Optimization of organics and nitrogen removal in intermittently aerated vertical flow constructed wetlands: Effects of aeration time and aeration rate

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Abstract
In this study, to optimize aeration for the enhancement of organics and nitrogen removal in intermit- tently aerated vertical flow constructed wetlands (VF CWs) for treating domestic wastewater, the experimental VF CWs were operated at different aeration time (1 h d^{-1}, 2 h d^{-1}, 4 h d^{-1}, 6 h d^{-1}, 8 h d^{-1} and 10 h d^{-1}) and aeration rate (0.1 L min^{-1}, 0.2 L min^{-1}, 0.5 L min^{-1}, 1.0 L min^{-1} and 2.0 L min^{-1}) to investigate the effect of artificial aeration on the removal efficiency of organics and nitrogen. The results showed that the optimal aeration time and aeration rate were 4 h d^{-1} and 1.0 L min^{-1}, which could create the appropriate aerobic and anoxic regions in CWs with the greater removal of COD (97.2%), NH_4-N (98.4%) and TN (90.6%) achieved simultaneously during the experiment. The results demonstrate that the optimized intermittent aeration is reliable option to enhance the treatment performance of organics and nitrogen at a lower operating cost.

Keywords: Constructed wetlands; Intermittent aeration; Denitrification; Nitrification;Organic matter removal

1. Introduction

With the rapid urbanization and economic growth especially in developing countries, the decentralized domestic wastewaters in vast rural areas is generally discharged directly into water bodies due to the inadequate investment of major municipal wastewater treatment infrastructures, thus resulting in negative environmental consequences (Ongley et al., 2010; Wu et al., 2011, 2015a; Shao et al., 2014). Constructed wetland (CW), as the one of the widely used ecological technologies, has been attracted more attention as the alternative solutions for wastewater treatment in these years, owing to the advantages of good efficiency, low cost and low maintenance (Vymazal, 2011; Feng et al., 2012; Chyan et al., 2013; Wu et al., 2015b). According to water level, CWs can be classified as free water surface (FWS) CWs and subsurface flow (SSF) CWs which could be further divided into vertical flow (VF) and hori- zontal flow (HF) CWs. Among those kinds of CWs, VF CWs have been widely used for decentralized sewage treatment because of a higher oxygen transfer rate and adaption to cold climate (García et al., 2010; Saeed and Sun, 2012; Wu et al., 2015b).

During wastewater treatment in CWs, biological removal pro- cesses (i.e. microbial processes) play an important role in the removal of organics and nitrogen (Saeed and Sun, 2012). Oxygen availability was recognized as the crucial influencing factor for or- ganics and nitrogen removal (Jia et al., 2011; Li et al., 2014). How- ever, limited oxygen supply and transfer capacity in traditional VF CWs cannot generally meet the requirement for the complete removal processes of organic matter and nitrogen, particularly for treating high-strength wastewaters (Saeed and Sun, 2012; Wu et al., 2015c). Therefore, in order to improve the availability of ox- ygen and in turn to enhance the removal efficiency of VF CWs, artificial aeration (including continuous aeration and intermittent aeration) has been proposed as a solution to enhance the oxygen availability in CWs. Boog et al. (2014) indicated that artificial aeration increased the oxygen concentrations in both continuously and intermittently aerated VFCWs, and aeration significantly increased the removal of organic matter and TN. Fan et al. (2013a) reported that VF CW with intermittent-aeration achieved high removal of organic pollutants, NH_4-N and TN simultaneously. Moreover, artificial aeration was applied in hybrid HF CW systems to enhance the treatment performance, which showed that aeration greatly improved organics and nitrogen removal than typical HF CW (Li et al., 2014). Several studies have also focused on the effects of aeration mode, aeration period, aeration position, hydraulic loading rate and C/N ratios on the pollutants removal in aerated CWs (Jia et al., 2011; Fan et al., 2013b; Dong et al., 2012; Wang et al., 2015; Wu et al., 2015c). Nevertheless, considering the concept of sustainability based on costebenefit analysis, CWs using artificial aeration requires additional energy input and increases the lifecycle cost, even though this approach can greatly improve treatment performance (Wu et al., 2015c). Therefore, the optimization of artificial aeration for the contaminant removal in CWs would be of great help to design more effective and sustainable CWs, and should be extensively investigated.

Due to the limited knowledge in previous literatures, the aim of this study was to investigate the optimization of organics and nitrogen removal in VF CWs for treating domestic wastewater by the use of intermittent aeration. Effects of aeration time and aeration rate on the oxygen distribution in CWs were evaluated. Moreover, the enhancing organics and nitrogen removal performance in aerated CWs was examined in detail. It is expected that the results from this study will offer a reference for successful application of intermittently aerated CWs at a low capital cost.

