Application of light-weight filtration media in an anoxic biofilter for nitrate removal from micro-polluted surface water

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ABSTRACT

The research investigated nitrate removal from micro-polluted surface water by the single-stage process of anoxic biofilter using light-weight polystyrene beads as filtration media. In this study, sodium acetate was used as an external carbon source and the nitrate removal efficiency under different regimes of hydraulic loading rate (HLR), water temperature, and C/N ratio was studied. In addition, the effect of backwash on denitrification efficiency was investigated. The results show that the biofilter achieved a high nitrate removal efficiency in 2 weeks at water temperatures ranging between 22 and 25 °C at a C/N ratio (COD: NO3⁻-N) of 6.1. Besides, the average removal efficiency of nitrate at HLRS of 5.66, 7.07 and 8.49 m³ m⁻² h⁻¹ were 87.5, 87.3 and 87.1%, respectively. The average removal efficiency of nitrate nitrogen was 13.9% at a HLR of 5.66 m³ m⁻² h⁻¹ at water temperatures of 12–14 °C, then it increased to 93.7% when the C/N ratio increased to 10. It suggests that the optimal hydraulic retention time is at water temperatures of 8–10 °C. The water consumption rate of backwash was about 0.2–0.3%, and denitrification efficiency returned to the normal level in 12 h after backwash.

Key words | backwash, biofilter, light-weight filtration media, micro-polluted surface water, nitrate removal

INTRODUCTION

The Chinese government is paying more and more attention to micro-polluted lakes, such as Taihu Lake, Chaohu Lake, Dianchi Lake, and West Lake. Constructed wetlands are mainly used for the treatment of micro-polluted wastewater. However, removal efficiency of nitrogen is always low in a single-stage wetland system (Cui et al. 2011), and it always fluctuates in a large range (Díaz et al. 2012). Meanwhile, there are some other shortcomings such as long hydraulic retention time (HRT), large footprint, high infrastructure investment, etc. Therefore, a combined process of biofilter-ecological gravel bed was applied to remove nitrate from micro-polluted surface water (Sheng-bing et al. 2013). Although high nitrate removal efficiency was achieved, there were still two shortcomings to overcome: the complexity and big footprint of the two-stage process, and the big amount of backwash water which accounted for 2–4% of treated water. Thus, a single-stage biofilter using light-weight filtration media was applied to treat nitrate-contaminated micro-polluted surface water in this study.

When it comes to application of biofiltration, filtration media play a very crucial role in removing nitrate from polluted water. A wide variety of natural and synthetic materials have been explored in previous studies: sludge-fly ash ceramic particles in an A/O combined biological aerated filter (Han et al. 2009; Yue et al. 2009), clay ceramic particles in an up-flow anaerobic biofilter (UAF) (Han et al. 2013), coal fly ash ceramic granules (Jing et al. 2012), combined media (granular activated carbon and ceramsite) (Liu et al. 2008), novel media-sludge ceramic (Wu et al. 2015) and self-made sludge-ceramsite (Zou et al. 2012). All these ceramsite filtration media have proved to have a great removal performance due to their larger specific surface area for microbial growth. However, the media would be agglomerated over time, which normally leads to a huge amount of backwash water and large energy consumption, and, in turn, high costs for management and maintenance.

In order to reduce the quantity of backwash water with a lower head loss and longer filtration cycle, filtration media with low density like polypropylene or polystyrene bead have
been applied. For example, some researchers used filters packed with light-weight filtration media mentioned above to reduce suspended solids, turbidity, apparent color, Fe, Mn, residual aluminum, iron hydroxide flocks and so on (Ngo & Vigneswaran 1995; Hsu et al. 2001; Šecérov Sokolović et al. 2009; Schöntag & Sens 2014). Besides, BOD removal using S-shaped polystyrene pieces as filtration media was carried out by Xie et al. (2004a) to determine the optimum operational parameters for an aerobic biofilter using light-weight filtration media for treating domestic wastewater (Xie et al. 2004b).

Although use of polystyrene beads as the filtration media is not an original innovation, few researchers applied the polystyrene beads to nitrate removal from micro-polluted surface water. Therefore, the main objectives of this research were to: (1) investigate start-up of an anoxic biofilter using polystyrene beads as the filtration media for nitrate removal from micro-polluted surface water on a pilot scale; (2) evaluate performance of nitrate removal under different regimes of hydraulic loading rate (HLR), water temperature and C/N ratio; and (3) investigate effects of backwash on denitrification efficiencies and the required volume of backwash water.

