Performance evaluation of wastewater treatment using horizontal subsurface flow constructed wetlands optimized by micro-aeration and substrate selection

Fei Zhong, Juan Wu, Yanran Dai, Dongfang Xiang, Shuiping Cheng and Hongjiu Ji

ABSTRACT

The effects of micro-aeration and substrate selection on domestic sewage treatment performance were explored using three pairs (with or without micro-aeration) of horizontal subsurface flow (HSSF) constructed wetlands (CWs) filled with zeolite, ceramsite or quartz granules. The individual and combined effects of micro-aeration and substrate selection on the purification performance of the experimental-scale HSSF CWs were evaluated. The results showed that micro-aeration significantly increased the treatment efficiencies for chemical oxygen demand, total nitrogen, total phosphorus (TP), ortho-phosphate (PO$_4$-P) and ammonium nitrogen (NH$_4$-N) using HSSF CWs, while the substrate selection significantly affected the TP, PO$_4$-P and NH$_4$-N removal efficiencies ($p < 0.05$). A two-way analysis of variance (ANOVA) indicated that there was a significant interaction term (i.e. micro-aeration × substrate selection) for NH$_4$-N removal ($p < 0.05$). Among the three substrates, ceramsite was the best substrate for the treatment of domestic sewage using HSSF CWs. Therefore, the results of this study suggest that a ceramsite-filled HSSF CW with micro-aeration could be the optimal configuration for decentralized domestic sewage treatment.

Key words | constructed wetlands, domestic sewage, substrate selection, micro-aeration

INTRODUCTION

Constructed wetlands (CWs) have been reported to treat a variety of wastewater types (Vymazal 2009; Calheiros et al. 2010; Zhong et al. 2011), providing a cost-effective and flexible option for wastewater treatment. Therefore, CWs are appealing for applications in developing countries (Dong et al. 2012). In China, domestic sewage treatment was proposed as the main purpose of CW projects (Zhang et al. 2012). Ammonium nitrogen (NH$_4$-N) is a predominant contaminant of domestic sewage and has negative effects on aquatic systems; previous studies have shown that it must be treated under aerobic conditions (Vymazal & Kröpfelová 2011; Sun et al. 2012). Unfortunately, the oxygen demand often exceeds the rate of oxygen transfer into CWs; this situation is most common in horizontal subsurface flow (HSSF) CWs (Nivala et al. 2012). The dominance of anaerobic or anoxic conditions inhibits the nitrifying activity and results in a relatively low CW nitrogen (N) removal rate (Paulwetter et al. 2009). The redox conditions in CWs should be manipulated to improve N removal efficiency. Aeration was recommended to optimize the redox conditions in CWs, and the effects of aeration location (Butterworth et al. 2013; Li et al. 2014), aeration intensity (Zhang et al. 2010; Zhong et al. 2014) and aeration pattern (Redmond et al. 2014) on the treatment efficiencies of CWs were reported. Taking account of the operational convenience and costs, front aeration with a low-power air pump might be a promising approach to establish heterogeneous redox conditions in HSSF CWs (Zhong et al. 2014). In HSSF CWs, phosphorus (P) removal mainly depends on the physical–chemical and hydrological properties of the substrate, because P is mainly absorbed by or precipitated in substrate (Vohla et al. 2011). Generally, the life expectancy of subsurface flow CWs, especially as concerns phosphorus removal, depends on the substrates used (Sakadevan & Bavor 1998; Gruneberg & Kern 2001). Selecting a substrate with a high P-sorption capacity is therefore important to
obtain a sustained P-removal by CWs (Arias et al. 2001). The P-binding capacities of various materials were compared, and some materials were reported to have profound effects on the treatment efficiencies of CWs separately (Brix et al. 2001). However, aeration and substrate selection have not been studied under controlled conditions to isolate their individual and combined effects.

The objective of this study was to assess the individual and combined effects of aeration and substrate selection on the purification performance of HSSF CWs. Related research would be beneficial for developing detailed design and operation criteria and to accelerate the effective application of CWs.

