Front. Environ. Sci. Eng. 2023, 17(3): 34
https://doi.org/10.1007/s11783-023-1634-z

SHORT COMMUNICATION

Impacts of backwashing on micropollutant removal and associated microbial assembly processes in sand filters

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HIGHLIGHTS

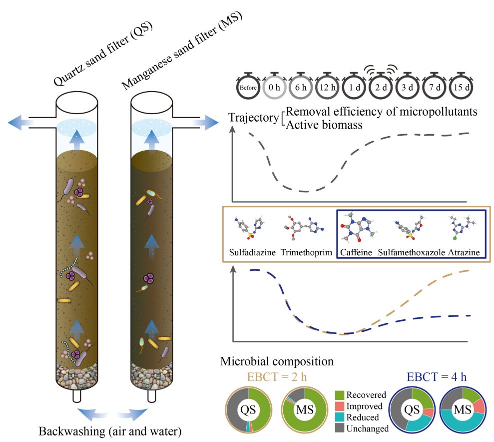
- Backwashing in sand filters with 2-h and 4-h EBCTs was simulated.
- Removal efficiency of five micropollutants recovered within 2 d at 2-h EBCT.
- Active biomass of sand filters recovered within 2 d under two EBCTs.
- Microbial composition gradually recovered to pre-backwashing level at 2-h EBCT.
- Recovered microbes only accounted for 15.55 %–25.69 % in the sand filters at 4-h EBCT.

ARTICLE INFO

Article history:
Received 19 April 2022
Revised 27 June 2022
Accepted 23 August 2022
Available online 7 October 2022

Keywords:
Sand filter
Backwashing
Recovery
Micropollutants
Community composition

GRAPHIC ABSTRACT



ABSTRACT

Backwashing is crucial for preventing clogging of sand filters. However, few studies have investigated the effect of backwashing on micropollutant removal and the dynamic changes in the microbial community in sand filters. Here, we used a series of manganese and quartz sand filters under empty bed contact times (EBCTs) of 2 h and 4 h to explore variations in micropollutant degradation and temporal dynamics of the microbial community after backwashing. The results showed that the removal efficiencies of caffeine, sulfamethoxazole, sulfadiazine, trimethoprim, atrazine, and active biomass recovered within 2 d after backwashing in both types of sand filters at 2-h EBCT, but the recovery of sulfadiazine and trimethoprim was not observed at 4-h EBCT. Moreover, the removal efficiency of atenolol increased after backwashing in the manganese sand filters, whereas maintained almost complete removal efficiency in the quartz sand filters at both EBCTs. Pearson correlation analysis indicated that microbial community composition gradually recovered to the pre-backwashing level (R increased from 0.53 to 0.97) at 2-h EBCT, but shifted at 4-h EBCT (R < 0.25) after backwashing. Furthermore, the compositions of the recovered, depleted, and improved groups of microbes were distinguished by applying hierarchical clustering to the differentially abundant amplicon sequence variants. The cumulative relative abundance of recovered microbes at 2-h EBCT was 82.76 % ± 0.43 % and 46.82 % ± 4.34 % in the manganese and quartz sand filters, respectively. In contrast, at 4-h EBCT, the recovered microbes dropped to 15.55 %–25.69 % in both types of sand filters.

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1 Introduction

Sand filters are commonly applied in drinking water treatment and can efficiently remove suspended solids, organic matter, and microorganisms from source water (EPA, 1990). During the process, particulate matter and microbes can attach to filter sands and develop a thick biofilm in both rapid and slow sand filters (Bai et al., 2013). To prevent clogging and restore pollutant removal efficiency, backwashing using air, water, or a combination of both is usually required for sand filters (Amirtharajah, 1993). Accordingly, backwashing strategies and modeling (Han et al., 2009; Piche et al., 2019), expansion of filter media during backwashing (Pizzolatti et al., 2015; Hunce et al., 2018), and backwashing water reuse (Hamoda et al., 2004; Tan et al., 2017) have been investigated to enhance performance. However, backwashing can induce a loss in biomass, thus decreasing the pollutant removal efficiency of sand filters. To the best of our knowledge, few studies have focused on microbial biofilm community dynamics after backwashing, which is crucial for pollutant removal by sand filters.

