



## Influence of intermittent aeration and organic loading rate on lab-scale constructed wetland systems treating synthetic wastewater

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### ABSTRACT

By coupling the intermittent characteristics of rural sewage discharge and solar energy density, the performance responses of lab-scale tidal flow constructed wetland (TF) and conventional vertical flow constructed wetland (VF) to different organic loading rates (OLRs) (166.3, 300.9, 451.2, 602.4, and 751.2 g/m<sup>2</sup> d) in synthetic domestic sewage treatment were investigated to study its feasibility to replace conventional aeration and reduce the dependence of wastewater treatment plants on the grid in this study. Artificial aeration was conducted based on simulating photovoltaic aeration. The increase in OLR promoted the removal performances for the chemical oxygen demand (COD) and ammonia–nitrogen (NH<sub>4</sub><sup>+</sup>-N) in TF, which could be up to 95.7 and 97.2%, respectively, under OLR of 751.2 g/m<sup>2</sup> d. Excess organic carbons inhibited NH<sub>4</sub><sup>+</sup>-N removal through competition for oxygen available in VF, where the COD removal efficiency could be 89.6% while low NH<sub>4</sub><sup>+</sup>-N removal (40%) was obtained under OLR 751.2 g/m<sup>2</sup> d. With the increase in OLR, the removal of total nitrogen (TN) was enhanced. A higher OLR can not only provide enough electron donors but also produce more anoxic regions for denitrification. The best TN removal was 77.5% in TF and 49.8% in VF, which occurred at OLR of 751.2 and 602.4 g/m<sup>2</sup> d, respectively. A drop of TN removal in VF was attributed to the lack of nitrate–nitrogen (NO<sub>3</sub><sup>-</sup>-N) when reaching OLR of 751.2 g/m<sup>2</sup> d. The removal efficiency for total phosphorus was increased gradually with the increase in OLR. This paper suggests that appropriate control of OLR can achieve the optimal effect of pollutants removal, especially the elimination of TN.

*Keywords:* Vertical flow; Tidal flow; Photovoltaic aeration; Organic loading rate; Oxygen supply

### 1. Introduction

The discharge of rural domestic sewage is increasing with the development of rural economy and the

improvement of living standard. However, the status quo in sewage treatment is not optimistic. By 2009, the proportions of domestic sewage treated are as low as 18.1% for county towns and 4.9% for rural villages [1]. The increasing discharge of untreated sewage leads to water pollution, worsening potable water, thus

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influencing rural living environment and threatening the health of residents.

Vertical flow constructed wetlands (VF) have been widely used for wastewater purification recently owing to the fact that they improve the deficiencies of vulnerability to climate in surface flow constructed wetlands and less effective for nitrogen removal in subsurface flow constructed wetlands [2]. VF has been used successfully in Europe for the sewage treatment over the last 10 years, which could receive 90% removal for chemical oxygen demand (COD) and ammonia–nitrogen ( $\text{NH}_4^+\text{-N}$ ) [3,4].

Tidal flow constructed wetlands (TF) are a relatively new technology with a novel method of oxygen transfer [5,6], which is characterized by multiple flood and drain cycles per day. During the drained phases,  $\text{NH}_4^+\text{-N}$  adsorbed to biofilms in the flooded phases is oxidized to nitrate ( $\text{NO}_3^-\text{-N}$ ) and nitrite ( $\text{NO}_2^-\text{-N}$ ) via nitrification. In subsequent flooded phases,  $\text{NO}_3^-\text{-N}$  and  $\text{NO}_2^-\text{-N}$  desorb to the bulk water. Researchers have demonstrated that TF has the advantage of requiring approximately half the power of aerated wetlands [7].

The treatment performance in wetlands can be influenced by various factors, such as run mode, dissolved oxygen (DO), pH, temperature, organic and hydraulic loading rate (OLR and HLR), etc. OLR plays a crucial role in nitrogen removal performance by nitrification–denitrification [8], which is always the main focus when evaluating the treatment ability of constructed wetlands [9,10]. Nitrification requires aerobic condition, so excessive organic matter will inhibit nitrifying bacteria competing for oxygen [11]. Denitrification is a heterotrophic process, so is often restricted by the lack of organic carbon source [12,13]. External carbon addition was then adopted to enhance the influent organic matter in some studies to support denitrification [14,15]. Zhao et al. [16] found that conventional VF obtained total nitrogen (TN) removal efficiency of 25–62% when OLR changed from 6.7 to 26.7  $\text{g}/\text{m}^2\text{ d}$ . Another research study by Fan et al. [17] in intermittently aerated VF obtained simultaneously high removals of COD (96%),  $\text{NH}_4^+\text{-N}$  (99%), and TN (90%) at OLR of 90.3  $\text{g}/\text{m}^2\text{ d}$ . However, most researches are paid attention to VF, so research on the effect of OLR on the treatment performance of TF is seldom.

Solar power applied to treat wastewaters has been demonstrated in several studies and projects [18]. As the discharge of rural sewage, solar energy density also has the analogous feature of changing periodically, which differs significantly between daytime and night. Aeration with photovoltaic (PV) power supply [19] is a common pattern utilizing solar energy, but batteries are involved in great majority of PV systems,

which will increase the investment and maintenance costs. Therefore, information on PV systems excluding batteries, in which current pumps are connected directly with PV panels is little. The application of PV aeration constructed wetlands in rural domestic sewage treatment will not only take full use of the wetland characteristics of easy operation, efficient removal, etc., but also improve energy efficiency and reduce infrastructure investment.

