Desalination and Water Treatment
www.deswater.com
doi: 10.5004/dwt.2019.24713

169 (2019) 191–197
November

Improving treatment performance of biofilters for heavily polluted surface water using combined organic/inorganic substrates and exogenous carbon source of wood pieces

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Received 7 April 2019; Accepted 22 July 2019

ABSTRACT

Effective treatment of external pollution inputs is highly important for the protection of receiving waters. In this study, three tidal flow (flooded/drained time ratio of 16/8 h) biofilters with combined substrates of ceramsite/volcanics and fibrous carrier were used to remediate heavily polluted river water. Good removal performance for NH_4^+–N, COD and P was achieved, with mean efficiencies of 77.8%–97.4%, 80.0%–81.0% and 62.3%–94.9%, respectively. But TN removal was inefficient (30.5%–36.5%) primarily because nitrate removal was restricted mainly by deficiency of organic carbon in the influent. Thus wood pieces were added to the biofilters as exogenous carbon source for heterotrophic denitrification. TN removal was largely enhanced with mean removal efficiency of 65.7%–70.5% during the initial 20 operation days because of decreased effluent nitrate concentration, despite of the gradual decrease of TN removal efficiency and release of pollutants from the wood during the first several operation days. The biofilter with combined substrates and additional plant biomass as carbon source is a promising eco-technology for the remediation of polluted surface water with high nitrate content and low C/N ratio.

Keywords: Biofilter; Combined substrates; River water; Low C/N; Wood pieces

1. Introduction

Eutrophication of water bodies as a consequence of receiving excessive nutrients and other pollutants can result in harmful algal blooms and quality deterioration of aquatic environments and is a worldwide environmental issue [1,2]. Most of the surface water bodies in China are suffering from eutrophication and ecosystem degradation due to rapid economic development and urbanization, albeit water qualities of many lakes in China have gradually become a little better in recent years owing to the implementation of a number of water remediation technologies [2]. As excessive imports of external pollutants can cause deterioration of water bodies, it is well recognized that effective treatment/control of exogenous influent pollution is critical for the protection of aquatic environment of receiving waters [1,3–5].

Among the various technologies for wastewater treatment, biofilter packed with varied materials as biofilm media has been widely applied to remediate polluted surface water because it is reliable for removal of pollutants even at low level, cost-effective and flexible/convenient to be built and

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operated and environmentally friendly [6–8]. Wastewater can passively flow through biofilters with pollutants transformed and/or removed primarily by microbiota inhabited on the media. Filled substrate and operation conditions are critical for the treatment performance and cost of biofilters, and many attempts have been made to enhance biofilter performance [9]. Certain inorganic materials with high surface area and porosity and adsorption capacity for pollutants, such as zeolite, ceramsite, volcanics, etc., were employed as preferable media in biofilters alternative of conventional sand and gravel in recent years [10,11]. Synthetic organic materials such as fibrous carrier and plastic ring are also used as biofilter media because they can provide abundant habitats for various microorganisms and prevent the media from clogging [7,8,12]. With regard to operation conditions, tidal operation and intermittent aeration are two commonly applied strategies to improve pollutant (especially nitrogen) removal performance of biofilters, because poor performance is frequently obtained in continuously saturated biofilter systems and these operation modes can create alternating aerobic and anoxic environments which are necessary for nitrogen removal by nitrification and denitrification in a single reactor [9,13]. Furthermore, addition of exogenous organic carbon source is usually needed for effective denitrification when treating nitrate-rich wastewater with low C/N ratio [14–16]. Further study is still required for optimization of biofilter to treat wastewaters more efficiently, and combination of the strategies mentioned above is likely to be a good solution. However, such attempt, especially for remediation of polluted surface water, is still scarce.

In this study, three laboratory-scale biofilters with combined substrates (ceramsite, volcanics and fibrous carrier) were established to remediate heavily polluted river water. They are operated in a tide-flow mode with flooding/draining time ratio of 16/8 h in an operating cycle, and during the second operation phase, wood pieces were added to promote nitrogen removal because of the poor nitrate removal. The main objective of this study is to investigate and compare the treatment performance of the tidal-operated biofilters with combined substrates and exogenous solid organic carbon for heavily polluted river water.

