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Nitrate removal from tail water by integrated vertical-flow constructed wetlands at a high hydraulic loading rate

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ABSTRACT

Nitrate removal rates of two pilot-scale integrated vertical-flow constructed wetlands (IVCWs) treating tail water, under a hydraulic loading rate (HLR) of 250 mm/d with a mean influent NO_3^-–N concentration of 24.4 $mg \cdot L^{-1}$, were evaluated. Mean NO_3^-–N removal efficiencies of 15.5 and 18.5% with mass removal rates of 1.01 $g \cdot m^{-2} \cdot d^{-1}$ and 1.16 $g \cdot m^{-2} \cdot d^{-1}$ for IVCW 1 (planted with *Canna indica and Pontederia cordata*) and 2 (planted with *Typha orientalis and Arundo donax var. versicolor*), respectively, were achieved. The removal rate constants as fitted by the first-order area-based model averaged 0.046 and 0.055 $m \cdot d^{-1}$, respectively. Since NO_3^-–N was the dominant nitrogen form in the effluent, denitrification was the limiting step in nitrogen removal despite of favorable pH and anaerobic conditions in the wetland beds. Low availability of carbon source, high HLR, and low temperature could be the probable influencing factors for the observed low NO_3^-–N removal efficiencies. However, IVCW could be used to treat tail water for nitrate removal at a comparable high loading rate.

Keywords: Integrated vertical-flow constructed wetlands (IVCWs); Tail water; NO_3^-–N removal; Denitrification; High loading rate

1. Introduction

Conventional wastewater treatment plants with activated sludge are usually effective in removing Chemical oxygen demand (COD) and ammonia in wastewaters, but the effluent, so-called tail water, is usually characterized by high nitrate concentrations [1–4]. In addition, nitrate is the predominant form of nitrogen pollutants in nonpoint drainage from fertile agricultural fields [5,6] and groundwater [7,8].

It is generally regarded that high content of nutrient is one of important causes for the eutrophication of aquatic ecosystem [9,10]. Serious algal blooms followed by hypoxia in waterbodies can be caused by high nitrate loads [11]. Besides, methemoglobinemia and bladder cancer in humans have also been demonstrated to be associated with high nitrate concentrations in drinking water [12].

In order to treat wastewater containing nitrate, a variety of technologies, including ion-exchange, electrodialysis, reverse osmosis, ultrafiltration, and biological denitrification, have been developed [13].

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Among them, biological treatment is markedly preferable due to the superiorities of low cost and simplicity [14].

Constructed wetlands (CWs) are generally regarded as a promising economic biological treatment system and have been widely applied for treatment of various types of wastewaters [15–18]. The anoxic zones and microsites predominantly presented in wetland substrates are favorable for denitrification [19]. Assimilation by wetland plants could also contribute to nitrate removal. Besides, plant productivity could be potentially utilized as carbon source to drive denitrification [20]. Some studies have focused on the treatment of various nitrate-contaminated wastewaters using free water surface (FWS) and horizontal subsurface flow (HSSF) CWs [20–25].

Integrated vertical-flow constructed wetlands (IVCWs), a new CW type composed of a vertically down-flow chamber followed by an up-flow chamber, have been applied to treat polluted surface water [26], domestic wastewater [27], and as a part of ecosystem restoration in China [28], owing to the advantages of relatively "compact" and high treatment performance. However, the performance and potential capacity of IVCW treating tail water were rarely reported.

In this study, two pilot-scale IVCW mesocosms were constructed in parallel to receive tail water with a mean influent nitrate concentration of 24.4 $mg \cdot L^{-1}$ and a hydraulic loading rate (HLR) of 250 mm/d. The aims were to (1) evaluate the performance and nitrate removal capacity of the IVCWs treating tail water at a high loading rate; (2) identify some factors affecting nitrate removal in the IVCW mesocosms.

