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Rechargeable stormwater biofilters: In situ regeneration of PFAS removal capacity by using a cationic polymer, polydiallyldimethylammonium chloride

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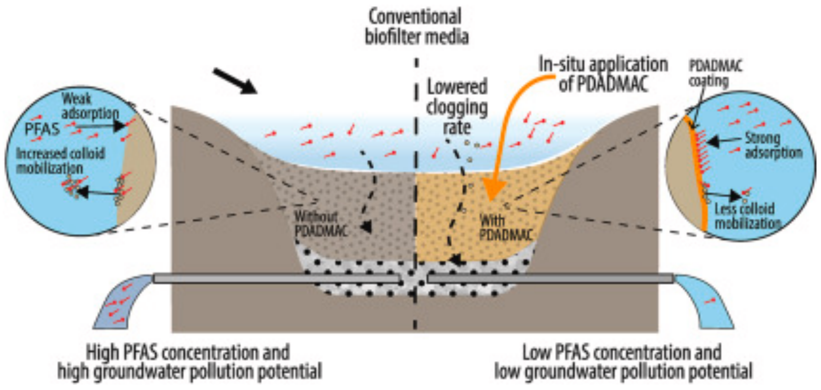
Highlights

- Polydiallyldimethylammonium chloride (PDADMAC) replenished PFAS removal capacity.
- The benefit of PDADMAC decreased with increases in PFAS carbon chain length.
- PDADMAC did not negatively affect the clogging rate of stormwater biofilters.
- PDADMAC application could reduce the colloid-facilitated release of PFAS.
- PDADMAC could last long in stormwater biofilters without biodegradation.

Abstract

Conventional stormwater biofilter media such as sand and compost have limited capacity to remove PFAS due to the exhaustion of attachment sites with pollutants and other stormwater constituents. Replacing exhausted filter media is expensive whereas in situ regeneration of their adsorption capacity can be cost-effective. This study demonstrates that in situ application of cationic polymers such as polydiallyldimethylammonium chloride (PDADMAC), a drinking water coagulant, could regenerate the filter media capacity to remove anionic PFAS. The benefit of PDADMAC increased with a decrease in PFAS carbon chain length: the removal of PFBA increased by 9.9±1.8 times while the removal of PFOA only increased by 0.3±0.03 times. The result indicates the application of the cationic polymer can help remove short-chain PFAS, the ones that are most difficult to remove by typical organic amendments such as activated carbon and biochar. Despite being used as coagulants, the addition of PDADMAC did not negatively affect the clogging rate of the biofilters in the presence of suspended sediments. Surprisingly, biofilters exposed to PDADMAC required 35.3% more suspended sediments than the biofilters without PDADMAC to reduce the hydraulic conductivity below half of the initial value. PDADMAC application reduced the release of colloids from the biofilter media during intermittent flow, thereby decreasing the potential of the colloid-facilitated release of adsorbed PFAS. The adsorbed PDADMAC in the biofilter was not degraded, indicating that applied PDADMAC could last long and continue to remove PFAS in the biofilter. Thus, in situ application of cationic polymers can help regenerate the PFAS removal capacity of conventional biofilters.

