsorption capacity (µmol/g)	Material Cost (USD/Ton)	Accessibility	Advantage	Challenges
Biochar	0.00008-795.73	350-1,200[a]	Locally produced; Easy to obtain	Cheap, easily available, environmentally friendly	Biochar feedstock and pyrolysis temperature can greatly impact adsorption capacity, hence quality control is difficult
GAC	0.01-300	2,750[b]	Widely available, can be easily sourced	Industry standard, hence quality control is maintained	Very poor removal for short chain PFAS
IX-Resin	89.7-5100	17,600[b]	Sophisticated infrastructure required for manufacturing	High removal capacity, regenerable	Expensive to manufacture and operate. Regeneration becomes a recurring cost

a131

b132

6. Challenges and opportunity

6.1. Variability in adsorption capacity

Although amendments show suitable performance in removing PFAS compared to a control of just sand or soil, their efficacy can be unpredictable **(Fig. 5)**. The ability of amendments to remove PFAS can vary significantly depending on the specific type of PFAS, the amendment used, and the environmental conditions of the biofilter system [133]**.** Consequently, when designing best management practices (BMPs), a combination of amendments tailored to the PFAS type at the site may be necessary [134]**.** Moreover, the removal capacity of each amendment may vary widely due to differences in preparation and the amount of amendment added to the biofilter [135].

6.2. Lack of guidance for amendment use

To our knowledge, there are no guidance for amendment use, when it comes to designing STS for remediation of PFAS, and we have considered multiple factors, such as site assessment to identify target PFAS, possible transport mechanisms to identify source, and suggested remediation techniques.

Our guide also gives an insight to pragmatic factors that are important to consider, such as recurring costs, availability, and regeneration capability, for designing cost prohibitive STS. On selection on amendment, it can be incorporated in the design to meet local design factors and regulations, before installation.

6.3. Disconnect between lab and field studies or lack of testing in field

In laboratory studies, amendments are often evaluated based on their ability to adsorb PFAS, which is measured as the maximum amount of pollutant that can be removed per unit mass of amendment before it is saturated. However, while this adsorption capacity can be used to compare different amendments, it may not accurately reflect their performance in bioretention systems, as the actual removal of contaminants can be influenced by factors such as hydraulic residence time and other constituents present in the stormwater [136]. If the hydraulic retention time is reduced or the infiltration rate of stormwater is increased, the ability of pollutants to be adsorbed may be significantly impaired, regardless of their adsorption capacity [137]. Along with that, amendments may experience reduced adsorption of PFAS and other contaminants in the presence of dissolved organic carbon and salts that are in a dissolved state [138]. Hence, it is important for laboratory experiments to take such factors into consideration, to replicate a practical functioning of the amendments to adsorb PFAS.

These factors make it important to have field conditioning for laboratory experimenting while designing BMP of STS amendments. For future studies, we recommend looking at pilot scale studies with standardized testing procedures.

6.4. Cost benefit analysis

The cost of amendments plays a critical role in their practical use for stormwater treatment systems, regardless of their effectiveness. Waste biomass-derived materials like compost or biochar are generally less expensive than surface-functionalized amendments such as IX resins (Table 1). Therefore, even if a novel amendment can efficiently remove contaminants, it may not be suitable for field applications if it is not cost-effective. Shipping costs also impact the overall cost of amendments, and using a local source for a specific amendment can considerably reduce its application costs [139]. Thus, conducting cost-benefit and life-cycle analyses is essential to inform regulatory agencies of the potential benefits that can be realized by using these amendments in stormwater management.

6.5. Future Studies

This review has looked at existing adsorbents available and tried to draw a comparison to choose the best option. Future studies can focus on new engineered materials such as Zeolite and try studying the sorption kinetics of PFAS onto them. Recommendations for frameworks to be set up based on soil chemistry, PFAS species, and adsorbent availability can also be suggested as we see from this study that PFAS adsorption can be highly variable based on certain factors, and finding correlations between the variables could be considered BMP for setting up stormwater biofilters. They should also investigate interfacial properties of PFAS, in stormwater infrastructure and desorption of PFAS species from the adsorbents, to prevent secondary sources of contamination. Another vital challenge we face in this area is, adsorption selectivity. NOM in water competes with adsorption of PFAS onto adsorbents [140]**,** along with presence of other salts and other anions that reduce the adsorption capacity of the amendments. To maximize removal of PFAS and other contaminants from stormwater runoff, conventional standalone media might not be enough to achieve high contaminant removal. Hence, more studies should be looked into composite engineered media, that utilizes coating, doping, or other surface enhancements to take advantage of not only hydrophobic interactions, but electrostatic interactions as well [141]. This may enhance the treatment life of the system as well, by requiring less replacement of amendment, once the adsorbent is oversaturated. The implementation of this new approach has the potential to convert conventional STS into a feasible and cost-effective solution for stormwater treatment.

7. Conclusion

The occurrence of PFAS in stormwater can be attributed to multiple factors, and its transport makes it ubiquitous and essential to be treated to prevent contamination. Current soil stormwater treatment system is ineffective in removing the PFAS species, especially short chain PFAS. Biochar, GAC, and IX Resins are investigated as potential amendments to add to STS, that significantly improves the removal of several PFAS species. Biochar is an inexpensive method that improves long chain PFAS removal but fails to remove short chain, whereas GAC shows high variability for removal which makes it unreliable. Ion-Exchange resins on the other hand shows high removal of PFAS for both short and long chains but is significantly more expensive to implement. These factors make it necessary for sites contaminated with PFAS to carry accurate investigation, which could help choosing any or few of these amendments in combination to treat PFAS in stormwater.

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