

Treatment of Polluted Urban Surface Waters by Sponge Based Aerobic Biofilm Reactor: Purification Performances and Resilience

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Research Article

Keywords: Surface waters, Polyurethane sponge, Filling ratios, Ammonium, Function resilience

Posted Date: October 1st, 2021

DOI: <https://doi.org/10.21203/rs.3.rs-837749/v1>

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Abstract

Surface waters are suffering continuous discharging of pollutions, and low DO and black-odorous were easily formed, especially in those slow-flowing urban lakes and enclosed small ponds. In-situ treatment by artificial aeration or water cycling with a combination of polyurethane sponge as biofilm carriers can disentangle this situation without any land occupation. Long-term continuous experiments (187 days) showed that indigenous microorganisms in urban surface waters could form biofilms in the novel style of sponge-based aerobic biofilm reactors (SABRs). In urban lake waters treatment, the purification performances of SABRs were stable and resilient as the $\text{NH}_4^+\text{-N}$ and $\text{NO}_2^-\text{-N}$ removals were steady, even facing the abrupt increase of $\text{NH}_4^+\text{-N}$ and $\text{NO}_2^-\text{-N}$ concentrations in influent. Enhancing the polyurethane sponge filling ratios in SABRs can reduce DO but did not affect $\text{NH}_4^+\text{-N}$ removal. SABRs were also able to treat enclosed surface waters or black-odorous waterbodies. Combined SABRs with water cycling, $\text{NH}_4^+\text{-N}$ removal time was shorter than the time needed by water cycling when biodegradable organic matter was not present. The massive biodegradable organic matter could inhibit nitrification and prolong the purification time. Further results showed that organic matter could be used as carbon sources to eliminate the produced $\text{NO}_3^-\text{-N}$ in SABRs. Therefore, the developed new bioreactor could act as one effective way for treating N-polluted urban surface waters.

Introduction

Due to rapid economic development and population increase, surface waters are suffering excess nutrition discharged from municipal and agricultural drainage (Nsenga Kumwimba et al. 2018). Continuous nutrition inflow to the surface waters can potentially damage the ecosystems, and lead to low dissolved oxygen (DO), submerged aquatic plant disappearance (Yan et al. 2019), algae bloom (Wu et al. 2016), especially in those slow-flowing surface waters like urban lakes and/or enclosed small ponds (Hobbie et al. 2017).

Phosphorus in surface waters can be removed by chemical adsorption and precipitation (Funes et al. 2018; Min et al. 2020; Wang et al. 2018a; Wu et al. 2019b; Xiong and Peng 2008). Nitrogen removal from actual surface waters was difficult as it needs to combine nitrification and denitrification (Xia et al. 2017). Abiotic environmental conditions will affect nitrification and denitrification (Austin et al. 2019; Yang et al. 2019; Zhang et al. 2014), such as DO, temperature, pH, and ammonium concentration ($\text{NH}_4^+\text{-N}$). DO in surface waters can be easily consumed by organic matter, then favoring denitrification and restraining the critical ammonium nitrification. Even more, stormwater runoff suddenly increases contaminant loading to surface water (Zhao et al. 2015), and the effluent of wastewater water treatments in cold seasons often contains relatively higher $\text{NH}_4^+\text{-N}$ than that in warm seasons (He et al. 2018), which may affect the surface water qualities and hydrobios.

Many physicochemical and biological approaches have been used to remove nutrients from polluted water before flowing to the surface waters. Wetlands (Austin et al. 2019; Ballantine et al. 2014; Saeed et al. 2019), vegetated drainage ditches (Nsenga Kumwimba et al. 2018), riparian buffer strips, and integrated buffer zones (Zak et al. 2018) can mitigate the adverse impacts of high nutrient loading with performance being better during the warm season. But these methods generally need a large scale of land (Liu et al. 2016) and limit their use in urban areas. Hence, in-situ treatment by floating beds was acceptable, and the combination of the plants with biofilm bacterial communities can enhance the removal rate without occupying land areas (Liu et al. 2016; Wang et al. 2018b; Wu et al. 2016; Zhang et al. 2015). However, floating beds were vulnerable to the season change (Liu et al. 2016), and most plants would die and the residues should be removed to avoid the second pollution during the cold season.

Polyurethane sponge is a synthetic polymer hydrophilic biological carrier and an ideal growth medium for biofilm formation (Feng et al. 2012; Guo et al. 2010; Zhang et al. 2016). It has been used in wastewater treatment to enhance nitrogen removal due to the high DO gradient in the cubic biofilms (Feng et al. 2012), and different filling ratios can affect

the removal effectivity (Feng et al. 2012; Guo et al. 2010; Zhang et al. 2016; Zhao et al. 2019). Anaerobic biofilms on polyurethane also can enhance the biodegradation of refractory compounds (Shi et al. 2019). When cultured on polyurethane sponges, the mix-aerobic denitrifiers can effectively remove nitrogen in actual reservoir source water (Zhou et al. 2016). But little is known about the use of polyurethane sponges in urban surface waters.

Functional resilience of a reactor is critical in practice use, as the environmental conditions often change and may affect the treatment abilities. Thus it is needed to determine how bioreactors are able to cope with disturbance and recover after disturbance (Boog et al. 2018; Cabrol et al. 2012; Cho et al. 2016; Sukias et al. 2018). Previous research found that it took 14 days to completely recover nitrification under ammonia shock loading for steel wastewater treatment (Cho et al. 2016). However, there were seldom reports focusing on the resilience of a reactor in treating urban surface waters.

In this study, a novel style of biofilm reactor was developed to purify urban surface waters under various conditions. Polyurethane sponges were used as carriers to enrich the indigenous microorganisms to form biofilms, and the micro-polluted urban lakes and enclosed surface waters such as those small ponds in sub-districts were taken as study objects. We prospected that cultured biofilms can benefit contaminant removal in urban surface waters. The objectives of this study were: 1) to develop aerobic biofilm reactors with polyurethane sponge as carriers; 2) to check the resilience of function by suddenly increasing NH_4^+ -N or NO_2 -N concentrations in influent; 3) to investigate the purification performances of biofilm reactors for enclosed-surface water treatment.

Materials And Methods

Aerobic biofilm reactor set-up

Cubic-shaped polyurethane sponges (10×10×10 mm) served as biofilm carriers in this study, purchased from AiQin Environmental Technology Co., Ltd., Jiangsu, China. The average specific surface area of a sponge cube was $91,000\text{m}^2\text{m}^{-3}$, and the porosity was 95%.