**MATERIALS AND METHODS**

**Experimental setup**

The experimental device was set up in the Yuhuang Mountain surface water pre-treatment plant located in Hangzhou, Zhejiang province, China. A schematic diagram and picture of the biofilter configuration are shown in Figure 1(a) and 1(b). The biofilter was made of polyvinyl chloride (PVC) with a diameter of 0.30 m, height of 5.20 m, total volume of 368 L and effective volume of 212 L. Polystyrene beads (3.4–3.8 mm in diameter) were purchased as the filtration media from Veolia Environment, Paris, and the characteristics of the polystyrene beads are shown in Table 1. The height of the filtration media layer was 3.0 m, and a buffer zone of 0.30 m was laid at the bottom of the biofilter to distribute water evenly. Besides, a circular plate made of PVC with a grid spacing of 3 mm was set to hold up the light-weight filtration media at the height of 4.30 m due to the low density of the light-weight filtration media.

The biofilter was operated in an up-flow mode. Raw water was mixed with external carbon sources in a static mixer and then pumped into the buffer zone through the inlet pipe (Table 2). Moreover, the biofilter was backwashed every 3 days with air scour and water to prevent clogging. The air was blown into the biofilter using an air compressor at a flow rate of 12 m/h, while the water was pumped into the biofilter in a down-flow mode with a flow rate of 60 m/h. One combined air and water backwash cycle consisted of the following steps: (1) air flushing for 30 s, and (2) water flushing for 30 s. Three cycles were conducted each time.

**Start-up and operation of the biofilter**

The experiment was conducted from May 25, 2014 to August 28, 2015, and the whole experimental period could be divided into three phases: start-up, phase II, and phase III. The biofilter was inoculated with the seed sludge from the
During the experiment, water samples were collected every 2 days, and the analyses of NO\textsubscript{2}-N, NO\textsubscript{3}-N and NH\textsubscript{4}-N were conducted according to Standard Methods (APHA 2005). Total nitrogen (TN) and total organic carbon (TOC) were determined by Muti N/C 3000 (Analytik Jena AG, Germany). Dissolved oxygen (DO) was measured using a DO meter (HACH HQ-40d, USA).

**RESULTS AND DISCUSSION**

**Nitrate removal efficiency at water temperatures ranging between 22 and 25 °C at startup**

As shown in Figure 2(a), during the initial period of start-up, the average removal efficiency of NO\textsubscript{3}-N was 51.7% at a HLR of 2.83 m\textsuperscript{3} m\textsuperscript{-2} h\textsuperscript{-1}. When HLR was raised to 4.24 m\textsuperscript{3} m\textsuperscript{-2} h\textsuperscript{-1}, the average removal efficiency of NO\textsubscript{3}-N increased progressively to 68.3%, with a maximum of 94.4% on the 14th day compared to 80% obtained by Sheng-bing et al. (2013). The difference might be attributed to the lower water temperature (16 °C) in their tests. Two weeks after the activation of the biofilter, the concentrations of nitrate showed no further change and the reactor was therefore considered to be at steady state. Sixteen days later, HLR was increased gradually, and nitrate removal efficiency always remained at high levels. Average nitrate removal efficiencies at HLRS of 5.66, 7.07 and 8.49 m\textsuperscript{3} m\textsuperscript{-3} h\textsuperscript{-1} were 87.5, 87.3 and 87.1%, respectively. The corresponding denitrification volumetric loading rates were 71.04, 82.20 and 82.29 gNO\textsubscript{3}-N m\textsuperscript{-3} d\textsuperscript{-1}, respectively. Compared to the HLR of 7.07 m\textsuperscript{3} m\textsuperscript{-2} h\textsuperscript{-1}, denitrification volumetric loading rate only increased by 0.09 gNO\textsubscript{3}-N m\textsuperscript{-3} d\textsuperscript{-1} at a HLR of 8.49 m\textsuperscript{3} m\textsuperscript{-2} h\textsuperscript{-1}, although the denitrification capacity of the biofilter increased remarkably with HLR. It may be that the biofilter approached a maximum volumetric loading rate at a HLR of 7.07 m\textsuperscript{3} m\textsuperscript{-3} h\textsuperscript{-1} (i.e., HRT of 0.4 h). Meanwhile, the removal efficiency of 87.1% was reached, while HRT dropped to the lowest value of 0.55 h (i.e. 21 min). Tsukuda et al. (2015) obtained the nitrate removal efficiency of 26.9% at a HRT of 15 min in a fluidized sand biofilter using a waste-based endogenous carbon source, suggesting the higher nitrate removal efficiency observed in our research might be attributed to a slightly longer HRT and application of a more easily utilized carbon source in this study.

Figure 2(b) shows the concentrations of NO\textsubscript{2}-N in influents and effluents. Nitrite accumulations on the 6th and 12th days were only 0.20 and 0.28 mg/L, respectively. There was little nitrite accumulation during the start-up phase, and the maximum value of nitrite accumulation was lower than 0.3 mg/L, which was comparable to the results of the previous research (Sheng-bing et al. 2013).