Slow sand filters can remove a fraction of the dissolved organic carbon, ammonium, and manganese through microbial transformation and degradation (Piche et al., 2019). Additionally, recent evidence suggests that some organic micropollutants can be removed by sand filters (Zearley and Summers, 2012; Carpenter and Helbling, 2017; Di Marcantonio et al., 2020), which is at least partly attributed to the biofilm formed in the filter sands (Wang et al., 2021). Owing to the limited micropollutant adsorption onto sand material, biodegradation of micropollutants by indigenous microbial populations is the most crucial removal mechanism of micropollutants in slow sand filters (Wang et al., 2022). Slow sand filters whose empty bed contact times (EBCTs) are typically longer than 1 h, have a higher removal efficiency of micropollutants than rapid sand filters, whereas clogging issues are more severe (Grace et al., 2016; Zhou et al., 2022). During backwashing, the water and air used can expand the volume of the filter media, and thus strip and remove the biofilm from the filter sands. Therefore, we hypothesized two scenarios after backwashing: 1) the micropollutant biodegradation capacity in sand filters would gradually recover along with the formation of new biofilm, and 2) micropollutant removal is improved because the old thick biofilm would be washed away; thus, functional microbes would be highly exposed to the bulk volume. The crucial question regarding periodic biofilm formation in slow sand filters, which has not been addressed, is how microbial community composition and functional dynamics are correlated with micropollutant removal in sand filters after backwashing.

Therefore, we conducted a laboratory column experiment to track the dynamics and resilience of the microbial biofilm community and the corresponding micropollutant removal after backwashing with slow sand filters. Two types of filter materials, manganese and quartz sand filters, were used to fully compare the influence of backwashing on the microbial community in slow sand filters under two different EBCTs. We aimed to examine the temporal dynamics of both the concentration of micropollutants and the microbial community after backwashing and to indicate the optimal intervals for backwashing slow sand filters for micropollutant removal.

2 Materials and methods

2.1 Micropollutant and feedwater preparation

Six commonly detected micropollutants were selected: atenolol, trimethoprim, sulfamethoxazole, sulfadiazine, caffeine, and atrazine. These are considered priority micropollutants because they are frequently found in source water for drinking water production (Daneshvar et al., 2012; Inoue-Choi et al., 2016; Eder et al., 2021; Kokoszka et al., 2021). Sodium acetate (3 mg/L organic carbon), ammonium chloride (0.5 mg/L NH_4^+-N), potassium nitrate (2 mg/L NO_3^--N), dipotassium hydrogen phosphate (0.15 mg/L P), and all six micropollutants (10 μg/L for each) were added to the dechlorinated tap water as feed water (Zhou et al., 2022).

2.2 Column experiment

The column structure was the same as that used in our previous study (Zhou et al., 2022). Commonly applied filter materials, that is, manganese and quartz sand, were used to compare the performance of micropollutant removal and microbial community composition. Two EBCTs were set up, 2 h and 4 h, as our previous research showed highly efficient micropollutant removal in the sand filters under these two EBCTs (Zhou et al., 2022); thus, the influence of backwashing was of high priority. Three columns were used for each group as biological replicates.

Backwashing was performed with filtered water at a loading rate of 0.9 mL/m^2 using a peristaltic pump and air at a loading rate of 100 mL/m^2 using an air compressor (Haili Co., China) to achieve ~33 % bed expansion for 5 min. During the experiment, sand from different depths in each column was collected and mixed (~5 g) for DNA extraction at each of the nine time points: before backwashing and at 0, 6 h, 12 h, 1 d, 2 d, 3 d, 7 d, and 15 d after backwashing and subjected to 16S rRNA sequencing. Detailed bioinformatics and statistical analyses are provided in the Supporting Information. Clean reads of all samples were accessed from the NCBI (PRJNA764307). In addition, 1 L each of influent and effluent water was collected at each time point for *in-situ* physicochemical measurements. Physicochemical parameters (Fig. S1), micropollutants, and microbial parameters (active biomass and metabolic rate) were measured according to previously described methods (Zhou et al., 2022).