Sponge-based aerobic biofilm reactors (SABRs) were set in 100L plastic buckets and simulated as treat urban surface waters (Fig. 1a). The effective volume of the plastic bucket was 80L. Each SABR consisted of an up-flow zone and three biofilm zones, all made by acrylics cylinder with 100 mm inner diameter. The up-flow zone was 600 mm in height, and biofilm zones were 280mm in length (consisting of a 100mm cylinder and a 180mm cone) (Fig. 1b). Three biofilm zones were connected with the up-flow zone and arranged 120° between themselves (Fig. 1c). The top of the up-flow zone was sealed with a conical acrylic plate to prevent water from flowing out of the up-flow zone and flow the surface water averagely into the three biofilm zones. Fine iron wires were arranged in the connection area to prevent the carriers from dropping off when introduced into biofilm zones. Small holes were made at the top of the biofilm area to circulate the surface water and prevent the internal carriers from being washed away. This special design considered that in practical application, the reactors will be placed in the middle of the surface water and reduce the interference of the surrounding people.

Polyurethane sponges were filled only in the biofilm zones, and the filling ratios in biofilm zones were the same in one SABR. Filling ratios of 20%, 40%, 60% and 80% were adopted and referred to as FR20, FR40, FR60, and FR80, respectively. The filling ratios in this study were based on the volume of the biofilm zone, which is different from previous studies as those filling rates were based on the whole reactor (Zhang et al. 2016; Zhao et al. 2019).

Reactor operating conditions

Purification performances and resilience treatment of urban lake waters

The urban lakes are the receiving waters for urban pollution, such as the effluents from the wastewater treatment plants. Thus the experiment was fed with continuous flow. Surface waters were collected to laboratory every week from Lake Xuanwu, located in Nanjing City (China). The operation was divided into three stages (Table 1). Influent was continuously fed to reactors by peristaltic pumps (6.95 mL min^{-1}), and hydraulic retention time (HRT) was 8 days as the HRT of Lake Xuanwu was about 7 days. Ammonium chloride (NH_4Cl) was added to the lake waters (collected from Xuanwu Lake) to prepare the influent in stage I and stage II, and simulate the sudden increase of $\text{NH}_4^+\text{-N}$ concentration in influent. No sediments were added into the buckets as dredging is often adopted in urban lakes, such as Lake Xuanwu. After dredging, the organic carbon contents in the surface layer of sediment were low and denitrification was inhibited (Zhong et al. 2009).

Stage I: This stage was used to enrich the indigenous microorganisms to form biofilms on bio-carriers and study the purification performances of SABRs for urban lakes. Actual surface water was continuously fed to SABRs without any nutrition addition. DO supply by controlled airflow with 60 mL min^{-1} and lasted for 70 days (Stage I-a). Submersible water pumps (Tian Xia Yu Jia, TA8800, 2000 L h^{-1}) were used in Stage I-b for enhancing the hydraulic condition and lasted for 37 days.

Stage II: This stage was used to study the function resilience of SABRs as rainfall may lead to non-point pollution flow into urban lakes, which often occurred in warm seasons. Nitrogen fertilizer is often used for urban landscaping. During the warm and rainy season, the nitrogen fertilizer will flow into the urban lake. Previous studies found that low organic loading was favorable for nitrification (Luo and Meng 2020). However, most organic matters in surface runoff are humic-like compounds and are difficult to biodegrade (Zhao et al. 2015). Moreover, the dissolved organic carbon in surface water was only $2.03\text{-}7.67 \text{ mg L}^{-1}$, and the bioavailable organic carbon only accounting for 6%-11% (Wu et al. 2019a; Xu et al. 2020). Thus only $\text{NH}_4^+\text{-N}$ concentrations in influent were intermittently improved to $2.00\text{-}4.00 \text{ mg L}^{-1}$ for 7 times.

Stage III: This stage was also used to study the functional resilience of SABR when facing a sudden increase of pollution in influent as $\text{NH}_4^+\text{-N}$ concentration in the effluent of industrial wastewater treatment plants can reach over 10 mg L^{-1} (He et al. 2018). The organic matters in wastewater treatment plants can be removed stably and effectively, and the organic matters in effluent were often refractory. Thus only $\text{NH}_4^+\text{-N}$ concentration increased suddenly to 15.00 mg L^{-1} in influent and smoothly declined for 16 days, followed by influent as in stage II ($3.26 \text{ mg L}^{-1} \text{NH}_4^+\text{-N}$) for 7 days.

Purification performances for treatment of enclosed surface waters

Synthetic waters were used to simulate the enclosed surface waters, which often contain high concentrations of organic matters and $\text{NH}_4^+\text{-N}$ with black-odorous appearances (such as those small ponds in subdistricts and parks). Due to no continuous discharge into those enclosed surface waters, batch experiments were adopted and to examine whether the biofilms cultured by urban lake waters can directly be used in those stagnant waters. Control and only hydraulic circulation were also set up (same plastic buckets with 80 L effective volume). As organic matter may not be present in some enclosed surface waters, two operating conditions were applied in the experiments.

No organic matters present: $\text{NH}_4^+\text{-N}$ concentrations were improved to about 14 mg L^{-1} through NH_4Cl addition. NO_3^- -N concentration was improved to $1.85 \pm 0.10 \text{ mg L}^{-1}$ to test whether denitrification can occur naturally in the control treatment. Before experiments, pH was adjusted to 7.68 ± 0.03 by NaHCO_3 addition (Table 1).

Organic matters present: Acetic acid and glucose (at a ratio of 1:1 by chemical oxygen demand) were added into water to simulate those black-odorous water bodies ($\text{TOC } 59.56 \pm 3.09 \text{ mg L}^{-1}$) before experiments. Other characters of surface waters were slightly higher than no organic matters present (Table 1).

Due to low DO and high organic matters in surrounding untreated surface waters, it is beneficial for denitrification and removing the produced NO_3^- -N by SABRs. Thus batch experiments were used to remove NO_3^- -N by mixing the effluent from SABR with the untreated surface waters. The mix ratios were 10%, 20%, and 50%. Two controls were also adopted (effluent of SABR and untreated surface waters). The mixed waters were not purged with N_2 and were open to air during the experiments. Each treatment was repeated two times.

Analytical methods

Samples were taken from the outflow and immediately filtered through 0.45 μm pore size filter then stored in -20°C until analysis. The concentrations of NH_4^+ -N, NO_2^- -N, NO_3^- -N, PO_4^{3-} -P, TN, and TP were determined in accordance with standard methods (SEPA 2002). The DO, water temperature, and pH were in situ measured by a dissolved oxygen analyzer (YSI 5000, USA) and a digital pH meter (PB-10, Sartorius, Germany), respectively. Dissolved organic carbon concentrations (DOC) were measured by a TOC analyzer (Torch- Teledyne Tekmar).

