Decentralized domestic wastewater treatment using intermittently aerated vertical flow constructed wetlands: Impact of influent strengths

Haiming Wu, Jinlin Fan, Jian Zhang, Huu Hao Ngo, Wenshan Guo, Zhen Hu, Shuang Liang

*College of Resources and Environment, Northwest A & F University, Yangling, Shaanxi 712100, PR China
bShandong Key Laboratory of Water Pollution Control and Resource Reuse, School of Environmental Science & Engineering, Shandong University, Jinan 250100, PR China
1National Engineering Laboratory of Coal-Fired Pollutants Emission Reduction, Shandong University, Jinan 250061, PR China
4School of Civil and Environmental Engineering, University of Technology Sydney, Broadway, NSW 2007, Australia

HIGHLIGHTS
- Intermittent aeration was effective in COD and TN removal from domestic sewage.
- Influent strength did not affect pollutant removal in intermittent aerated VFCWs.
- Removal performance was higher in aerated VFCWs compared with non-aerated VFCWs.

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ABSTRACT
In this study, the removal performances of organic pollutants and nitrogen in vertical flow constructed wetlands (VFCWs) with and without intermittent aeration fed with different strengths of influent were evaluated as a possible treatment for decentralized domestic wastewater in northern China. The intermittent aeration strategy not only significantly increased removal efficiencies of organic pollutants and ammonium nitrogen (NH₄-N), but also successfully created alternate aerobic and anaerobic conditions resulting in high total nitrogen (TN) removal. Moreover, increasing influent strength did not affect the removal efficiencies of organic matters and nitrogen in aerated VFCWs. Compared with non-aerated VFCWs, much higher removal of organic pollutants (96%), NH₄-N (98%), and TN (85%) was obtained simultaneously in intermittent aeration VFCWs, especially at high influent strengths. The results suggest that the intermittent aeration could be an appropriate strategy for achieving the high removal performance in VFCWs, especially for in-situ treatment of high strength decentralized domestic wastewaters.

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1. Introduction
In China, the decentralized domestic wastewaters in vast villages, small towns, scattered residential areas and expressway service areas were, in most cases, beyond the coverage of major municipal wastewater treatment systems. They were often poorly collected and directly discharged, greatly threatening the natural environment (Zhang et al., 2005; Ye and Li, 2008; Wu et al., 2012). Constructed wetlands (CWs) have long been acknowledged as a promising technology for on-site treatment of decentralized domestic wastewaters due to their easy management and maintenance. In particular, vertical flow constructed wetlands (VFCWs) appears to be a preferred mode because of their small footprint (Zhang et al., 2009) and good efficiency (Jia et al., 2010). Although satisfactory organic removal performances can be achieved in VFCWs treating low-strength wastewaters, the total nitrogen (TN) removal efficiency always remains as a major challenge for conventional CWs, including VFCWs. For instance, the TN removal efficiency in conventional VFCWs was reported to be merely around 50% under the nitrogen loading of around 2 g m⁻² day⁻¹ (Vymazal, 2007), which was far from satisfactory and needed for further improvement.

Nitrification and denitrification is widely acknowledged as the main pathway for biological nitrogen removal in CWs. It needs both aerobic and anaerobic environments, which, however, can hardly be achieved simultaneously in CWs. Continuous artificial aeration strategies has been adopted to achieve complete nitrification in many studies (Malais-Landry et al., 2009; Org et al., 2010).
However, the higher dissolved oxygen (DO) concentration and the greater depletion of organic pollutants, the side-effect of continuous aeration, significantly limiting the denitrification process performed by anaerobic heterotrophic microorganisms. Consequently, TN removal efficiency remains far from satisfactory (Maltas-Landry et al., 2009; Nivala et al., 2007). Instead of continuous artificial aeration, intermittent aeration mode appeared to be a more cost-effective strategies (Fan et al., 2013a), which not only saved operating cost but also greatly increased TN removal efficiency by creating favorable conditions (i.e. alternate aerobic and anaerobic conditions) for nitrification and denitrification simultaneously.

The treatment performance of CWs, in terms of both organic pollutants and nitrogen, is heavily dependent on influent strength (Saeed and Sun, 2012). Nevertheless, the influent concentration was actually changing rather than stable in most CWs. In particular, when conventional CWs were applied for the treatment of high-strength wastewater, the effluent quality was often not satisfied and far below the discharge standards (Knight et al., 2000). So far, however, few attempts have been made to evaluate the effect of influent strength on the treatment performance in VFCWs, especially in intermittent aerated VFCWs. It is hypothesized that the intermittent aeration strategy would bring more advantages for the treatment of high-strength wastewater due to the adequate oxygen supply.

