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Removal of pharmaceutical active compounds in multi-module biochar filter (MmBF) for post-septic tank treatment

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ABSTRACT

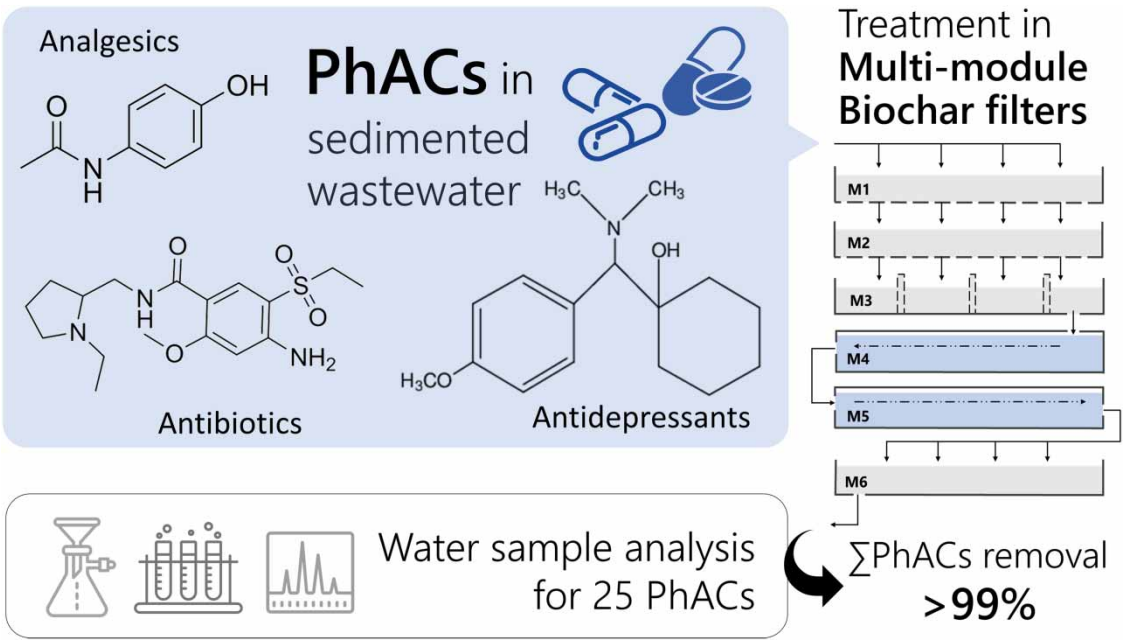
Pharmaceutically active compounds (PhACs) in wastewater pose significant environmental risks due to their persistence and potential to disrupt aquatic ecosystems. Onsite wastewater treatment systems (OWTS) often fail to adequately remove PhACs. This study investigated the efficiency of a multi-module biochar filter (MmBF) system as secondary treatment in OWTS for removing PhACs. Two parallel MmBF systems, each comprising six sequential modules filled with biochar, were evaluated for their removal of 25 detected PhACs across multiple pharmaceutical classes. The MmBFs were operated with municipal wastewater as influent for over one year, after which wastewater samples were collected and analysed from the influent and effluent of each module. The MmBFs showed consistent reduction in the aggregated PhACs concentrations by >99% over several sampling occasions, with only six PhACs having less than 95% removal. The first aerobic module M1 contributed to more than 92% of the total removal. The subsequent aerobic modules (M2–M3) provided additional reduction, resulting in over 98% of PhACs removal in the aerobic modules. In contrast, the anoxic modules (M4–M5) had a lower overall contribution, but the removal of specific compounds was observed, suggesting potential anaerobic degradation. This study demonstrates the potential of biochar-based systems as a sustainable option for OWTSs.

Key words: biochar, micropollutants, multi-module biochar filter, onsite wastewater treatment systems, pharmaceutically active compounds

HIGHLIGHTS

- Multi-module biochar filter achieved >99% PhACs removal from municipal wastewater.
- First aerobic module (M1) contributed over 92% of the total PhACs removal.
- Sequential aerobic–anoxic modules showed potential for PhACs biodegradation.
- Sample analysis after one year of operation indicated long-term MmBF stability.

GRAPHICAL ABSTRACT



1. INTRODUCTION

Pharmaceuticals and personal care products (PPCPs) are ubiquitous in our daily lives and are routinely discharged into wastewater streams. Upon entering wastewater treatment systems, PPCPs persist and can accumulate (Salgado Costa *et al.* 2023). Research has shown that PPCPs, even at low concentrations, can have negative effects on both aquatic ecosystems and human health (Gonzalez-Rey & Bebianno 2012; Reyes *et al.* 2021). These compounds have the potential to disrupt the endocrine system in organisms and contribute to the development of antibiotic resistance (Ebele *et al.* 2017). A Swedish case study by Björklund & Svahn (2021) found that municipal wastewater treatment plants (WWTPs) discharge about 71 kg of pharmaceutical active compounds (PhACs) annually, including metoprolol, diclofenac, and carbamazepine, into natural recipients.

A range of PhACs, including antibiotics, analgesics, antimicrobials, pharmaceuticals, hormones, and phenolic substances, has been detected in the effluent from onsite wastewater treatment systems (OWTSs) (Ejhed *et al.* 2018), with some compounds, such as simvastatin and estradiol, exceeding Swedish freshwater quality standards and posing risks to aquatic life (Fick *et al.* 2010; SwAM 2019). These findings align with previous studies that have identified OWTSs as significant sources of emerging organic contaminants in groundwater and surface water, posing environmental and human health risks (Gao *et al.* 2019; Hägglund 2021). For instance, Godfrey *et al.* (2007) detected carbamazepine, sulfamethoxazole, and nicotine in septic effluent and the underlying aquifer in Montana, USA. Similarly, Schaider *et al.* (2016) found higher concentrations of organic wastewater compounds, including sulfamethoxazole and carbamazepine, in Massachusetts drinking water wells near OWTSs. Gao *et al.* (2019) also reported 44 detected pharmaceuticals in Swedish groundwater near soil infiltration systems.

The importance of this matter has been raised in the European Green Deal (European Commission 2021), where the European Commission proposes a recast of the Urban Wastewater Treatment Directive to strengthen micropollutant removal requirements (European Commission 1991). The new directive mandates >80% removal for 12 PhACs that pose environmental risks even at low concentrations. These include amisulpride, carbamazepine, citalopram, clarithromycin, diclofenac, hydrochlorothiazide, metoprolol, venlafaxine, benzotriazole, candesartan, irbesartan, and a mixture of 4-methylbenzotriazole and 5-methylbenzotriazole (European Parliament and Council 2024). While these regulations primarily target large-scale WWTPs, improving PhACs removal efficiency in OWTSs remains equally crucial to limit environmental contamination from decentralized sources.