Statistical analyses

To compare the removal efficiencies of different treatments, one way-ANOVA followed by Tukey HSD post-hoc test was conducted by using SPSS software with significance considered at $P < 0.05$ (SPSS Statistics version 20.0).

Results And Discussion

Performance of SABRs in treating urban lake waters

During stage I, the surface of polyurethane sponges in four SABRs changed from white to yellow and then to brown (Fig. S1), indicating that biofilms were formed by the indigenous microorganisms in urban surface waters. Artificial aeration and water cycling were often used in surface water purification, as these methods can improve the DO and change the hydraulic condition (Cao et al. 2020). DO values in four buckets were improved to over 5.00mgL^{-1} (Fig. 2a), which benefited NH_4^+ -N removal. With artificial aeration, DO concentrations in reactors did not show significant difference among the different filling ratios. Wherever, DO concentrations declined with the filling ratios when water cycling was applied to change hydraulic conditions (Fig. 2a).

Within 20 days of experiments, NH_4^+ -N and NO_2^- -N concentrations in effluent of four reactors increased to $0.40\text{--}0.60\text{mgL}^{-1}$ and $0.05\text{--}0.10\text{mgL}^{-1}$, respectively, and then declined close to 0mgL^{-1} (Fig. 3a, b). Filling fraction had only a slight influence on NH_4^+ -N removal, which consisted with using polyurethane sponges in MBBR for treating synthetic domestic wastewater (Zhang et al. 2016), but contradicted with the effect of packing rates on NH_4^+ -N removal (Feng et al. 2012), due to the characters of synthetic wastewater with high COD in that study.

NO_3^- -N concentrations of the four SABRs increased from 0.79mgL^{-1} to $0.95\text{--}1.13\text{mgL}^{-1}$ (Fig. S2a), and no TN removals were observed (Fig. S2b), which means denitrification was inhibited during the experiment. Recently, some studies suggested that denitrification can occur in oxic waters, which ascribed to the heterogeneity microstructure of suspended sediments (Jia et al. 2016; Liu et al. 2013; Xia et al. 2017) and the abundant appearance of periphytic biofilms (Wu et al. 2014). However, denitrification was not observed in this study because of the high DO and low nutrient level in lake waters. Firstly, denitrification could occur when DO concentration was up to $4.00\text{--}5.00\text{mgL}^{-1}$, and the denitrification rate decreased with an increase of DO levels (Wang and Chu 2016). In the continuous flow stages, DO values in the bulk liquid were higher than 5.00mgL^{-1} , which could restrain denitrification. Secondly, traditional denitrification used organic compounds as carbon sources (Wang and Chu 2016), and simultaneous nitrification/denitrification can be achieved in wastewater treatment under continuous aeration when carbon source was enough (Macedo et al. 2019). But TOC in the

water of Lake Xuanwu was only 2.03mgL^{-1} (Xu et al. 2020), and most of the TOC in lake waters were recalcitrant to biodegradation (Koehler et al. 2012). Thirdly, though aerobic denitrifying microbes have been isolated and identified from lakes, reservoir sediments (Huang et al. 2013; Wen et al. 2019; Yao et al. 2020; Zhou et al. 2016; Zou et al. 2014), and aerobic denitrification was observed in a drinking water reservoir by indigenous aerobic denitrifiers via in situ oxygen enhancement (Zhou et al. 2016; Zhou et al. 2020). But aerobic denitrifiers also need carbon sources for denitrification (Wen et al. 2019; Xia et al. 2020). Our previous studies have shown that *Penicillium tropicum*, an aerobic denitrifying fungus, requires 5.80mgL^{-1} TOC to remove 1.00mgL^{-1} NO_3^- -N in aerobic condition (Yao et al. 2020). Lastly, phosphorus deficiency would also inhibit denitrification (Fan et al. 2018; Zhou et al. 2016), as the phosphorus concentrations in influent were only about 0.05mgL^{-1} (Fig. S2c).

Resilience of SABRs with sudden increase of NH_4^+ -N concentrations in influent of urban lake waters

With a slight increase of NH_4^+ -N concentrations in influent in stage Ⅱ, DO in the four SABRs was lower than that in stage Ⅰ and declined with the filling ratios (Fig. 2a). With the first sudden increase of NH_4^+ -N concentration in influent (in stage Ⅱ), NH_4^+ -N and NO_2^- -N concentrations in effluent increased slightly then declined close to 0 within 4-6 days and 4-8 days (Table 2), which were similar to the result under shorting HRT (Chang et al. 2019), aeration interruption (Boog et al. 2018; Murphy et al. 2016), and shock loading of influent (Cabrol et al. 2012; Cho et al. 2016; Sukias et al. 2018).

Subsequently, the concentration of NH_4^+ -N and NO_2^- -N concentrations in the influent discontinuously increased abruptly for six times, also the concentrations of NH_4^+ -N and NO_2^- -N in the effluent firstly increased abruptly and then decreased. The periods for NH_4^+ -N and NO_2^- -N concentrations to return to the previous stable state were shortened from 4-6 days to 0-4 days and from 4-8 days to 0-3 days, respectively (Table 2). One exception was that it took 8 days to reduce NO_2^- -N concentration to below 0.01mgL^{-1} in FR20 because NO_2^- -N concentrations in the 4th shock loading were much higher than previous shocking loadings (Fig. 3b; Table 2). These results were similar to the nitrification reactor treating steel wastewater. After the first shock loading, it took 14 days to recover nitrification, but only needed 1 day after the second and third NH_4^+ -N shock loading (Cho et al. 2016). When NH_4^+ -N concentrations in influent suddenly increased to 15.00mgL^{-1} in stage Ⅲ, it took 7-8 days to recover the function of nitrification (Table 2). This result further supported that the SABR function was robust to various disturbances, which could be related to adaptation to fluctuating environmental conditions (Berga et al. 2017). In addition, the influent nitrifier may be related to the resilience of SABRs, as the response of bacterial communities to disturbances can be affected by the dispersal (Yu et al. 2018), and the long HRT may also play an important role as it can dilute the NH_4^+ -N concentration in surface water.

Similar to stage Ⅱ, the NO_3^- -N concentrations in effluent were higher than those in influents (Fig. S2a; Fig. 2b), and no TN removal was observed (Fig. S2), indicating that denitrification was still inhibited, regardless of increasing NH_4^+ -N concentrations.

Performances of SABRs in treating enclosed surface waters

Condition without biodegradable organic matter

Water cycling within SABRs can improve the DO in enclosed surface waters (Fig. 4a). DO in control declined slightly from 6.00mgL^{-1} to 3.50mgL^{-1} during the initial 10 days, then decreased to 0.50mgL^{-1} within 3 days. However, DO values by water cycling were above 9.00mgL^{-1} during the whole stage, and DO values in four SABRs all declining from 7.90 - 8.90mgL^{-1} to 3.50 - 5.50mgL^{-1} within 8 days, then slowly increased to 10.00mgL^{-1} .