Graphical abstract



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Introduction

Subsurface soil or subsurface stormwater treatment systems filter pollutants from surface runoff and protect groundwater (Alam et al., 2021). However, many emerging pollutants such as per- and polyfluoroalkyl substances (PFAS) could pass through these systems and contaminate groundwater (Spahr et al., 2020). In fact, nearly 72% of drinking water treatment facilities that use groundwater as the drinking water source were contaminated with PFAS (Guelfo and Adamson, 2018). Stormwater conveys PFAS from contaminated areas such as industries, landfills, and airports to surface waters and groundwater via subsurface infiltration (Anderson et al., 2016; Dauchy et al., 2019a; Podder et al., 2021). Dissolved PFAS concentration in stormwater ranges between 14.3 and 96 $ng L^{-1}$ (Page et al., 2019; Saifur and Gardner, 2021), although particulate matter in stormwater could contain significant amounts of PFAS (Borthakur et al., 2021c; Xiao et al., 2012). Infiltration-based stormwater treatment systems such as biofilters have been used to treat stormwater pollutants (Grebel et al., 2013), but conventional biofilter media such as sand and compost have low adsorption capacity for PFAS (Aly et al., 2018; Hale et al., 2017). Amending the conventional filter media with carbon adsorbents such as biochar and activated carbon could improve PFAS removal (Askeland et al., 2020; Mukhopadhyay et al., 2021; Park et al., 2020). However, adsorption sites on all media would eventually become exhausted due to the adsorption of the myriad of pollutants and constituents present in stormwater, thereby requiring cost-prohibitive media replacement (Tirpak et al., 2021). In contrast, in situ regeneration of the adsorption capacity of the exhausted filter media can be cost-effective.

In situ regeneration typically requires an addition of a chemical or mixture of chemicals that would coat existing filter media with material with high adsorption capacity (Charbonnet et al., 2018; Demeestere et al., 2002; Dusenbury and Cannon, 2004). Filter media can adsorb PFAS by either ion exchange or hydrophobic interactions or both (Park et al., 2020). Typical stormwater amendments such as biochar or activated carbon remove PFAS by hydrophobic interaction (Fabregat-Palau et al., 2022), but they have limited capacity to remove short-chain PFAS such as perfluorobutanoic acid (Xiao et al., 2017), which are most challenging to remove from stormwater. In this case, cationic polymers such as polydiallyldimethylammonium chloride (PDADMAC) could be added to improve removal by the electrostatic attraction (Aly et al., 2018, 2019; Liu et al., 2020; Ramos et al., 2022; Ray et al., 2019). These cationic polymers can easily adsorb onto most natural and engineered media such clay (Ray et al., 2019), activated carbon (Liu et al., 2020; Ramos et al., 2022), and aquifer soil (Aly et al., 2018, 2019). However, most of these studies added filter media pre-coated with PDADMAC. In practice, in situ application is desirable to recharge conventional biofilters without any amendments.

Despite the known benefit of PDADMAC to enhance PFAS removal, other potential unintended consequences of the application PDADMAC in stormwater biofilters have not been explored. First, most of the previous studies with PDADMAC were tested in groundwater or saturated systems (Aly et al., 2018, 2019; Liu et al., 2020; Ramos et al., 2022; Ray et al., 2019), whereas stormwater systems are unsaturated systems with exposure to dry-wet cycles. In unsaturated soil, the presence of air packets could cause preferential flows and limit a complete filter media coverage of PDADMAC during in situ application of the polymer. Second, unlike groundwater, stormwater contains a high concentration of suspended particles. The presence of PDADMAC, a drinking water coagulant, can increase the aggregation of suspended sediments, which could increase the clogging rate of the biofilters (Li et al., 2021; Nguyen et al., 2019). Yet, no study to date has examined changes in the clogging potential of biofilters after in situ application of PDADMAC. Third, for PDADMAC to remove PFAS consistently, the applied PDADMAC should last long in the biofilter media without rapid degradation. A previous study (Simcik et al., 2019) showed that microbial communities from a groundwater aquifer did not degrade PDAMAC, but microorganisms present in activated sludge did. Yet, no study to date has examined the degradation of PDADMAC in stormwater biofilters, where the microorganisms would be different from that of activated sludge.

This study examines the effect of in situ application of PDADMAC into biofilters on their capacity to remove PFAS and maintain infiltration capacity and evaluate the potential of biodegradation of PDADMAC by biofilter's microbial community. Conventional biofilter media (sand and compost) were used to evaluate changes in infiltration capacity and PFAS removal capacity after exposure to PDADMAC. Four types of PFAS with increasing carbon chain length were used to evaluate the effect of carbon chain length on the improvement in adsorption capacity by the application of PDADMAC. Furthermore, the persistence of sequestered PDADMAC was tested by quantifying the degradation of PDADMAC in biofilters during 2–7 days of flow interruption. The results have practical significance in turning conventional biofilters into regenerative biofilters for enhanced removal of PFAS from stormwater.