In the onsite wastewater treatment process, PhACs are mostly removed by adsorption and biodegradation (Nguyen *et al.* 2024). In nature-based systems, such as constructed wetlands, biodegradation is often part of a larger bioremediation process,

where plants and the surrounding ecosystem further enhance pollutant removal (Zhang *et al.* 2023). OWTSs with septic tanks and soil infiltration fields usually remove PhACs by adsorption and aerobic biodegradation in a soil infiltration field, which could be similar in function to the activated sludge and other biological treatment in WWTPs (Schaider *et al.* 2017). The extended hydraulic retention time (HRT) promotes the biodegradation of PhACs (Ejhed *et al.* 2018). Clyde *et al.* (2021) reported similar or higher removal efficiencies of PPCPs in nitrogen-removing biofilters with longer HRT compared with conventional OWTSs. Furthermore, the removal of certain PPCPs was greater when treated in both aerobic and anoxic parts of the biofilters, indicating various mechanisms contributing to overall performance (Clyde *et al.* 2021). While aerobic conditions generally support higher degradation rates, certain organic micropollutants have shown potential for biotransformation under anoxic conditions as well, likely depending on compound characteristics and biomass composition (Burzio *et al.* 2022).

Conventional OWTSs rely on natural soil filtration. Their performance is influenced by soil properties, hydraulic loading rates, and biological processes that can lead to clogging, affecting retention time and limiting adsorption capacity (McKinley & Siegrist 2011; Baykuş *et al.* 2021). To address these challenges, alternative filter media are needed to enhance treatment performance and long-term system reliability. While WWTPs use costly materials like activated carbon to remove PhACs (Kårelid *et al.* 2017; Cai *et al.* 2018), OWTSs require low-cost, sustainable alternatives. Biochar is a carbon-rich material with a high specific surface area due to its porous structure (Anand *et al.* 2023). Together with its range of surface functional groups (e.g., –OH, –C–O, –C = O, –COOH), this makes biochar capable of adsorbing various pollutants, including heavy metals, organic pollutants, and nutrients, from wastewater (Afrooz & Boehm 2016; Chang *et al.* 2021; Xu *et al.* 2024). Ion exchange or electrostatic interactions, driven by the charge from functional groups, and π–π interactions between PhACs and biochar's aromaticity have been shown to influence the removal efficiency (Hamadeen & Elkhatib 2022). The production method, feedstock type, and pyrolysis conditions significantly affect the biochar's properties and its efficiency in water treatment (Anand *et al.* 2023). Dalahmeh *et al.* (2018) found high removal of carbamazepine and metoprolol in biochar filters due to adsorption, while metoprolol was poorly removed in sand filters. This study indicates that biochar might be a suitable filter material for the removal of PhACs in small and onsite systems. However, long-term evaluations of unmodified biochar systems under extended, pilot-scale conditions remain scarce. Most existing studies use short-term setups, small-scale columns, or modified biochar. This study addresses that gap by assessing the removal efficiency of 25 PhACs in a multi-module, unmodified biochar filter (MmBF) over a full year of operation. It builds on earlier work that demonstrated the MmBF's potential for nutrient and organic matter removal (Shigei *et al.* 2024), expanding its evaluation to include micropollutant removal and module-specific performance.

In the current study, we investigated the removal of PhACs using a pilot-scale MmBF system having four aerobic and two anoxic modules (Shigei *et al.* 2024). This study builds on previous research about the application of the MmBFs for the removal of organic matter, nitrogen, phosphorus, and *Escherichia coli* (*E. coli*) as an alternative OWTS (Shigei *et al.* 2024). Two MmBF systems receiving wastewater after primary treatment were operated for more than one year, and the removal of PhACs present in the wastewater was evaluated to (i) assess the removal efficiency of the MmBF system after extended operation time (270–367 days) and (ii) examine the roles of the six modules for the removal. We hypothesize that the sequential aerobic and anoxic modules in the MmBF would facilitate PhACs removal, with the aerobic modules (M1–M3) providing higher removal efficiencies through aerobic degradation and adsorption on biochar. In contrast, the anoxic modules (M4–M5) and the final aerobic module (M6) may facilitate reductive transformations of certain compounds.

2. METHODS

2.1. Filter operation and sample collection

Two identical MmBFs were used, each with six polyvinyl chloride plastic modules (36 × 56 × 20 cm). Modules M1–M5 contained biochar, while M6 had a layer of pine bark on top of the biochar, all media layers were 15–17 cm height. The modules M1–M3, and M6 had aerobic conditions with unsaturated downflow. The modules M4–M5 have anoxic conditions with saturated horizontal-flow mode, to promote denitrification (Figure 1, Supplementary Table S1). The detailed setup and operational conditions of the MmBFs are described by Shigei *et al.* (2024). The MmBFs were installed at the Uppsala wastewater treatment plant (Kungsängsverket) to access real wastewater and were operated continuously with intermittent loading from 25 January 2021 to 16 February 2022 (381 days). The hydraulic loading rate (HLR) applied to each MmBF was 50 L m^{-2} day^{-1}, with an organic loading rate of 36 g COD m^{-2} day^{-1}.

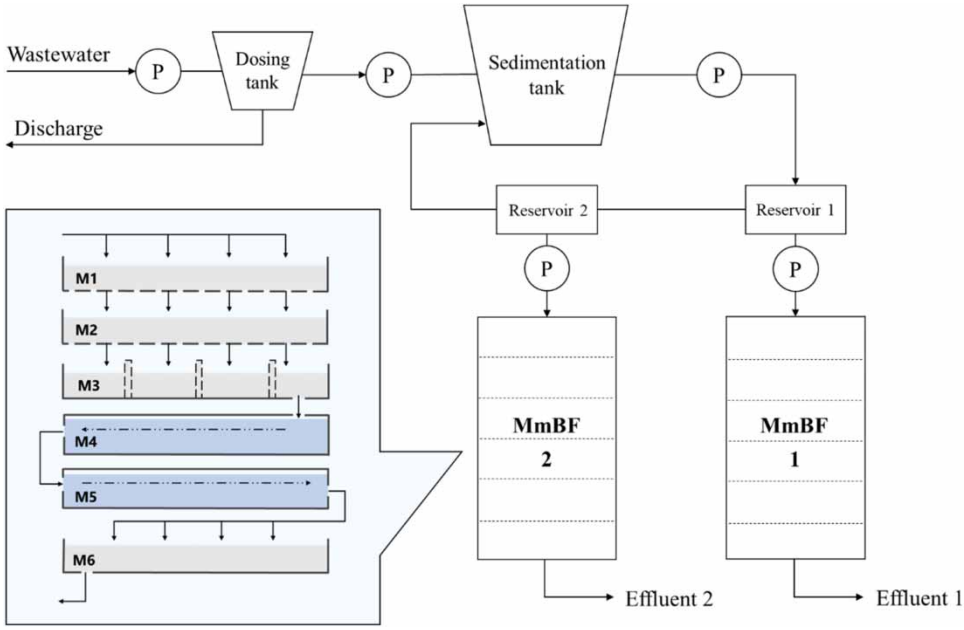


Figure 1 | Schematic diagram of the multi-module biochar filters.

