

RESEARCH ARTICLE OPEN ACCESS

Using Biochar in Static and Dynamic Flow Systems to Remediate Per- and Polyfluoroalkyl Substances From Contaminated Stormwater Runoff

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Received: 17 June 2025 | **Revised:** 19 September 2025 | **Accepted:** 23 September 2025

Funding: This study was financially supported by the Swedish Research Council for Sustainable Development FORMAS (project number 2020-01107, SAFESTORM) and the project “Residues for best use – Biochar as solution and product in the circular and climate positive society (Rest-to-Best)” funded by the Swedish Innovation Agency (Vinnova) (project number 2021-01589).

Keywords: biochar | per- and polyfluoroalkyl substances (PFAS) | physico-chemical characterization | remediation | stormwater

ABSTRACT

Biochar has recently been identified as a potential solution for the remediation of organic micropollutants from contaminated water. Herein, we have assessed the potential mitigation of per- and polyfluoroalkyl substances (PFAS) by means of biochar adsorption as a green alternative to coal-based sorbents for PFAS-polluted stormwater systems. For this purpose, 13 biochar materials (originating from diverse feedstocks as well as intended for both commercial and research purposes) were initially screened for PFAS remediation capabilities in static flow systems. These experiments pointed to biochar sorption as a promising strategy for PFAS remediation, with some materials showing removal efficiencies of around 99% after 7 days of exposure. Though not all of the biochar materials tested performed equally, differences could be observed. As a next step, five biochar materials were studied under constant-flow column experiments for a duration of 69 days using real stormwater spiked with PFAS. Results showed that vast differences could be observed for the retention rates of the tested PFAS contaminants, with estimated bed volumes for an 80% breakthrough ranging from, for example, 13–60 for per-fluorobutanesulfonic acid and from 4 to 53 for perfluoropentanoic acid. In terms of the PFAS backbone, both the static and dynamic flow experiments highlighted that long-chain PFAS showed stronger sorption onto the biochar surface than short-chain PFAS; however, no relevant impact could be identified in terms of the PFAS functional group. Overall, biochar is emerging as a promising and environmentally friendly approach for removing PFAS from contaminated stormwater.

1 | Introduction

Stormwater runoff has been identified as a major source of toxic chemicals in the aquatic ecosystem through its direct discharge

into the environment (Müller et al. 2020; Masoner et al. 2019; Charters et al. 2016; Gasperi et al. 2022) or after rudimentary treatment, such as natural and constructed wetlands (Kondor et al. 2024; Istenič et al. 2011; Sébastien et al. 2014). With the

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current shift to increasingly more fluctuating weather patterns (Glaas et al. 2024), the accumulation of organic micropollutants in streets, roads, and so on is exacerbated by long periods without any precipitation. Additionally, with heavy rainfall afterwards, the amount of organic micropollutants mobilized in stormwater suddenly increases by flushing off paved surfaces in cities, resulting in peak contaminant loadings into receiving water bodies (Reoyo-Prats et al. 2018). Moreover, during cold seasons, snow precipitation, with its prolonged exposure time, retains environmental contaminants for a long time. With eventual melting, the accumulated pollutants are mobilized from the accumulated snow and released into the environment in acute events (Boom and Marsalek 1988; Björklund et al. 2011; Marklund et al. 2005). Thus, there is an urgent need for innovative techniques for the treatment and control of stormwater pollution.

Per- and polyfluoroalkyl substances (PFAS) have been identified as an emerging group of chemicals that can be mobilized by stormwater and, thus, be a major contributor to stormwater pollution (Chen et al. 2023; Codling et al. 2020; Tsou et al. 2024). PFAS are typically characterized as short- and long-chain compounds depending on the number of fluorinated carbons. Thus, short-chain PFAS have < 6 fluorinated carbons, whereas long-chain PFAS have ≥ 6 fluorinated carbons (Buck et al. 2011). PFAS, either short or long chain, have been shown to be extremely persistent and potentially bioaccumulative and toxic to ecosystems and humans (Fenton et al. 2021; Ahrens and Bundschuh 2014). Thus, remediation strategies should be evaluated to identify efficient stormwater treatment technologies. To this end, some studies have tested the use of activated carbon (Pritchard et al. 2023a), phytoaccumulation by native plants (Awad et al. 2022), or implementation of polymeric-clay composite sorbents in urban ornamental gardens to remove PFAS from stormwater runoff (Ray et al. 2019).

Biochar matter is generated through anaerobic pyrolysis of biomass, either plant-based waste (e.g., timber, wood chips, agricultural residues, etc.) or wastewater sludge (Gray et al. 2014; Boehm et al. 2020), that confers several properties to this carbon-rich material. Its high porosity, abundance of amorphous carbon, and the presence of interspersed voids (Gray et al. 2014) result in a large surface area and, thus, strong potential for chemical interactions in aqueous solutions (Boehm et al. 2020). Hence, biochar has been identified as a promising alternative to mitigate the harm of pollutants from polluted water bodies (Reddy et al. 2014; Ghavanloughajari et al. 2020; Gwenzi et al. 2021; Blum et al. 2019), with particular attention paid to its application in stormwater as well (Boehm et al. 2020; Kaya et al. 2022; Okaikue-Woodi et al. 2020; Tan et al. 2015; Hawkins et al. 2024; Pritchard et al. 2023b). Unlike most sorbent material studies in the literature, including other carbon-based options, such as granular activated carbon, biochar has been shown to be more cost-efficient, with the added advantage of regeneration ability through methods such as re-pyrolysis, heat-activated chemical desorption, or microwave treatment of spent materials (Baaloudj et al. 2025; Alsawy et al. 2022), helping to achieve both material regeneration as well as catalyzed destruction of contaminants (Sun et al. 2024; Xiao et al. 2020). This makes biochar a sustainable and cost-effective solution for environmental remediation applications with net environmental benefits (Sparrevik et al. 2011).

