Confirmation the optimal aeration parameters for nitrogen removal and nitrous oxide emission in wastewater ecological soil infiltration systems with brown earth

Yafei Sun, Junling Pang, Shiyao Wang, Tingting Tao, Xun Fu, Ying Zhang, Bo Sun and Jing Pan

ABSTRACT

Nitrogen removal is an obstacle for the wide application of wastewater ecological soil infiltration (WESI) system in domestic wastewater treatment. In this study, matrix dissolved oxygen (DO), nitrogen removal and nitrous oxide (N₂O) emission in aerated pilot WESI systems were investigated under different aeration times (1, 2, 3, 4 and 6 h/d) and aeration rates (1, 2, 3 and 4 L/min). The results showed that aerobic conditions in upper matrix and anoxic or anaerobic conditions in the subsequent matrix were developed in an aerated/non-aerated cycle at the optimal aeration condition of aeration time of 4 h/d and aeration rate of 3 L/min. Simultaneously, high removal efficiency of chemical oxygen demand (COD) (97.9%), NH₄⁺-N (98.2%), total nitrogen (TN) (90.7%) and low N₂O emission rate (13.2 mg/(m² d)) were obtained. The results would provide optimal aeration parameters for application of intermittent aerated WESI systems.

Key words | aeration, N₂O, nitrogen, wastewater ecological soil infiltration system

INTRODUCTION

The wastewater ecological soil infiltration (WESI) system is one of onsite domestic wastewater treatment methods, which have been widely used in rural areas such as tourist resorts, villa districts and decentralized villages of Japan, American, Australia and China because it is designed, located, operated and maintained satisfactorily (Jiang et al. 2011). In a WESI system, wastewater inflows into the distribution layer (30–50 cm below surface), slowly rises up and spreads into the surrounding matrix under capillary action of the infiltration layer (above 30–50 cm), and then infiltrates to the lower layer under the action of gravity (USEPA 2002). In this process, suspended substances, organic matter and nutrients were removed by physical, chemical and biological reaction in the matrix (Zhang et al. 2005; Wang et al. 2010). Among these, biological oxidation, biological nitrification and denitrification are the main mechanisms for organic matter and nitrogen removal (Lance & Whisler 1976; Lance 1986). During wastewater treatment in WESI systems, dissolved oxygen (DO) concentration is an important factor affecting organic matter and nitrogen removal (Wang et al. 2010; Song et al. 2016).

Many studies have been conducted to improve DO conditions of WESI systems by intermittent operation (Li et al. 2017; Wu et al. 2019), bioaugmentation technology (Zou et al. 2009; Kong et al. 2014, 2015; Wang et al. 2015), adjust infiltration system configuration (Wang et al. 2010; Llorens et al. 2011; Pan et al. 2015) and aeration of WESI system matrix (Pan et al. 2015; Yang et al. 2016). Intermittent aeration had been approved as the most effective way to develop appropriate DO conditions for organic matter and nitrogen removal (Song et al. 2016; Sun et al. 2018). Several studies have investigated on the effects of influent hydraulic loading rate, surface organic loading, chemical oxygen demand/nitrogen (COD/N) ratio, and aerated mode on pollutants removal in aerated WESI systems (Wu et al. 2015; Song et al. 2016; Yang et al. 2016; Jiang et al. 2017). Yang et al. (2016) reported that short
aeration time was disadvantageous to organic matter and ammonia nitrogen (NH₄-N) removal, while long aeration time could change anaerobic environment in WESI systems and restrain denitrification (Wu et al. 2016). Therefore, it is necessary to investigate the effects of aeration time and aeration rate on pollutants removal before this strategy applying in WESI systems.