Figure 2(c) shows TOC concentrations in influents, effluents and raw water at different HRTs. The effluent TOC was about 0.35–0.75 mg/L more than that in raw water, which indicates that the external carbon source was utilized to a large extent, and that the addition of external carbon sources might not cause the secondary pollution.
Effect of C/N ratio on nitrate removal efficiency at low water temperatures

The results suggest that denitrification efficiency was high at the end of the start-up phase. The reactor had matured in this period and the nitrate removal efficiency was high and stable because of favorable condition of water temperature and C/N ratio. The results were consistent with the previous research (Sheng-bing et al. 2013), so this research further focused on how to adjust C/N ratio to improve denitrification efficiency at low water temperatures.
Figure 3(a) shows that the nitrate removal efficiency increased with C/N ratio at water temperatures between 12 and 14 °C. The nitrate removal efficiency reached an average value of 13.9% at a C/N ratio of 6, and remained approximately the same level throughout the rest of this test. Cooler water temperatures and low nitrate concentrations entering the biofilter observed here might be the main reason for lower removal efficiencies. However, it increased rapidly to 93.7% when C/N ratio increased to 10, which was much higher than the value of 50% reported by Shengbing et al. (2015) at water temperatures ranging between 13 and 17 °C. The finding in this study was consistent with the previous studies suggesting that sufficient carbon sources could enhance denitrification performance by improving biomass and microbial activity at low water temperatures (Qin et al. 2015; Zhang et al. 2015; Karanasios et al. 2016). However, a relatively high C/N ratio of 10 was required, which indicates that influent DO consumed part of the carbon sources (Tsukuda et al. 2015). The average removal efficiency of NO$_3^-$-N decreased to 19.6% at water temperatures between 8 and 10 °C. It suggests that the denitrification performance was not necessarily improved by the increase in the amount of external carbon source when water temperature decreased to 10 °C or below. Besides, one study suggests that denitrification would be greatly restricted when water temperature decreased to 10 °C or below (Li et al. 2016).

Because of the low NO$_3^-$-N removal efficiency, no nitrite accumulation was observed at a C/N ratio of 6 at water temperatures of 12–14 °C (Figure 3(b)). Although the concentration of effluent nitrite was higher than that in the influent during the initial period, the maximum of nitrite accumulation was only 0.07 mg/L after the C/N ratio increased to 10. Besides, there was no nitrite accumulation observed either during the later period or at water temperatures of 8–10 °C.

As shown in Figure 3(c), the average concentrations of effluent TOC were 0.47, 0.70 and 1.84 mg/L higher than that of raw water TOC under three different operational conditions, respectively. It indicates that the external carbon source was utilized to a large extent at C/N ratios of 6 and 10 at water temperatures between 12 and 14 °C, while the utilization rate was relatively lower at a C/N ratio of 10 at water temperatures ranging between 8 and 10 °C.

Effect of HLR on nitrate removal when water temperatures ranged between 8 and 10 °C

According to the above results, it suggests that not only a significant reduction in nitrate removal efficiency, but also a relative lower utilization rate of carbon source were observed when water temperature decreased to 10 °C or below. Therefore, simply increasing the amount of the external carbon source could not improve the removal efficiency of NO$_3^-$-N effectively.

HLR and HRT are two relevant parameters. HLR decreases with increase in HRT. Generally speaking, an appropriate extension of HRT may improve denitrification performance at low water temperature. Thus, it is important to determine the optimal HLR at low water temperatures, and the results are shown in Figure 4.

Figure 4(a) shows that denitrification efficiency increased with HRT. The average nitrate removal efficiencies at HRTs of 0.5, 1 and 2 h were 19.6, 53.3 and 85.3%, respectively, while the denitrification volumetric loading rates were 19.01, 22.18 and 17.32 gNO$_3^-$-N m$^{-3}$ d$^{-1}$, respectively. On the basis of the above experimental data, the rate reached a maximum value at a HRT of 1 h. One previous study suggests the nitrate removal efficiency of 50% was observed at a HRT of 5 h when the water temperature was in the range of 5–10 °C in a sulfur-limestone autotrophic denitrification (SLAD) biofilter (Zhou et al. 2013). Compared with the SLAD biofilter, a higher nitrate removal efficiency of 85% was achieved at a shorter HRT of 2 h when water temperatures ranged between 8 and 10 °C in this research. It may be attributed to a longer lifecycle of autotrophic bacteria compared to heterotrophic bacteria. Thus, a longer HRT was needed and lower nitrate removal efficiency was observed at low water temperatures in the SLAD biofilter. Besides, it was exciting that 85.3% nitrate removal efficiency obtained in this study was comparable to the nitrate removal performance observed in a denitrifying biofilter with polycapro lactone as a carbon source and filtration medium when water temperatures ranged between 8 and 10 °C (Li et al. 2016). Moreover, there was no nitrite accumulation at HRTs of 0.5, 1 and 2 h according to Figure 4(b).