In the absence of organic matter, $\text{NH}_4^+\text{-N}$ can be effectively removed by water cycling. It took 16 days to remove $\text{NH}_4^+\text{-N}$ by water cycling. When combined with the SABRs, the $\text{NH}_4^+\text{-N}$ removal time can be reduced to 9 days (Fig. 4c). $\text{NH}_4^+\text{-N}$ concentrations in control declined slightly from 14.22 mgL^{-1} to 8.14 mgL^{-1} without $\text{NO}_2^-\text{-N}$ accumulations. On the contrary, both water cycling and SABR treatments had $\text{NO}_2^-\text{-N}$ accumulations, and $\text{NO}_2^-\text{-N}$ concentrations increased firstly and then declined rapidly (Fig. 4d). The highest $\text{NO}_2^-\text{-N}$ concentration by water cycling was 8.56 mgL^{-1} (16th day), and much higher than those by SABRs (5.08 mgL^{-1} on 7th day, 7.49 mgL^{-1} on 8th day, 6.31 mgL^{-1} on 9th day, and 8.27 mgL^{-1} on 8th day, respectively). These results revealed that biofilms in SABRs promoted nitrification and reduced purification time.

Although the cultured biofilms had nitrification ability, it still needed 2 days to activate nitrification to treat surface water containing a high concentration of $\text{NH}_4^+\text{-N}$ (Fig. 4c, d, e). This result consists of that nitrification started after 46 hours when the initial $\text{NH}_4^+\text{-N}$ concentration increased to 8.10 mg L^{-1} in Tay Ninh River water (Le et al. 2019). In other words, the biofilms cultured by urban lake waters can be used in the stagnant waters treatment, and it needed time to activate the microbial activity as the pollutant concentration in enclosed surface water was much higher than urban lake waters, even though the biofilms had a sudden increase of $\text{NH}_4^+\text{-N}$ concentrations. Thus the limited nitrification capacity of biofilms may be the reason for high DO values at the beginning of the experiment (Fig. 4a).

Condition with biodegradable organic matter present

In control, the organic matter led to a decline of DO to 0 mgL^{-1} during the initial 5 days, while DO in four SABRs increased slightly and then declined to $2.00\text{-}3.10 \text{ mgL}^{-1}$ within 2 days, followed by an increase to 8 mgL^{-1} (Fig. 5a). Within the same time, DOC concentrations in control decreased from 59.56 mgL^{-1} to 8.57 mgL^{-1} within 11 days, while it took only 5-6 days to remove the biodegradable organic matters by water cycling and SABRs (Fig. 5b).

In the presence of organic matter, the time for $\text{NH}_4^+\text{-N}$ removal by SABRs needed more than 22 days, which was much longer than that without organic matter (Fig. 5c). By FR20, FR40, FR60 filling ratios, $\text{NH}_4^+\text{-N}$ concentrations increased slightly within initial 2 days, then declined and became stable at $10.04 \pm 0.41 \text{ mgL}^{-1}$ (lagged for 8-11 days), ultimately decreased to $0.46\text{-}2.59 \text{ mgL}^{-1}$. However, $\text{NH}_4^+\text{-N}$ concentration in FR80 was stable and lagged for 7 days before declining to 0.54 mgL^{-1} . In control, $\text{NH}_4^+\text{-N}$ concentration stable at $11.13 \pm 0.88 \text{ mgL}^{-1}$ at the end of the experiment, and only 21.73% was removed (Fig. 5c). Surprisingly, $\text{NH}_4^+\text{-N}$ was also removed by water cycling under organic conditions, which may be because the organic matters benefited microbe growth, and attached biofilms were formed on the wave zone. In summary, the organic matters in enclosed surface waters inhibited $\text{NH}_4^+\text{-N}$ removal and prolonged the $\text{NH}_4^+\text{-N}$ removal time.

In control, $\text{NO}_2^-\text{-N}$ concentration straightly declined to 0 within 6 days. By water cycling, $\text{NO}_2^-\text{-N}$ concentration increased to 2.29 mgL^{-1} within 17 days then declined to 0 in 5 days (Fig. 5d). While the changes of $\text{NO}_2^-\text{-N}$ concentrations in SABRs showed a bimodal pattern, with accumulations during initial 6 days and then declined, followed by a second increase and decline tendency, which were different from the results in the condition without organic matters (Fig. 4d). Previous studies showed that low organic loading was favorable for nitrification (Luo and Meng 2020). But $\text{NH}_4^+\text{-N}$ and $\text{NO}_2^-\text{-N}$ concentrations in FR20, FR40, and FR60 increased during initial 4 days, which was in accordance with dissimilatory nitrate reduction to ammonium (DNRA) (Carlson et al. 2020; Srinandan et al. 2012). Thus, DNRA was suspected to occur when there were abundant biodegradable organic matters in this study. When organic matters were used up after 6 days, the subsequent decline of $\text{NO}_2^-\text{-N}$ concentration was due to the transformation of $\text{NO}_2^-\text{-N}$ into $\text{NO}_3^-\text{-N}$ (Fig. 5d). The second increase and decline of $\text{NO}_2^-\text{-N}$ concentrations in four SABRs were similar to the result of no organic matter condition (Fig. 4d) and ascribed to the nitrification.

In control, biodegradable organic matters led to low DO, accompanied by 100% removal of NO_3^- -N in 5 days. While NO_3^- -N concentrations in SABRs only declined in the initial few days when DO were low (Fig. 5a, e). TN in control was removed by 37.56 %, and much higher than by using water cycling and SABRs (removed by 8.62 %, 11.79 %, 7.78 %, 8.02 %, 9.94 % in FR20, FR40, FR60, FR80, and water cycling, respectively) (Fig. S3b). These results revealed that denitrifying microbes were abundant in actual surface water, and denitrification can be easily achieved when DO was low. It also implied that the addition of nitrate can induce denitrification in black-odorous waterbodies without any other treatment.

The results in Fig. 6 showed that mixing the effluent of SABR with the untreated surface waters could eliminate the produced NO_3^- -N (Fig. 6b, c, d). But different mix ratios have different results. By low mix ratios (10% and 20%), NO_3^- -N were 100% removed within 1 day (Fig. 6b, c). However, by the mix ratio of 50% treatment, NO_3^- -N concentration decreased rapidly from 5.94 mg/L to 3.66 mgL⁻¹ within 1 day and stabilized at 3.20 mgL⁻¹ (Fig. 6d), simultaneously the NO_2^- -N concentration increased from 0.48 mgL⁻¹ to 4.07 mgL⁻¹, as the organic matter was not enough for denitrification. Thus TN removal by 50% mix ratio (9.9%) was lower than 20% mix ratio (19%) and 10% mix ratio (15%) (Fig. 6). Therefore, in order to remove TN to the maximum extent, it is necessary to quantify the biodegradable organic matter in closed surface waters.