2. Materials and methods

2.1. Experimental setup and operations

Two parallel pilot-scale IVCW systems, each comprising a down-flow chamber (1 m × 1 m × 1 m) in series with an up-flow chamber (1 m × 1 m × 1 m) (Fig. 1), were established outside near Donghu Lake of Wuhan, China. Two layers of gravel in different diameters were filled as the wetland media, with the depth of the lower layers (diameter 10–20 mm) being both 45 cm, followed by an upper layer (diameter 2–10 mm) of 40 and 30 cm for down-flow and up-flow chamber, respectively. The average porosity of the substrate was 0.40 and the effective volume of each mesocosm was 0.6 m^3.

Based on our previous study, four species of macrophytes were chosen as wetland plants in the experiment. *Canna indica* (*Canna* L.) and *Pontederia cordata* (*Pontederia* L.) were transplanted carefully into the down-flow chamber and up-flow one of IVCW 1. Likewise, *Typha orientalis* (cattail) and *Arundo donax var. versicolor* (*Arundo donax*) were transplanted into the IVCW 2. The plant density was 8 stems/m^2.

Synthetic wastewater was used to minimize variability of the influent characteristics. The tail water (Table 1) was simulated according to the effluent characteristics of a continuously aerated membrane bioreactor in a previous study [3].

Identical operation conditions were conducted for the two IVCW mesocosms. Wastewater was introduced to each mesocosm in 0.5 m^3/d twice a day, yielding a HLR and nominal hydraulic retention time (HRT) of 250 mm/d and 1.2 d, respectively.

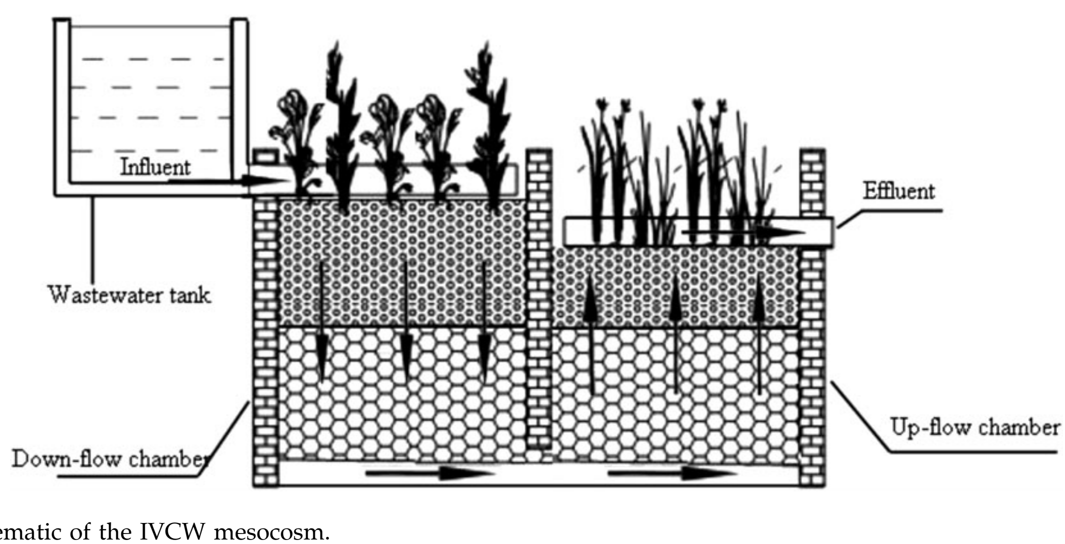

Fig. 1. Schematic of the IVCW mesocosm.