Therefore, eight batch-operated VFCWs with and without intermittent aeration were operated in parallel for the treatment of simulated domestic wastewaters with different influent strengths in this study. The effects of intermittent aeration and influent strength on the removal performances of COD, NH₃-N, and TN in VFCWs were systematically investigated. The acquired results may contribute toward a more complete understanding of wastewater treatment performance in VFCWs, particularly in the combination of intermittent aeration and different influent strengths.

2. Methods

2.1. Characterization of microcosm wetlands

The experiments were carried out in Baihua Park in Jinan, northern China (36°40′36″N, 117°03′42″E), with sub-humid continental climate. The climate is characterized by an annual precipitation of 670.7 mm and an average temperature of 14.7 °C. Eight microcosm VFCWs were performed in parallel under a transparent rain shelter in March 2013. Fig. 1 shows schematic diagram of the experimental VFCW. Each mesocosm was 85 cm in height and 20 cm in diameter. Multi-dimensional gradation of the substrate was adopted to favor oxygen diffusion within the substrate with an average gravel bed porosity of 40% (Fan et al., 2013a). A vertical perforated PVC pipe (65 cm in length and 3 cm in diameter) was inserted into the substrate in the center of VFCWs in order to measure various physical and chemical parameters in situ. There were porous air spargers installed at the bottom of VFCWs for oxygen supply. In this study, the emergent plants employed were Phragmites australis at a density of eight rhizomes per unit. After planting, VFCWs were kept flooded for about 1 month until wetland plants were well established. The experiment began in May and lasted for more than 4 months. During the whole experimental period, temperature was 14–29 °C.

2.2. Experimental procedure

In order to minimize variability in the experiment, synthetic wastewater was used in this study. The influent strength was manipulated by changing the content of sucrose, (NH₄)₂SO₄ and KH₂PO₄, and formulated four different influent strengths (A, B, C and D). Synthetic wastewater composed of 215, 387, 559, 860 mg L⁻¹ sucrose, 94, 188, 282, 376 mg L⁻¹ (NH₄)₂SO₄, 8.75, 17.5, 26.3, 35 mg L⁻¹ KH₂PO₄, 10 mg L⁻¹ MgSO₄₆H₂O, 10 mg L⁻¹ FeSO₄ and 10 mg L⁻¹ CaCl₂ was used in this study. The characteristics of the influents with different strengths were shown in Table 1. The eight VFCWs were divided into four groups fed with different strengths of influent. Every two VFCWs were operated with the same influent strength, one of which was intermittently aerated with an airflow rate of 31 L m⁻² min⁻¹ for 4 h (hours 0–1, 6–7, 12–13 and 18–19) each day. Hydraulic retention time (HRT) was 72 h according to preliminary experiments. 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. Each wetland unit held 6 L wastewater when filled, and effluent was discharged from the outlets at the bottom of VFCWs.

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 eight reactors. The samples were taken to the laboratory and analyzed immediately for NH₃-N, NO₃-N, NO₂-N and TN according to standard methods (APHA, 2005). COD was measured by a HACH DR 2000™ Spectrophotometer, USA. Dissolved oxygen (DO) was measured in situ at the mid-point of the water depth from the vertical perforated PVC pipe by a DO meter (HQ 304 53LED™ HACH USA).

2.4. Statistical analysis

All statistical analyses were performed through the statistical program SPSS 11.0 (SPSS Inc., Chicago, USA). Two-sample t-tests were used to evaluate the significance of differences between means. Concentration profiles describing the removal of the pollutants as a function of time in VFCWs were fitted to the first-order k–C model (IWA, 2000; Lin et al., 2008). In all tests, differences
and correlations were considered statistically significant when $P < 0.05$.