2. Materials and methods

2.1. Experimental biofilters

Cylindrical columns with inner diameter and height of 0.1 and 1.1 m, respectively, were used as biofilters for polluted water treatment in this study. The columns were made of opaque polyethylene and filled with combined inorganic and organic materials as media. Three groups of combined substrates were packed in the columns: (BF I) clay ceramsite + fibrous carrier (v:v = 1:1); (BF II) volcanic + fibrous carrier (v:v = 1:1) and (BF III) clay ceramsite + volcanic + fibrous carrier (v:v:v = 1:1:1) from the bottom up to a height of 1.0 m. A layer (0.05 m) of gravel with grain size of 15–20 mm was placed at the bottom for better effluent collection. Ceramsite and volcanics were effective for ammonium and phosphorus removal and fibrous carrier-filled biofilter was favorable for nitrate removal and had larger working volume for wastewater compared with those filled with inorganic materials [17]. Furthermore, ceramsite, volcanics and fibrous carrier are all cost-efficient materials commonly applied in biofilm technology for water treatment [7]. Therefore, the combined substrates were expected to be able to achieve high overall pollutant removal performance. The particle sizes of ceramsite and volcanics were 6–10 mm (circular/oval) and 5–8 mm (rhombic/polygonal), respectively, and the fibrous carriers are polyethylene ring (Ø = 8 cm) with dense filamentous fibers on the edge. The biofilters with combined substrates were illustrated schematically in Fig. 1.

Clay ceramsite and volcanics were obtained from a local market and fibrous carrier was purchased from Hongpeng Environmental Protection Equipment Co., Ltd. (Yixing, China). Seed sludge collected from the aeration tank of a small wastewater treatment plant in Yunnan university was introduced with simulated wastewater into the biofilters in a batch mode for several times for biofilm development on the media, and approximately 6 weeks of acclimation until relatively stable treatment performance was allowed for the systems before formal experiment. Furthermore, after 2 months of formal operation, 200 g of wood pieces (2–8 cm) with C/N/P contents of 45.5%/0.12%/0.008% was added to each system from the top as carbon source for denitrification because the nitrate removal was greatly restricted and wood is cheap and has high carbon content [3]. The wood pieces were collected from a local wood factory and washed with distilled water and dried naturally before addition. The wood pieces were added in the top layer of the biofilters for convenient addition and following replacement. Subsequently, the systems continued to run for 6 weeks.

2.2. Operation of the biofilters

The influent of the biofilters was simulated based on the water qualities of a heavily polluted urban river in Kunming city which flows into Dianchi Lake, a famous eutrophic lake in Yunnan, China. Simulated wastewater was used to minimize the variability of water qualities and

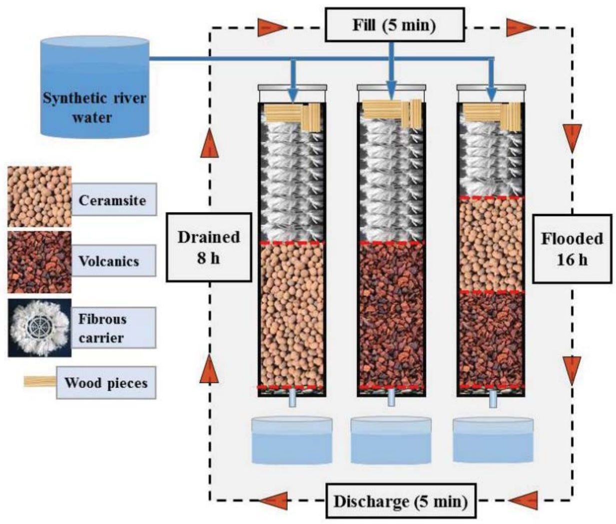

Fig. 1. Schematic illustration of the experimental hybrid biofilters.

be free from suspended solids which is detrimental for biofilters and should be removed before feeding. Glucose, NH_4Cl, KNO_3 and KH_2PO_4 were dissolved in tap water as sources of chemical oxygen demand (COD), NH_4^+–N, NO_3^-–N and phosphorus (P) in the influent, with the corresponding concentrations of 56.1 ± 9.8, 6.2 ± 0.4, 13.0 ± 0.7 and 0.9 ± 0.1 mg L^{-1}, respectively. NO_3^-–N was the dominant nitrogen form in the influent and the mean ratio of COD/N was 2.9, which was lower than the proposed values [18,19] for effective nitrogen removal.