Table 1
Characteristics of the influent (shown as mean value ± standard deviation)

Parameter	Value	Parameter	Value
DO ($mg\,L^{-1}$)	4.9 ± 1.2	TN ($mg\,L^{-1}$)	30.4 ± 3.2
pH	7.5 ± 0.2	TN loading rate ($g\,m^{-2}\,d^{-1}$)	7.61 ± 0.80
Conductivity (μs/cm)	498 ± 73	NH_4^+–N ($mg\,L^{-1}$)	1.3 ± 0.5
NO_3^-–N ($mg\,L^{-1}$)	24.4 ± 2.8	NO_2^-–N ($mg\,L^{-1}$)	0.5 ± 0.2
NO_3^-–N loading rate ($g\,m^{-2}\,d^{-1}$)	6.10 ± 0.70	TP ($mg\,L^{-1}$)	3.2 ± 0.4
COD ($mg\,L^{-1}$)	46.9 ± 6.4	TP loading rate ($g\,m^{-2}\,d^{-1}$)	0.79 ± 0.10
COD/NO_3^-–N	2.0 ± 0.3	COD/TN	1.6 ± 0.2

Note: $n = 60$ for pH, DO, and conductivity; $n = 20$ for pollutant concentration.

2.2. Water quality analysis

The trial was carried out from 10 August 2010 to 10 January 2011. Water samples were collected from the inlet and outlet once a week, and analysis was performed immediately for dissolved oxygen (DO), pH, electric conductivity, and temperature using an Orion 5-star portable multimeter (Thermo Fisher Scientific Company, USA). COD, total nitrogen (TN), ammonia nitrogen (NH_4^+–N), nitrate nitrogen (NO_3^-–N), nitrite nitrogen (NO_2^-–N), and total phosphorus (TP) were determined according to the standard methods [29].

Pollutant removal efficiency was calculated by the percentage of deduction in concentration for each pollutant as follows: Removal efficiency $= (1 - C_{\mathrm{eff}}/C_{\mathrm{inf}}) \times 100\%$ where C_{inf} and C_{eff} are the influent and effluent concentrations in $mg{\cdot}L^{-1}$, respectively.

2.3. Removal rate constants

In order to evaluate the kinetics of nitrogen removal in the IVCWs, first-order area-based removal rate constants (k) were calculated according to Kadlec and Knight [21]: $k = \mathrm{HLR}(\mathrm{In}(C_{\mathrm{inf}}/C_{\mathrm{eff}}))$ where HLR is hydraulic loading rate ($m{\cdot}d^{-1}$), C_{inf} and C_{eff} are the influent and effluent concentrations in $mg{\cdot}L^{-1}$, respectively.

2.4. Data analysis

Statistical analysis was carried out using SPSS 17.0 software package for Windows. Two-sided paired sample t test was conducted to detect any significant differences in water qualities between the influent and effluent, and independent sample t test for pollutant removal efficiencies between the two IVCW mesocosms, with $p < 0.05$ defined as a statistically significant difference.

Table 2
Water quality characteristics of the effluent of the IVCW mesocosms (shown as mean value ± standard deviation)

Parameter	IVCW 1	IVCW 2
pH	7.6 ± 0.1	7.6 ± 0.2
DO ($mg\,L^{-1}$)	1.1 ± 0.45	1.0 ± 0.4
Conductivity (μs/cm)	592 ± 54	575 ± 54
T (°C)	18.6 ± 8.4	18.7 ± 8.3
NO_3^-–N ($mg\,L^{-1}$)	20.4 ± 2.6	19.8 ± 3.3
TN ($mg\,L^{-1}$)	25.5 ± 2.8	24.8 ± 3.1
NO_2^-–N ($mg\,L^{-1}$)	1.3 ± 0.8	1.4 ± 0.8
NH_4^+–N ($mg\,L^{-1}$)	1.7 ± 2.0	2.0 ± 1.5
COD ($mg\,L^{-1}$)	13.2 ± 8.8	12.9 ± 6.4
TP ($mg\,L^{-1}$)	2.3 ± 1.1	2.2 ± 1.0

Note: $n = 60$ for pH, DO, conductivity, and T; $n = 20$ for pollutant concentration.

3. Results

The effluent water quality characteristics and mean nitrogen removal efficiencies of the IVCW mesocosms were exhibited in Tables 2 and 3, respectively. Fig. 2 showed the temporal variations of nitrogen removal efficiencies and concentrations of NH_4^+–N and NO_2^-–N during the experimental period.