3. Results and discussion

3.1. DO distribution during experimental cycle

The cyclic distribution of DO in the eight microcosm wetlands is shown in Fig. 2. The difference was distinct between the VFCWs with intermittent aeration and without aeration. Sharp decrease of the DO concentration was observed once the influent was pumped in. For non-aerated VFCWs, fast consumption of DO mainly occurred in the first 4 h, and then DO concentration changed slightly before drainage. As shown in Fig. 2a, DO concentration of the effluent in non-aerated VFCWs decreased with the increasing influent strength, but no significant difference was found, which were all below 0.35 mg L$^{-1}$. As a result, anaerobic region could be well developed in the four VFCWs in the most time. Limited oxygen supply was a restriction to nitrification and organic biodegradation. Although vegetable had been reported to contribute to oxygen supply (Cui et al., 2009; Taylor et al., 2010), while Dong et al. (2011) indicated that most oxygen released by roots was consumed by root respiration. As shown in Fig. 2b, in intermittent aerated VFCWs, the DO concentration could be 5–8 mg L$^{-1}$ during aeration time and lower than 0.3 mg L$^{-1}$ when artificial aeration was turned off. The DO fluctuation proved the successful formation of cyclic anaerobic and aerobic conditions within the substrate favoring nitrification and denitrification cyclically. The effluent DO concentrations in intermittent aerated VFCWs also decreased with the increasing influent strength, which was mainly due to the degradation of more nutrients and organic matters.

3.2. Impact of influent strength on organics removal

The average effluent concentrations of COD, NH$_4$-N, NO$_3$-N, TN and TP in the eight VFCWs during the experimental period are shown in Table 2. There were significant differences in VFCWs with various influent strength and operation strategies. For intermittent aerated VFCWs, organic matters degradation and nutrient removal efficiencies were much better than that of non-aerated VFCWs. In order to further probe the impact of intermittent aeration and influent strength on contamination removal, influent and effluent samples were taken at certain intervals during one cycle for the analysis of the transformation of organic matters and nitrogen.

Fig. 3 shows the COD profiles in different VFCWs. Similar to the DO profile, the COD concentration decreased rapidly in all the VFCWs immediately after feeding and changed slightly from 12 h after the beginning of the cycle. In non-aerated VFCWs, the average effluent COD concentrations were 30.83, 113.49, 196.35 and 363.87 mg L$^{-1}$, with the removal rates of 85.49%, 73.59%, 68.08% and 62.68% for various influent strengths. Limited oxygen supply always restricted organic matters degradation (Vymazal and Kröpfelová, 2000). With the increasing of influent strength, the limitation of oxygen became more obvious, resulted in decrease of COD removal rates in non-aerated VFCWs. While for intermittent aerated VFCWs, COD removal rates were 98.17%, 98.49%, 96.42% and 96.69% under the different influent strengths (A, B, C and D). Increasing influent strength did not affect the efficiency of organic matters degradation, which was significantly different with that of non-aerated VFCWs. This was also proved by the values of the reaction coefficient based on the kinetic models. The estimated first-order removal rate constants ($k$) for COD removal in intermittent aerated VFCWs were 0.27–0.41 d$^{-1}$, and increased with the influent strength rising, while $k$ value in non-aerated VFCWs was effected by the influent concentrations.

Organic matters could be degraded aerobically and anaerobically in subsurface flow constructed wetlands (Sayed and Sun, 2012). The aerobic heterotrophic bacteria played an important role.
Table 2
Characteristics of effluent and respective removal efficiencies (mean ± SD, n = 20).

<table>
<thead>
<tr>
<th>Parameters</th>
<th>Influent strengths</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>A</td>
</tr>
<tr>
<td></td>
<td>IA</td>
</tr>
<tr>
<td>COD (mg/L)</td>
<td>4.52 ± 4.11</td>
</tr>
<tr>
<td>(%)</td>
<td>99.17 ± 4.66</td>
</tr>
<tr>
<td>NH₄-N (mg/L)</td>
<td>0.34 ± 0.52</td>
</tr>
<tr>
<td>(K)</td>
<td>98.05 ± 2.02</td>
</tr>
<tr>
<td>NO₃-N (mg/L)</td>
<td>1.96 ± 1.55</td>
</tr>
<tr>
<td>(mg/L)</td>
<td>0.002 ± 0.01</td>
</tr>
<tr>
<td>TN (mg/L)</td>
<td>2.38 ± 1.86</td>
</tr>
<tr>
<td>(%)</td>
<td>69.14 ± 7.73</td>
</tr>
<tr>
<td>TP (mg/L)</td>
<td>0.17 ± 0.12</td>
</tr>
<tr>
<td>(mg/L)</td>
<td>7.29 ± 0.21</td>
</tr>
<tr>
<td>DO (mg/L)</td>
<td>7.22 ± 5.58</td>
</tr>
</tbody>
</table>

IA means intermittent aeration and NA means non-aeration.