MATERIALS AND METHODS

CW

A schematic diagram of the HSSF CW used in this study is shown in Figure 1. The wetland cells were rectangular tanks constructed of polyvinylchloride. The dimensions were 1.2 m in length, 0.4 m in width and approximately 0.7 m in depth. Along the water flow, each wetland cell was separated into four chambers, i.e. the water inflow chamber, the distribution chamber, the filtration chamber and the outflow chamber. The water inflow chamber preceded the distribution chamber and served as a retention basin for suspended solid. The water distribution chamber, which was filled with gravel, provided a uniform distribution of water to the filtration chamber. The filtration chamber was filled with substrate to a depth of 65 cm and was planted with Iris sibirica and Thalia dealbata. Water was maintained at approximately 5 cm below the surface using a standpipe at the outflow chamber.

A feeding tank with a 1,000 L capacity was used to store the mechanically pretreated wastewater collected from a nearby wastewater treatment plant. Peristaltic pumps (BT300–2, Longer Precision Pump Co., Ltd, Baoding, China) were used to transfer the wastewater to the wetlands to achieve a hydraulic loading rate of 100 mm/day. Before the experiment, the treatment system had been stabilized for 5 months.

To investigate the effects of micro-aeration and substrate selection on the performance of HSSF CWs, six experimental-scale HSSF CWs filled with three different substrate types (i.e., zeolite, ceramsite and quartz granules, with diameters ranging from 3 to 8 mm) were operated over 3 months. The substrate materials were obtained from the Shengshi Water Purification Material Co. Ltd, Henan, China.
group of CWs (filled with zeolite (AEZE), ceramicsite (AECE) and quartz (AEQU) granules) was micro-aerated using a low-power air pump (BOYU, s-2000) with a capacity of 4 L/min. The other group of CWs (i.e., ANZE, ANCE and ANQU) was operated without aeration. Aeration was performed at the inflow chamber of the CWs.

**Physico-chemical analysis**

Water samples were collected weekly from the feeding tank and the effluent between 9 and 10 a.m. Dissolved oxygen (DO), pH, the oxidation-reduction potential (ORP) and water temperature (T) were measured in situ using a Thermo-Orion 5 Star portable meter (Thermo-Orion Inc., Waltham, MA, USA). Concentrations of NH$_4^+$-N, NO$_3^-$-N, NO$_2^-$-N and PO$_4^{3-}$-P were measured using flow injection analysis (Lachat Instruments, Loveland CO, QC8500). Chemical oxygen demand (COD) was determined using COD digestion vials (HACH, Loveland CO, 2125825) and a portable spectrophotometer (HACH, Loveland CO, DR2800). Furthermore, total nitrogen (TN), total phosphorus (TP) and total suspended solids (TSS) were measured according to the Standard Method (State Environmental Protection Administration of China 2002). The concentrations of K, Ca, Mn, Fe, Cu and Zn in the substrates were determined using a mobile X-ray fluorescence spectrometer (EPX-50, Innov-X System Co., Woburn, MA, USA).

**Statistical analysis**

Statistical analysis was performed using the SPSS 19.0 software package for Windows. The difference in substrate compositions, treatment efficiencies between HSSF CWs with and without aeration, and the treatment efficiencies among the HSSF CWs using different substrates were evaluated using one-way analysis of variance (ANOVA). Two-way ANOVA was used to determine the individual and combined effects of micro-aeration and substrate selection on the purification performance of the studied HSSF CWs.