The NO_3^-–N was obviously removed by the IVCW regardless of the high loading rate ($p < 0.05$), with mean effluent concentrations of 20.4 and 19.8 $mg{\cdot}L^{-1}$ for IVCW 1 and 2, respectively (Table 2). Mean removal efficiencies of 15.5 and 18.5% for IVCW 1 and 2 were achieved, with mean removal rate constants of 0.046 and 0.055 $m{\cdot}d^{-1}$, and mass removal rate of 1.01 and 1.16 $g{\cdot}m^{-2}{\cdot}d^{-1}$, respectively (Table 3).

As for TN, mean effluent concentrations of 25.5 and 24.8 $mg{\cdot}L^{-1}$ for IVCW 1 and 2 were obtained, with dominant form of NO_3^-–N (Table 2). The removal efficiencies and mass removal rates averaged 15.5 and 18.0%, 1.23 and 1.41 $g\,m^{-2}{\cdot}d^{-1}$ for IVCW 1 and 2,

Table 3
Nitrogen removal efficiencies of the IVCW mesocosms (shown as mean value ± standard deviation, $n = 20$)

Item	IVCW 1	IVCW 2
Nitrate		
Removal efficiency (%)	15.5 ± 11.2	18.5 ± 13.3
Removal rate constant ($m\,d^{-1}$)	0.046 ± 0.043	0.055 ± 0.048
Mass removal rate ($g\,m^{-2}\,d^{-1}$)	1.01 ± 0.90	1.16 ± 0.86
TN		
Removal efficiency (%)	15.5 ± 11.2	18.0 ± 10.5
Removal rate constant ($m\,d^{-1}$)	0.044 ± 0.033	0.052 ± 0.031
Mass removal rate ($g\,m^{-2}\,d^{-1}$)	1.23 ± 0.87	1.41 ± 0.81

respectively, with mean removal rate constants of 0.044 and 0.052 $m{\cdot}d^{-1}$ (Table 3). Higher nitrogen removal was achieved in IVCW 2 although no significant difference was detected ($p > 0.05$).

The concentrations of NO_2^-–N and NH_4^+–N in the effluent, with average values of 1.3 and 1.8 $mg{\cdot}L^{-1}$, were both significantly higher than those in the influent especially after the end of November ($p < 0.05$, Fig. 2(B)).

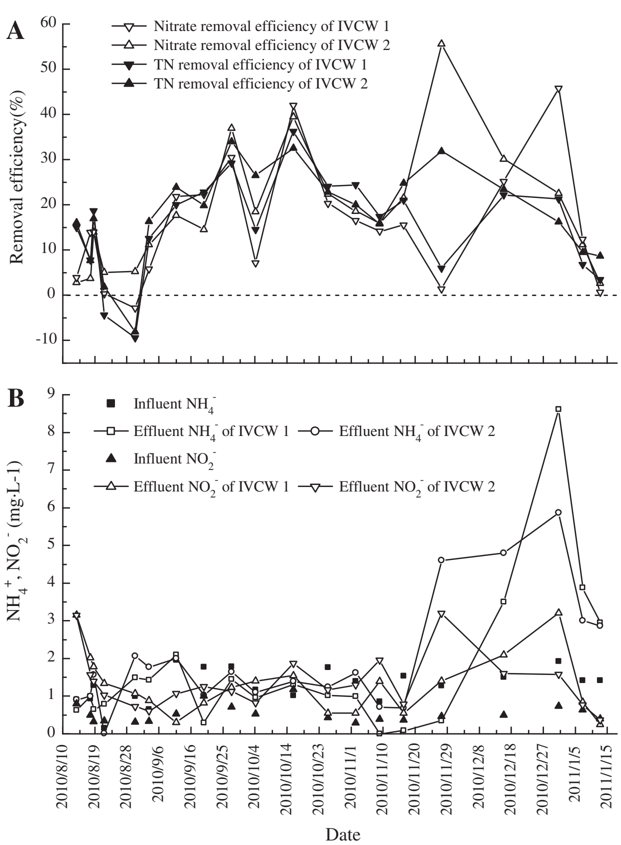

Fig. 2. Temporal variations of (A) nitrogen removal efficiencies and (B) NH_4^+–N and NO_2^-–N concentrations of the IVCW mesocosms.