In addition, WESI systems would produce considerable amount of N₂O in biological nitrification and denitrification processes. N₂O accounts for approximately 5% of the total greenhouse effect, whose greenhouse effect is about 150–200 times of CO₂ (Tol 2002). At the same time, the decomposition product of N₂O could react with O₃ to destroy the ozone layer and cause harm to human beings. Oxygen environment, pH, influent COD/N ratio and loading rate could affect N₂O emission in a WESI system (Kong et al. 2002; Li et al. 2017, 2018; Sun et al. 2018). Kong et al. (2016) and Li et al. (2017, 2018) researched N₂O emission and spatial distribution in non-aerated WESI systems. Jiang et al. (2017) investigated the effect of organic loading rate on N₂O emission in an intermittent aeration WESI system with aeration time of 4 hours. The study by Sun et al. (2018) reported that intermittent aeration WESI system amended with biochar and sludge could improve nitrogen removal and reduce N₂O emission. Zheng et al. (2018) evaluated the influence of shunt ratio on N₂O emission in non-aerated WESI systems and intermittent aeration WESI systems with aeration time of 4 h and aeration rate of 4 ± 0.2 L/min. However, few studies in literature are focused on the effects of aeration time and aeration rate on N₂O emission of an intermittent aeration WESI system.

The key aim of this study was to explore the effects of aeration time and aeration rate on nitrogen removal, N₂O emission and identify the optimal aeration parameters for high nitrogen removal and low N₂O emission in intermittent aeration WESI systems. Hopefully, the results of this study will provide a reference for successful application of intermittent aeration WESI systems in decentralized wastewater treatment.

**MATERIAL AND METHODS**

**System description and operation**

Nine pilot WESI systems made of polyvinyl chloride column (height of 120 cm and internal diameter of 30 cm) were carried out in Shenyang Normal University in China. Figure 1 shows the schematic diagram of a pilot-scale WESI system. Each system was filled from top to bottom with 110 cm of mixed matrix (80% of brown earth and 20% of coal slag by mass proportions, respectively) and 10 cm of gravel (5–10 mm in diameter). Wastewater was distributed through a polyvinyl chloride pipe at the depth of 50 cm. The effluent was collected by outlet pipe at the bottom. An air pipe and air diffuser was installed at the depth of 40 cm of each system for oxygen supply.

Domestic wastewater family area of Shenyang normal University after sedimentation process was continuously pumped into each WESI system under the hydraulic loading of 0.12 m³/(m² d) and organic loading of 25.4 g COD/(m² d). Hydraulic retention times were monitored by using NaCl as a tracer, which were about 24 h. Table 1 shows daily variations of the influent during the experimental period. Each WESI system was operated for 50 days under the same conditions before this study to allow systems maturation. Two experiments were conducted to determine the optimal aeration conditions for pollutant removal and N₂O emission in intermittent aeration WESI systems. The first experiment was to confirm the optimal aeration time. In this experiment, five systems were operated with intermittent aeration under aeration rate of 3.0 ± 0.2 L/min and for aeration time 1 h/d (the system were firstly subject to aeration for an hour, and then have 23 h interval without aeration.), for aeration time 2 h/d, for aeration time 3 h/d, for aeration time 4 h/d, and for aeration time 6 h/d, respectively. DO of the matrix was monitored by DO electrodes which were placed at 50 cm (upper layer), 80 cm (middle layer) and 110 cm (bottom layer) depths in nine pilot WESI systems. The second experiment was to ascertain the optimal aeration rate. The other four systems were operated with intermittent aeration under aeration rate of 1, 2, 3 and 4 L/min with the optimal aeration time from the first experiment.

**Sample collection and analytical methods**

Every five days, 50 mL water samples of the influent and effluent were collected to evaluate nitrogen and organic matter removal performance according to standard analytical methods of APHA (2005). Twelve water and gas samples were collected for a 60-day measurement period in each system. DO signals were collected online every 15 min by MDA-501 data acquisition system (Tuopu Co. Ltd, Shunde, China). Gas was sampled using static-stationary chamber to investigate N₂O. The details about N₂O sampling and measuring methods were listed in a previous study (Zheng et al. 2018).
All statistical analyses were performed by using SPSS 12.0 software (SPSS Inc., Chicago, IL, USA). Two-sample t-tests were used to estimate the significance of differences between means. Differences were considered statistically significant at $P < 0.05$.