In this study, we aimed to explore the potential for the use of biochar materials in the remediation of PFAS contamination in stormwater. To this end, two different strategies were consecutively used. In the first step, the removal efficiency (RE) of biochar was evaluated in static flow experiments using PFAS-contaminated stormwater, and, in the second step, a dynamic flow system was set up to evaluate the long-term performance of biochar as a PFAS adsorption approach in stormwater treatment facilities. This is, to the best of the authors' knowledge, the first evaluation of biochar efficiency for PFAS remediation that has been conducted with field-collected stormwater in static and dynamic flow systems for a diverse set of 13 biochar materials.

2 | Materials and Methods

2.1 | Chemicals and Materials

In total, 15 PFAS were included in this study. The target compounds comprised of 10 perfluoroalkyl carboxylic acids (PFCAs): perfluorobutanoic acid (PFBA), perfluoropentanoic acid (PFPeA), perfluorohexanoic acid (PFHxA), perfluoroheptanoic acid (PFHpA), perfluorooctanoic acid (PFOA), perfluorononanoic acid (PFNA), perfluorodecanoic acid (PFDA), perfluoroundecanoic acid (PFUnDA), perfluorododecanoic acid (PFDoDA), and perfluorotetradecanoic acid (PFTeDA); three perfluoroalkyl sulfonic acids (PFASs): perfluorobutanesulfonic acid (PFBS), perfluorohexanesulfonic acid (PFHxS, linear (LPFHxS) and branched (BPFHxS)), and perfluorooctane sulfonic acid (PFOS, linear (LPFOS) and branched (BPFOS)); one sulfonamide: perfluorooctanesulfonamide (FOSA); and one fluorotelomer: 6:2-fluorotelomersulfonic acid (6:2 FTSA) (Wellington Laboratories). A standard solution containing 15 isotopically labeled internal standards (ILIS), purchased from Wellington Laboratories, was used. The solution contained $^{13}\text{C}_4$ -PFBA, $^{13}\text{C}_5$ -PFPeA, $^{13}\text{C}_5$ -PFHxA, $^{13}\text{C}_4$ -PFHpA, $^{13}\text{C}_8$ -PFOA, $^{13}\text{C}_9$ -PFNA, $^{13}\text{C}_6$ -PFDA, $^{13}\text{C}_7$ -PFUnDA, $^{13}\text{C}_3$ -PFDoDA, $^{13}\text{C}_2$ -PFTeDA, $^{13}\text{C}_3$ -PFBS, $^{13}\text{C}_3$ -PFHxS, $^{13}\text{C}_8$ -PFOS, $^{13}\text{C}_2$ -6:2 FTSA, and $^{13}\text{C}_8$ -FOSA. Additionally, a native PFAS mixture standard (PFAC-24PAR, Wellington Laboratories, containing all target compounds) was used for a method recovery test.

Methanol (LiChrosolv, hypergrade for LC-MS, Supelco) was purchased from Merck and used for sample extraction in both static and dynamic flow experiments as well as for the preparation of standards. Other materials used for sample preparation and analysis included MilliQ (MQ) water (MilliQ IQ7000 system with a connected LC-Pack polished filter, Merck), acetic acid (100%, Suprapur for trace analysis, Supelco, Merck), ammonium hydroxide solution (ACS reagent, 28.0%–30.0% NH_3 basis, Supelco, Merck), and ammonium acetate (LiChropur, eluent additive for LC-MS, Supelco, Merck).

2.2 | Biochar Materials

In this study, 13 different biochar materials (labeled BC-A to BC-M) were tested. In order to cover a spectrum as wide as possible of biochar composition and feedstocks, both commercially available and research biochar products were evaluated (Table 1). Acquired

commercially available materials are often sold as soil improvers for farming purposes but also in retail shops for gardening ends. However, research biochar materials are often designed to accomplish a certain goal, for example, to increase the water-holding capacity or to enhance the remediation of a particular group of chemicals. In the collection of biochar materials, biochar materials BC-A, BC-B, BC-C, BC-D, BC-E, and BC-G were commercially available. From another perspective, the materials were also selected to cover a wide variety of feedstocks. Thus, although the biochar suite is mainly comprised of plant-based materials, BC-J and BC-M consisted of digestate and sewage sludge, respectively.

In order to evaluate the potential impact of the physico-chemical characteristics of the biochar materials, physical surface characterization (Brunauer, Emmett, and Teller [B.E.T.]—surface area and pore size) was carried out; for details, see Section S1 in the Supporting Information.

2.3 | Stormwater Collection

Grab stormwater samples were collected in 25L capacity high-density polypropylene bottles (pre-rinsed with ethanol and MilliQ water) at the *Kunsängsdammen* stormwater pond (59° 50' 37" N; 17° 40' 2.8" W) in the municipality of Uppsala, Sweden, which gathered stormwater mainly from an industrial and commercial area during the period from September to November 2023. Naturally occurring concentrations of PFAS in collected stormwater were below the limit of detection in most cases or at negligible concentrations compared to the spiking levels for the experiments described below.

2.4 | Static Flow Experiments Design

Static flow experiments were intended as fast screening of the biochar performance for the remediation of PFAS from stormwater in a semi real-world scenario, without aiming for

complete chemical equilibrium, and served as a pre-experiment in order to select biochar materials for the subsequent dynamic flow experiments.