4. Discussions

4.1. Influencing factors for nitrate removal in the IVCWs

It has been well documented that denitrification and plant uptake are the predominant pathways for nitrate removal in CWs [7,30]. In addition, nitrate ammonification could also contribute to nitrate removal [31]. Among them, the microbial denitrification is recognized to be the dominant long-term mechanism for nitrate removal, especially at a high nitrate loading rate [8,20]. Several hydraulic factors, including HLR, water depth, and HRT, together with carbon source supplied, characteristics of wetland substrate, macrophyte species and density, microbial communities, etc. could all affect the denitrification and thus NO_3^-–N removal in CWs [11,32,33]. The higher NO_3^-–N removal rate observed in IVCW 2 probably suggested the influence of plant species on NO_3^-–N removal.

In this trial, a significant drop of DO concentration from inlet to outlet was observed (Tables 1 and 2), which might be resulted from the severe restraint of atmosphere aeration by consistent saturation of the wetland bed, relatively large depth of the substrate, the low feeding frequency and oxidation degradations of organics and ammonia. The pH value (7.65 ± 0.14) and DO concentration (1.08 ± 0.44 $mg{\cdot}L^{-1}$) of the effluent indicated that the IVCW beds were favorable for denitrification. However, the dominant nitrogen form of NO_3^-–N in the effluent indicated that denitrification was still the limiting step for nitrogen removal in the IVCW systems. Furthermore, the significantly higher NO_2^-–N concentration in the effluent compared to the influent ($p < 0.05$) could also demonstrate denitrification in the wetland beds was incomplete [34]. The possible reasons could be deduced as follows:

4.1.1. Insufficient carbon source

Carbon source plays a very important role in nitrate removal, because it can promote the growth of denitrifying bacteria and be utilized as electron donor to fuel denitrification. It was reported that removal efficiencies for NO_3^-–N and NO_2^-–N increased with C/N ratio [8,20,35]. A significant effect of carbon addition on the nitrate removal was informed for the treatment of nitrate-contaminated wastewater using CWs [8,36,37]. Gagnon et al. [37] reported that mean nitrate removal efficiency was just 7% in HSSF CW mesocosms treating hydroponics wastewater without carbon addition, but achieved about 70% in those with sucrose addition at a COD/NO_3^-–N ratio of 3.5:1. Ingersoll and Baker [20] reported that nearly complete nitrate removal could be achieved at a critical C/NO_3^-–N of around 5:1 in FWS CW microcosms treating nitrate-contaminated wastewater. Narváez et al. [34] reported a COD/NO_3^-–N ratio of 7.7:1 for a complete denitrification in HSSF CWs-treating leachates from nurseries. By addition of fructose, removal efficiency of greater than 90% for nitrate in groundwater was obtained in FWS CWs when the COD/NO_3^-–N > 3.5 [8].

In the present experiment, mean value of COD/NO_3^-–N in the fed wastewater was only 2.0, and a significant fraction of the organic matter would be biodegraded within the upper layer of the down-flow chamber [38,39]. Therefore, limited carbon source for denitrification and accumulations of NO_3^-–N and NO_2^-–N in the wetland beds could be expected, which was in accordance with other references [20,36,40,41].