**RESULTS AND DISCUSSION**

**Effect of aeration time on DO profiles in an aerated/non-aerated cycle**

Figure 2 shows average DO distribution in an aerated/non-aerated cycle. Intermittent aeration did not change anoxic conditions at 80 and 110 cm depths, which made DO concentrations of 50 cm depth more than 7.8 mg/L for all WESI systems during aeration. DO concentrations of 50 cm depth decreased immediately when aeration finished because of pollutants degradation consuming oxygen. Nevertheless, DO concentrations fluctuations were significantly distinct at 50 cm depth in the non-aerated period with different aeration times. Long periods of anoxic conditions (DO lower than 1.0 mg/L) were created at 50 cm depth by short aeration times (1 and 2 h/d) due to inadequate oxygen supply. When aeration time was 3 h/d, average DO concentration of 50 cm was less than 1.4 mg/L for 6 h per day. DO concentration higher than 1.5 mg/L is the essential condition for nitrification (Ye & Li 2009). Aeration time of 1, 2 and 3 h/d was deficient which could not achieve ammonia nitrogen elimination completely. Aerobic environment (DO more than 2.0 mg/L) at 50 cm depth has been developed with long aeration times (4 and 6 h/d) in the non-aerated period. It could conclude that intermittent aeration with aeration time of 4 or 6 h/d achieved

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**Table 1** | Daily variations of the influent

<table>
<thead>
<tr>
<th>Parameters</th>
<th>COD</th>
<th>NH$_4^+$-N</th>
<th>TN</th>
<th>TP</th>
<th>pH</th>
</tr>
</thead>
<tbody>
<tr>
<td>Influent (mg/L)</td>
<td>185.3–245.3</td>
<td>33.5–38.2</td>
<td>35.8–41.8</td>
<td>3.9–4.8</td>
<td>6.8–7.2</td>
</tr>
</tbody>
</table>
aerobic conditions at 50 cm depth and anaerobic conditions at 80 and 110 cm depths in an aerated/non-aerated cycle, which would favor nitrification and denitrification. Aeration time cannot be too long, which cause energy consumption increasing (Foladori et al. 2015).

Effect of aeration time on pollutants removal and N₂O emission

As shown in Figure 3 and Table 2, average COD removal efficiencies were 86.7%, 90.4%, 93.8%, 98.1% and 99.0% with aeration time of 1, 2, 3, 4 and 6 h/d at aeration rate of 3 L/min, respectively. And average COD concentration of the effluents were less than 30 mg/L, which were below Class I (Level A ≤ 50 mg/L) of Chinese Criterion for Water Discharge from Municipal Wastewater Treatment Plant (GB18921-2002). COD removal efficiency enhanced with the increase of aeration time. In WESI systems, organic matter is decomposed by aerobic and anaerobic heterotrophic bacteria. However, aerobic degradation is usually more crucial (Saeed & Sun 2012). Sufficient oxygen supply improved the performance of aerobic biochemical oxidation with aeration time being prolonged. The highest COD removal efficiency was achieved with aeration time of 6 h/d, which was in accordance with DO concentration at 50 cm depth. Average COD removal efficiency was not significantly different when aeration time increased from 4 to 6 h/d (P > 0.05). A similar conclusion has been drawn in previous studies (Wu et al. 2016; Zhou et al. 2018).