As stated above, denitrification could occur naturally by mixing the effluent of SABR with the surrounding-untreated surface waters. Thus partition the enclosed-surface waters into two parts, with one part treated by SABR to remove organic matters and NH_4^+ -N, then transfer the produced NO_3^- -N to another part without aeration, and this sequencing treatment may be an economical way to tackle those black-odorous waterbodies when it contains high organic matters. Moreover, the produced NO_3^- -N can promote the biodegradation of hazardous organic chemicals (Wang et al. 2019) and improve the oxidation-reduction potential of sediment (Li et al. 2019) in another part of surface waters.

Other water quality parameters

Biological conversion of NH_4^+ -N to NO_x -N often led to pH decline, but in the continue experiment in this study, pH in effluents was higher than influent (Fig. 2b). It may due to the low flow influent or high HRT, and buffer effect of surface waters. Moreover, the fine wires used in this study can lead to a pH increase due to Fe corrosion (Di Capua et al. 2019). While in enclosed surface waters, nitrification led to the decrease of pH (Fig. 4b; Fig. S3a), promoted the release of Fe^{2+} and Fe^{3+} from fine wires then precipitates with phosphorus. Thus the removal of PO_4^{3-} -P in the batch experiments was due to the fine wires (Fig. 4f; Fig. S3c).

Conclusions

A novel biofilm reactor (SABR) was constructed for urban surface water purification. Biofilms were formed on polyurethane sponges by the indigenous microorganisms in urban surface waters. SABRs could improve DO and efficiently remove NH_4^+ -N in lake waters, but NH_4^+ -N was only transformed into NO_3^- -N. Increasing the filling ratios of bio-carriers reduced DO but did not affect NH_4^+ -N and TN removal. SABRs were stable to the continued influent and showed resilience when facing various intermittent increases of NH_4^+ -N and NO_2^- -N concentrations in influent. While biodegradable organic matters in enclosed-surface waters can inhibit NH_4^+ -N removal by SABRs and prolong the purification time. The produced NO_3^- -N in SABRs was rapidly removed through mixing with the untreated surface waters when biodegradable organic matters were present abundantly. Therefore, the developed new bioreactor provided one practical and efficient way for treatment of urban surface waters.

Declarations

Ethics approval and consent to participate

Not applicable.

Consent for publication

Not applicable.

Availability of data and materials

All data generated or analyzed during this study are included in this published article.

Competing interests

The authors declare no competing interests.

Funding

This study was supported by grants from the National Special Program of Water Environment (2017ZX07204005).

Author contributions

Shangwei He: Methodology, Investigation, Formal analysis, Writing -Original Draft. Na Song: Writing - Review &Editing. Zongbao Yao: Writing - Review &Editing. Helong Jiang: Conceptualization, Resources, Writing - Review &Editing, Funding acquisition.

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Tables

Table1 Operation conditions at different stages

	Influent water characters					DO supplied by	Mode
	NH ₄ ⁺ -N (mg/L)	NO ₂ ⁻ N(mg/L)	NO ₃ ⁻ N(mg/L)	PO ₄ ³⁻ -P(mg/L)	T(°C)		
Stage Ia (70 days)	0.05-0.49	0.00-0.13	0.09-1.50	0-0.10	14.4-25.8	Air	Continue
Stage Ib (37 days)	0.05-0.56	0.01-0.19	0.34-1.79	0.01-0.13	22.4-25.8	Water cycling	Continue
Stage II (55 days)	0.34-4.69	0.00-1.08	0.74-5.49	0.01-0.12	20.5-26.2	Water cycling	Continue
Stage III* (23 days)	15.00	0.15	1.53	0.06	20.4-24.6	Water cycling	Continue
Stage III* (20 days)	14.22±1.15	0.05±0.00	1.85±0.10	0.04±0.00	11.4-18.2	Water cycling	Batch
Stage III* (22 days)	14.22±0.30	0.43±0.10	3.57±0.10	0.43±0.06	6.5-14.6	Water cycling	Batch

*Influent water characters at the beginning of the experiments.

Table2 Function resilience of four SABRs (Days needed to recover by increased suddenly of NH₄⁺-N and NO₂⁻-N in influent)

	NH ₄ ⁺ -N (days needed to recover to below 0.1 mg/L)							NO ₂ -N (days needed to recover to below 0.05 mg/L)								
	Stage								Stage							
	1st	2nd	3rd	4th	5th	6th	7th		1st	2nd	3rd	4th	5th	6th	7th	
FR20	6	2	1	0	4	1	2	8	8	3	0	8	3	0	2	4
FR40	4	2	1	0	4	1	1	8	7	2	0	1	1	1	0	4
FR60	6	4	1	0	4	1	2	7	6	2	0	3	0	0	0	4
FR80	4	3	1	0	4	1	1	7	4	0	0	2	0	0	0	6