The biochar materials were dried overnight at 110°C to remove inherent humidity due to the biochar production process. After drying, 18 g of biochar was mixed with 180 mL of stormwater (L/S ratio 10, similar to previous studies (Ceranek et al. 2024)), followed by gentle shaking by hand for 30 s. Samples were left still for 7 days at 8°C to enable chemical interactions between the liquid and solid phases without impacting the physical structure of the biochar. Abrasion of the physical structure of biochar was avoided in order to mimic the material that would subsequently be used for the dynamic flow experiments. After 7 days, samples were filtered through glass microfiber filters (47 mm diameter, 1.2 µm pore size, Whatman, China), to remove any biochar particulate and to stop the experiment. From the filtered aqueous phase, three 40 mL aliquots were collected for solid-phase extraction (SPE) and subsequent ultrahigh performance liquid chromatography tandem mass-spectrometry (UHPLC–MS/MS) analysis (for details, see Section 2.6).

For this purpose, stormwater was artificially spiked with the 15 target PFAS at a concentration of 1.25 ng mL⁻¹. The spiking level was selected to be consistent with environmentally relevant concentrations (Chen et al. 2023). Additionally, a procedural blank (consisting of Milli-Q water with no biochar), a stormwater blank (consisting of nonspiked stormwater with no biochar), spiked stormwater (with no biochar), and gravel tests (consisting of spiked stormwater in contact with gravel) were performed. The RE was calculated as the % of PFAS remaining in the spiked stormwater after the treatment compared to the experimental control (spiked stormwater with no biochar).

2.5 | Dynamic Flow Experiment Design

Column experiments were performed in tailor-made transparent PVC columns with an internal diameter of 3.6 cm and a

TABLE 1 | Biochar coding, feedstock, and physico-chemical surface properties.

Biochar	Feedstock	BET surface area (m ² /g)			Adsorption average pore size (nm)
		External	Microporous	Total	
BC-A	Agricultural residues	8.10	6.34	14.4	7.98
BC-B	Bark from pine and spruce trees	30.2	44.5	74.7	2.78
BC-C	Spruce	41.5	238	279	2.27
BC-D	Wood/forest residues	20.3	64.7	85.0	4.12
BC-E	Garden residues	44.6	177	221	2.82
BC-F	Forest biomass	83.8	251	335	2.37
BC-G	Wood chips	49.5	124	174	2.58
BC-H	Waste timber	13.0	23.7	36.8	5.42
BC-I	Demolition wood	12.6	37.1	49.7	5.11
BC-J	Digestate sludge	68.2	10.1	78.3	5.74
BC-K	Conifer and broadleaf trees	73.6	188	262	3.69
BC-L	Conifer and broadleaf trees	19.4	40.8	60.1	2.82
BC-M	Sewage sludge	21.52	14.94	36.47	6.87

length of 15 cm. As suggested in previous studies, this type of construction has a length to diameter ratio greater than 4, which simulates field conditions and minimizes potential transversal dispersity (Niarchos et al. 2022; Banzhaf and Hebig 2016). Flow direction was set upward to facilitate the removal of potentially trapped air bubbles and to prevent the formation of favorable channels in the biochar packing and, thus, ensure proper contact and interaction between the sorbent and stormwater. Additionally, chambers (2×3.6 cm) at the inlet and outlet of the columns were filled with gravel to ensure a homogeneous flow distribution. A nylon mesh (pore diameter = $50 \mu\text{m}$) was placed at the inlet and outlet to prevent biochar particles from escaping the treatment system and keeping them securely in place with tailor-made perforated PVC holders (see Supporting Information S1: Figure S1 for more details).

For the column experiments, five biochar materials were selected based on the coverage of different biochar materials (BC-M, sewage sludge), commercial availability (BC-B and BC-G), and materials that performed well (BC-B, BC-F, and BC-G) and marginally during static flow experiments (BC-L and BC-M). The five selected biochar materials were packed in duplicate columns to account for potential experimental artifacts. Additionally, duplicate control columns in which no treatment was applied were filled with inert gravel to mimic a column saturated with sorbing material. Thus, in total, 12 column experiments were monitored in parallel using a 12-channel peristaltic pump (ISM932D, Ismatec IPC, Masteflex, Barrington, IL, USA). Overall, 228 ± 14 g of gravel, 21.7 ± 0.9 g of BC-B, 45.1 ± 0.7 g of BC-F, 21.4 ± 1.4 g of BC-G, 54 ± 4.8 g of BC-L, and 100 ± 0.15 g of BC-M were used. After packing, the columns were equilibrated by pumping Milli-Q water through them for 24 h before starting the treatment.

Artificially spiked stormwater with a set of 15 PFAS at 10 ng mL^{-1} per compound was used for the column sorption tests. A constant water flow of 12 mL h^{-1} was applied for 69 days. Water samples (6 mL) were collected every 8 h, which corresponds to approximately 0.66 bed volumes (BV), in polypropylene (PP) tubes during the first 7 days of the experiment. From Day 8 to Day 14, water samples were collected every 12 h (≈ 1 BV), from Day 15 to Day 21, water samples were collected every 24 h (≈ 2 BV), and from Day 22 until the end of the experiment, water samples were collected every 48 or 72 h (≈ 4 or 6 BV). In total, 63 water samples were collected for each experimental column (756 in total). The high-resolution sampling strategy at the beginning of the column experiments allowed better traceability of the breakthrough in the initial stages of the experiment. The experiment was stopped when no more significant changes in compounds' breakthrough were observed over the course of several weeks. The maximum BV measured was 140 BV.

For each of the time points collected, individual concentrations of PFAS were measured in the treatment columns (C), averaged, and normalized to the average of the measured concentration in the control columns used during the same sampling event (C_0). This permitted the evaluation of the RE (or breakthrough) of the different biochar materials (C/C_0) as well as the construction of breakthrough curves to estimate their overall efficiency.