2. Material and methods

2.1. Site and system description

The experiment was carried out in Baihua Park in Jinan, China as described in detail by Wu et al. (2015b). Eleven parallel microcosm VF CWs were constructed for treating simulated domestic wastewater in this study. Six systems were for the optimization experiment of aeration time and the others were for the optimization experiment of aeration time. Each system was made of PVC plastic pipe with a height of 65 cm and a diameter of 20 cm. Multi-dimensional gradation of the substrate and porous air spargers were used for oxygen supply and oxygen diffusion as described in detail by Fan et al. (2013c). A vertical perforated PVC pipe was installed into the substrate in the center of VF CWs in order to measure various physical and chemical parameters in situ. Each system was planted in May 2014 with healthy P. australis (at a density of eight rhizomes per system). The systems were submerged in tap water immediately after planting to allow the development of plants and microbes until June 2014, and then the systems started operation.

2.2. Experimental procedure

In June 2014, the systems were mainly mature, and were continuously fed with wastewater to start the experiment. To minimize the influence of the fluctuation of influent water quality, the influents were synthetically prepared using sucrose, $(NH_4)_2SO_4$, KH_2PO_4, $MgSO_4$, $FeSO_4$ and $CaCl_2$ in this study. Table 1 shows the composition of the synthetic influent in terms of COD, NH_4-N, NO_3^--N, NO_2^-N,TN and TP. In the optimization experiment of aeration time, six systems were operated and intermittently aerated with an airflow rate of 1.0 L min^{-1} for 1 h d^{-1}, 2 h d^{-1}, 4 h d^{-1}, 6 h d^{-1}, 8 h d^{-1} and 10 h d^{-1}, respectively. Based on the optimal aeration time, the other systems were operated and intermittently aerated with the aeration rate of 0.1 L min^{-1}, 0.2 L min^{-1}, 0.5 L min^{-1}, 1.0 L min^{-1} and 2.0 L min^{-1} for the optimization experiment of aeration time. The hydraulic retention time (HRT) was 72 h, which just a cycle in this study, and the depth of water in each system was approximately 60 cm. Sequencing fill-and-draw batch mode was used for influent mode. At about 8:00 am on the first day of each cycle, the influent was supplied in batch mode into each VFCW within 15 min. Effluent was discharged from the outlets at the bottom of VFCWs. The average air temperature generally ranged from 23 ^{o}C to 32 ^{o}C during the experimental period in this study.

2.3. Sampling and analysis

Water samples of influent and effluent at different time were taken to analyze the transformation of organics and nitrogen in eleven reactors. The samples were taken to the laboratory and analyzed immediately for NH_4-N, NO_3^--N, NO_2^--N and TN according to standard methods (APHA, 2005). COD was measured by a HACH DR 2008™ Spectrophotometer, USA. In each cycle, after the influent was supplied into each CW, dissolved oxygen (DO) was measured at the midpoint of the water depth from the vertical perforated PVC pipe by a DO meter (HQ 30d 53LED™ HACH USA) until effluent was discharged from the CW at the end of the cycle.

2.4. Statistical analysis

All statistical analyses were performed by the statistical program SPSS 11.0 (SPSS Inc., Chicago, USA). The tables and figures show the results of averaged data, Two-sample t-tests were used to evaluate the significance of differences between means. In all tests, differences and correlations were considered statistically significant when $P < 0.05$.

3. Results and discussion

3.1. Optimization of aeration time for treatment performance

3.1.1. DO distribution in systems during experimental cycle

Fig. 1 shows the cyclic distribution of DO in the intermittently aerated VF CWs with different aeration times. According to Fig. 1, intermittent aeration enhanced the oxygen availability for all wetland systems during the period of aeration with various aeration times, however, the variation and fluctuation of DO concentrations was observed to be significantly distinct in different systems during the course of different aeration times. The influent DO concentration was approximately 7.65 mg L^{-1}, and the DO concentrations in all systems decreased immediately in the initial phase due to fast consumption of oxygen for the removal of degradable pollutants. While the alternate increase (6e8 mg L^{-1}) and decrease of DO concentrations were developed as the sequence of intermittent aeration was applied, indicating that the cyclic anaerobic and aerobic conditions were formed successfully in

Table 1
Water quality parameters of the influent during the experimental period.