Figures

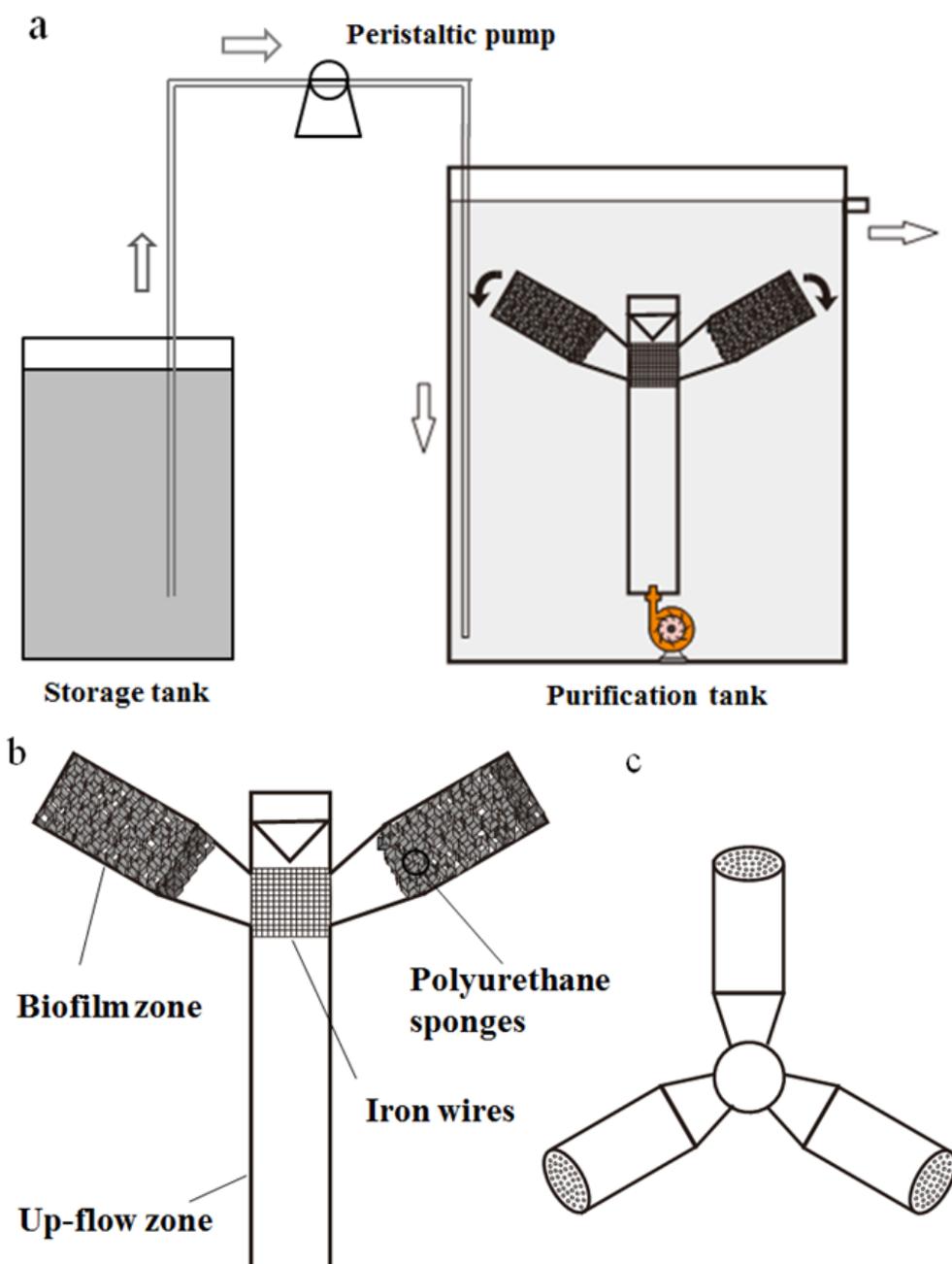


Figure 1

Schematic diagram of the experimental set-up (a) and configurations of the sponge-based aerobic biofilm reactor (b, c)

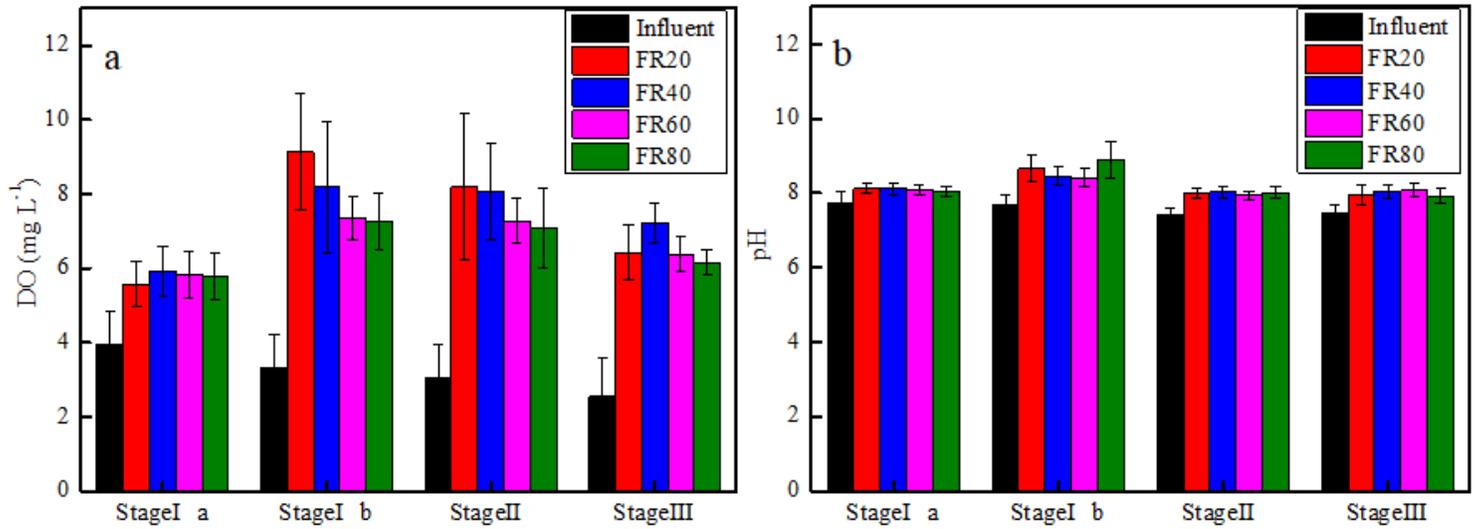


Figure 2

DO and pH from stage I to stage III (Filling ratios of SABRs by 20%, 40%, 60%, 80% were referred to as FR20, FR40, FR60, and FR80, respectively)