Average NH₄⁺-N removal efficiency was significantly improved from 81.4% to 98.8% when aeration time increased from 1 to 6 h/d in WESI systems (Table 2). NH₄⁺-N removal efficiency increased with aeration time increasing. Average NH₄⁺-N concentration of the effluents were 6.2, 4.6, 2.2, 0.7 and 0.4 mg/L with aeration time of 1, 2, 3, 4 and 6 h/d at aeration rate of 3 L/min, respectively. Sufficient oxygen supply could promote the reduction of NH₄⁺-N by ammonia-oxidizing bacteria (Saleem & Moe 2014). DO result showed that the oxygen concentration of 50 cm matrix was above 2.1 mg/L through intermittent aeration with aeration time of 4 and 6 h/d at the aeration rate of 3 L/min, which was favorable for the nitrification process. When aeration time increased from 4 to 6 h/d, NH₄⁺-N removal efficiency increased slightly in WESI systems. There was no significant difference between NH₄⁺-N removal efficiency with aeration time of 4 and 6 h/d (P > 0.05).
Average total nitrogen (TN) removal efficiencies were 73.6%, 78.8%, 87.5%, 90.2% and 89.7% with aeration times of 1, 2, 3, 4 and 6 h/d at aeration rate of 3 L/min, respectively (Table 2). TN removal efficiency increased with aeration time increasing between 1 and 4 h/d. However, TN removal efficiency dropped as aeration time increased from 4 to 6 h/d. In this study, aerobic conditions at 50 cm depth and anoxic or anaerobic conditions in the subsequent section were created when aeration times were 4 and 6 h/d, which obtained nearly complete nitrification. After effective nitrification, NO3-N could be eliminated only if sufficient organic carbon was offered as electron donor. Limited available organic matter would inhibit denitrification process in WESI systems (Wang et al. 2013; Wu et al. 2013). More organic matter was available with aeration time of 4 h/d because of lower organic matter degradation compared with aeration time of 6 h/d, which resulted in higher TN removal.

N2O emits from incomplete nitrification and denitrification processes (Sun et al. 2018). In this study, average N2O conversion ratios (0.19–0.38%) were coincident with previous studies (Kong et al. 2002; Li et al. 2018) (in Figure 3). Average N2O emission rates with aeration times of 1, 2, 3, 4 and 6 h/d at aeration rate of 3 L/min were 26.4, 21.6, 15.3, 15.2 and 15.7 mg/(m² d), respectively. Average N2O emission rate dropped with aeration time increasing (from 1 to 4 h/d). Average N2O emission rate rose as aeration time increased from 4 to 6 h/d. More oxygen was available with aeration time of 6 h/d which improved nitrification process and aerobic biochemical oxidation of organic matter. Meanwhile, more carbon source was required for N2O to N2 reduction after efficient nitrification. However, organic matter was not enough compared with aeration time of 4 h/d, which restrained part of N2O from transforming to N2.

![Figure 3](https://iwaponline.com/wst/article-pdf/80/1/144/600026/wst080010144.pdf)

**Table 2** Pollutants removal and N2O emission with different aeration time

<table>
<thead>
<tr>
<th>Item</th>
<th>1 h/d</th>
<th>2 h/d</th>
<th>3 h/d</th>
<th>4 h/d</th>
<th>6 h/d</th>
</tr>
</thead>
<tbody>
<tr>
<td>Effluent of COD (mg/L)</td>
<td>29.3</td>
<td>21.1</td>
<td>13.7</td>
<td>4.3</td>
<td>2.1</td>
</tr>
<tr>
<td>COD removal efficiency (%)</td>
<td>86.7</td>
<td>90.4</td>
<td>93.8</td>
<td>98.0</td>
<td>99.0</td>
</tr>
<tr>
<td>Effluent of NH4-N (mg/L)</td>
<td>6.2</td>
<td>4.6</td>
<td>2.2</td>
<td>0.7</td>
<td>0.4</td>
</tr>
<tr>
<td>NH4-N removal efficiency (%)</td>
<td>81.4</td>
<td>86.2</td>
<td>93.4</td>
<td>97.9</td>
<td>98.8</td>
</tr>
<tr>
<td>Effluent of TN (mg/L)</td>
<td>9.7</td>
<td>7.8</td>
<td>4.6</td>
<td>3.6</td>
<td>3.8</td>
</tr>
<tr>
<td>TN removal efficiency (%)</td>
<td>73.6</td>
<td>78.7</td>
<td>87.5</td>
<td>90.2</td>
<td>89.7</td>
</tr>
<tr>
<td>N2O emission rate (mg/(m² d))</td>
<td>26.4</td>
<td>21.6</td>
<td>15.3</td>
<td>13.2</td>
<td>13.7</td>
</tr>
<tr>
<td>N2O conversion ratio (%)</td>
<td>0.38</td>
<td>0.31</td>
<td>0.22</td>
<td>0.19</td>
<td>0.20</td>
</tr>
</tbody>
</table>
It is necessary to service aerobic conditions in upper layer for nitrification and anaerobic conditions in lower layer for denitrification. Intermittent aeration with 4 h/d was recommended for WESI systems in light of nitrogen removal, N₂O emission and operation costs.