**RESULTS AND DISCUSSION**

**Water quality in the influent and effluent of the six experimental-scale HSSF CWs**

The characteristics of the influent, effluent and areal removal rates of the six experimental-scale HSSF CWs are summarized in Table 1. During the experimental period, the CWs received inlet loadings of NH$_4^+$-N: 3.0 ± 0.7 g/m$^2$/d, TN: 5.3 ± 0.7 g/m$^2$/d, TP: 0.34 ± 0.03 g/m$^2$/d, COD: 9.0 ± 2.8 g/m$^2$/d and TSS: 3.0 ± 2.2 g/m$^2$/d. The influent was characterized by a high NH$_4^+$-N:TN ratio (0.89), a low COD:TN ratio (2.7) and a low DO concentration (0.20 ± 0.14 mg/L), which will cause poor N removal in the CW system without optimization. In this study, optimization measures, including micro-aeration and substrate selection, were considered. Higher ORP values were observed in the effluent of the HSSF CWs with micro-aeration (AEZE, AECE and AEQU) compared with those without aeration (ANZE, ANCE and ANQU). The effluent NH$_4^+$-N, TN, PO$_4^{3-}$-P, TP and COD concentrations were lower in the AEZE, AECE and AEQU configurations compared with the ANZE, ANCE and ANQU setups, respectively. The significant influence of micro-aeration on the HSSF CW treatment of these parameters is shown in Table 2 (p < 0.05).

Among the six experimental-scale HSSF CWs, the highest NH$_4^+$-N, TN, PO$_4^{3-}$-P, TP and COD areal removal rates were observed in the AECE configuration. The TN and TP areal removal rates by the AECE configuration are similar to the results reported by Chang et al. (2012) using an integrated vertical-flow CW. However, the COD areal removal rates found in this study are substantially lower than reported by Chang et al. (2012), which might be due to the low organic loading rate (9.0 ± 2.8 g/m$^2$/d) in this study. The average AECE effluent TP, COD and TSS concentrations satisfied the Grade I (A) discharge standard for pollutants in municipal wastewater treatment plants in China. The average effluent TN and NH$_4^+$-N concentrations satisfied the Grade I (B) and Grade II discharge standards, respectively. This result indicates that a ceramicsite-filled HSSF CW with micro-aeration has great potential for applications in decentralized domestic sewage treatment operations.

**Effects of micro-aeration and substrate selection on the treatment performances of HSSF CWs**

The effects of micro-aeration and substrate selection on the treatment performances of the HSSF CWs are shown in Figure 2. HSSF CWs have been previously reported to be efficient at TSS removal (Vymazal 2011). In this study, the average TSS removal efficiencies using the six experimental-scale CWs ranged from 82 to 90%. Neither substrate selection nor micro-aeration contributed to further enhancements in the TSS removal efficiencies.

Although organic compounds can be degraded both aerobically and anaerobically by micro-organisms in the
Table 1 | Water quality in the influent and effluent of the HSSF CWs