Regarding the PhACs analyses, approximately 400 mL of influent and effluent from two MmBFs were collected in plastic containers during four sampling campaigns from 29 October 2021 to 3 February 2022 (total $n = 8$). The effluent from each module of M1–M5 was collected twice from the two MmBFs, at the beginning and end of each sampling campaign (total $n = 19$). All samples were stored in the freezer at −20 °C until analyses. The samples collected for the PhACs analyses represented a period of stable operation.

2.2. Pharmaceutically active compounds

A set of 30 pharmaceutical compounds covering a wide range of therapeutic effects was analysed. The therapeutic effects and corresponding substances included: analgesics (diclofenac, ibuprofen, naproxen, paracetamol, tramadol), antibiotics (amisulpride, amoxicillin, ciprofloxacin, clarithromycin, erythromycin, sulfamethoxazole, trimethoprim), antidepressants (carbamazepine, citalopram, oxazepam, sertraline, venlafaxine, zolpidem), antifungals (fluconazole, ketoconazole), antihypertensives (atenolol, hydrochlorothiazide, irbesartan, losartan, metoprolol, propranolol), disinfectants (benzotriazole, Σ(6- and 4-methylbenzotriazole)), diuretics (furosemide), and immunosuppressive (methotrexate). The list of substances and their properties is shown in Supplementary Table S2.

2.2.1. Extraction of PhACs

Wastewater samples were thawed at room temperature overnight and then filtered using glass microfiber filters (0.7 μm). Sample preparation included weighing an empty bottle, adding approximately 250 mL of the sample, spiking it with 50 μL of a 1 ng μL^{-1} of isotopically labelled internal standard solution (Supplementary Table S3), and adding 30 mL of 0.1 M Na_2EDTA solution. The pH of the sample was adjusted to 3.0 using sulphuric acid. Samples were extracted on hydrophilic-lipophilic balance (HLB) solid-phase extraction (SPE) cartridges (Oasis HLB cartridges; 200 mg, 6 cc; Waters Corporation, Manchester, UK), which were conditioned with 6 mL of methanol followed by 6 mL of Milli-Q water. Samples were added gradually to ensure a slow passage through the cartridges, followed by rinsing with 6 mL Milli-Q water and drying by using a vacuum. Extracts were eluted into a glass tube with 4 mL methanol twice, which was passed slowly through the cartridges. The methanol extracts were concentrated under a gentle nitrogen gas stream to a small volume (<2 mL) and transferred to 2 mL vials. The extracts were evaporated to dryness in the vials and then reconstituted in a 1 mL methanol–water mixture (1:9, v/v). The detailed protocol follows the methodology described by Dalahmeh *et al.* (2018, 2019).

2.2.2. Instrumental analyses

The PhACs were analysed by IVL Swedish Environmental Research Institute in Stockholm using high-performance liquid chromatography coupled to a triple quadrupole mass spectrometer (MS/MS) equipped with a Shimadzu electrospray ionization source (ESI). The analysis was performed in multiple ion monitoring (MRM) mode with a capillary voltage of 3 kV using both positive and negative ionization. Chromatographic separation of the analytes was carried out on a biphenyl core–shell column (3.0 × 100 mm, 2.6 μ, 100 Å) from Phenomenex using a gradient elution programme. The mobile phases consisted of (A) Milli-Q water with 2 mM acetic acid, which helps maintain pH stability and improve ionization efficiency in ESI-MS analysis, and (B) methanol, pumped at a flow rate of 0.4 mL min^{-1} at 40 °C. The gradient started at 5% B with a steep increase to 70% B within 1 min, followed by a second increase to 95% B after 7 min, which was held for 3 min. After plateauing at 95% B, the gradient dropped to 5% B after 10 min, followed by an equilibration period at 5% B for 1.9 min. An injection volume of 1 μL was used, and each transition was scanned for 5 s. The samples were kept at 5 °C in the autosampler during analysis. Analyte concentration was quantified using a 7-point calibration curve (500, 200, 100, 50, 20, 10, and 5 ng mL^{-1}). The mass to charge ratios (m/z) of the precursor, fragment ions, and retention times for each compound are listed in Supplementary Table S5. All analyte concentrations in the samples were subtracted from the blank value and corrected for recovery (Supplementary Table S6).

2.3. Quality control

All compounds were identified with two transition ions except for ibuprofen. Limit of detection (LOD) and limit of quantification (LOQ) were estimated from 3× and 10× the noise level quantified in the blank sample. The accuracy of the quantification was estimated from a control sample spiked with 100 ng of analyte standards, analysed together with the samples. The accuracy of the quantification ranged from 78 to 100% for all compounds except for ciprofloxacin, which had an accuracy of 48%. The linearity of the calibration curves was evaluated for all compounds that had $R^2 > 0.99$. The recovery of the extraction protocol was assessed by spiking 100 ng of analyte standards mix of PhACs (Supplementary Table S4) into 200 mL of Milli-Q water and processing it using the same procedure as the actual samples, including filtration, SPE, evaporation to dryness, and reconstitution. The recovery ranged from 68 to 126% (Supplementary Table S6). However, five compounds (amoxicillin, atenolol, ciproflaxacin, erythromycin, and ketoconazole) exhibited poor recovery (<60%) and were excluded from further analysis to ensure data reliability. The remaining 25 PhACs belong to diverse pharmaceutical classes and physicochemical properties. While the exclusion of some compounds may limit the comprehensiveness of the study, the retained set still supports the generalizability of the findings to commonly encountered PhACs in municipal wastewater.

2.4. Calculations and statistical analysis

A two-tailed t-test (95% confidence level) was used to compare the effluent PhACs concentrations between the two filters to assess whether they had a significant difference in treatment performance. Normality was assessed using Q-Q plots and the Shapiro–Wilk test, with 10 compounds showing deviations from normality. Given the small sample size ($n = 4$ per test), statistical power was limited.

