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Removal of Pharmaceutical Active Compounds (PhACs) in Multi-module Biochar Filter (MmBF): Secondary Treatment for the Septic Effluent

Shigei, M.*, Persson, F.**, and Dalahmeh, S.S**

*Department of Earth Sciences, Uppsala University, Villavägen 16, 752 36 Uppsala, Sweden **Division of Water Environment Technology, Chalmers University of Technology, 412 96 Gothenburg, Sweden

* Department of Sustainable Development, Environmental Science and Engineering, The KTH Royal Institute of Technology, Teknikringen 10B, SE 100 44, Stockholm, Sweden.

Highlights:

- MmBFs showed >96% removal for a total of 30 Pharmaceutical active compounds (PhACs).
- Removal efficiency of Anti-hypertensives was slightly lower than other therapeutic groups (94 ± 11%).
- More than 93% of the influent PhACs were removed in the upper aerobic modules of the MmBFs.

Keywords: Multi-module biochar filter; Biochar; Pharmaceutical active compounds;

INTRODUCTION

Pharmaceutical active compounds (PhACs) have become ubiquitous in our daily lives and are routinely discharged into wastewater streams. PhACs have the potential to disrupt the endocrine system in organisms and contribute to the development of antibiotic resistance [1]. Monitoring studies have identified a variety of PhACs in the effluent of onsite wastewater treatment systems (OWTSs), due to their incomplete removal during the treatment process [2]. Therefore, it is imperative to develop and implement strategies aimed at minimizing the release of PhACs into effluent water by improving their removal in OWTSs, to meet future requirements [3].

Biochar is a waste-derived carbon-rich material that is gaining attention as a promising filter medium with a large surface area to adsorb wastewater pollutants and promote biodegradation. In this study, a pilot-scale Multi-module Biochar Filter (MmBF) system was investigated for OWTS. A previous study has shown the potential of the MmBF system for the removal of organic matter and nitrogen [4] Here, we focus on the role of the MmBF system in PhACs removal. The specific objectives are to (i) evaluate the removal efficiency of PhACs in the MmBFs; (ii) investigate the impact of aerobic and anoxic treatment conditions by analysing the performance across the modules.













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METHODOLOGY

Two identical MmBFs, consisting of six modules (M1 to M6; $36 \times 56 \times 20$ cm each), were built. M1 to M5 contained biochar (Vindelkol AB, Sweden), while M6 had a pine bark layer (Plantagen Sverige AB, Sweden) on top of the biochar to promote bacterial die-off. The media layer was 15-17 cm deep in each module. M1 to M3 and M6 were unsaturated and aerated, while M4 and M5 were saturated and non-aerated for anoxic conditions. The two MmBFs were installed at the municipal wastewater treatment plant for studies with real wastewater. They have been operated continuously, using intermittent loading starting from 25th January 2021 to the present, with few interruptions during the period. The hydraulic loading rate was 50 L m⁻² day⁻¹.

For the PhACs analyses, influent and effluent from the two MmBFs were collected four times between 29th October 2021 to 3rd February 2022. The effluent from each module of M1- M5 was collected twice for the two MmBFs at the beginning and end of the sampling campaign. The water samples for PhACs analysis were prepared using solid-phase extraction. Subsequently, the samples were analysed for 30 compounds using high-resolution liquid chromatography (HPLC) coupled with a triple quadrupole mass spectrometer (MS/MS) equipped with an electrospray ionization (ESI) source from Shimadzu. All analyte concentrations in the samples were adjusted by subtracting the blank values and corrected based on recovery rates.

RESULTS AND CONCLUSIONS

The MmBF influent showed a diverse range of pharmaceutical residues with a total concentration of all PhACs (Σ PhACs) of $1.3 \times 10^6 \pm 3.8 \times 10^5$ ng L⁻¹. Paracetamol (8.0×10^5 ng L⁻¹) and Naproxen (2.9×10^5 ng L⁻¹) were the main analgesics and Losartan (6.8×10^5 ng L⁻¹) was the main anti-hypersensitive drug, together making up 83% of Σ PhACs. Antidepressants and antibiotics were detected at lower concentrations in the influent compared to analgesics and anti-hypertensives (Figure 1). The MmBFs decreased the Σ PhACs to $4.1 \times 10^3 \pm 2.9 \times 10^3$ ng L⁻¹ as a mean effluent concentration of MmBF1 and MmBF2, which corresponds to the 96% removal. No significant difference was found between MmBF 1 and 2 in the removal of the PhACs (p >0.05). The removal efficiency varied slightly between the therapeutic groups. Analgesics showed the highest mean removal of 99 \pm 1%, followed by antidepressants (99 \pm 1%). Antibiotics and antihypertensives had lower efficiencies, at 97 \pm 3% and 94 \pm 11%, respectively.

The two MmBFs demonstrated a great reduction of \sum PhACs within the first aerobic module M1, where the \sum PhACs decreased by 93 ± 4%. The subsequent modules, M2 through M6, accounted for the removal of <7% of the \sum PhACs. M1 is an aerobic module and had the highest organic loading rate from the influent wastewater (36 gCOD m² day⁻¹). The high organic matter content and nitrification could enhance the biodegradation of the PhACs [5].

In conclusion, the MmBFs showed prominent efficiency in removing a wide range of PhACs, including analgesics, antihypertensives, antidepressants, and antibiotics, with overall removal efficiencies of >96%. This suggests that MmBFs could be a viable option for alternative OWTS reducing the













10th–14th November, 2024 Curitiba-Brazil

environmental impact of PhACs on the recipients. The samples for this study were taken after one year of operation of the MmBFs, which demonstrated the long-term efficiency of the MmBF treatment system. However, this experiment used municipal wastewater. The quality and hydraulic loading rate of domestic wastewater could vary. Seasonal temperature differences could also affect treatment efficiency. Future research should focus on investigating the MmBF microbial community and its performance under different hydraulic loading rates and temperatures.



Figure 1. Influent and effluent concentration of PhACs after MmBFs treatment shown by different therapeutic categories; (A) analgesic medicines, (B) anti-hypertensives, (C) antidepressants, and (D) antibiotics. The unit of the Y-axis is ng L^{-1} by logarithmic scale.













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