The biofilters were operated in a tidal downflow mode. The simulated heavily polluted river water (3.5 L) was introduced into the systems from the top and retained for wastewater-substrate contact for a flooding time of 16 h. After that, the treated wastewater was completely discharged from the outlet at the bottom of the biofilters and then a draining period of 8 h was followed (Fig. 1), yielding a hydraulic retention time of 1 d. The periodic flooding and draining operating schemes were applied to create alternative oxic and anoxic conditions in the biofilters, and the flooding/draining time ratio of 16/8 h was set because it was more favorable for pollutant removal from polluted river water according to our previous study [17]. The biofilters were put indoors and the experiment was carried out from October 2017 to February 2018 with air temperatures of 7°C–17°C.

2.3. Evaluation of treatment performance of the biofilters

Composite influent and effluent water samples from the biofilters were periodically collected every 2–3 d, and COD, NH_4^+–N, NO_3^-–N, NO_2^-–N, total nitrogen (TN) and total phosphorus (TP) concentrations in the samples were analyzed in accordance with the standard methods [20]. The pH and dissolved oxygen (DO) content were measured in situ using portable meters (INESA, China). Approximately 15 sampling campaigns were carried out before (phase I) and after (phase II) wood piece addition, respectively, to evaluate pollutant removal performance of the biofilters. Significant differences in pollutant removal efficiencies among the biofilters were detected by one-way analysis of variance (ANOVA) followed by the Tukey test at a significant level of 0.05 using SPSS 19.0 for Windows.

3. Results and discussion

3.1. Treatment performance of biofilters with combined substrates for polluted river water

The influent and effluent concentrations as well as removal efficiencies for nitrogen, COD and TP of the biofilters are shown in Figs. 2a–f. Effective NH_4^+–N removal (77.8%–97.4%) was achieved in these biofilters, especially in biofilter III with three materials packed, with average effluent NH_4^+–N concentrations of 0.16–1.38 mg L^{-1}. Water-saturated biofilter/constructed wetland is usually subjected to oxygen deficiency due to very low air diffusion into the substrate pores, thus ammonium removal in such systems is often limited as a result of nitrification restraint in reducing conditions [21,22]. Supplying DO is a feasible way to improve ammonium removal in biofilter, and tidal flow operation was proved to be an effective and economical strategy [13,23]. In the tidal flow biofilters, air could be passively sucked/diffused into the matrix when/after draining treated wastewater, and NH_4^+ and organic matters adsorbed on ceramsite and volcanics in the flooding period could be oxidized, with adsorption capacity of the media regenerated for the next operation cycle [11,23]. The lower NH_4^+ removal efficiency of system II ($p < 0.05$) was attributed to the lower adsorption capacity for NH_4^+ [10] and abundance of attached nitrifying bacteria [17] in volcanics than those in ceramsite, and more volume of inorganic substrate in biofilter III (2/3) allowed highly efficient NH_4^+ removal (Fig. 2a). Moreover, satisfactory COD removal (80.0%–81.0%) was achieved in these systems with average effluent concentration of 10.0–10.9 mg L^{-1} and there was no significant difference ($p > 0.05$) among them, indicating that the three biofilters all could remove organics effectively. Organic substances in the polluted river water were degraded/utilized by various microorganisms in the systems, including denitrifying bacteria. In addition, the biofilters effectively removed phosphorus from the influent, with average effluent TP concentration of 0.05–0.34 mg L^{-1} and high efficiencies averaging 94.9% and 89.3% achieved in systems II and III, respectively. Substrate sorption has been widely recognized as the predominant pathway responsible for phosphorus removal in the biofilters [4,24,25]. Volcanics is a porous natural mineral rich in Mg, Ca and Al which can bind a large amount of P [26], thus can be employed as a promising agent for efficient P removal and recovery from wastewater. P removal efficiency of biofilter I (62.3%) was significantly lower ($p < 0.05$), which was primarily resulted from the low P adsorption capacity of ceramsite compared with volcanics [4]. Thus the combined substrates of ceramsite and volcanics are beneficial for simultaneous efficient removal of NH_4^+ and P.