3 Results and discussion

3.1 Recovery trajectories of micropollutant removal after backwashing

The details of the recovery trajectories of all sand filters are shown in Fig. 1. At 2-h EBCT, the removal efficiencies of five micropollutants (caffeine, sulfamethoxazole, trimethoprim, sulfadiazine, and atrazine) recovered to the same level as that before backwashing within 2 d in both sand filters. However, after 4-h EBCT, the removal of caffeine, sulfamethoxazole, and atrazine recovered within 2 d after backwashing, whereas a descending removal efficiency was observed when treating trimethoprim and sulfadiazine in both types of sand filters. Specifically, after backwashing, the removal efficiency of atenolol in the manganese sand filter increased rapidly from 0 %–25 % to 75 %–99 %, whereas remained at a high level (almost 100 % removal efficiency) in the quartz sand filter. The same improved recovery also appeared at both EBCTs in the two types of sand filters for atrazine treatment.

3.2 Recovery trajectories of microbial biomass and carbon metabolism

First, the change in active biomass (indicated by ATP concentration) and metabolism rate after backwashing was profiled. As shown in Fig. 2(a), for both types of sand filters, at the two EBCTs, the active microbial biomass decreased rapidly after backwashing (near 0 nmol/L) but gradually recovered to the pre-backwashing level within 2 d. We found that the quartz sand filters possessed 1.99 times more active biomass than the manganese sand filters. The inoculum of the quartz sand filters was from drinking water treatment of surface water, whereas that of the manganese sand filter was adapted from groundwater treatment, whose active biomass is relatively low. Notably, at the EBCT of 2 h, the average active biomass in both types of sand filters was up to 3.99 times more than that at 4-h EBCT. The EBCT altered the influent load, and an extended EBCT induced a low load. Subsequently, the active biomass of both types of sand filters at 4-h EBCT was less than that at 2-h. The recovery trajectories of the metabolic rate are depicted in Fig. 2(b). At an EBCT of 2 h, the metabolic