Parameters	COD	NH_4^+-N	NO_3^--N	TN	TP	DO	pH
Influent (mg L^{-1})	426.23 ± 13.83	39.65 ± 0.61	4.35 ± 0.17	44.12 ± 0.79	4.52 ± 0.35	7.65 ± 1.12	7.58 ± 0.82

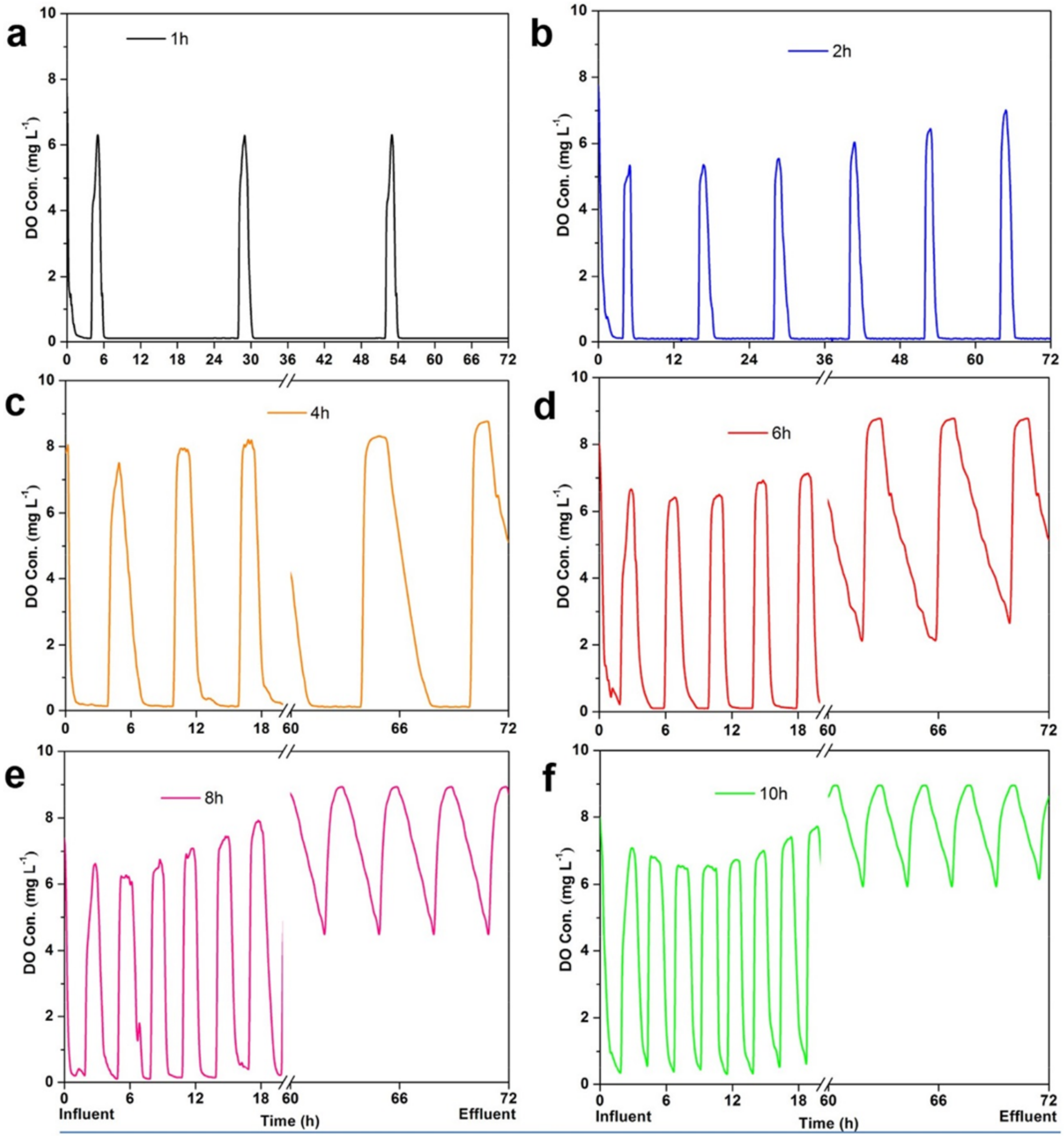

Fig. 1. DO profile in the typical cycle in intermittently aerated VF CWs with different aeration times (a: 1 h d^{-1}, b: 2 h d^{-1}, c: 4 h d^{-1}, d: 6 h d^{-1}, e: 8 h d^{-1} and f: 10 h d^{-1}) at the airflow rate of 1.0 L min^{-1}.