<table>
<thead>
<tr>
<th></th>
<th>Influent</th>
<th>Effluent</th>
<th>AEZE</th>
<th>ANZE</th>
<th>AECE</th>
<th>ANCE</th>
<th>AEQU</th>
<th>ANQU</th>
</tr>
</thead>
<tbody>
<tr>
<td>T (°C)</td>
<td>14.4 ± 7.4</td>
<td>13.9 ± 7.4</td>
<td>13.8 ± 7.4</td>
<td>13.9 ± 7.4</td>
<td>14.0 ± 7.6</td>
<td>14.2 ± 7.4</td>
<td></td>
<td></td>
</tr>
<tr>
<td>pH</td>
<td>8.01 ± 0.33</td>
<td>7.71 ± 0.27</td>
<td>7.81 ± 0.33</td>
<td>8.36 ± 0.53</td>
<td>8.61 ± 0.29</td>
<td>7.54 ± 0.42</td>
<td>7.44 ± 0.48</td>
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<tr>
<td>ORP (mV)</td>
<td>−71.20</td>
<td>−15.4 ± 31</td>
<td>−108.4 ± 19.3</td>
<td>57.1 ± 25.6</td>
<td>−90.6 ± 28.3</td>
<td>36.9 ± 51.2</td>
<td>−117.2 ± 60.3</td>
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<tr>
<td>DO (mg/L)</td>
<td>0.20 ± 0.14</td>
<td>0.17 ± 0.06</td>
<td>0.12 ± 0.02</td>
<td>0.22 ± 0.10</td>
<td>0.13 ± 0.04</td>
<td>0.26 ± 0.12</td>
<td>0.15 ± 0.04</td>
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</tr>
<tr>
<td>NO₂-N (mg/L)</td>
<td>0.25 ± 0.07</td>
<td>0.22 ± 0.16</td>
<td>0.29 ± 0.26</td>
<td>0.22 ± 0.13</td>
<td>0.14 ± 0.10</td>
<td>0.14 ± 0.19</td>
<td>0.19 ± 0.15</td>
<td></td>
</tr>
<tr>
<td>NO₃-N (mg/L)</td>
<td>0.16 ± 0.12</td>
<td>0.36 ± 0.58</td>
<td>0.14 ± 0.10</td>
<td>1.37 ± 1.54</td>
<td>0.09 ± 0.06</td>
<td>0.27 ± 0.25</td>
<td>0.15 ± 0.13</td>
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<tr>
<td>NH₄-N (mg/L)</td>
<td>29.69 ± 7.20</td>
<td>19.30 ± 5.15</td>
<td>23.91 ± 6.82</td>
<td>14.40 ± 4.86</td>
<td>24.35 ± 4.48</td>
<td>22.42 ± 8.00</td>
<td>25.42 ± 7.24</td>
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</tr>
<tr>
<td>NH₃-N areal removal rate (g/m² d)</td>
<td>1.04 ± 0.46</td>
<td>0.58 ± 0.36</td>
<td>1.53 ± 0.49</td>
<td>0.53 ± 0.41</td>
<td>0.73 ± 0.33</td>
<td>0.43 ± 0.36</td>
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<tr>
<td>TN (mg/L)</td>
<td>33.34 ± 6.58</td>
<td>22.34 ± 5.53</td>
<td>26.81 ± 6.21</td>
<td>19.23 ± 7.96</td>
<td>28.32 ± 4.06</td>
<td>25.72 ± 7.85</td>
<td>28.72 ± 6.47</td>
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<tr>
<td>TN areal removal rate (g/m² d)</td>
<td>1.10 ± 0.40</td>
<td>0.65 ± 0.52</td>
<td>1.41 ± 0.59</td>
<td>0.50 ± 0.46</td>
<td>0.76 ± 0.51</td>
<td>0.46 ± 0.46</td>
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<tr>
<td>PO₄⁻-P (mg/L)</td>
<td>2.90 ± 0.27</td>
<td>2.48 ± 0.59</td>
<td>2.91 ± 0.31</td>
<td>0.73 ± 0.43</td>
<td>1.70 ± 0.21</td>
<td>2.31 ± 0.97</td>
<td>2.61 ± 0.30</td>
<td></td>
</tr>
<tr>
<td>PO₄⁻-P areal removal rate (g/m² d)</td>
<td>0.04 ± 0.07</td>
<td>0.00 ± 0.03</td>
<td>0.22 ± 0.06</td>
<td>0.12 ± 0.04</td>
<td>0.06 ± 0.10</td>
<td>0.05 ± 0.03</td>
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<tr>
<td>TP (mg/L)</td>
<td>3.38 ± 0.32</td>
<td>2.78 ± 0.70</td>
<td>3.10 ± 0.35</td>
<td>0.87 ± 0.50</td>
<td>1.94 ± 0.28</td>
<td>2.46 ± 1.00</td>
<td>2.80 ± 0.26</td>
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<tr>
<td>TP areal removal rate (g/m² d)</td>
<td>0.06 ± 0.08</td>
<td>0.03 ± 0.05</td>
<td>0.25 ± 0.07</td>
<td>0.14 ± 0.04</td>
<td>0.09 ± 0.10</td>
<td>0.06 ± 0.04</td>
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<tr>
<td>COD (mg/L)</td>
<td>90.4 ± 27.6</td>
<td>22.0 ± 9.0</td>
<td>31.8 ± 20.9</td>
<td>13.7 ± 8.9</td>
<td>29.3 ± 12.4</td>
<td>20.7 ± 6.7</td>
<td>31.7 ± 12.6</td>
<td></td>
</tr>
<tr>
<td>COD areal removal rate (g/m² d)</td>
<td>6.84 ± 2.58</td>
<td>5.87 ± 2.33</td>
<td>7.68 ± 2.21</td>
<td>6.11 ± 2.35</td>
<td>6.98 ± 2.60</td>
<td>5.88 ± 2.54</td>
<td></td>
<td></td>
</tr>
<tr>
<td>TSS (mg/L)</td>
<td>30.2 ± 21.6</td>
<td>3.9 ± 4.7</td>
<td>4.6 ± 3.2</td>
<td>2.7 ± 2.8</td>
<td>4.7 ± 3.3</td>
<td>3.9 ± 4.8</td>
<td>3.7 ± 2.8</td>
<td></td>
</tr>
<tr>
<td>TSS areal removal rate (g/m² d)</td>
<td>2.63 ± 1.93</td>
<td>2.55 ± 1.97</td>
<td>2.75 ± 2.02</td>
<td>2.54 ± 1.98</td>
<td>2.62 ± 1.92</td>
<td>2.65 ± 2.13</td>
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</tbody>
</table>