**Effect of aeration rate on DO profiles in an aerated/non-aerated cycle**

Average DO concentrations at 50 cm depth were 2.8, 4.4, 8.0 and 9.8 mg/L during aeration period with aeration rate of 1, 2, 3 and 4 L/min at aeration time of 4 h/d, respectively (in Figure 4). When artificial aeration finished, DO concentration in the upper layer deceased quickly because organic matter degradation consumed oxygen, which was higher than 0.5, 1.1, 2.3 and 3.1 mg/L at the end of each aerated/non-aerated cycle. Average DO concentrations at 50 cm depth were below 1.5 mg/L for 4 h and 2 h with aeration rate of 1 and 2 L/min, respectively, which would restrict COD and NH₄-N elimination. While average DO concentrations in upper layer were more than 2.3 and 3.1 mg/L all the time with aeration rate of 3 and 4 L/min, respectively, which favored for organic matter degradation and nitrification simultaneously.

**Effect of aeration rate on pollutants removal and N₂O emission**

Average COD removal efficiencies were 90.3%, 94.4%, 97.9% and 98.9%, with average effluent COD concentrations of 21.2, 12.2, 4.5 and 2.3 mg/L under aeration rate of 1, 2, 3 and 4 L/min at aeration time of 4 h/d, respectively (in Figure 5 and Table 3). Average COD removal efficiency in the intermittent aeration WESI system was significantly higher than that of a WESI system without aeration (lower than 84% (Pan et al. 2015)) within aeration rate of this study (P < 0.05). WESI systems with aeration time of 4 h/d and aeration rate of 1, 2, 3 and 4 L/min supplied extra oxygen for organic matter degradation. COD removal efficiency was raised with aeration rate increasing due to more oxygen supply by aeration. This result was in agreement with previous reports that adequate oxygen supply would improve COD elimination (Yang et al. 2016; Jiang et al. 2017). The highest average COD removal efficiency was obtained at aeration rate of 4 L/min. There was no significant differences between COD removal efficiencies with aeration rate of 3 and 4 L/min (P > 0.05).
Average NH$_4^+$-N removal efficiencies were 90.4%, 95.5%, 98.2% and 99.1% with aeration rate of 1, 2, 3 and 4 L/min at aeration time of 4 h/d, respectively (in Figure 5 and Table 3). Average NH$_4^+$-N removal efficiency increased with the increase of aeration rate owing to more DO supply which was consistent with previous reports (Wu et al. 2016; Zhou et al. 2018). No significant differences were observed in NH$_4^+$-N removal efficiencies between aeration rate of 3 and 4 L/min ($P > 0.05$).

Average TN removal efficiency significantly increased from 80.4% to 90.7% as aeration rate increased from 1 to 3 L/min, which decreased from 90.7% to 88.8% when aeration rate increased from 3 to 4 L/min. COD and NH$_4^+$-N removal efficiencies were improved with aeration rate increasing, which resulted in organic matter lacking in denitrification process and restricted TN removal under higher aeration rate (Saeed & Sun 2012; Fan et al. 2015). The decrease of TN removal efficiency with aeration rate of 4 L/min was attributed to unsatisfactory denitrification. The result was different from previous research (Zhou et al. 2018). Biochar as substrate was used in Zhou’s study, which provides carbon source for denitrification under high aeration rate.