The mass of PhACs retained in the biochar in MmBFs was estimated for the entire modules and each module by the following formula:

$$\Delta q = \frac{\{(C_0 - C_e) \times Q\}}{m} \times t$$

where Δq is the amount of PhACs removed in the biochar between the sampling events (mg g^{-1}), C_0 is the influent concentration of PhACs (mg L^{-1}) to the pilot or its modules, C_e is the effluent concentration of PhACs (mg L^{-1}) to the pilots or its modules, Q is the flow rate (L day^{-1}), t is the number of days since the last sampling event (day), and m is the mass of the biochar (g).

The cumulative retention of PhACs over the entire duration of filter operation = $\Sigma\Delta q$ for all PhACs.

Since the samples were taken after long-term operation (270 days) of MmBFs, this amount (Δq) retained on the biochar for different sampling times does not account for non-equilibrium conditions, which could be present in pilot-scale MmBFs

operation, where varying PhACs concentrations can impact adsorption efficiency. It is important to note that this estimation is an approximation for the MmBFs performance of PhACs removal.

2.5. Kinetic model fitting

To analyse the adsorption kinetics of PhACs in the MmBFs, concentration data through the modules were fitted to pseudo-first-order and pseudo-second-order kinetic models. These models describe the rate of PhACs removal as wastewater passes through the system, rather than equilibrium adsorption behaviour.

Pseudo-first-order model:

$$C_t = C_0 e^{-k_1 t}$$

where C_t is the effluent concentration at module t, C_0 is the influent concentration, and k_1 is the first-order rate constant.

Pseudo-second-order model:

$$\frac{1}{C_t} = \frac{1}{C_0} + k_2 t$$

where k_2 is the second-order rate constant.

Pseudo-kinetic models were chosen because the influent concentrations change progressively across sequential treatment modules, which reflect a dynamic removal process rather than an equilibrium adsorption state. Langmuir and Freundlich adsorption isotherms typically require batch experiments, where equilibrium concentrations are measured after a fixed contact time for different initial concentrations (Ho & McKay 1999). Since the MmBF system operates under continuous flow conditions with changing influent concentrations, kinetic modelling was more appropriate. A plug flow assumption was applied in the kinetic model to represent directional flow through the sequential modules. Although this is a simplification, a tracer test conducted in a previous study (Shigei *et al.* 2024) reported average HRTs of 168 h (M1–M3), 21 h (M4–M5), and 135 h (M6), indicating relatively stable and sequential flow behaviour across the functional zones. This supports the validity of the plug flow approximation used in the modelling.

Non-linear regression was performed in RStudio using the nlsLM function from the minpack.lm package, and the best-fit model was determined based on the coefficient of determination (R^2).

3. RESULTS

3.1. Concentration and overall removal of PhACs

The filters MmBF1 and MmBF2 demonstrated effective removal of conventional wastewater pollutants, including COD (94–95%), Inorganic Nitrogen (57–86%), and *E. coli* (1.2–2.0 $\log_{10}$ reduction) (Supplementary Table S7). The influent of the MmBF presents a diverse range of pharmaceutical residues where the total concentration of all PhACs ($\sum$PhACs) was 1.6×10^6 ng L^{-1} (see Table 1 for the data with SD). Among these, certain compounds were found in high concentrations, such as paracetamol (7.4×10^5 ng L^{-1}), losartan (4.1×10^5 ng L^{-1}), and naproxen (1.7×10^5 ng L^{-1}). In contrast, zolpidem and methotrexate were detected at lower concentrations of 1.3×10^2 and 1.2×10^2 ng L^{-1}, respectively.

Cumulatively, the MmBF system decreased the $\sum$PhACs in the influent from 1.6×10^6 to 5.2×10^3 ng L^{-1} (Table 1). These reductions led to an overall average removal efficiency of 99.7% in MmBF1 and MmBF2 (Table 1). Statistical analysis showed no significant differences in effluent PhACs concentrations between MmBF1 and MmBF2 ($p > 0.05$). The distribution of 10 compounds deviated from normal distribution, but results remained consistent across tests (Supplementary Table S8). The total amount of PhACs removed on biochar was 1.6×10^5 ng g^{-1} at the first sampling (Day 270) and 2.1×10^5 ng g^{-1} at the final sampling (Day 367).

Among the analgesics, paracetamol concentrations decreased dramatically from 7.4×10^5 to 1.3×10^2 ng L^{-1}, with a removal efficiency surpassing 99% (Table 1 and Figure 2 analgesics). For the antihypertensives, naproxen also exceeded 99% removal. Losartan demonstrated a comparable removal efficiency, exceeding 99% from an initial 4.1×10^5 ng L^{-1} to the effluent concentration of 5.9×10^2 ng L^{-1} (Table 1 and Figure 2 antihypertensive). Among the antidepressants (Figure 2), carbamazepine showed slightly lower removal efficiency, at 96 and 97% in MmBF1 and MmBF2, respectively (Supplementary Table S9). Notably, antibiotic compounds (Figure 2) were present in low initial concentrations in the influent and

Table 1 | Mean concentration ($\times 10^2$ ng L^{-1}) ± standard deviation (SD) of the 25 PhACs in the influent ($n = 4$) and effluent ($n = 8$) of MmBF1 and 2, the cumulative retention (ng $g^{-1}_{Biochar}$), and the mean removal efficiency ± SD (%)