3.3. Impact of influent strength on nitrification performance

In CWS, the transformation and removal of nitrogen could be accomplished by both classical and novel routes. The classical routes included nitrification–denitrification, plant uptake, adsorption etc. and the novel routes mainly involved partial nitrification–denitrification, Anammox etc. (Sared and Sun, 2012). While nitrification (NH₄⁺ → NO₂⁻ → NO₃⁻) coupled with canonical denitrification (NO₃⁻ → NO₂⁻ → NO → N₂O → N₂) was the major pathway of nitrogen removal. Because of limited oxygen supply in CWS, nitrification, as an aerobic chemo-autotrophic microbial process, usually became the limiting step for nitrogen removal (Maltais-Landry et al., 2009). As shown in Table 2, the average effluent NH₄-N concentration in non-aerated VFCWs was greatly higher than that of intermittent aerated VFCWs at the same influent strength. It is widely accepted that nitrification could occur with DO concentration above 1.5 mg L⁻¹ (Vymazal, 2007; Ye and Li, 2009). While in non-aerated VFCWs, the DO was rapidly consumed after feeding, and then the DO concentration was below 0.35 mg L⁻¹ in most time of the cycle causing an anaerobic environment in substrate, thus the NH₄-N removal was severely limited. The average effluent NH₄-N concentrations and NH₄-N profile in typical cycles were shown in Table 2 and Fig. 4a, respectively. It is shown that nitrification mainly occurred in the first 12 h, and the NH₄-N concentrations were still high in the effluent, which were 14.70, 29.11, 47.65 and 62.67 mg L⁻¹ for the four influent strengths, respectively. In non-aerated VFCWs, as the influent NH₄-N concentration increased from 20 to 80 mg L⁻¹, the average NH₄-N removal rate decreased from 31.05% to 21.0%, this might because increased organic matters consumed more available oxygen and further limited the nitrification process.

In order to improve the DO availability in VFCWs, artificial aeration appeared to be the most effective alternative to guarantee sufficient oxygen supply (Maltais-Landry et al., 2009). In particular, artificial aeration seemed to be the only option to realize complete nitrification when dealing with high-strength wastewater (Hu et al., 2012). Former study also found that more nitrifying bacteria and other viable bacteria were detected in intermittent aerated VFCWs than non-aerated VFCWs by FISH (fluorescence in situ hybridization analysis) (Fan et al., 2013c). Their results also indicated that the growth of ammonia-oxidizing bacteria (AOB) in VFCWs could be greatly enhanced by intermittent aeration operation. As shown in Table 2 and Fig. 4b, in intermittent aerated VFCWs, the average effluent NH₄-N concentrations were 0.34,

![Influent strength (K-C model)](image)

Fig. 3. COD profiles in VFCWs without aeration (a) and with intermittent aeration (b).

in the aerobic degradation of organic matters. A previous study (Tao et al., 2007) investigated the effects of influent strength on heterotrophic bacterial activity in mesocosm CWS, and demonstrated that a stronger influent did not show a significant inhibition on heterotrophic activity. On the other hand, Öz et al. (2010) reported that sufficient oxygen supply could greatly increase the performance of aerobic biochemical oxidation and improved the COD removal rate. This study also indicated that sufficient oxygen supply could greatly increase the organic matters degradation with high COD removal rates of above 95%, even in high influent strength VFCWs (such as influent strength C and D).
0.49, 0.58 and 0.68 mg L\(^{-1}\) for the different influent strengths, and the calculated \(k\) values for NH\(_4\)N removal in aerated VFCSs were 0.05–0.12 d\(^{-1}\). Because of the efficient oxygen supply, excessive organic matters and other oxygen-demanding nutrients seemed not to be a limitation for nitrification process. The NH\(_4\)N removal rates were all above 95% in the intermittent aerated VFCSs, which were consistent with the previous study of Fan et al. (2013a). Artificial intermittent aeration could greatly improve nitrification performance in various influent strength VFCSs.