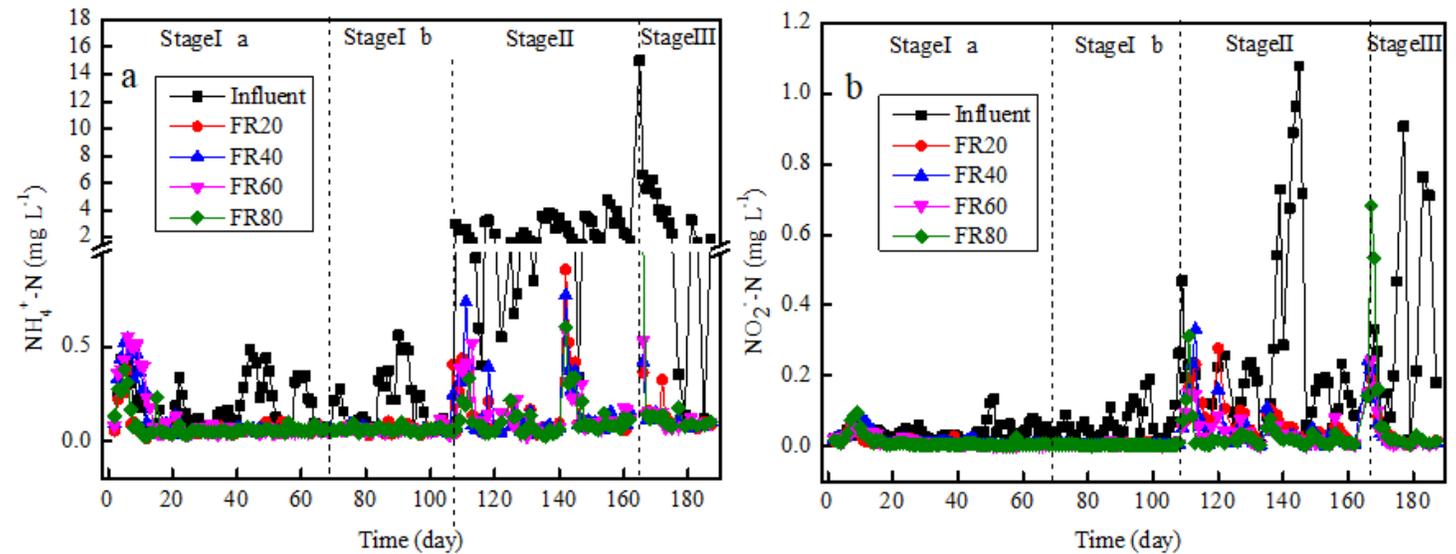


Figure 3

NH₄⁺-N and NO₂⁻-N concentrations in continuous experiments by using SABRs (Filling ratios by 20%, 40%, 60%, 80% were referred to as FR20, FR40, FR60, and FR80, respectively)

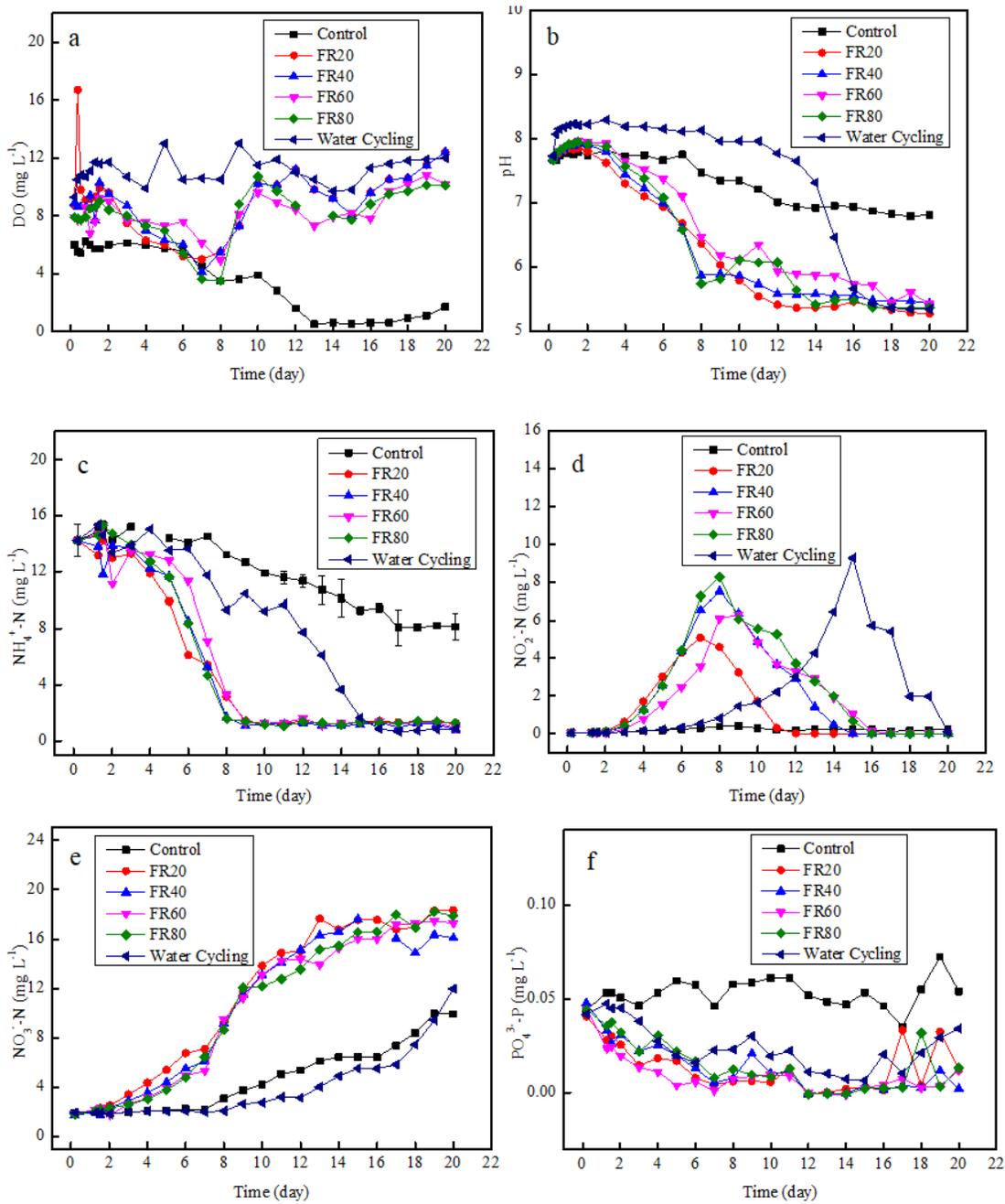


Figure 4

Purification performances by different treatments (no organic matters presented)