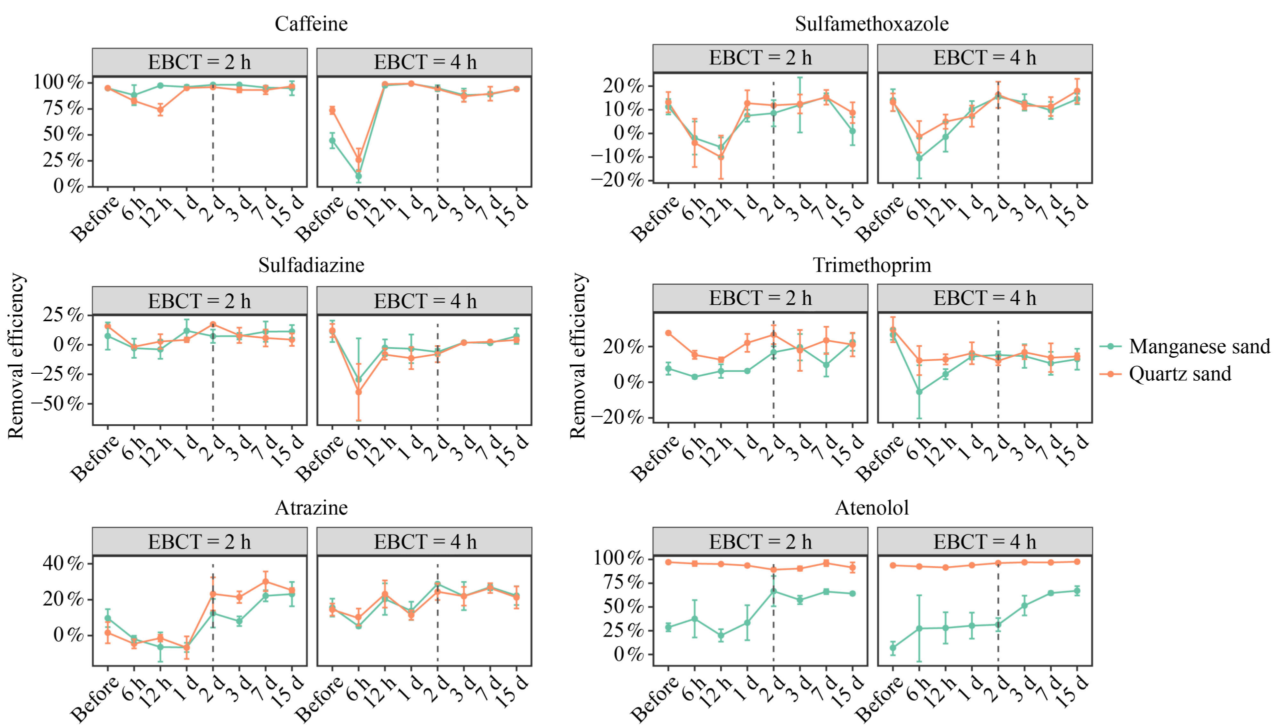

Fig. 1 Removal efficiency of six selected micropollutants in manganese and quartz sands filters before and after backwashing. "Before" refers to before backwashing; "6 h", "12 h", "1 d", "2 d", "3 d", "7 d", and "15 d" refer to time points after backwashing. The negative removal efficiency may be due to the exposure of micropollutants accumulated in the filter media or biofilm to bulk volume after backwashing, leading to higher concentrations of micropollutants in effluent than the influent.

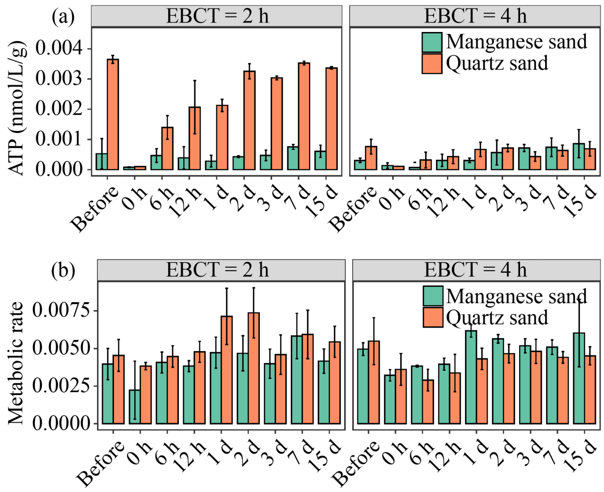

Fig. 2 Recovery trajectories of (a) active microbial biomass and (b) carbon metabolism rate after backwashing.

rates of both sand filters decreased sharply after backwashing, but recovered rapidly after 6 h and later reached a peak at 2 d. When the EBCT was prolonged to 4 h, the metabolic rate decreased immediately after backwashing but recovered within 1 d. Thus, a shortened EBCT required less recovery time for the metabolic rate but occurred within 1 d with both EBCTs.

3.3 Recovery trajectories of microbial community composition

The recovery trajectories of the microbial community after backwashing, as revealed by Pearson's correlation analysis, varied with EBCT (Fig. 3). The Pearson correlation coefficients showed increasing trends after backwashing of the two filters (Fig. 3(a)). At an EBCT of 2 h in the manganese sand filters, the correlation coefficient was 0.55 at 0 h after backwashing, and slightly decreased to ~0.4 at 6–12 h but gradually increased to 0.97 from 1 d to 15 d. In the quartz sand filters, the correlation coefficient gradually increased from 0.53 (0 h) to 0.95 (15 d) after backwashing. This suggests that most of the microbes in the sand filter recovered after backwashing under 2-h EBCT. In contrast, at 4-h EBCT, the correlation coefficients were less than 0.25 at all-time points after backwashing, demonstrating that a noticeably change in the microbial community was induced by backwashing. We further identified the proportion of the recovered microbial community after backwashing in both types of sand filters at the two EBCTs through clustering of differentially abundant ASVs (Fig. 3(b), Fig. S2). At 2-h EBCT, the cumulative relative abundances of the recovered microbes were 82.76 % ± 0.43 % and 46.82 % ± 4.34 % for the manganese and quartz sand filters, respectively, which were much higher than those for the depleted (0.32 %–3.43 %) and improved (2.54 %–7.57 %) microbe groups. Accordingly, the ASVs in the recovered group belonged to *Pseudomonas*, *Arthrobacter*, and *Bacillus*, which have been reported to biodegrade caffeine (Woolfolk, 1975; Yu et al., 2008; Summers et al., 2012), sulfonamides (e.g., sulfamethoxazole and sulfadiazine) (Deng et al., 2016), and trimethoprim (Liu et al., 2018), respectively (Fig. S3). At 4-h EBCT, there was a significant shift in the microbial community. The depleted microbes had the highest relative abundance (35.40 % ± 13.12 %) compared to that of the other two groups (recovered: 19.84 % ± 3.79 % and improved: 9.68 % ± 2.57 %). ASVs belonging to *Arthrobacter* were included in the depleted groups, which may have been responsible for the reduced removal of trimethoprim after backwashing. Notably, ASVs affiliated with *Nitrospira*, which were documented as atenolol degraders (Xu et al., 2017), were included in the improved group at both EBCTs, which corresponded to the improved removal of atenolol in manganese sand filters and high stable removal efficiency in quartz sand filters (Fig. 1).