2.6 | Sample Extraction and Analysis

2.6.1 | Static Flow Experiments' Sample Preparation

The extraction protocol used for static flow experiment samples is reported in detail elsewhere (Smith et al. 2022). All equipment used for sample preparation were thoroughly rinsed with methanol. The glassware as well as glass fiber filters were burned overnight at 400°C before further use. After the sample filtration step, described in Section 2.4, PFAS were extracted from the filtrate using Oasis WAX cartridges (6 cc, 150 mg, Waters). Briefly, all blanks and samples were spiked with an absolute amount of 5 ng of the ILIS ($100 \mu\text{L}$ of the $0.05 \mu\text{g mL}^{-1}$) after the filtration step and before SPE. The recovery test samples were in addition spiked with 5 ng ($20 \mu\text{L}$ of 250 ng mL^{-1}) of the native PFAS mixture standard. Methanol-rinsed 50 mL PP centrifuge tubes (Falcon, conical bottom, Corning Science Mexico, Mexico) containing the samples were vigorously shaken and sonicated for 5 min (Branson 5800, Emerson Electric Co., USA) after the addition of internal standards. Meanwhile, cartridges were pre-conditioned with 4 mL of 0.1% ammonium hydroxide in methanol, followed by 4 mL of methanol, and, finally, 4 mL of MQ water. Samples were then loaded by gravity at approximately 1 drop per second. Cartridges were washed afterwards with 4 mL of 25 mM ammonium acetate buffer in MQ water and dried under high-vacuum conditions. PFAS were eluted from the cartridge sorbent using 4 mL of methanol, followed by 4 mL of 0.1% ammonium hydroxide in methanol, and collected in a 15 mL PP tube. The samples were then concentrated to about 1 mL under a gentle stream of nitrogen, after which they were transferred to a 1.5 mL PP vial (Short Thread Vial, transparent with filling lines, Scantec Nordic, Sweden). The 15 mL PP tube was then rinsed three times with methanol, the rinse was added to the PP vial, and the sample in the vial was then concentrated to 1 mL and stored in a freezer until the UHPLC/MS-MS analysis.

2.6.2 | Dynamic Flow Experiment Sample Preparation

The samples were collected over the course of the column test experiment in 15 mL PP tubes and prepared for PFAS analysis using the direct injection method. Sample aliquots of $500 \mu\text{L}$ were filtered using $0.2 \mu\text{m}$ regenerated cellulose syringe filters (17 mm Thermo Scientific, Rockwood, TN, USA), after which they were directly transferred to 1.5 mL PP vials, and $400 \mu\text{L}$ of methanol and $100 \mu\text{L}$ of the $0.05 \mu\text{g mL}^{-1}$ ILIS were added. The vials were then vortexed and kept in a freezer until the UHPLC/MS-MS analysis.

2.6.3 | Instrumentation

For both static and dynamic flow experiments, samples were analyzed using a SCIEX Triple Quad 3500 UHPLC/MS-MS system equipped with a Phenomenex Gemini C18 HPLC column ($1.7 \mu\text{m}$), a Phenomenex Kinetex C18 precolumn ($1.7 \mu\text{m}$), and a Phenomenex KJ0-4282 analytical guard column (for details, see [Smith et al. 2022]). The injection volume was set to $10 \mu\text{L}$ for an 11-min chromatographic run consisting of mobile

phase A: 10 mM ammonium acetate in MQ and mobile phase B: methanol at a flow rate of 0.6 mL min^{-1} . The gradient was initially set to 5% B, with an increase within the first 0.1 min to 55% B, reaching 99% over 4.4 min, and maintained at 99% for 3.5 min. After this, the gradient was lowered back to 5% B over 0.5 min, where it was kept for 2.5 min for column re-equilibration. The tandem MS was operated under multiple reaction monitoring (MRM) mode and negative electrospray ionization. Detailed information about the selected transitions for each monitored PFAS can be found in Smith et al. (2022). For the static flow experiment samples, a calibration curve (range $0.01\text{--}100 \text{ ng mL}^{-1}$) of native PFAS in pure methanol was used, whereas the same series of calibration curve standards, prepared in a 50:50 methanol to MQ water ratio, was used for direct injection analysis of PFAS from the column test experiment. The instrument was operated using *SCIEX Analyst* (v1.7.3) software, whereas the data extraction, processing, and evaluation were carried out using *SCIEX OS* software (v3.3.1.43).

3 | Results and Discussion

3.1 | Static Flow Experiments

Figure 1 shows the RE for the 15 investigated PFAS in the tested materials. As indicated by the violin plots, BC-B, BC-F, BC-G, and BC-K clearly performed better in terms of the sorption of individual PFAS, with average removal efficiencies ranging from 96% to 99% (Supporting Information S1: Table S1). However, based on the results, BC-H and BC-I were clearly not appropriate materials for the removal of PFAS from contaminated stormwater. Whereas some PFAS were efficiently removed from the aqueous phase (up to 99% RE), the vast majority of the other PFAS did not show satisfactory results, with RE for some chemicals down to 0% (for BC-I). On the contrary, BC-G showed the narrowest individual PFAS distribution with all data points (except for one) within the range 98.5%–100%. However, most biochar materials showed a wide distribution of RE, with efficiencies as low as approximately

30%. Thus, the variation in RE was large, although most of the biochar materials are of similar origin (plant based), except for BC-J (digestate sludge) and BC-M (sewage sludge-based).