wetland systems. In addition, it was found that the short aeration times (1 h d^{-1} and 2 h d^{-1}) resulted in a relatively long period of anoxic conditions (DO < 1 mg/L) due to inadequate oxygen supplement. A predominantly aerobic environment (DO > 2 mg/L) has been maintained in CWs with the long aeration times (6 h d^{-1}, 8 h d^{-1} and 10 h d^{-1}) during the later operating period when artificial aeration was stopped. It can be suggested that under those aeration intensity in this study was excess which was unsuitable for achieving complete nitrogen elimination. By contrast, the variation of DO concentration in the system under the aeration time of 4 h d^{-1} could be the proper oxygen distribution for creating aerobic, anoxic, and anaerobic zones that might favor nitrification and denitrification cyclically.

3.1.2. Organics removal

Organics removal in the intermittently VF CWs aerated with an airflow rate of 1.0 L min^{-1} for different aeration times was monitored during the course of this study. As shown in Fig. 2a, all the aerated wetlands significantly reduced COD concentration to be less than 30 mg L^{-1} when the influent COD concentration was 426.23 mg L^{-1} (Table 1). The average COD removal efficiencies of six different VF CWs all exceeded 90%, that is, 93.6%, 94.9%, 96.8%, 98.6%, 98.9%, and 99.3% for the aeration time of 1 h d^{-1}, 2 h d^{-1}, 4 h d^{-1}, 6 h d^{-1}, 8 h d^{-1} and 10 h d^{-1}, respectively, indicating that artificial aeration can improve COD removal efficiency remarkably. However, it is clearly shown that the COD removal efficiency was improved slightly with the increasing of aeration time. No significant differences ($p > 0.05$) were found between the COD removal efficiency of the experimental CWs with aeration time set at 6 h d^{-1}, 8 h d^{-1} and 10 h d^{-1}. A similar result of organics removal in CWs using artificial aeration has also been reported in many studies (Dong et al., 2012; Fan et al., 2013b; Wang et al., 2015). Organic matters could be mainly degraded aerobically and anaerobically in VF CWs (Saeed and Sun, 2012; Wu et al., 2015c). The high COD

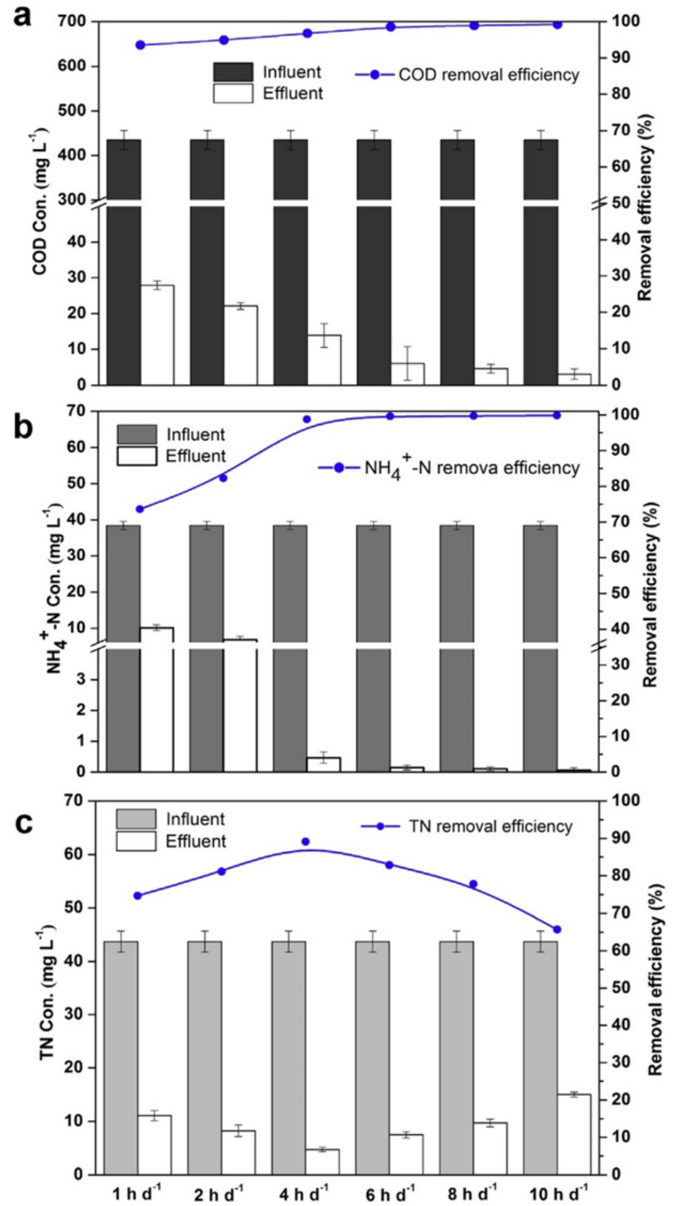

Fig. 2. Characteristics of COD, NH_4-N and TN removal in intermittently aerated VF CWs with different aeration times (1 h d^{-1}, 2 h d^{-1}, 4 h d^{-1}, 6 h d^{-1}, 8 h d^{-1} and 10 h d^{-1}) at the airflow rate of 1.0 L min^{-1}.