*All data are presented as the means ± standard deviation (SD); n = 10 for all samples.

Table 2 | F-values and significance of a two-way ANOVA for the effects of aeration and substrate selection on HSSF CW treatment performance

<table>
<thead>
<tr>
<th>Substrate selection</th>
<th>Aeration</th>
<th>Aeration &gt; substrate selection</th>
</tr>
</thead>
<tbody>
<tr>
<td>COD</td>
<td>1.92</td>
<td>19.99b</td>
</tr>
<tr>
<td>TSS</td>
<td>0.14</td>
<td>2.97</td>
</tr>
<tr>
<td>TN</td>
<td>2.43</td>
<td>20.58a</td>
</tr>
<tr>
<td>TP</td>
<td>29.77b</td>
<td>11.38a</td>
</tr>
<tr>
<td>NH₄-N</td>
<td>6.67a</td>
<td>41.53b</td>
</tr>
<tr>
<td>PO₄⁻-P</td>
<td>32.09b</td>
<td>11.65a</td>
</tr>
</tbody>
</table>

*p < 0.01.

CWs (Ong et al. 2010), the results indicated that micro-aeration significantly enhances the COD removal efficiencies in the HSSF CWs filled with ceramsite and quartz granules (p < 0.05). Nivala et al. (2007) also indicated that supplementary aeration could be used in addition to macrophytes to yield better results in reducing organic contaminants in wastewater. There was no significant difference in COD removal among the CWs filled with different substrates without micro-aeration. This result is consistent with Zou et al. (2012), who reported similar COD removal efficiencies among four biofilters using different substrates. With micro-aeration, the COD removal efficiency of the ceramsite-filled HSSF CW was significantly higher than removal efficiencies of the CWs filled with zeolite and quartz (p < 0.05). Previous work has shown that the microstructure of ceramsite when sintered at 1,000 °C has many unevenly distributed pores (0.5 μm < pore size < 10.0 μm) and rough surfaces with many agglomerated crystals (Xu et al. 2008), which is theoretically suitable for the attachment and growth of micro-organisms. Micro-aeration might...
encourage the growth of organic-degrading micro-organisms attached to ceramsite.