Comparing individual PFAS, the variations of the RE (Figure 1) can be explained by the different sorption behaviors of PFAS showing a general decrease in RE with decreasing perfluoroalkyl chain length and stronger sorption of PFSA and FOSA compared to PFCA, as observed previously (Söregård et al. 2020). In contrast to previous studies (McClellan et al. 2017), in this study, no major differences were observed between PFCA, PFSA, FOSA, and 6:2 FTSA, on comparing the same perfluoroalkyl chain length, indicating limited impact of the PFAS functional group on RE. For example, short-chain PFCA and PFSA showed RE ranging from 6% to 75% and from 25% to 76% in BC-H, respectively, or long-chain PFCA and PFSA ranging from 71% to 99% and from 92% to 98%, respectively, in BC-J. However, on comparing short- and long-chain PFCA and PFSA, clear differences in RE were observed (e.g., RE ranging from 21% to 87% for short-chain PFCA, from 88% to 92% for long-chain PFCA, from 42% to 92% for short-chain PFSA, and from 91% to 95% for long-chain PFSA in BC-C), suggesting that the length of the alkyl chain had a clear influence on the RE (Figure 2). In summary, long-chain PFCA and PFSA showed better RE than short-chain compounds. This behavior is in line with the fact that biochar is a highly hydrophobic material due to its unusually high content of alkyl chains as well as aromatic rings on the surface (Kinney et al. 2012; Wiedemeier et al. 2015). Thus, the adsorption capacity of biochar materials is highly favored toward molecules with a low to nonpolar moiety (Fabregat-Palau et al. 2022). In contrast, molecules that do not have a high degree of strong low-polarity moiety, such as short-chain PFAS, show limited sorption onto biochar surfaces due to repulsion forces between the negatively charged carboxylic or sulfonic acid moieties with the generally negatively charged biochar surface. Although this repulsion still exists for long-chain PFAS, the hydrophobic interactions between the fluoroalkyl tail and the surface are strong enough to overcome the repulsion (Söregård et al. 2020; Sörmo et al. 2021; Du et al. 2014; Lei et al. 2023). Additionally, other studies have identified that

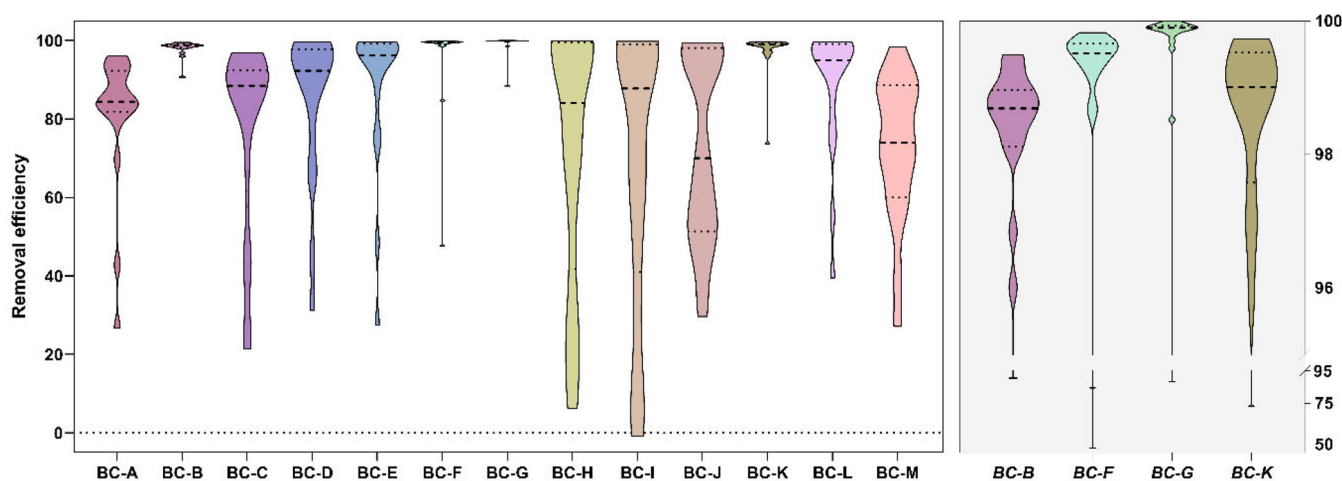


FIGURE 1 | Removal efficiency (RE) for individual PFAS in the different biochar materials tested in the static system. On the right panel (gray-shaded), violin plots for BC-B, BC-F, BC-G, and BC-K are plotted with a magnified y-axis for better visualization of the data. In the violin plots, dashed lines indicate the average value for the RE and dotted lines indicate the 25th and 75th quartiles. [Color figure can be viewed at [wileyonlinelibrary.com](https://onlinelibrary.wiley.com)]