Tidal operated biofilters with inorganic media (ceramsite and volcanics) were inefficient in nitrate removal from polluted river water, thus fibrous carrier which is favorable for the attachment of denitrifying bacteria aggregation was packed in these systems [17]. It showed that the average TN removal efficiencies were 30.5%–36.5% (Fig. 2d), which were higher than those of ceramsite- or volcanics-filled biofilter recorded in our previous study [17] despite that the relatively low ambient temperature during the experiment was detrimental for nitrogen removal. The biofilter I performed better than the other two systems in TN removal ($p < 0.05$), probably because ceramsite and fibrous carrier are relatively effective in nitrification and denitrification [17], respectively, and their combination could achieve the best nitrogen removal. Nevertheless, TN removal efficiency of the biofilters was still low due to the poor nitrate removal, even with nitrate accumulation in some cases (Fig. 2b) resulted from NH_4^+ nitrifying. Only a little amount of NO_2^-–N (0.04–0.43 mg L^{-1}), which is an intermediate product in nitrification and denitrification processes, was accumulated in the systems, particularly in biofilter II. But the average NO_3^-–N concentration in the effluent of the biofilters was as high as 11.2–12.9 mg L^{-1}, suggesting largely restricted denitrification process primarily as a consequence of low organic carbon content. Classical nitrification followed by denitrification is the most common pathway for biological nitrogen removal from various types of wastewater,