For the removal of NH$_4^+$-N and TN, the results presented herein are consistent with Zhang et al. (2010), i.e. aeration can significantly enhance the removal efficiencies of HSSF CWs (filled with zeolite and ceramsite) ($p < 0.05$). There was no significant difference in NH$_4^+$-N and TN removal efficiencies among the HSSF CWs filled with different substrates without aeration. With micro-aeration, the NH$_4^+$-N removal efficiencies of the ceramsite-filled HSSF
CW were significantly higher than the CWs filled with zeolite and quartz; the TN removal efficiencies of the ceramsite-filled HSSF CW were significantly higher than the quartz-filled CW (p < 0.05). The microstructure of ceramsite likely provides a diverse niche for both nitrifying and denitrifying bacteria in the aerated HSSF CW, which is beneficial for removing NH$_4$-N and TN. Previous work has suggested that zeolite should be helpful for ammonia removal (Stefanakis & Tsihrintzis 2012). In this study, there was no significant difference in the NH$_4$-N and TN removal efficiencies between the HSSF CWs filled with zeolite and quartz granules. Because of the variety of available zeolite materials, the ammonia adsorption by zeolite varies widely in the literature (Zhu et al. 2011). Thornton et al. (2007) reported that the maximum theoretical saturated adsorption of ammonia is 47.92 mg/g. However, Wang et al. (2006) found that the maximum theoretical saturated adsorption of ammonia is 2.33 mg/g for zeolite at an ammonia concentration of 100 mg/L. The cation exchange capacity is directly linked to the ability of zeolites to remove ammonia. At cation exchange sites, NH$_4$-N uptake occurs by ion exchange with ions in the following order: Na$^+$ > K$^+$ > Ca$^{2+}$ > Mg$^{2+}$ (Sprynskyj et al. 2005). The composition of the substrate is vital for the NH$_4$-N removal capacity. Although the Na$^+$ concentration was not provided, the concentrations of K$^+$ and Ca$^{2+}$ (Table 2) in the zeolite were lower than those in the quartz and ceramsite samples in this study and in other reports (Zhu et al. 2011). The zeolite composition in the CWs might be the reason for the poor ammonia removal rates.

The removal efficiencies of both PO$_4^{3-}$-P and TP were largest in the ceramsite-filled HSSF CW with aeration. Phosphorus is removed via sorption and precipitation processes. Moreover, the calcium (Ca), iron (Fe) and aluminum (Al) contents are important for efficiently removing P (Vohla et al. 2011). The most important substrate characteristic for determining their P-sorption capacity is the Ca-content (Brix et al. 2003). Heating Ca-rich media at high temperatures is a common way to improve the material. Heating will mostly likely form CaO, which has a more reactive Ca-phase than the more common CaCO$_3$ (Vohla et al. 2011). The highest Ca content (Table 3) and the heating process to produce ceramsite might be the reasons for the higher PO$_4^{3-}$-P and TP removal efficiencies of the ceramsite-filled HSSF CW, compared with the CWs using the other two substrates. Micro-aeration significantly enhanced the PO$_4^{3-}$-P and TP removal efficiencies in ceramsite-filled HSSF CWs (p < 0.05), which might be due to the appearance of ferric iron under aerobic conditions.

Two-way ANOVA showed that both micro-aeration and the substrate selection significantly affected the HSSF CW treatment efficiencies of TP, PO$_4^{3-}$-P and NH$_4$-N (p < 0.05). In addition, micro-aeration significantly increased the COD and TN removal efficiencies (p < 0.05). This result indicated a significant interaction term (i.e. micro-aeration × substrate selection) for the removal of NH$_4$-N (p < 0.05). This combined effect is consistent with the expectations that employing micro-aeration will further improve the NH$_4$-N removal efficiency in HSSF CWs using an optimized substrate.

**CONCLUSIONS**

Six experimental-scale HSSF CWs were investigated to determine the effects of micro-aeration and substrate selection on HSSF CW treatment performances. The average TSS removal rates of the HSSF CWs ranged from 82 to 90% and were not affected by micro-aeration or substrate selection. According to the results of this study, both micro-aeration and substrate selection can significantly influence the HSSF CW treatment efficiencies of TP, PO$_4^{3-}$-P and NH$_4$-N. In addition, micro-aeration can significantly increase the COD and TN removal efficiencies.
Among the three substrates used in this work (i.e. zeolite, ceramsite and quartz granules), ceramsite was recognized as the best substrate for HSSF CWs because of its porous structure and high Ca and Fe contents. Employing micro-aeration further improved the NH$_4^+$-N removal efficiency in the ceramsite-filled HSSF CWs.

**ACKNOWLEDGEMENTS**

The study was supported by the National Natural Science Foundation of China (51108335, 51278355), the Natural Science Foundation of Jiangsu Province (BK20130398) and the earmarked fund for Modern Agro-industry Technology Research System.

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First received 13 December 2014; accepted in revised form 10 February 2015. Available online 21 February 2015.