PhACs compounds	Mean concentration ± SD ($\times 10^3$ ng L^{-1})		Cumulative retention of PhACs (ng $g^{-1}_{Biochar}$)		Mean removal ± SD (%)
	Influent	Effluent	MmBF1	MmBF2	Both MmBFs
Paracetamol	736 ± 118	0.13 ± 0.13	87,353	87,361	99.98 ± 0.04
Losartan	412 ± 178	0.60 ± 0.36	64,029	64,044	99.8 ± 0.30
Naproxen	171 ± 59.6	0.76 ± 0.64	27,018	27,048	99.4 ± 1.11
Σ (6- and 4-methylbenzotriazole)	46.7 ± 45.2	0.21 ± 0.27	3,858	3,881	98.8 ± 3.24
Metoprolol	44.0 ± 21.5	0.23 ± 0.24	5,384	5,403	99.1 ± 2.21
Tramadol	36.1 ± 29.5	0.40 ± 0.17	4,186	4,195	98.1 ± 2.73
Venlafaxine	31.7 ± 14.1	0.38 ± 0.13	3,779	3,785	98.3 ± 2.47
Citalopram	23.4 ± 18.9	0.02 ± 0.02	2,197	2,245	99.7 ± 0.62
Ibuprofen	17.3 ± 6.3	0.40 ± 0.52	2,811	2,847	97.7 ± 6.12
Oxazepam	16.9 ± 7.1	0.13 ± 0.05	2,127	2,130	98.9 ± 1.49
Amisulpride	10.5 ± 8.1	0.06 ± 0.06	886	891	99.2 ± 1.77
Carbamazepine	10.3 ± 3.0	0.33 ± 0.04	1,535	1,533	96.6 ± 2.77
Propranolol	8.30 ± 7.3	0.01 ± 0.01	611	614	99.7 ± 0.65
Irbesartan	6.24 ± 2.4	0.11 ± 0.03	1,033	1,029	97.9 ± 1.73
Clarithromycin	5.20 ± 3.1	0.06 ± 0.01	956	957	98.3 ± 1.79
Trimethoprim	4.64 ± 1.1	0.04 ± 0.04	399	400	99.1 ± 2.11
Diclofenac	3.30 ± 1.1	0.04 ± 0.02	240	235	98.6 ± 2.18
Sertraline	3.12 ± 2.7	0.003 ± 0.001	4	29	99.8 ± 0.22
Fluconazole	1.90 ± 0.74	0.11 ± 0.02	45	50	93.6 ± 4.41
Sulfamethoxazole	0.39 ± 0.37	0.01 ± 0.001	11	16	96.3 ± 2.14
Furosemide	0.30 ± 0.35	0.05 ± 0.06	17	26	81.2 ± 17.97
Benzotriazole	0.21 ± 0.00	0.01 ± 0.003	746	746	93.0 ± 2.48
Hydrochlorothiazide	0.18 ± 0.08	0.03 ± 0.03	14	19	74.6 ± 50.55
Zolpidem	0.13 ± 0.08	0.001 ± 0.00	1	13	98.7 ± 1.87
Methotrexate	0.12 ± 0.00	0.008 ± 0.001	228	228	93.0 ± 2.48
ΣPhACs	1,590 ± 528.0	4.1 ± 2.7	2.1×10^5	2.1×10^5	99.7 ± 0.3

The removal efficiency (%) for each sampling event was calculated individually based on paired influent and effluent concentrations, and the mean removal efficiency ± SD (%) was determined by averaging these event-specific values.

remained below detection limits in the effluent, which may indicate the MmBF's capacity for antibiotic removal (Figure 2). During the sampling campaigns, stable removal was observed for most substances. However, exceptions were noted with furosemide, hydrochlorothiazide, and ibuprofen, which exhibited unstable removal, resulting in larger standard deviations (Supplementary Table S9). In addition, a reduction in removal efficiency across the four samplings was observed for several compounds (carbamazepine, clarithromycin, diclofenac, fluconazole, furosemide, hydrochlorothiazide, oxazepam, tramadol, venlafaxine, and zolpidem) and, to a lesser extent, for the total PhACs as well (Supplementary Figure S1).

3.2. Contribution of the different modules to the overall removal of ∑PhACs

The aerobic unit, composed of M1–M3, was observed to be the main module removing the ∑PhACs in the MmBF system (>99% contribution to the overall removal). The upper module (M1) demonstrated the largest contribution to the overall removal of ∑PhACs (93 ± 4%; Table 2 and Figure 3), while the subsequent modules, M2 through M6, accounted for <7% of the overall ∑PhACs removal. In M1, the concentration of ∑PhACs decreased significantly from 1.85×10^6 ng L^{-1} in the influent to 1.2×10^5 ng L^{-1} in the effluent. As the concentration of ∑PhACs entering each module decreased, the removal

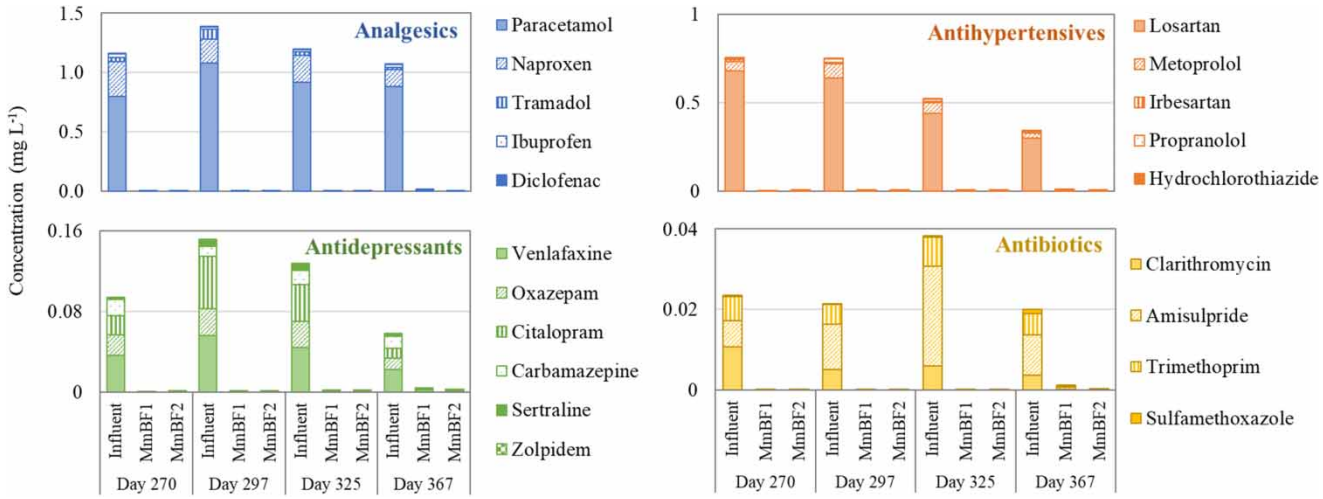


Figure 2 | Concentrations of pharmaceuticals in the influent ($n = 4$) and effluent ($n = 8$) (mg L^{-1}) of MmBF1 and 2 categorized by therapeutic categories: analgesics, antihypertensives, antidepressants, and antibiotics.