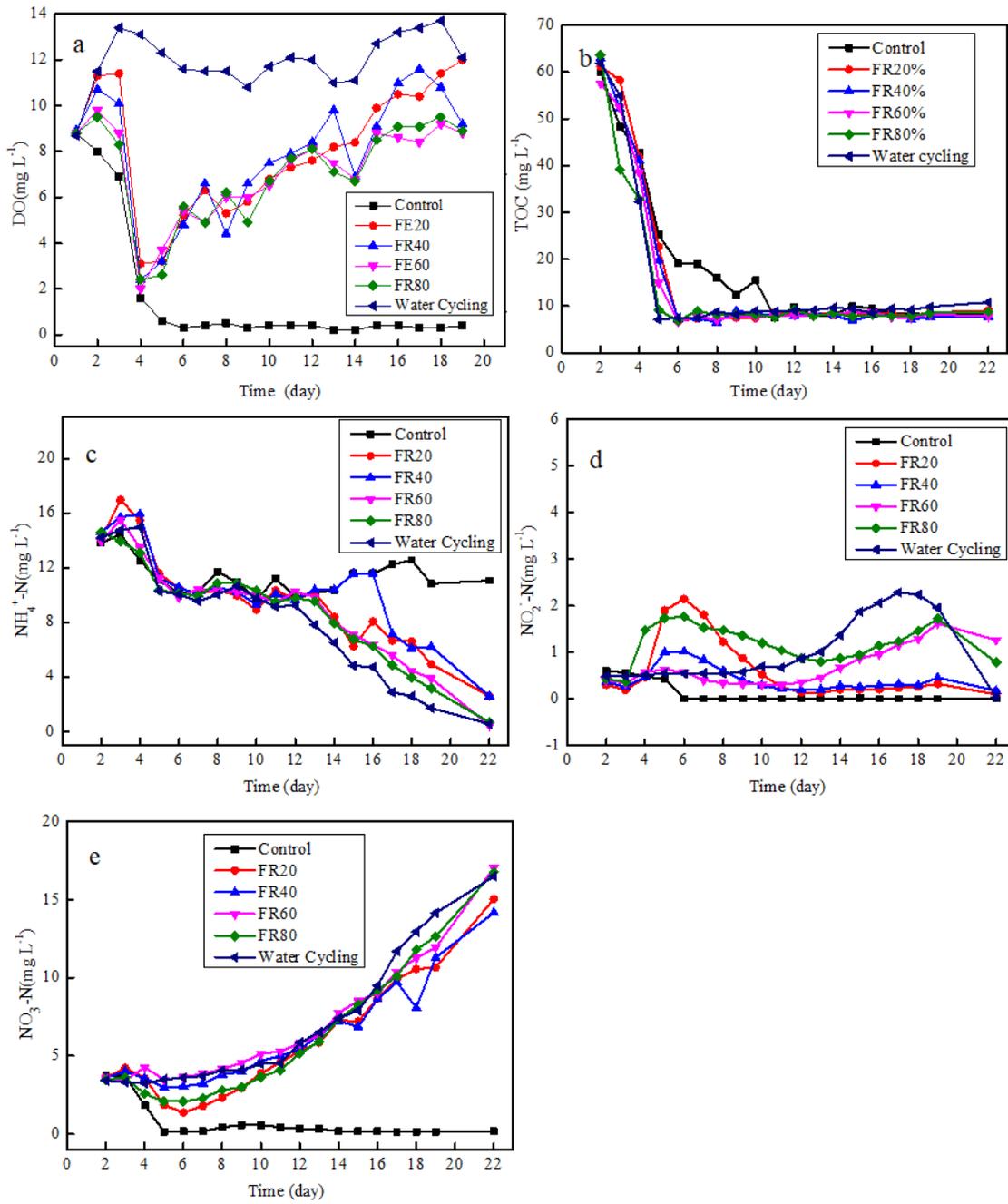


Figure 5

Purification performances by different treatments (organic matters presented)

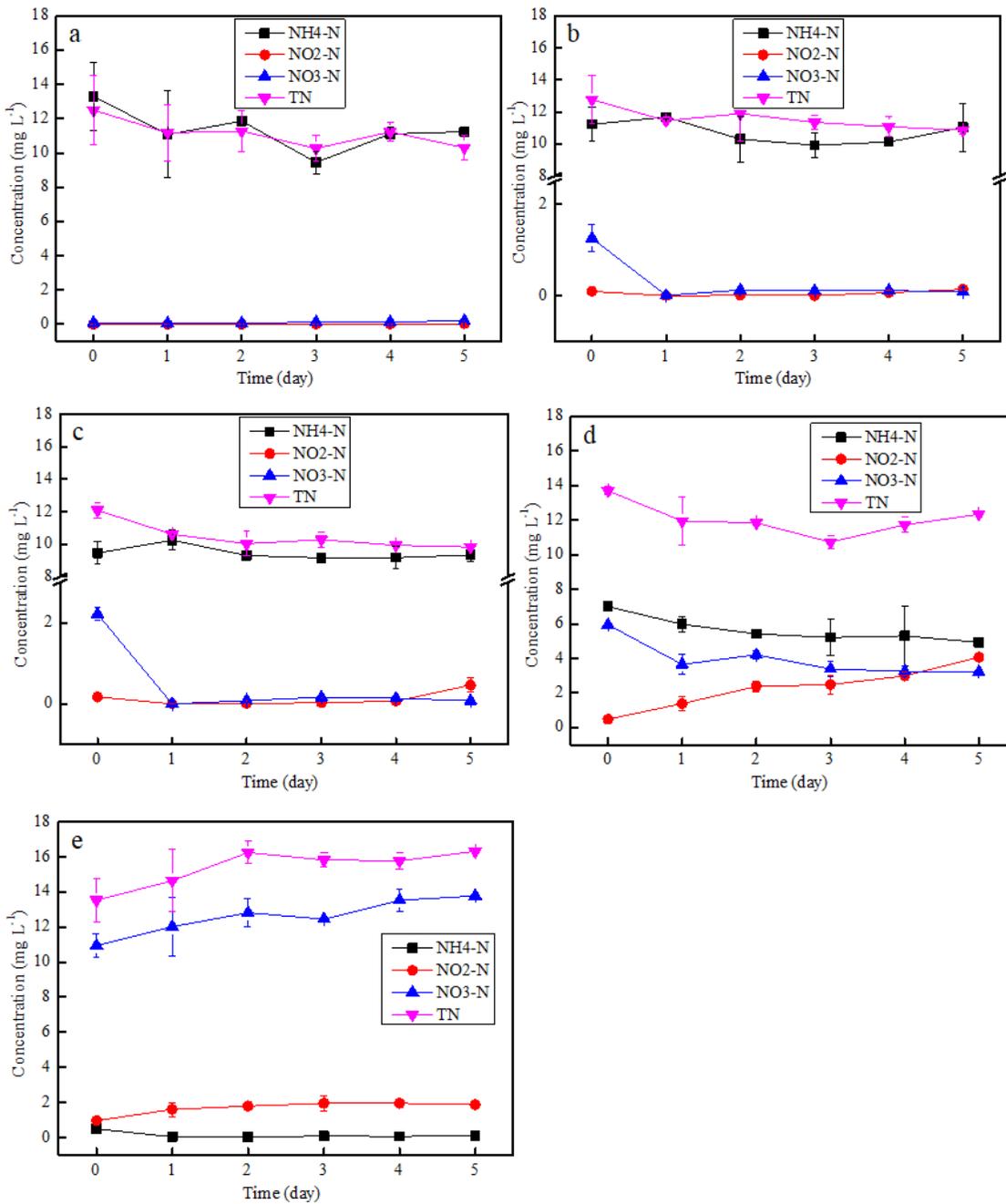


Figure 6

Performances of different volume mix ratios by effluent of SABR/untreated surface water (a. 0; b, 10%; c, 20%; d, 50%; e, 100%)

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