As shown in Figure 4(c), the average concentrations of effluent TOC were 1.84, 1.33 and 1.31 mg/L higher than that of TOC in the raw water at HRTs of 0.5, 1 and 2 h, respectively. The effluent TOC decreased with the extended of HRT, which indicates that the utilization rates of carbon sources increased with HRTs. However, the value of $\Delta$TOC (C$_{\text{effluent}}$ - C$_{\text{raw water}}$) was only decreased by 0.02 mg/L at a HRT of 2 h compared to 1 h. Hence, considering the denitrification volumetric loading rate, it suggests that the optimal HRT was 1 h.

Effect of backwash on denitrification performance

The amount of micro-organisms increased over time during the operation of the biofilter and the aging biofilm remained


Figure 3 | Nitrate removal efficiencies (a), concentrations of nitrite (b) and TOC (c) in influents and effluents at different C/N ratios and water temperatures.
in the filtration media layer, which would lead to the increase in hydraulic head loss. Besides, it might be easier just to penetrate filter layer, which, however, would lead to deterioration of water quality. Therefore, backwash was necessary to maintain the performance of the biofilter.

There is no doubt that backwash would inevitably lead to the loss of some viable micro-organisms, which might lead to decrease of denitrification efficiency over a period of time after backwash. Therefore, it is necessary to study the effect of backwash on denitrification efficiency. The results

Figure 4 | Nitrate removal efficiencies (a), concentrations of nitrite (b) and TOC (c) in influents and effluents at different HRTs with water temperature of 8–10 °C.
are shown in Figure 5 under the operational conditions of the biofilter were as follows: water temperature of 25 °C, C/N ratio of 6, HRTs of 0.5, 0.4 and 0.35 h, respectively.

As shown in Figure 5, nitrate removal efficiency was nearly 100% before backwash at a HRT of 0.5 h, and then it decreased sharply to 51.0% at a running time of 0.5 h after backwash. Nitrate removal efficiency fluctuated more at running times of 0.5–12 h after backwash. The removal efficiency of 95.8% was achieved at a running time of 12 h after backwash. Nitrate nitrogen removal efficiency was 95.6% before backwash at a HRT of 0.4 h, and there was no variation of denitrification efficiency at running times of 0.5–6 h after backwash. The removal efficiency decreased to 53.5% at a running time of 12 h after backwash. However, it increased rapidly to its original level afterwards. The nitrate removal efficiency was 97.2% before backwash at a HRT of 0.35 h with no decline observed in denitrification efficiency at running times of 0.5–3 h after backwash. The nitrate removal efficiency decreased to 37.7% at a running time of 6 h after backwash, but increased to its original level later with no fluctuation.

The biofilter was most likely to have been affected by backwash when operated at an HRT of 0.5 h, nonetheless denitrification efficiency returned to the normal level in 12 h after backwash. Little effect of backwash was observed on denitrification efficiency when the biofilter operated at HRTs of 0.4 and 0.35 h. It may be attributed to lower total biomass in the biofilter when the biofilter operated at a HRT of 0.5 h. Meanwhile, the loss of biomass was comparable at three different HRTs, thus the loss of biomass accounted for a larger proportion of the total biomass at a HRT of 0.5 h. The consumption of energy and backwash water was far lower than that of a conventional biofilter using sand, ceramsite or anthracite as filtration media, because the duration and intensity of air and water backwash were far less than that of the conventional biofilters applied by previous research (Schöntag & Sens 2014; Schöntag et al. 2015). The operational cycle of the biofilter was about 3 days in this study, suggesting that the water consumption rate of backwash was about 0.2–0.3%.

**CONCLUSIONS**

Nitrogen removal in micro-polluted surface water using polystyrene beads as the filtration media in an anoxic biofilter on a pilot scale was investigated. To reach the same treatment level under different regimes of temperature and HRT, the land area required for the single-stage biofilter was much smaller than the biofilter-ecological gravel bed combined process in the previous study. Besides, there was no secondary pollution caused by the addition of the external carbon source in the single-stage biofilter process. Finally, the water consumption rate of backwash was approximately one order of magnitude smaller than that of a biofilter equipped with ceramsite. Therefore, the single-stage biofilter process using polystyrene bead as filtration media had proved to be a promising alternative for removing nitrate from micro-polluted surface water.

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