Table 2 | The concentration (ng L^{-1}), removal percentage (%), and retention (ng g^{-1}) of ∑PhACs in the modules M1–M6 of the MmBFs

	Concentration of ∑PhACs (ng L^{-1})		Removal of ∑PhACs in relation to PhACs entering each module (%)		Contribution of each module to the overall ∑PhACs removal (%)		Cumulative retention of ∑PhACs (ng g^{-1})	
	Mean	SD	Mean	SD	Mean	SD	Mean	SD
Influent	1.85×10^6	3.9×10^5	–	–	–	–	–	–
M1	1.2×10^5	4.6×10^4	92.8	3.9	92.8	3.9	1.5×10^6	3.8×10^5
M2	6.6×10^4	4.1×10^3	46.6	19.6	4.4	3.6	4.3×10^4	4.5×10^4
M3	2.6×10^4	1.8×10^4	53.7	24.0	2.2	1.2	3.8×10^4	2.0×10^4
M4	2.0×10^4	6.0×10^3	−2.0	63.1	0.3	0.6	5.4×10^4	1.3×10^4
M5	1.3×10^4	7.1×10^3	24.3	70.5	0.3	0.7	7.4×10^3	1.2×10^4
M6 (Effluent)	8.4×10^3	9.7×10^3	31.0	69.9	0.2	0.7	7.9×10^3	2.0×10^4

The mean values are the average of the corresponding data of MmBF1 and 2. The mean removal and contribution values were calculated as the arithmetic mean of the four individual data points obtained from two sampling events for each of the two filters (MmBF1 and MmBF2).

efficiency also declined (Table 2). Modules M2, M3, and M5 contributed further but with smaller concentration differences, removing 47, 54, and 60% of the incoming ∑PhACs, respectively (Table 2). In contrast, M4 showed a negative removal efficiency ($-2.0 \pm 63.0\%$), which is likely due to the transition from aerobic to anoxic conditions interfering with removal processes, compounded by analytical uncertainty or sampling variability. Given the small concentration differences involved and the limited sampling occasions, the negative removal observed in M4 may reflect random variation and should be interpreted with caution. A slight difference in the removal efficiency through the modules was observed for diclofenac (Supplementary Figure S2). As for the contribution of each module to the overall removal, diclofenac showed the highest removal in M1, followed by removal in M4 and M5.

3.3. Removal kinetics of ∑PhACs

To assess the removal behaviour of PhACs across the MmBFs, the influent and effluent concentrations for each module were fitted to pseudo-first-order and pseudo-second-order kinetic models (Supplementary Figure S3). The results indicated that pseudo-first-order kinetics provided the best fit to the observed data, with R^2 values of 0.99 and 0.95 for MmBF1 and

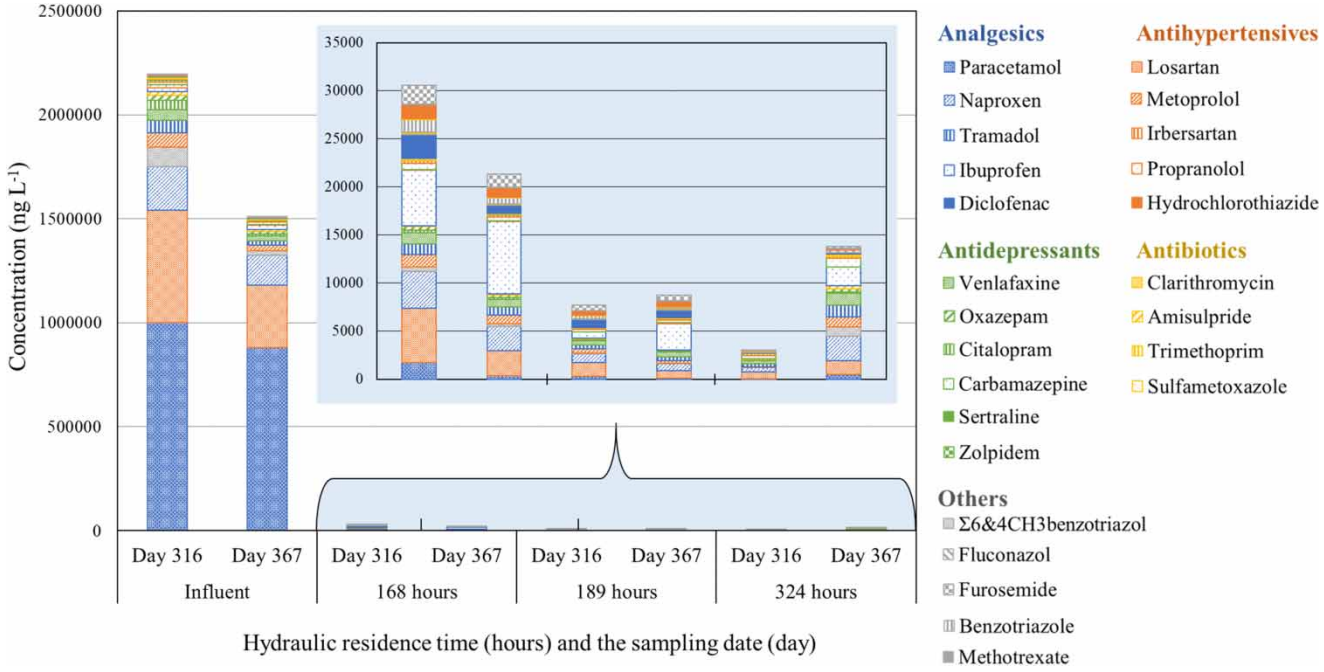


Figure 3 | Effluent PhACs concentrations in each functional unit of the MmBFs, corresponding to the total hydraulic residence time (M1–M3: 168 h, M4–M5: 189 h, M6: 324 h), based on averaged data of MmBF1 and 2 during two sampling events (Day 316 and Day 367).

MmBF2, respectively. The pseudo-second-order model showed lower agreement with the observed values, with R^2 values of 0.96 and 0.88 for MmBF1 and MmBF2, respectively. This suggests that the removal rate primarily depends on the concentration of PhACs in the wastewater rather than being controlled by surface site limitations.

4. DISCUSSION

4.1. Influent quality and PhACs properties

In the present study, 25 out of 30 analytes were detected in the wastewater sample, with analgesics and antihypertensives being predominant. The influent quality in terms of PhACs aligns well with findings from other Swedish WWTPs, as reported by Falås *et al.* (2012), who identified 70 non-antibiotic PhACs with concentrations ranging from 2.0 to 5.2×10^4 ng L^{-1}. Compounds, such as paracetamol, naproxen, and ibuprofen, were among the most abundant compounds in both studies, indicating their widespread use and persistence in wastewater.