removal efficiency in this experiment may be explained by the following reasons. First, compared with the real domestic wastewater, organics in the influent during our experiment was simplex and easily biodegraded. On the other hand, only short-term artificial aeration promoted the decomposition of organics in all the systems immediately in the initial operating phase. Therefore, increasing artificial aeration time did not enhance the efficiency of COD significantly due to the reduction of organic matters (Saeed and Sun, 2012), which was also consistent with the change of DO concentration.

3.1.3. Nitrogen removal

The effects of aeration times on the removal of NH_4-N in VF CWs were also investigated, and as shown in Fig. 2b, nitrification was well achieved in wetland systems by intermittent aeration. However, there were distinct differences among systems with different aeration times. The effluent NH_4-N concentrations in various systems were 10.14 mg L^{-1}, 6.82 mg L^{-1}, 0.47 mg L^{-1}, 0.15 mg L^{-1}, 0.11 mg L^{-1} and 0.06 mg L^{-1} for the aeration time of 1 h d^{-1}, 2 h d^{-1}, 4 h d^{-1}, 6 h d^{-1}, 8 h d^{-1} and 10 h d^{-1}, respectively. As the aeration time increased from 1 h d^{-1} to 4 h d^{-1}, NH_4-N removal efficiency in the intermittently aerated CWs was significantly improved from 73.6% to 98.8%. Whereas NH_4-N removal efficiency rose slightly with the continually increase of the aeration time. Concerning TN removal (Fig. 2c), differences between systems with different aeration times were even more evident. Speciously, average TN removal efficiencies in different systems were within the range of 65.7%e89.2%. The TN removal efficiency in CWs was significantly improved to be 89.2% with the increasing aeration time (from 1 h d^{-1} to 4 h d^{-1}), but as the aeration time increased from 6 h d^{-1} to 10 h d^{-1}, the TN removal efficiency dropped from 82.9% to 65.7%. This phenomenon could also be explained by the transformation of nitrogen in the systems (Fig. 3). With regard to nitrites and nitrates, the lower concentrations were found in the effluents when aeration time was 4 h d^{-1} in this experiment, clearly indicating that the proper aeration was the key factor for enhancing nitrification and denitrification simultaneously. According to the classical transformation and removal routes of nitrogen, nitrification ($NH_4 \rightarrow NO_2^- \rightarrow NO_3^-$) coupled with canonical denitrification ($NO_3^- \rightarrow NO_2^- \rightarrow NO \rightarrow N_2O \rightarrow N_2$) was the major pathway of nitrogen removal (Saeed and Sun, 2012; Wu et al., 2015c). Rising oxygen availability in CWs might favor aerobic chemo-autotrophic microbial process of nitrification (Maltais-Landry et al., 2009; Fan et al., 2013a). However, the higher oxygen supply to the wetland reduced the anoxic environment for denitrification to take place, which could be proved from the detected result in Fig. 3 that NO_3^--N increased to be the dominant form as the aeration time increased from 6 h d^{-1} to 10 h d^{-1} due to excessive aeration. Additionally, limited available organic matters might restrain the denitrification process (Saeed and Sun, 2012; Wu et al., 2015b).

On the whole, based on the present data, the appropriate aerobic and anoxic regions could be created in the aerated systems particularly in CWs with an aeration time of 4 h d^{-1}, thus strengthening the effectiveness of the microbes in reducing degradable organics and facilitating nitrification and denitrification, and thereby achieving simultaneous organics reduction and nitrogen elimination. The aeration time of 4 h d^{-1} might be suitable for further direct comparison of different aeration rates regarding organics and nitrogen removal.