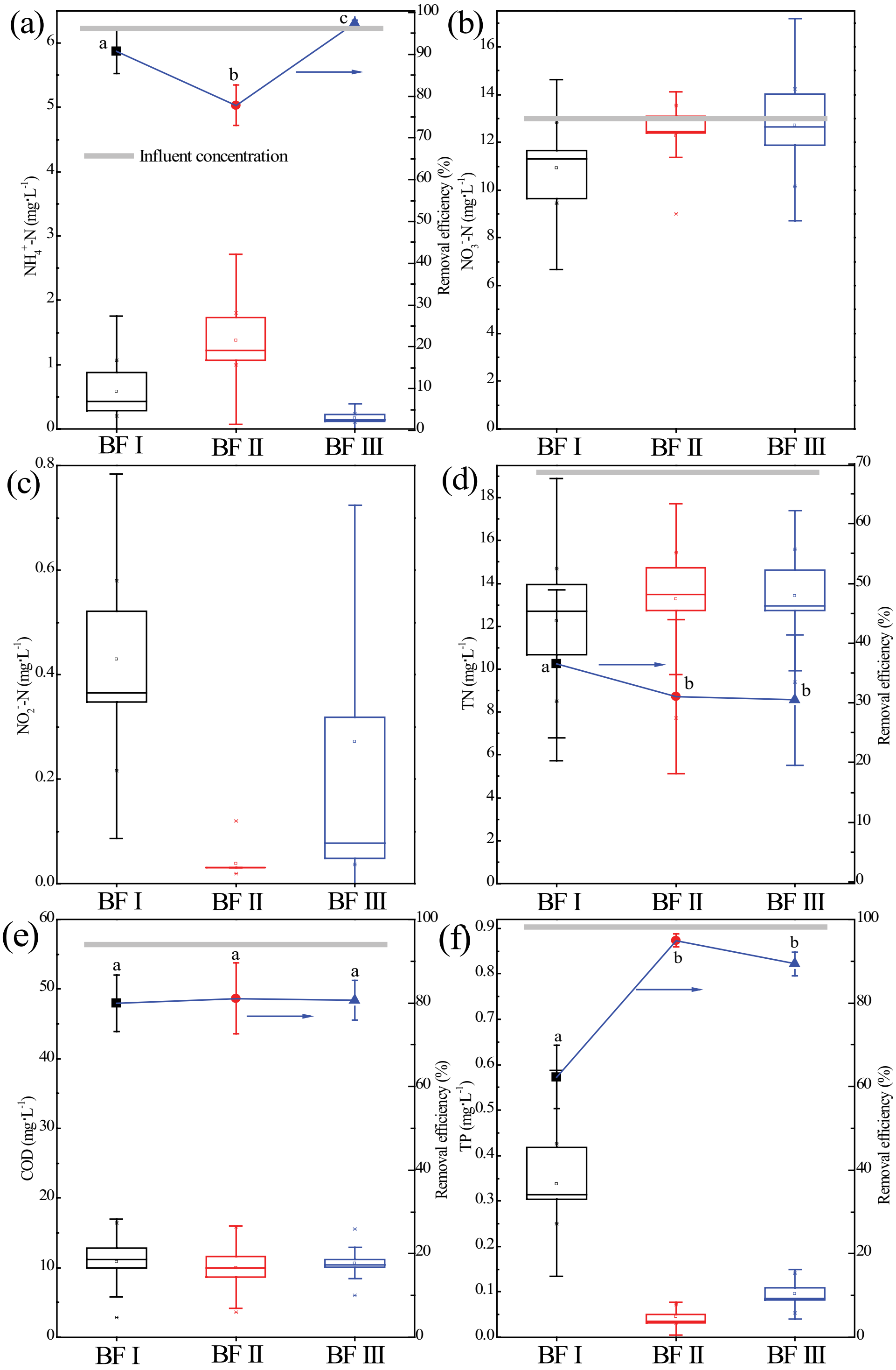

Fig. 2. Influent and effluent concentrations and removal efficiencies for (a) NH_4^+–N, (b) NO_3^-–N, (c) NO_2^-–N, (d) TN, (e) COD and (f) TP of the biofilters ($n = 15$). BF I: ceramsite + fibrous carrier; BF II: volcanic + fibrous carrier; BF III: ceramsite + volcanic + fibrous carrier.

and the presences of organic electron donors and reducing environment are required for heterotrophic denitrification [27,28]. DO (averaging 5.0 mg L^{-1} in the influent) in the biofilters was consumed in the initial flooding period (2–3 h) and then anoxic/anaerobic condition prevailed. But organic carbon source in the influent (C/N = 2.9) was insufficient for nitrate denitrifying. Lack of available carbon source and low temperature for effective nitrogen removal by denitrifying bacteria are frequently reported when treating nitrate-rich but low C/N wastewaters such as secondary effluent from wastewater treatment plant, agricultural drainage and eutrophic surface water [3,7,8,11,18,19,28]. Addition of cellulose-rich natural solid carbon with low/no cost and easy availability is a feasible strategy to enhance nitrogen removal cost-effectively in these wastewaters [14–16,29], and thus wood pieces with size of 2–8 cm were added to the biofilters from the top (Fig. 1) after 2 months of operation.

3.2. Treatment performance of biofilters with wood piece addition

Temporal variations of effluent concentrations and removal efficiencies for nitrate and TN of the biofilters added with wood pieces are presented in Fig. 3. TN removal in the biofilters was largely promoted by the wood addition, with the average and highest efficiencies of 65.6%–70.5% and 80.7%–85.1% obtained, respectively, during the first 20 d of operation. This promotion was attributed to that a large portion of nitrate in the systems was consumed by heterotrophic denitrifying bacteria using soluble organic compounds released from the wood, with much lower effluent NO_3^-–N concentrations (mean values of 5.7–6.6 mg L^{-1}) obtained. Natural lignocellulosic solid biomass and their fermentation broth are popular exogenous carbon sources to intensify nitrogen removal from wastewaters with high nitrate content and/or low C/N ratio [14–16,18]. After operating over 20 d, effluent nitrate concentration of the biofilters increased and TN removal efficiency decreased notably as expected because of a reduction in release of dissolved organics from the wood pieces for denitrifiers. Release of organic substances from plant biomass usually has two stages, fast dissolution of soluble compounds from structural polymers such as hemicelluloses and cellulose, and then slow leaching of insoluble organic matters mainly depending on microbial degradation [8,29,30]. Thus more operation time might be required for continuous supply of insoluble organic carbon from the wood pieces for denitrification. Moreover, low temperature (7°C–12°C) at this period was also partly be responsible for the low nitrate removal through affecting microbial activities involved in organics biodegradation and denitrification [15,22,27,28].