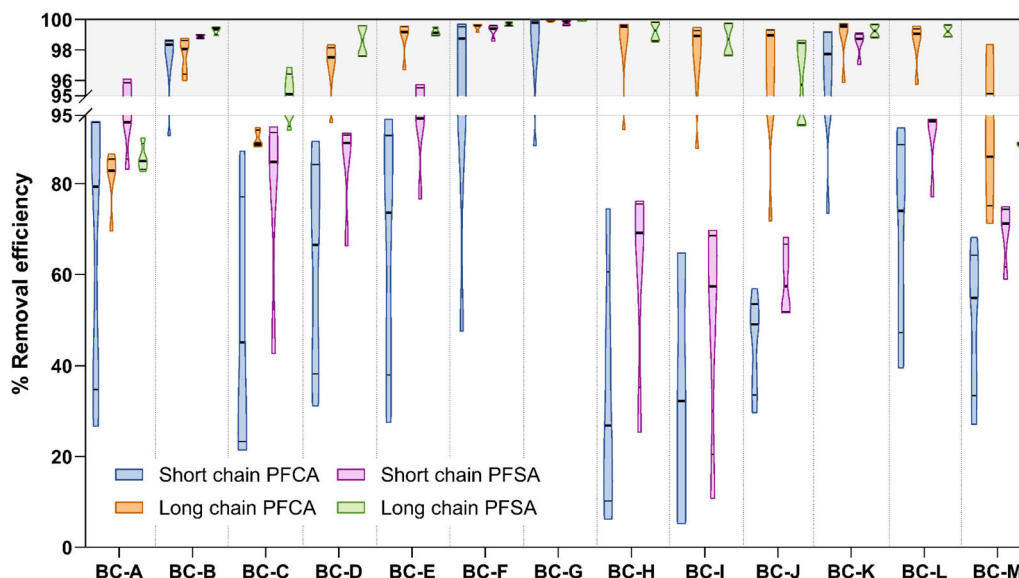


FIGURE 2 | Individual removal efficiencies in batch sorption experiments grouped by the length of the alkyl chain. Short-chain PFCA: PFBA, PFPeA, PFHxA, PFHpA, and PFOA; long-chain PFCA: PFNA, PFUnDA, PFDoDA, and PFTeDA; short-chain PFSA: PFBS, LPPHxS, BPPHxS, and 6:2-FTSA; and long-chain PFSA: LFOSA, BFOSA, LPFOS, and BPFOS. [Color figure can be viewed at [wileyonlinelibrary.com](https://onlinelibrary.wiley.com)]

coexisting long-chain PFAS can inhibit the sorption of short-chain PFAS in granular activated carbon by competing for the sites and pores (Zhang et al. 2023). However, recent studies have also identified that tailoring biochar materials for PFAS removal could help improve their sorption capacity. Surface modifications, such as the inclusion of ferric ions on the biochar surface to generate positively charged moieties, promote electrostatic attraction to the negatively charged carboxylic or sulfonic functional groups of PFAS (Liang et al. 2024). Additionally, use of biochar with larger pore-filling capacity has also demonstrated potential to improve effectiveness in PFAS remediation (Liang et al. 2024). In any case, additional research to evaluate the individual sorption of PFAS onto biochar would be needed to clearly identify certain trends.

From another perspective, and considering the differences in RE for the different materials, physicochemical characterization of the biochar material surface was conducted. To this end, B.E.T. surface area (external, microporous, and total) and average surface pore size analysis was carried out. In general, large differences can be observed in the total B.E.T. surface area of the tested materials, ranging from 14.4 to 335 m²/g (Table 1). Although other studies have found a correlation between the material surface area and RE in carbon-based materials (Sørmo et al. 2021; Saeidi et al. 2020; Krahn et al. 2023), no such conclusions could be drawn from the data presented herein. As reported in the above-mentioned studies, the larger the surface area, the better the RE; however, in the static flow experiments, biochar materials with similar RE rates such as BC-B and BC-K showed very different surface area values: 74.7 and 262 m²/g, respectively. Similarly, no trend could be observed for the average adsorption pore size measurement. Several studies have pointed out competition effects for effective sorption sites in environmental samples (Liang et al. 2024; Krebsbach et al. 2023). In the present study, real stormwater was used for the experiments and, therefore, coexisting contaminants in the matrix as well as stormwater ionic strength could have influenced PFAS sorption

into the biochar and its correlation with the sorbent physicochemical properties. However, it should also be noted at this point that the static flow experiments were conducted for a period of 7 days and, as a consequence, the chemical sorption equilibrium might not have been attained for all materials, as it heavily depends on the material-analyte composition. In any case, material characterizations and the data collected from 7-day experiments provided an estimation of the sorption capabilities for application in dynamic flow experiments.

3.2 | Columns' Sorption Experiments

Dynamic flow column tests were conducted to evaluate the sorption capacity and estimate the potential long-term efficiency of biochar to remediate PFAS from contaminated stormwater. To this end, proper tracing of the breakthrough curve was needed. Although other studies sampled the column outlet at equally distributed times or at a given time span throughout the experiment (Niarchos et al. 2022; Høisæter and Breedveld 2022), for this experiment, we decided to conduct high-resolution sampling of the column outlets to ensure comprehensive tracing of the breakthrough as described in Section 2.5.

Figure 3 shows the corresponding breakthrough curves for the individual PFAS (PFBA, PFPeA, PFHxA, PFHpA, PFOA, PFNA, PFDA, PFBS, LPPHxS, BPPHxS, LPFOS, FOSA, and 6:2 FTSA), except for PFUnDA, PFDoDA, and PFTeDA, which had to be excluded from the analysis because of sorption to the material used for the experiment resulting in low concentrations even in the control columns. In addition, BPFOS showed inconsistent data, most probably due to being heavily impacted by matrix interferences and also due to the fact that this branched isomer was present as an impurity from the PFOS reference standard. Thus, BPFOS breakthrough curves could not be obtained and, consequently, no conclusions could be drawn for