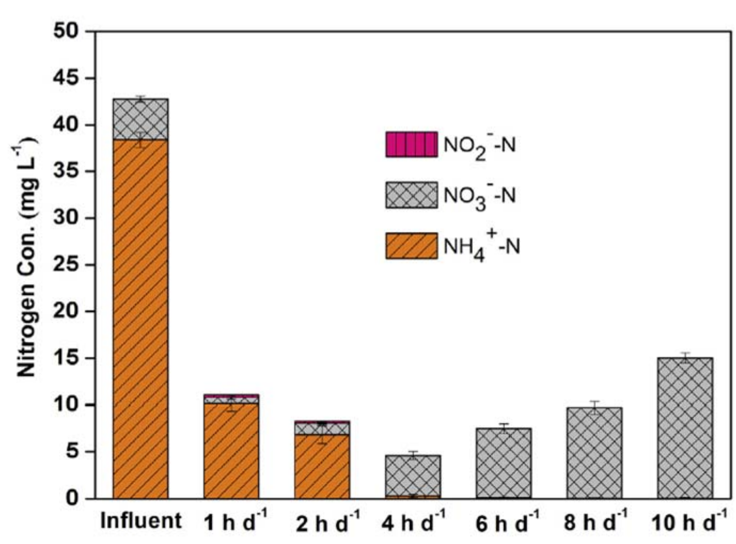

Fig. 3. Dynamic transformations of nitrogen (NH_4^+-N, NO_3^--N and NO_2^--N) in intermittently aerated VF CWs with different aeration times (1 h d^{-1}, 2 h d^{-1}, 4 h d^{-1}, 6 h d^{-1}, 8 h d^{-1} and 10 h d^{-1}) at the airflow rate of 1.0 L min^{-1}.

3.2. Optimization of aeration rate for treatment performance

3.2.1. DO distribution in systems during experimental cycle

The cyclic distribution of DO in the intermittently aerated VF CWs with different aeration rates is shown in Fig. 4. When aeration time was set at 4 h d^{-1}, intermittent aeration under various aeration rates significantly increased the oxygen availability in all wetland systems, and the variation also proved the successful formation of cyclic anaerobic and aerobic conditions within the substrate. However, the DO fluctuation range under various aeration rates was observed to be distinct, as shown in Fig. 4, Steep decrease of the DO concentration occurred in the first 4 h mainly due to fast consumption of DO for organic matters degradation, and then the DO concentration could be 2e9 mg L^{-1} during aeration time and lower than 0.3 mg L^{-1} when artificial aeration was turned off. It should be noted that the DO concentrations in intermittent aerated systems increased to be about 2 mg L^{-1} at the aeration rates of 0.1 L min^{-1} and 0.2 L min^{-1} which would be not enough to remove organics and NH_4-N. While the aeration rate was set at of 2 L min^{-1}, the DO concentrations in the system peaked at 9 mg L^{-1}, thus limiting nitrification and denitrification simultaneously. Nitrification takes place in aerobic region while denitrification occurs with anaerobic/anoxic condition (Kadlec and Wallace, 2009).

3.2.2. Organics removal

Organics removal in VF CWs intermittently aerated at different aeration rates (0.1 L min^{-1}, 0.2 L min^{-1}, 0.5 L min^{-1}, 1.0 L min^{-1} and 2.0 L min^{-1}) was shown in Fig. 5a, and the COD removal efficiency was above 90% in all systems, which indicated that the aeration of 4 h d^{-1} at different airflow rates entailed significant increment of dissolved oxygen and a consequent increase in COD removal. As shown in Fig. 5a, the average effluent COD concentrations were 38.47 mg L^{-1}, 25.31 mg L^{-1}, 18.73 mg L^{-1}, 12.33 mg L^{-1} and 6.96 mg L^{-1}, with the removal efficiencies of 91.2%, 94.2%, 95.7%, 97.2% and 98.4% for various aeration rates. It is obvious that with the increasing of airflow rate, the COD removal efficiencies were gradually promoted because of more oxygen supply, with the highest removal efficiency achieved at the aeration rate of 2.0 L min^{-1}. This phenomenon was consistent with the previous results that sufficient and efficient oxygen supply always enhanced organic matters degradation (Vymazal and Kröpfelová, 2009; Saeed and Sun, 2012). Ong et al. (2010) also reported that sufficient