4.1.2. High loading rate

Loading rate was reported to be an important factor influencing nitrate removal in CW. Nitrate removal efficiency decreased with the increase of loading rate [20,23]. In this experiment, the HLR of the IVCW mesocosm was 250 mm/d, yielding a nitrate loading rate of $6.10\,g{\cdot}m^{-2}\,d^{-1}$, which was significantly higher or in the high range of the values compared with the references [20,21,23], implying short retention time for nitrate elimination. Lane et al. [42] suggested that over 90% of nitrate removal efficiency could be obtained for wetlands at a nitrate loading rate of less than $0.1\,g{\cdot}m^{-2}\,d^{-1}$, but the removal efficiency decreased at higher loading rates. Furthermore, the lack of availability of organic carbon was more severe at high nitrate loading rates [23].

4.1.3. Low temperature

Biological denitrification is highly depended on temperature [1,25,31,36,43], and it was reported to slow down below 15℃ and nearly cease below 5℃ due to the drastically dropped activities of denitrifying bacteria [11]. In our experiment, the sharp decrease of NO_3^-–N removal rate observed in January (Fig. 2(A)) might be caused by sharply declining denitrification at low temperature.

Additionally, the low NO_3^-–N removal rate obtained at the beginning of the operation (August, Fig. 2(A)) might be attributed to the low maturity of the mesocosms, which meant low plant productivity

Table 4
Nitrate removal efficiencies and mass removal rates of different CW systems operated at similar nitrate loading rates

Location	Wetland type	Removal efficiency (%)	Mass removal rate ($g\,N\,m^{-2}\,d^{-1}$)	Reference
Wuhan, China	IVCW	15.5 and 18.5	1.01 and 1.16	This study
Montreal, Canada	HSSF CW	7	0.5	[37]
Phoenix, USA	FWS CW	Around 10–15	0.14–0.43	[20]
Taiwan, China	FWS CW	10.0	0.67	[23]
	HSSF CW	8.5	0.61	
Plönninge, Sweden	FWS CW	5–7	0.192–0.233	[24]
Southern California, USA	FWS CW	9.7	0.131–0.902 (0.554[a])	[1]

[a]Mean value.

Table 5
Nitrate removal rate constants ($k_{nitrate}$) of different CW systems

Location	Wetland type	$k_{nitrate}$ ($m\,d^{-1}$)	Reference
Wuhan, China	IVCW	0.046 and 0.055	This study
Taiwan, China	FWS CW	0.024–0.138	[23]
	HSSF CW	0.029–0.137	
Plönninge, Sweden	FWS CW	0.05–0.07	[24]
Idaho, USA	FWS CW	0.021–0.030	[44]
USA	FWS CW	0.028–0.173 (0.041[a])	[21]
	HSSF CW	0.018–0.232 (0.079)	
USA	FWS CW	0.07–0.15 (0.09)	[45]

[a]Mean value.

for nitrate assimilation and undeveloped denitrifying bacteria communities in the wetland substrates [23,24].

4.2. Nitrate removal rate constants

Although the nitrate removal rates of the IVCW systems were not ideal, they performed better in term of efficiency and rate in comparison to FWS and SSF CWs operated at a comparable high nitrate loading rate (Table 4). With regard to nitrate removal rate constant ($k_{nitrate}$), it was near the low range of literature values (Table 5). This could be attributed to the high HLR and low availability of carbon source in the influent [20,23]. However, higher $k_{nitrate}$ was achieved in the IVCW systems compared with those of around 0.025–0.030 $m\,d^{-1}$ obtained in FWS and SSF CWs at a similar nitrate loading rate [23,44]. This could be attributed to the relatively larger treatment volume on the basis of a same surface area and the configuration of the IVCW which forces the wastewater to flow down and then up, providing a longer treatment pathway and thus allowing more contact between wastewater and substrate, in comparison to FWS and SSF CWs which possess less treatment volume and single horizontal flow for wastewater traveling from inlet to outlet.

5. Conclusions

Based on this study, it could be a promising way to treat tail water with high concentration of nitrate by IVCW. HLR, carbon source, temperature, and wetland plant species were the main influencing factors for nitrate removal among them; insufficient carbon source might be the most important limiting factor. In order to optimize its nitrate removal performance, further researches are greatly needed.

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