According to the comprehensive review by Verlicchi *et al.* (2012) on studies of PhACs in 224 WWTPs, the average influent concentrations of paracetamol (reported as acetaminophen), ibuprofen, and naproxen were 3.8×10^4, 3.7×10^4, and 6.0×10^3 ng L^{-1}, respectively. The influent concentrations observed in our study were slightly higher than these averages but still within the reported range, indicating that compounds present at higher concentrations follow a similar trend across different countries. Consistent with these observations, the influent PhACs profile in our study was similar to that reported for OWTSs across Sweden by Gros *et al.* (2017). Their study detected 44 PhACs in both OWTSs and WWTPs across Sweden, with comparable concentrations, as confirmed by a *t*-test showing no significant difference ($p > 0.05$). In both the present study and the data from Gros *et al.* (2017), compounds, such as losartan, ibuprofen, and diclofenac, were found at concentrations exceeding 10^3 ng L^{-1}. Seasonal variability may also influence wastewater flows and PhACs concentrations and hence system performance. In Sweden, higher concentrations of certain pharmaceuticals – such as analgesics, antihistamines, and antidepressants – have been observed during spring, likely due to seasonal consumption patterns and reduced biodegradation in colder months (Bendz *et al.* 2005; Rehrl *et al.* 2020). Our sampling campaign covered four time points from late autumn to mid-winter (October to February), which partially captures potential seasonal trends. Although no major shifts in influent concentrations or treatment performance were observed during this period, future studies covering a full annual cycle would help assess the impact of broader seasonal variability on system performance.

4.2. Overall performance of the MmBFs in comparison with other OWTSs

Nearly complete removal was observed for the analgesics paracetamol, naproxen, and ibuprofen, demonstrating the MmBF's efficiency in treating these commonly used pharmaceuticals. Their accumulation within the biochar was significant, with amounts of 8.7×10^4 ng g^{-1} for paracetamol, 6.4×10^4 ng g^{-1} for naproxen, and 2.7×10^4 ng g^{-1} for ibuprofen. These results align with previous studies showing high biodegradability of paracetamol, ibuprofen, and naproxen in conventional WWTPs (Verlicchi *et al.* 2012).

Long-term operation of the MmBF, with samples analysed after more than one year, confirmed its operational stability. The minor variations in treatment efficiency between Day 270 and Day 367 suggest that biochar does not require frequent replacement. The extended HRT of 14 days (Shigei *et al.* 2024) may have contributed to enhanced PhACs removal, as extended contact time is known to facilitate adsorption and degradation (Ejhed *et al.* 2018).

The removal efficiency of the MmBF was superior to that observed in constructed wetlands (Ávila *et al.* 2015; Ilyas & van Hullebusch 2020) and slow sand filters (D'Alessio *et al.* 2015). Studies on PhACs removal in constructed wetlands reported efficiencies ranging from 25 to 83%, depending on the compound and system design, while slow sand filters removed less than 10% of certain PhACs, such as carbamazepine and gemfibrozil (Ávila *et al.* 2015; D'Alessio *et al.* 2015; Ilyas & van Hullebusch 2020; Zhang *et al.* 2023). The total PhACs retained on the biochar in this study (210 μg g^{-1}; Supplementary Tables S10 and S11) was similar to the specific adsorption capacity of a pilot-scale experiment using powdered activated carbon (230 μg g^{-1}) (Kårelid *et al.* 2017). Their study analysed 20 compounds and achieved 92–98% removal. The results from the MmBFs were obtained after one year of operation without replacing or backwashing the biochar. The inflow volume of the study by Kårelid *et al.* (2017) (86–113 L h^{-1}) was approximately 200 times higher than that of the MmBF system.

The HLR applied in this study was 50 L m^{-2} day^{-1}, which is within the reported range for OWTS (US EPA 2002). Previous studies have shown that OWTS typically operate at HLRs between 40 and 80 L m^{-2} day^{-1} (Lowe & Siegrist 2008), further supporting the relevance of the applied HLR in this study. Given that similar loading rates have been successfully tested in decentralized wastewater treatment systems, the extended HRT in the MmBF system is unlikely to be a major limitation under real-world conditions. However, further research is needed to evaluate the system's performance under increased HLRs to assess its scalability for larger applications. These findings indicate the great potential of biochar as a long-term solution for wastewater treatment.

In laboratory-scale studies, biochar with various modifications has been investigated and reported to achieve high removal efficiencies (Muter *et al.* 2019). However, to the best of our knowledge, this study is among the first to evaluate the performance of unmodified (raw) biochar in a pilot-scale experiment over an extended period.

4.3. Kinetic models of PhACs removal

Kinetic models revealed that pseudo-first-order kinetics best described PhACs removal in the MmBF system, with R^2 values of 0.99 for MmBF1 and 0.95 for MmBF2. This indicates that removal is primarily concentration-dependent rather than being limited by surface adsorption sites. The strong pseudo-first-order fit suggests that contact time and influent concentration control are critical design factors. However, it is important to note that the kinetic analysis was based on only two sampling occasions per module, conducted on Day 316 and Day 367 of operation. Although both samplings occurred after nearly one year of continuous operation – likely reflecting steady-state conditions – the limited temporal resolution constrains the robustness of the estimated rate constants and may not capture long-term variability or gradual system changes. Therefore, the derived kinetic parameters should be interpreted as indicative, and future studies should include more frequent and extended sampling to improve model reliability.

4.4. Mechanisms of PhACs removal in the MmBFs

The removal of PhACs in the MmBF system is likely governed by a combination of adsorption, microbial biodegradation, and chemical transformation processes (Chaturvedi 2020). While adsorption is likely the primary mechanism, particularly during the initial stages of operation, the consistently high removal efficiencies (>99%) observed during monthly samplings over a 4-month period suggest that biodegradation also plays a significant role in maintaining long-term performance. Adsorption onto the biochar matrix is facilitated by its highly porous structure, which provides a large surface area for PhACs to adhere to, thereby reducing their concentration in the effluent (Al-Gheethi *et al.* 2019). Hydrophobic interactions and electrostatic attraction between PhACs molecules and biochar contribute to the adsorption process, particularly for compounds

with high octanol–water partition coefficients (log K_{ow} >4), which tend to bind more strongly to non-polar surfaces (Matamoros & Salvadó 2013). Higher attenuation (>50%) by hydrophobic contaminants (log K_{ow} >4) through the infiltration system was also observed in the study by Gao *et al.* (2019).

In addition to adsorption, biodegradation by microbial communities colonizing the biochar surface may play a crucial role in PhACs removal, particularly during extended operational periods that allow sufficient time for biofilm development. Certain microorganisms can metabolize PhACs, or even oxidize them completely to CO_2 (Betsholtz *et al.* 2024). Biodegradation is influenced by environmental conditions, particularly the oxygen availability. Aerobic degradation is likely dominant in the first modules (M1–M3), where oxygen is more readily available, while anaerobic transformation pathways may contribute in the later modules (M4–M5). The presence of microbial activity in the biochar system suggests that biodegradation could complement adsorption, potentially extending the biochar's effective lifespan by reducing saturation (Cheng *et al.* 2017). Although microbial community analysis was not conducted in this study, the sequential aerobic–anoxic design of the MmBF, along with evidence of nitrification and denitrification (Shigei *et al.* 2024), suggests microbial contributions to PhACs degradation.