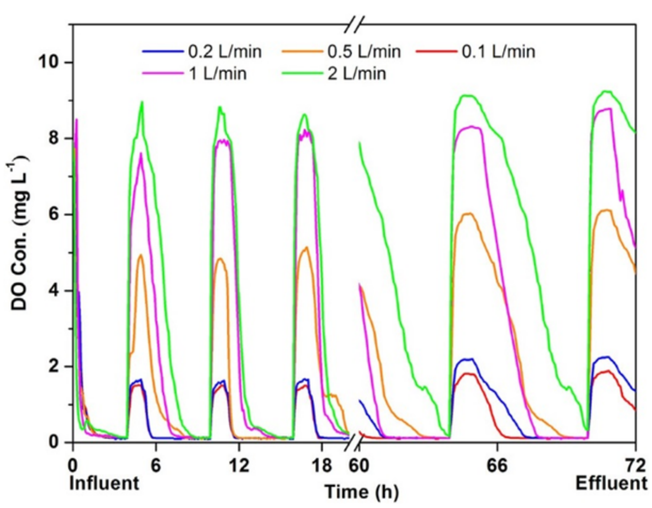

Fig. 4. DO profile in the typical cycle in intermittently aerated VF CWs with different aeration rates (0.1 L min^{-1}, 0.2 L min^{-1}, 0.5 L min^{-1}, 1.0 L min^{-1} and 2.0 L min^{-1}) at the aeration time of 4 h d^{-1}.

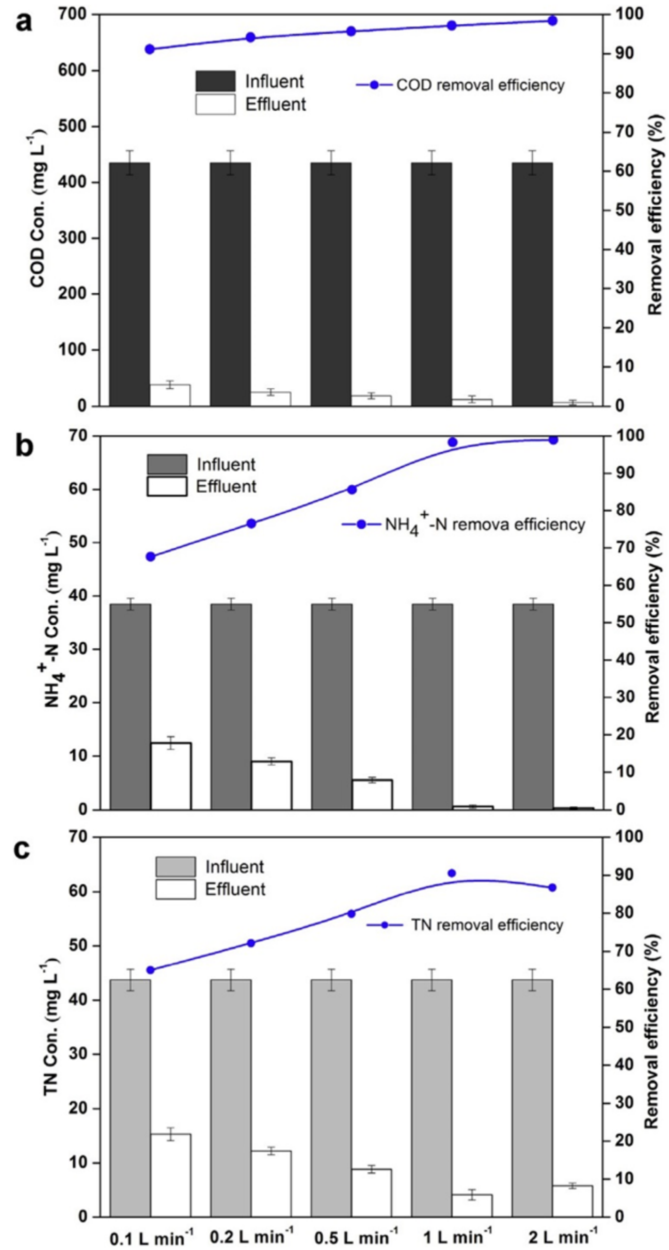

Fig. 5. Characteristics of COD, NH_4-N and TN removal in intermittently aerated VF CWs with different aeration rates (0.1 L min^{-1}, 0.2 L min^{-1}, 0.5 L min^{-1}, 1.0 L min^{-1} and 2.0 L min^{-1}) at the aeration time of 4 h d^{-1}.

oxygen supply could greatly increase the performance of aerobic biochemical oxidation and improved the COD removal efficiency.