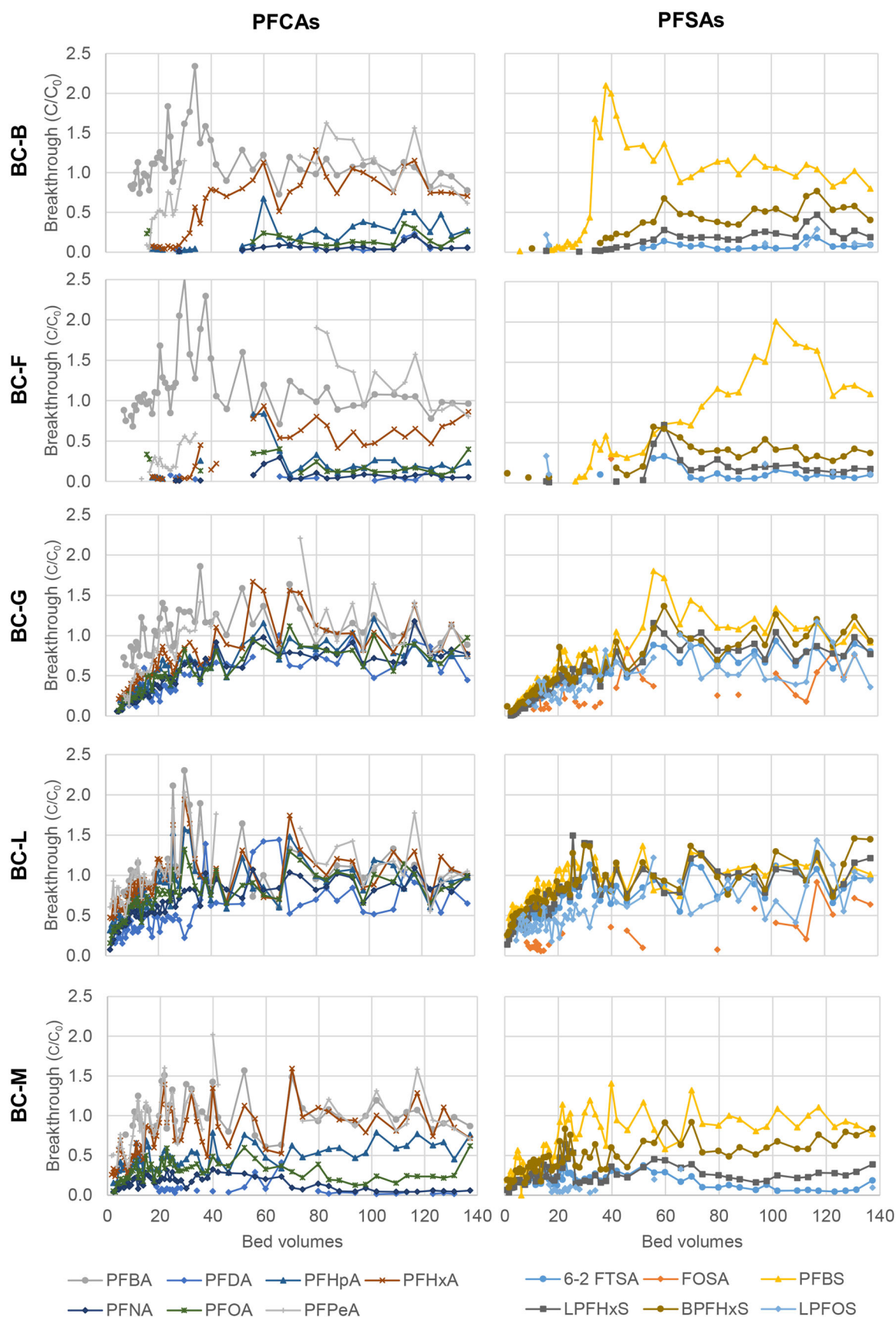


FIGURE 3 | Breakthrough curves for the studied biochar-PFAS pairs. PFCAs are shown on the left panel and PFSA are shown on the right panel. Each horizontal alignment of the figures corresponds to the same biochar material. [Color figure can be viewed at [wileyonlinelibrary.com](https://onlinelibrary.wiley.com)]

this specific compound. In general, the rapid breakthrough of short-chain PFAS such as PFBA or PFBS should be highlighted (Figure 3). Although biochar materials sorbed PFBS for a longer time than PFBA, the latter was almost breaking through from the very beginning of the experiment in all biochar materials tested. Similar observations were reported by Yu et al. (2009), indicating that the presence of a sulfonic group on PFBS results in a longer sorption time compared to PFBA (Yu et al. 2009). Additionally, these results align with the observations from the batch experiments, in which both PFBA and PFBS were not efficiently removed from the stormwater, as well as other studies in which PFBA and PFBS were poorly remediated using carbon-based materials (Zhang et al. 2021; Shahrokhi et al. 2024). On the contrary, other PFAS, such as PFHpA, PFOA, PFNA, PFDA, 6:2 FTSA, LPFHxS, and BPFHxS, were efficiently removed by some of the biochar materials, even reaching an equilibrium state in which changes were no longer observed in the RE, for example, 6:2 FTSA, LPFHxS, and BPFHxS in BC-F, reaching a plateau RE at approximately 70 BV. However, for the other compounds, such as FOSA or LPFOS, the scarcity of data points did not allow an evaluation of their RE.

On comparing the different biochar materials, BC-L clearly stood out due to a quicker breakthrough and less efficient performance, with the vast majority of PFAS rapidly escalating to $C/C_0 \approx 1$. On the contrary, BC-B and BC-F performed most efficiently in terms of PFAS removal and a steady state was reached at approximately 60–80 BV for most PFAS. Additionally, BC-M showed unexpectedly good removal of PFAS (e.g., PFOA, PFNA, 6:2 FTSA, and LPFHxS), although these PFAS were not as effectively removed in the batch tests. The good performance could be explained by the material of the biochar (sewage sludge), which was different compared to the other biochar materials used for the column tests, which were mainly plant-based materials.

In order to better estimate the RE, logarithmic curves were fitted for the individual pairs' biochar–PFAS data (Supporting Information S1: Table S2, Supporting Information S1: Figure S2), and the BV required to achieve an 80% removal (BV_{80}) was calculated (Table 2). In many cases, the lack of sufficient data points hindered proper logarithmic curve fitting and hence the calculation of BV_{80} (indicated as noncalculable (n.c.) in Table 2 for data transparency). However, for those cases in which the calculated BV_{80} exceeded the interpolation range, the value was converted into > 140 or even < 1 for those cases in which breakthrough occurred at beginning of the experiment. Yet, it is clear from Table 2 as well as Figure 3 that the biochar material with the most efficient and sustained sorption capacity for the studied PFAS was BC-F. For this material, many PFAS could not be detected and, if detected,

the BV_{80} was among the highest calculated values. From another perspective, BV_{80} could be interpolated only in the cases of BC-G and BC-L in the whole series of PFCA. Yet, Figure 4 clearly shows how the efficiency of remediation of the PFCA by these materials increased with the number of carbons (nC) in the alkyl chain, which is also in line with the findings in the batch experiments and previous studies (McCleaf et al. 2017).