Furthermore, biodegradation within the biochar matrix may involve the transformation of previously adsorbed compounds. Microbial communities can access PhACs not only from the aqueous phase but also ones bound to the biochar surface, metabolizing these substances and potentially freeing up adsorption sites. This process, often referred to as bioregeneration, helps maintain adsorption capacity and delay media saturation (Smolin *et al.* 2020; Zou *et al.* 2024). Such regenerative biodegradation has been observed in similar biofiltration systems, where microbial activity not only reduced the contaminant load but also supported long-term filter performance (Smolin *et al.* 2020). To fully identify the mechanisms occurring in the biochar filters, future work characterizing the microbial community and establishing the roles of biodegradation, adsorption, and bioregeneration by, for instance, ^{14}C-labeled organic micropollutants (Betsholtz *et al.* 2024), would be needed.

Apart from adsorption and biodegradation, abiotic chemical processes, such as redox reactions, may also contribute to PhACs removal. Biochar showed redox-active properties, acting as an electron shuttle that facilitates electron transfer between organic contaminants and redox-active species, such as Fe(III)/Fe(II) or quinon–hydroquinone couples (Yu *et al.* 2015; Deng *et al.* 2022). Some PhACs undergo abiotic transformation through these interactions with biochar surface functional groups or due to redox conditions within the biochar matrix. In this study, redox activity and chemical transformations were not directly measured, nor were significant pH changes observed. However, the sequential aerobic–anoxic conditions within the filter may create localized redox gradients that facilitate such reactions. The observed removal of recalcitrant compounds like diclofenac, especially in anoxic modules, indicates the potential for biodegradation or abiotic transformation.

In summary, the high and sustained PhACs removal efficiencies in the MmBF system likely result from a combination of physical adsorption, microbial biodegradation, and minor contributions from abiotic chemical processes. Adsorption may dominate during early operation, especially for hydrophobic compounds, but microbial processes appear to be key to maintaining long-term removal performance.

4.5. Module-specific removal patterns and variability based on compound properties

The first three modules (M1–M3), operating under aerobic conditions, contributed to over 98% of total removal. The most significant reductions occurred in M1, where higher organic load and aerobic microbial activity (Shigei *et al.* 2024) may have facilitated efficient degradation. Although sludge retention time (SRT) was not measured in this study, the high organic load in M1, combined with the prolonged HRT of 14 days, may have supported microbial processes similar to those associated with longer sludge retention time systems, where increased microbial diversity and the presence of slow-growing microorganisms, such as ammonia-oxidizing bacteria, enhance PhACs degradation (Dawas-Massalha *et al.* 2014; Rattier *et al.* 2014). Interestingly, M4 showed a slight increase (−2% removal) in total PhACs concentration (Table 2), possibly due to desorption of previously adsorbed compounds or shifts in microbial activity. This was followed by a 60% removal in M5, suggesting that anaerobic degradation processes may contribute to PhACs transformation in later modules.

Aerobic or anaerobic preferences vary across compounds (He *et al.* 2018). In this study, most compounds were predominantly removed by aerobic modules M1–M3. However, diclofenac also showed removal in the anoxic modules (M4 and M5, <20%) (Supplementary Figure S2). Its overall removal efficiency across all modules was 99.3 ± 0.6%. Diclofenac, recognized as a compound with low biodegradability and often challenging to remove in conventional WWTPs with activated sludge (29% removal) (Verlicchi *et al.* 2012), was effectively treated in the MmBF system. While sampling in these modules was limited, their effectiveness suggests potential microbial-driven degradation in anoxic conditions.

In this study, there was a variation in the removal efficiency across different types of PhACs (Figure 2). Analgesic medications, such as paracetamol and ibuprofen, showed significant reductions, consistent with international trends observed in various wastewater treatment systems (Verlicchi *et al.* 2012). However, the removal of certain compounds, such as atenolol, metoprolol, and furosemide, remained challenging, with reported efficiencies generally below 50% across different treatment plants (Verlicchi *et al.* 2012). Similar patterns have also been reported in Sweden, where effluent concentrations of most PhACs typically range between 1 and 500 ng L^{-1} (Falås *et al.* 2012). The MmBFs also showed lower removal efficiency for furosemide (81%); however, metoprolol was effectively removed (99%) with a high retention (5.4×10^3 ng g^{-1}), indicating biochar's potential to retain fewer biodegradable compounds. In addition to furosemide, a few other compounds showed slightly lower removal efficiencies, including fluconazole (94%), hydrochlorothiazide (75%), and benzotriazole (93%) (Supplementary Table S9). These compounds share characteristics such as a low octanol–water partition coefficient (log K_{ow} <4) and high-water solubility (>1,000 mg L^{-1}), making them more challenging to treat in adsorption-based systems (Supplementary Table S2). Nonetheless, the removal efficiencies observed in this study were substantially higher than those typically achieved in WWTPs. For example, hydrochlorothiazide generally shows removal efficiencies of 50% or lower in conventional WWTPs (Verlicchi *et al.* 2012). In contrast, the MmBFs demonstrated excellent removal efficiencies, even for compounds that are typically poorly removed (<30%) in WWTPs, such as tramadol, clarithromycin, trimethoprim, and propranolol, all of which exceeded 94% removal in this study.

5. CONCLUSIONS

This study investigated the efficiency of MmBFs in the removal of PhACs from municipal wastewater. The MmBF system showed high efficiency in removing a wide range of PhACs, including analgesics, antihypertensives, and antidepressants, with overall removal efficiencies >99% across multiple sampling occasions. This high level of treatment was achieved even after one year of operation, with the first aerobic module (M1) contributing over 92% of the total removal. The effluent quality achieved by the MmBF system not only surpassed typical OWTSs but also exceeded that of large-scale conventional WWTPs with activated sludge systems, particularly for persistent compounds, such as diclofenac, metoprolol, tramadol, and propranolol. The MmBFs demonstrated excellent ability to treat these compounds, with removal efficiencies exceeding 94%. This significant improvement could be attributed to the high adsorption capacity and enhanced biodegradation within the biochar modules, offering a sustainable alternative for micropollutant removal in onsite wastewater treatment. However, further studies are warranted to investigate the contribution of the bacterial community to the removal in the MmBF and maintenance requirements of biochar-based treatment systems related to the lifetime of the system.

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DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.

CONFLICT OF INTEREST

The authors declare there is no conflict.

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