3.4. Impact of influent strength on TN removal

Nitrification could convert nitrogen into various forms but did not achieve nitrogen removal from the wastewater (Wu et al., 2009). The nitrified nitrogen must be processed via anaerobic and heterotrophic microbial denitrification in order to be permanently removed from CWs. Many factors such as insufficient organic matters, excess oxygen and the lack of nitrate could limit denitrification process (Tanner et al., 2002; Fan et al., 2011a). As shown in Table 2, the TN removal efficiency was far from satisfactory in non-aerated VFCSs, and the effluent was still dominated by high concentration NH\(_4\)N due to limited nitrification. The average effluent TN concentrations were 13.63, 27.72, 46.15 and 61.06 mg L\(^{-1}\) at various influent strengths, and no obvious NO\(_3\)-N was detected in the four non-aerated VFCSs due to anaerobic conditions. TN profile of non-aerated VFCSs was shown in Fig. 4a, and it was similar to NH\(_4\)N profile in Fig. 4a. This result further proved that nitrification was the limiting step of nitrogen removal in CWs.

Non-aerated VFCSs could not achieve a high NH\(_4\)N removal rate with low DO concentration, which would greatly inhibit denitrification due to the insufficient supply of NO\(_3\)-N as electron acceptors. Due to more DO depletion with the increasing influent strength, the TN removal rate dropped from 43.32% to 27.06% with the increasing of TN concentration from 25 to 85 mg L\(^{-1}\), which was similar to the variation of NH\(_4\)N removal rate in VFCSs with various influent strengths.

Nitrogen removal rates in most CWs were low due to their inability to provide aerobic and anoxic conditions for nitrification and denitrification simultaneously. Malatais-Landry et al. (2009) reported that artificial aeration could increase nitrogen removal by 11–86%. While in many studies with continuous artificial aeration strategy, TN removal rates were still not satisfactory although nitrification had been complete conducted (Fan et al., 2013a; Liu et al., 2013). With the sufficient oxygen supply, denitrification usually became the limiting step for effective TN removal due to the lack of anoxic conditions and the depletion of the influent organic matters (Liu et al., 2012). In this study, intermittent aeration (4 h per day) was adopted to achieve complete nitrification. Furthermore, as shown in Fig. 2b, alternative aerobic and anaerobic conditions were well developed for nitrification and denitrification in one cycle simultaneously. After effective nitrification and the existence of favorable anaerobic conditions, an adequate amount of organic matters were needed for TN removal. Fan et al. (2013b) indicated that influent COD/N ratios would significantly influence the TN removal rates in intermittent aerated CWs, high-efficiency nitrification and denitrification simultaneously occurred in...
intermittent aerated VFHWs with high COD/N ratios, and the average TN removal rate could reach 90% with COD/N ratio of 10. In this study, the influent COD/N ratios were all above 10. High COD/N ratio could supply sufficient organic materials as electron donor for complete denitrification, and achieved high TN removal rates. As shown in Table 2 and Fig. 3b, the average effluent TN concentrations were low in all the intermittent aerated VFHWs, which were 2.38, 4.69, 6.65 and 8.56 mg L⁻¹ at four influent strengths. The k value calculated by fitting the k-β model illustrated that existence and type of aeration was likely to influence the decay coefficient. Moreover, results obtained from the kinetic models also indicated that TN removal occurred faster in aerated VFHWs (0.08–0.17 d⁻¹) although the influent concentrations were similar. As favorable anaerobic conditions and sufficient organic matters were supplied for complete denitrification, there were no obvious NO₃⁻N and NO₂⁻N accumulation in the four intermittent aerated VFHWs. The average TN removal rates were all above 85% in the intermittent aerated VFHWs with various influent strengths, these results were comparable or superior to many previous studies (Fan et al., 2013a,c; Dong et al., 2012). The differences in TN efficiencies between intermittent aerated CWs and non-aerated CWs could be partially explained by the different bacterial community compositions in CWs. Zhong et al. (2014) analyzed the shifts in bacterial community diversity and composition in different CWs, and showed that nitrate-oxidizing bacteria (NOB) and ammonia-oxidizing bacteria (AOB) had much higher abundances in aerated CWs, whereas more sequences related to sulfate-reducing bacteria and anaerobic sulfur-oxidizing bacteria were detected in the non-aerated CW.

4. Conclusions

The application of intermittent aeration in VFHWs not only significantly increased the DO concentration, but also successfully created alternate aerobic and anaerobic conditions. Regardless of influent strengths, excellent removal efficiencies of COD, NH₃-N, and TN were successfully achieved in intermittent aerated VFHWs, and significantly higher than conventional VFHWs. The results suggest that the intermittent aeration could be a valuable strategy for further increasing the treatment efficiencies and thus expanding the application of VFHWs, especially for the in-situ treatment of high strength decentralized domestic wastewaters.

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