3.2.3. Nitrogen removal

The removal of NH_4-N and TN in VF CWs with different aeration rates were also investigated, and the results were illustrated in Fig. 5a and b, respectively. As the oxygen availability and distribution was improved, the removal of NH_4-N and TN in CWs with various aeration rates was greatly enhanced. However, a significant varying trend could be found between removal efficiencies of NH_4-N and TN. The removal of NH_4-N occurred mainly by means of nitrification, and this process required the establishment of oxygen-enriched zones (Jia et al., 2011). Similar to the COD removal, the higher aeration rate resulted in the increase of NH_4-N removal efficiency. The concentration of NH_4-N in the effluent ranged

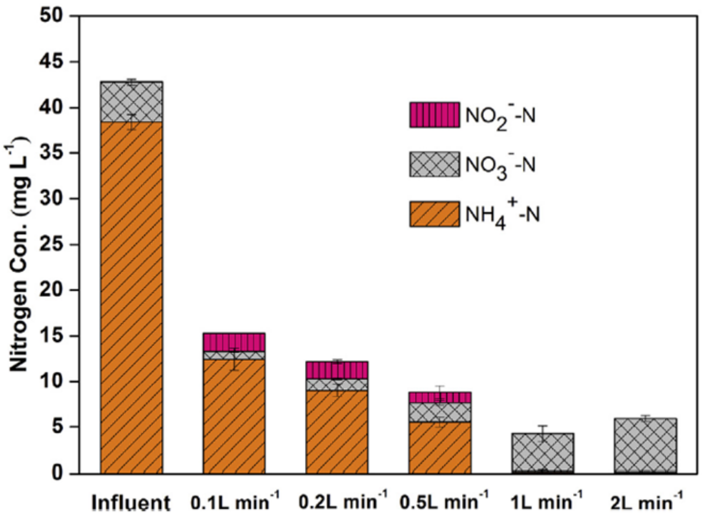

Fig. 6. Dynamic transformations of nitrogen (NH_4^+-N, NO_3^--N and NO_2^--N) in intermittently aerated VF CWs with different aeration rates (0.1 L min^{-1}, 0.2 L min^{-1}, 0.5 L min^{-1}, 1.0 L min^{-1} and 2.0 L min^{-1}) at the aeration time of 4 h d^{-1}.

between 0.38 mg L^{-1} and 12.44 mg L^{-1} due to variations in the aeration rate, and correspondingly the removal efficiencies of NH_4-N were in the range of 67.6e99.0%. No significant differences ($p > 0.05$) were detected between the NH_4-N removal of systems with the aeration rate of 1.0 L min^{-1} and 2.0 L min^{-1}. While concerning the TN removal, variation was contrary to that of NH_4-N removal. The removal efficiencies of intermittent aerated systems were significantly promoted to be 90.6% as aeration rate increased from 0.1 L min^{-1} to 1.0 L min^{-1}, and then a decline was observed at the aeration rate of 2.0 L min^{-1}. This phenomenon could be explained by the fact that higher oxygen concentration and lack of organic matter might restrain denitrification even though NH_4-N removal was enhanced due to excess aeration (Saeed and Sun, 2012; Dong et al., 2012). It was also reported that the low TN removal could be caused by the unsatisfactory denitrification even though nitrification was performed well (Li et al., 2014). Fig. 6 illustrates the variation of NH_4-N, NO_3^--N and NO_2^--N in different systems to determine the extent of nitrification and denitrification at different aeration rates. As indicated in Fig. 6, the effluent NH_4-N and NO_2^--N concentrations of the aerated CWs with the aeration rate of 1.0 L min^{-1} decreased to a very low level, which was attributed to the provided intermittent aeration. However, NO_3^--N concentrations in the experimental wetland at the aeration rate of 2.0 L min^{-1} increased slightly, which clearly showed the extra aeration would cause the incomplete nitrification (Tanner et al., 2002; Fan et al., 2013a). This means that, the proper aeration is necessary to maintain aerobic and anaerobic zones, particularly when simultaneous nitrification-denitrification is desirable in a single wetland system (Hu et al., 2012; Saeed and Sun, 2012). Therefore, on the basis of aeration efficiency, operating costs and treatment performance, VF CWs intermittently aerated for 4 h d^{-1} at the rate of 1.0 L min^{-1} would be an optimal strategy to reduce organics and nitrogen in wastewaters.

4. Conclusions

The optimization of intermittent aeration in VF CWs significantly increased the oxygen availability, but also successfully created alternate aerobic and anaerobic conditions, and thereby achieving simultaneous organics reduction and nitrogen elimination. High removal efficiencies of COD (97.2%), NH_4-N (98.4%) and TN (90.6%) were obtained simultaneously in VF CWs intermittently aerated for 4 h d^{-1} at the rate of 1.0 L min^{-1}. This study provided encouraging strategy for reducing organics and nitrogen in wastewaters for intensified CWs. Nevertheless, further research is needed in order to evaluate the long-term treatment performance in full scale systems for successful application.

Acknowledgments

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