The effect of wood pieces addition on removal performance of the biofilters for NH_4^+, COD and TP was also investigated and the results are shown in Fig. 4. Removals of these pollutants, especially COD, were adversely impacted in the initial several days after the wood addition because released refractory organics containing N and P were not be fully biodegraded/utilized and eventually washed out of the systems. However, high performance of NH_4^+, TP and COD removals recovered in a short operation period (5–10 d), demonstrating the feasibility of wood addition for the effective enhancement of nitrogen removal in biofilter treating low C/N wastewater. In addition, wood is easily accessible with low cost and contains high carbon content, thus is a good option of carbon source for nitrate removal [3,30]. Similar variation patterns of pollutant removals were observed in the three biofilters. The results were consistent with those documented previously [14,30,31], and adding wood (or other plant biomass and fermentation broth) in a batch mode might be a good strategy. The recovered NH_4^+ removal efficiency (mean values of 68.9%–74.0%) was lower

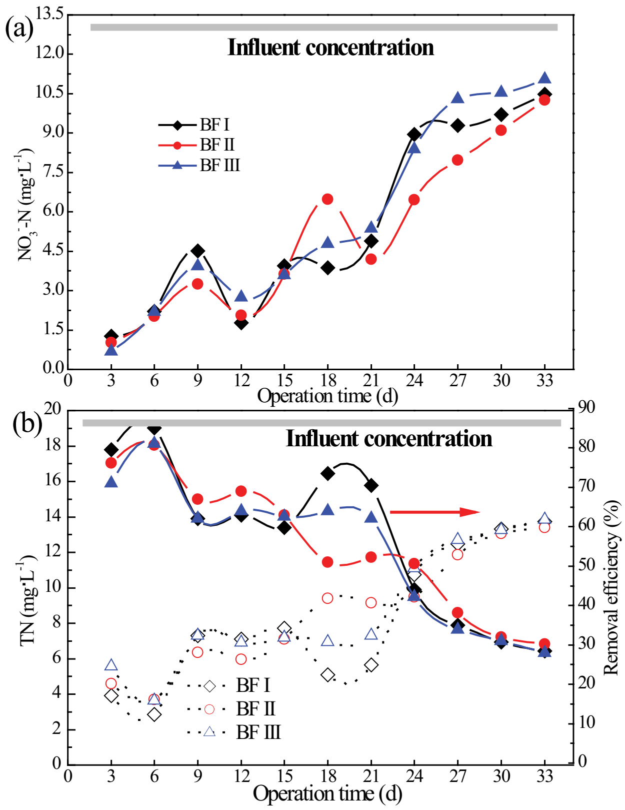

Fig. 3. Temporal variations of effluent concentrations and removal efficiencies of the biofilters with wood pieces for (a) NO_3^-–N and (b) TN.

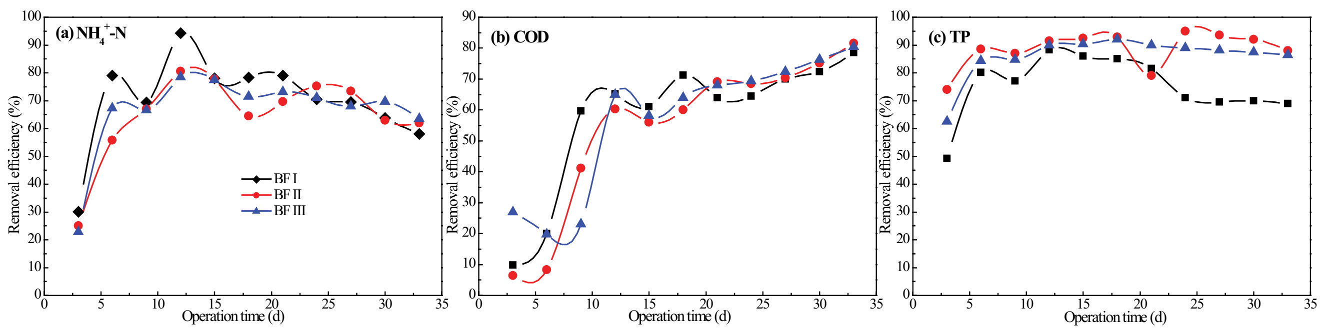

Fig. 4. Temporal variations of (a) NH_4^+–N, (b) COD and (c) TP removal efficiencies of the biofilters with wood pieces addition.