Therefore, we concluded that for both manganese and quartz sand filters, at 2-h EBCT, the microbial community was less affected by backwashing and recovered rapidly within 2 d. When the EBCT was extended to 4 h, although active biomass and four out of six micropollutants took 2 d to recover, the microbial community changed drastically, resulting in the loss in biodegradability of some micropollutants (e.g., sulfadiazine and trimethoprim). Considering that the average run times of slow sand filters varied from 4 to 14 d in different studies (De Souza et al., 2021), the 2-d recovery time should not affect the operation of slow sand filters with short EBCT, whereas for longer EBCT (> 4 h), the removal efficiency of target micropollutants after backwashing, such as sulfadiazine and trimethoprim, should be carefully evaluated.

4 Conclusions

The effects of backwashing on the concentration of representative micropollutants (caffeine, sulfamethoxazole, sulfadiazine, trimethoprim, atrazine, and atenolol) and microbial community composition in the slow sand filter were explored. The temporal dynamics of micropollutant removal and microbial community composition after backwashing were tracked, and the removal efficiencies of caffeine, sulfamethoxazole, sulfadiazine, trimethoprim, and atrazine gradually recovered within 2 d in both sand filters at 2-h EBCT, whereas declining trends in sulfadiazine and trimethoprim degradation were found at 4-h EBCT. After backwashing, the removal efficiency of atenolol in the manganese sand filter increased rapidly but remained at a high level (almost 100%) in the quartz sand filter for both EBCTs. Correspondingly, the active biomass recovered within 2 d under all conditions. Microbial community composition gradually recovered to