4 | Conclusions

Overall, several biochar materials with significant differences in their physico-chemical surface characteristics were tested for the static and dynamic flow remediation of PFAS from contaminated stormwater. Although other studies have also focused on assessing sorption materials, this study is, to the best of the authors' knowledge, the first to test 13 different biochar materials in actual stormwater. The static flow experiments revealed biochar as a promising alternative to mitigate PFAS from contaminated stormwater, with some biochar sorbents showing removal efficiencies of up to 99% in 7 days. Long-chain PFAS were more efficiently removed from the aquatic media than short-chain PFAS.

For the dynamic flow system, the performance of the biochar materials was shown to differ somewhat from the static portion of the study. Although some biochar rapidly showed breakthrough in most of the PFAS studies, other materials, such as sludge biochar, stood out as highly efficient in the long-term dynamic system evaluation. For the behavior of the different PFAS, similar results as in the static experiment were observed.

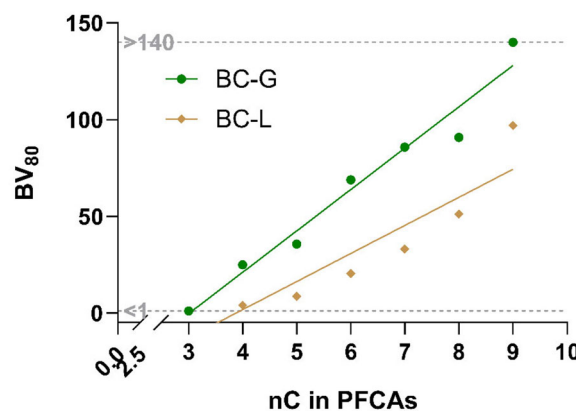


FIGURE 4 | Comparison of BV required to achieve 80% removal (BV_{80}) for the series of studied PFCA in BC-G and BC-L, respectively. [Color figure can be viewed at [wileyonlinelibrary.com](https://onlinelibrary.wiley.com)]

TABLE 2 | Estimated bed volumes (BV) for 80% breakthrough.

	PFBA	PFPeA	PFHxA	PFHpA	PFOA	PFNA	PFDA	PFBS	LPFHxS	BPFHxS	LPFOS	6:2 FTSA	FOSA
BC-B	< 1	45	n.c.	n.c.	n.c.	n.c.	n.c.	46	> 140	> 140	n.c.	n.c.	n.c.
BC-F	< 1	53	n.c.	n.c.	n.c.	n.c.	n.c.	60	n.c.	n.c.	n.c.	n.c.	n.c.
BC-G	< 1	25	36	69	86	91	> 140	34	81	55	> 140	104	n.c.
BC-L	< 1	4.1	8.7	20	33	51	97	13	26	19	92	46	n.c.
BC-M	< 1	9.4	29	> 140	> 140	n.c.	n.c.	38	> 140	> 140	n.c.	n.c.	n.c.

Note: Biochar–PFAS cases in which no curve could be interpolated (i.e., values could not be estimated) are indicated as noncalculable (n.c.); cases in which the calculated value was beyond the interpolation range are indicated as > 140 (maximum BV tested); and < 1 indicates breakthrough observed from the beginning of the experiment.

Short-chain PFAS usually broke through at an earlier stage, tending to remain in the stormwater. However, dynamic flow experiments showed efficient removal of long-chain PFAS even after 3 months of constant operation. Yet, further investigations under equilibrium conditions are needed to elucidate the mechanisms of PFAS adsorption onto biochar materials. Additionally, tailored biochar materials designed for enhanced PFAS remediation are expected to achieve increased PFAS removal efficiencies while maintaining cost-effectiveness. Overall, plant- and sewage-based biochar materials, a green and sustainable treatment strategy, have been proven to be promising materials for the removal of PFAS from contaminated stormwater.

Author Contributions

Alberto Celma: conceptualization, investigation, formal analysis, data curation, writing – original draft. **Aleksandra Skrobonja:** investigation, formal analysis, data curation, writing – review and editing. **Santosh Govind Khokarale:** investigation, formal analysis, writing – review and editing. **Jyri-Pekka Mikkola:** investigation, writing – review and editing. **Erlend Sørmo:** investigation, writing – review and editing. **Gerard Cornelissen:** investigation, writing – review and editing. **Karin Wiberg:** conceptualization, funding acquisition, writing – review and editing. **Lutz Ahrens:** conceptualization, investigation, data curation, funding acquisition, writing – review and editing.

Acknowledgments

The authors acknowledge the substantial assistance provided by Marija Lukaric (SLU) during the dynamic flow column test in sample collection, as well as Anna-Karin Dahlberg and Johan Eriksson for their efforts toward funding acquisition and for discussions during the duration of the project. Also, the authors thank the collaborators from the Norwegian Geological Institute, Skanska Sverige (Stockholm, Sweden), as well as the different participants of the Rest-to-Best project for their kind donations of biochar materials. This study was financially supported by the Swedish Research Council for Sustainable Development FORMAS (project number 2020-01107, SAFESTORM) and the project “Residues for best use – Biochar as solution and product in the circular and climate positive society (Rest-to-Best)” funded by the Swedish Innovation Agency (Vinnova) (project number 2021-01589). Aleksandra Skrobonja acknowledges the Ministry of Science, Technological Development and Innovation, Republic of Serbia (Contract number 451-03-66/2024-03/200017). The authors also wish to thank Bio4Energy, a strategic research environment appointed by the Swedish government and the Wallenberg Wood Science Center under the auspices of the Alice and Knut Wallenberg Foundation. This study is also related to the activities of the Johan Gadolin Process Chemistry Centre at Åbo Akademi University.

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Supporting Information

Additional supporting information can be found online in the Supporting Information section.
Celma et al. PFAS columns_SI.