Section snippets

Stormwater and biofilter media preparation

Natural stormwater was used to simulate the adsorption of PFAS onto biofilter media under field conditions as organic carbon, nutrients, and other pollutants typically present in natural stormwater could compete with PFAS for adsorption sites on the filter media. Stormwater was collected from Ballona Creek, Los Angeles, and their basic water quality parameters were measured and reported (Table S1). The stormwater was spiked with perfluorobutanoic acid (PFBA, Acros Organics), perfluorohexanoic …

PDADMAC injection replenished the biofilter attachment sites for PFAS

Coating the biofilters with PDADMAC replenished the exhausted PFAS adsorption sites and increased overall PFAS adsorption capacity of compost-sand biofilters (Fig. 1a). A comparison of the PFAS removal capacity of biofilters after exposure to the same amount of contaminated stormwater revealed that the increase in PFAS removal due to PDADMAC application depended on the carbon chain length (Fig. 1b). The benefit of the PDADMAC application to enhance PFAS removal decreased with an increase in the …

Enhanced PFAS removal in conventional biofilters by PDADMAC application

Most biofilters contain a mixture of sand and compost, which have a limited capacity to remove PFAS from stormwater (Fabregat-Palau et al., 2022). This study demonstrated a simple method to recharge the capacity of these biofilters without replacing the conventional media with more expensive amendments. The result shows that the application of cationic polymers such as PDADMAC into biofilters is particularly useful to remove the short-chain PFAS. The improvement in PFAS removal was attributed …

Conclusions

Compost biofilters have limited ability to remove PFAS and could become exhausted rapidly. This study demonstrated a simple method to regenerate their adsorption capacity by applying cationic polymers such as PDADMAC in situ. The application of PDADMAC not only replenished the exhausted PFAS adsorption sites but also improved the removal capacity for short-chain PFAS, which are typically difficult to be removed by amendments such as activated carbon or biochar. Moreover, the PDADMAC application …

CRediT authorship contribution statement

Annesh Borthakur: Conceptualization, Methodology, Investigation, Formal analysis, Visualization, Writing – original draft. **Tonoy K. Das:** Methodology, Investigation, Formal analysis. **Yuhui Zhang:** Investigation. **Silvi Libbert:** Investigation. **Samantha Prehn:** Investigation. **Pia Ramos:** Investigation. **Gregory Dooley:** Formal analysis, PFAS analysis or sample measurement. **Jens Blotevogel:** Writing – review & editing. **Shaily Mahendra:** Writing – review & editing, Funding acquisition. **Sanjay K. Mohanty:** …

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper. …

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…To sustainably manage groundwater resources, managed aquifer recharge (MAR) technologies have been implemented in many places (Alam et al., 2021; Levintal et al., 2023), where treated municipal wastewater or stormwater is applied to replenish groundwater (Cáñez et al., 2021; Das et al., 2023; Zhang et al., 2020). However, both stormwater and treated wastewater contain perfluoroalkyl and polyfluoroalkyl substances (PFAS) (Borthakur et al., 2022; Cáñez et al., 2021; Page et al., 2019). Most PFAS can adsorb on subsurface soil and capillary fringe in recharge basins (Adamson et al., 2020; Maizel et al., 2021), thereby decreasing groundwater pollution.…

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2023, Journal of Cleaner Production

Citation Excerpt :

…The creek receives dry-weather irrigation runoff from 318 km2 of the urban area, with 82% developed and 61% impervious surface. The detailed characteristics of stormwater from the sampling area were reported elsewhere (Borthakur et al., 2022b; Ghavanloughajar et al., 2021). The specific water quality parameters for the samples used in this study are detailed in Table S1.…

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