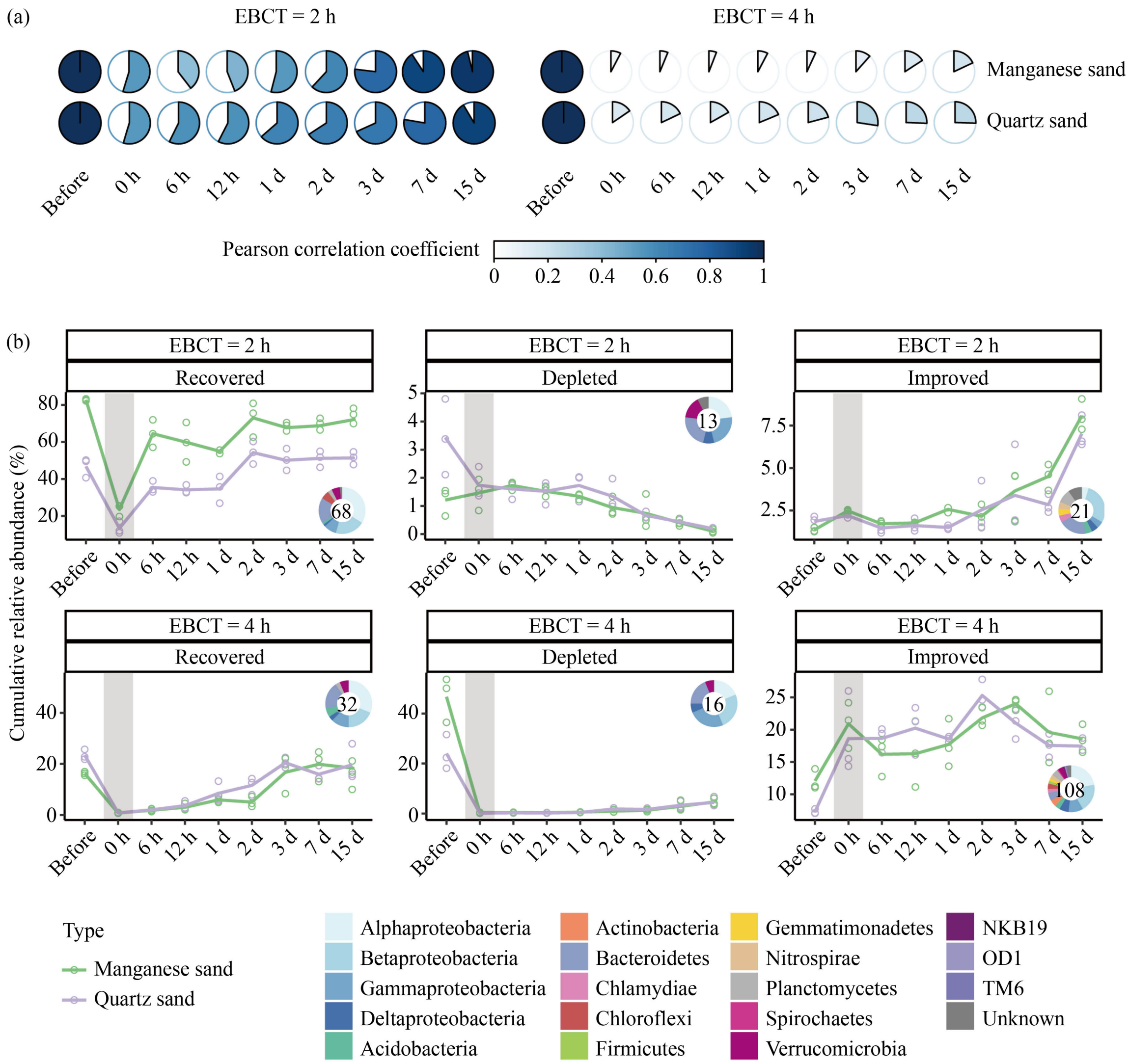

Fig. 3 Recovery trajectories of microbial community: (a) Recovery trend reflected by Pearson correlation analysis between microbial community composition before and at each time point after backwashing (0 h, 6 h, 12 h, 1 d, 2 d, 3 d, 7 d, and 15 d) in two types of sand filters. (b) Longitudinal changes in relative abundances of groups detected through hierarchical clustering of differentially abundant ASVs ($P_{FDR} < 0.05$) in manganese and quartz sand filters at 2- and 4-h EBCTs, respectively. Statistical significance was determined by negative binomial generalized linear models and pairwise Wald tests (two-sided) corrected with Benjamini-Hochberg procedure. Trend lines represent mean values throughout the experiment and inset donut plots display size (number of ASVs) and taxonomic composition of each group. Detailed taxonomy is provided in Fig. S3.

the pre-backwashing level at 2-h EBCT, and the recovered microbes accounted for 82.76 % ± 0.43 % and 46.82 % ± 4.34 % in the manganese and quartz sand filters, respectively, at 2-h EBCT. In contrast, at 4-h EBCT, the community composition in sand filters did not recover to the pre-backwashing level ($R < 0.25$), and the depleted microbes were the major group in both types of sand filters. Therefore, a 2-d optimal interval for the recovery of micropollutant removal after backwashing should not affect the operation of slow sand filters with a short EBCT.

Acknowledgements This work was supported by the National Key R&D Program of China (No. 2018YFE0204101), the National Natural Science Foundation of China (No. 51778603), and the Alliance of International Science Organizations (ANSO-CR-KP-2020-05). The authors would also like to thank Hongyan Li, Zhiyong Yu, and Yali Cheng from the Laboratory of Environmental Aquatic Chemical Analysis in the Research Center for Eco-Environmental Science, Chinese Academy of Sciences, for technical support.

Electronic Supplementary Material Supplementary material is available in the online version of this article at https://doi.org/10.1007/s11783-023-1634-z and is accessible for authorized users.

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