Average N$_2$O emission rates with aeration rate of 1, 2, 3 and 4 L/min at aeration time of 4 h/d were 21.4, 15.6, 13.2 and 13.9 mg/(m$^2$ d), respectively (in Figure 5 and Table 3). Average N$_2$O emission rate decreased with aeration rate increasing from 1 to 3 L/min and increased when aeration rate increased from 3 to 4 L/min. Highest N$_2$O emission rate with aeration rate of 1 L/min was attributed to low nitrification and denitrification. Carbon source of lower part was not enough with aeration rate of 4 L/min due to high COD degradation which restricted N$_2$O transforming to N$_2$ compared with aeration rate of 3 L/min. Sufficient carbon

![Figure 5](https://iwaponline.com/wst/article-pdf/80/1/144/600026/wst080010144.pdf)  
**Figure 5** | Pollutants removal and N$_2$O emission at aeration rate of 1 L/min, 2 L/min, 3 L/min and 4 L/min with aeration time of 4 h/d.

<table>
<thead>
<tr>
<th>Item</th>
<th>1 L/min</th>
<th>2 L/min</th>
<th>3 L/min</th>
<th>4 L/min</th>
</tr>
</thead>
<tbody>
<tr>
<td>Effluent of COD (mg/L)</td>
<td>21.2</td>
<td>12.2</td>
<td>4.5</td>
<td>2.3</td>
</tr>
<tr>
<td>COD removal efficiency (%)</td>
<td>90.3</td>
<td>94.4</td>
<td>97.9</td>
<td>98.9</td>
</tr>
<tr>
<td>Effluent of NH$_4^+$-N (mg/L)</td>
<td>3.2</td>
<td>1.5</td>
<td>0.6</td>
<td>0.3</td>
</tr>
<tr>
<td>NH$_4^+$-N removal efficiency (%)</td>
<td>90.4</td>
<td>95.5</td>
<td>98.2</td>
<td>99.1</td>
</tr>
<tr>
<td>Effluent of TN (mg/L)</td>
<td>7.2</td>
<td>5.5</td>
<td>3.4</td>
<td>4.1</td>
</tr>
<tr>
<td>TN removal efficiency (%)</td>
<td>80.4</td>
<td>85.0</td>
<td>90.7</td>
<td>88.8</td>
</tr>
<tr>
<td>N$_2$O emission rate (mg/(m$^2$ d))</td>
<td>21.4</td>
<td>15.6</td>
<td>13.2</td>
<td>13.9</td>
</tr>
<tr>
<td>N$_2$O conversion ratio (%)</td>
<td>0.31</td>
<td>0.23</td>
<td>0.19</td>
<td>0.21</td>
</tr>
</tbody>
</table>

Table 3 | Nitrogen removal and N$_2$O emission with different aeration rate

Average COD removal efficiency in this study increased from 90.3% to 94.4% when aeration rate increased from 1 to 2 L/min, and decreased from 94.4% to 97.9% when aeration rate increased from 2 to 3 L/min. High aeration rate favors nitrification which was consistent with previous reports (Gu et al. 2016; Zhou et al. 2018). Higher aeration rate decreases dissolved oxygen (DO) content in the system which leads to lower denitrification rate and thus lower N$_2$O emission rate.
source supply after efficient nitrification could greatly decrease N$_2$O emission in WESI systems (Zheng et al. 2018; Liang et al. 2019).

According to the results and considering aeration efficiency, operating costs, WESI systems with aeration time of 4 h/d and aeration rate of 3 L/min would be an optimal choice to improve nitrogen removal and reduce N$_2$O emission simultaneously.

CONCLUSION

Aeration with optimal aeration time of 4 h/d and aeration rate of 3 L/min could create aerobic conditions for nitrification in upper matrix and did not change anoxic or anaerobic conditions for denitrification in the subsequent matrix in an aerated/non-aerated cycle simultaneously, which achieved high COD removal efficiency of 97.9%, NH$_4^+$-N removal efficiency of 98.2%, TN removal efficiency of 90.7% and low N$_2$O emission rate of 15.2 mg/(m$^2$.d) in a WESI system with brown earth. Intermittent aeration is an optional method to improve organic matter removal, nitrogen removal and reduce N$_2$O emission for WESI systems.

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