Table 1
Pollutant mass removal rates (g m^{-3} d^{-1}) of the biofilters

Pollutant index	Before wood addition			After wood addition		
	BF I	BF II	BF III	BF I	BF II	BF III
NH_4^+–N	3.09 ± 0.23	2.66 ± 0.26	3.33 ± 0.24	2.39 ± 0.56	2.21 ± 0.52	2.26 ± 0.52
TN	3.93 ± 1.43	3.35 ± 1.48	3.27 ± 1.25	6.07 ± 2.11	5.86 ± 1.82	5.78 ± 1.88
COD	24.9 ± 6.02	25.4 ± 7.06	25.1 ± 5.49	17.8 ± 6.80	16.8 ± 7.88	17.5 ± 6.89
TP	0.31 ± 0.06	0.47 ± 0.04	0.44 ± 0.04	0.35 ± 0.06	0.44 ± 0.04	0.43 ± 0.04

than that before wood addition and decreased during the last period of the experiment (Fig. 4a), which might be resulted from the lower temperature in this period.

3.3. Pollutant mass removal capacity of the biofilters

As listed in Table 1, mass removal rate of the biofilters for NH_4^+–N, TN, COD and TP were 2.66–3.33, 3.27–3.93, 24.9–25.4 and 0.31–0.47 g m^{-3} d^{-1} on average, respectively, before addition of wood pieces. Then the mean TN mass removal rate was remarkably enhanced to 5.78–6.07 g m^{-3} d^{-1} after wood addition, although the average NH_4^+–N, COD and TP mass removal capacity decreased to 2.21–2.39, 16.8–17.8 and 0.35–0.44 g m^{-3} d^{-1}, respectively, primarily due to organics release in the initial stage after wood addition. The pollutant removal capacities of these biofilter systems were comparable with or higher than those obtained in some other systems with the similar influent properties [14,18,19,29,30] despite the varied system configurations and operation conditions in these studies. Accordingly, these findings indicated that the proposed tidal flow biofilters packed with combined ceramsite/volcanics and fibrous carrier as media and added with appropriate dosage of exogenous solid carbon source were efficient for synchronous removal of nutrients and organics in heavily polluted river water with low C/N ratio and high nitrate proportion in TN, and thus had high potential for remediation of polluted surface water.

4. Conclusion

Biofilters operated in a tidal mode with a flooding/draining time ratio of 16/8 h and filled with combined ceramsite/volcanics and fibrous carrier as media can efficiently remove NH_4^+–N (77.8%–97.4% on average), COD (80.0%–81.0%) and P (62.3%–94.9%) in heavily polluted river water. But TN removal was limited (30.5%–36.5%) as a result of low C/N ratio in the influent. Addition of wood pieces could effectively promote TN removal of the biofliters, although TN removal efficiency decreased gradually with operation time and removal performances for NH_4^+–N, COD and P were negatively impacted in the initial several days. The best overall treatment performance was obtained in the biofilter packed with three materials (ceramsite, volcanics and fibrous carrier). Tidal-operated biofilter with combined substrates and exogenous solid organic carbon is a promising eco-technology with high potential to treat polluted surface water with low C/N.

Acknowledgments

This work was financially supported by the Major Research and Development Project of Yunnan Province, China (2018BC001), Project of Science and Technology Program of Yunnan Province, China (2016FD014), Open Research Fund from Yunnan Key Laboratory for Plateau Mountain Ecology and Restoration of Degraded Environments (2018DG005), Yunnan University's Research Innovation Fund for Graduate Students (2018Z093) and the Undergraduate Innovation and Entrepreneurship Training Program of China (201810673010).

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