By coupling the intermittent characteristics of rural sewage discharge and solar energy density, lab-scale simulating PV aeration VF and TF were carried out in this study. The effect of OLR on their treatment performance in the presence of aeration was comprehensively studied.

## 2. Materials and methods

### 2.1. Experimental setup and operation

Two lab-scale cylindrical wetland systems were operated on horizontal ground in the experiment. They were composed of one set of tidal flow unit, the other set of conventional vertical flow unit. As shown in Fig. 1, each wetland unit had a height of 1,700 mm and an inner diameter of 150 mm. Six sampling points situated at 0, 30, 60, 90, 120, 150 cm from the bottom were tapped along the reactors.

Multiple layers of substrate are arranged, to allow the sizes of particles to increase progressively from the top layer to the bottom layer [20]. However, plenty of studies have reported the occurrence of clogging when wetlands with this conventional medium arrangement are operated [21]. Therefore, an

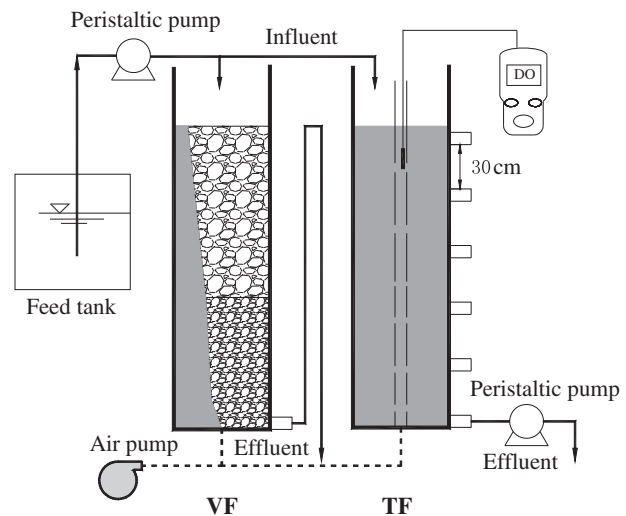


Fig. 1. Schematic diagram of the lab-scale wetland system.

alternative arrangement of media was adopted. Volcanic rocks with 8–10 mm in diameter were used in the top layer (800 mm), and ones were filled in the bottom layer (700 mm) with 3–5 mm in diameter. The porosity of the columns was 39.8%. The main components of volcanics were shown as follows: 53.82% SiO<sub>2</sub>, 8.36% CaO, 2.46% MgO, 9.08% Fe<sub>2</sub>O<sub>3</sub>, 1.12% FeO, 16.89% Al<sub>2</sub>O<sub>3</sub>, 0.06% TiO<sub>2</sub>, 2.3% K<sub>2</sub>O, 2.55% Na<sub>2</sub>O. A vertical perforated PVC pipe (1,700 mm in length and 30 mm in diameter) was inserted into the substrate in the middle of wetlands to measure various physical and chemical parameters.

Two units were installed with aeration systems and time controllers. To simulate the difference of solar energy density between daytime and night, the wetlands received aeration at an intensity of 1.53 m<sup>3</sup>/m<sup>2</sup> d, which began at 8 AM, and stopped at 5 PM with the aeration duration of 9 h/d.

The synthetic wastewater was prepared daily by mixing 168.3, 336.7, 505, 673.3, 841.7 mg/L C<sub>6</sub>H<sub>12</sub>O<sub>6</sub> (200, 400, 600, 800, 1,000 mg/L COD), 125 mg/L NH<sub>4</sub>Cl (30 mg/L TN), and 40 mg/L K<sub>2</sub>HPO<sub>4</sub>·3H<sub>2</sub>O (5 mg/L total phosphorus) into tap water. The synthetic wastewater was stored in a feeding tank, and fed into two systems at the same time at a flow rate of 25 mL/min by a peristaltic pump. The running mode was shown in Table 1. The HLR of each column was 0.76 m/d. Effluent of TF was pump out from the bottom outlet, at the rate of 27 mL/min, while it was discharged as the principle of U-tube in VF. TF was sampled after 23:00 every day while VF was collected after 17:00. The samples were stored in fridge of 4°C. After effluent, TF was kept dry for 9 h while VF was kept wet for 15 h.

## 2.2. Sampling and water quality analysis

In order to allow the microbial communities to acclimate, the two wetlands were pre-operated for 40 d. Effluents were collected every day. Laboratory analysis was performed on the water samples for COD, NH<sub>4</sub><sup>+</sup>-N, NO<sub>3</sub><sup>-</sup>-N, NO<sub>2</sub><sup>-</sup>-N, TN, and TP. NO<sub>3</sub><sup>-</sup>-N,

NO<sub>2</sub><sup>-</sup>-N, TN, and TP were carried out in accordance with standard APHA methods [22]. COD was determined using fast digestion-spectrophotometric method. NH<sub>4</sub><sup>+</sup>-N was measured using Nessler's reagent colorimetric method. All absorbances were analyzed by the spectrophotometer DR 5000 HACH. HACH HQ40d meter was used in the DO testing.

## 3. Results and discussions

### 3.1. The DO profiles in wetlands

DO is directly related to the degree of organic matter degradation, nitrification, and denitrification [23]. The profiles of DO concentration in 120–150 cm region above the reactor bottom were shown in Figs. 2 and 3. According to Fig. 2, the overall 360 min tested in TF consisted of three phases: phase I (0–100 min), phase II (100–300 min), and phase III (300–360 min). The time taken to note DO change before the DO meter merged into bulk water was defined as phase I. Phase II was the period of DO meter merged till the termination of aeration (influent), followed by one hour after stopping aeration (phase III). Note that there were no data of DO in TF under OLR 166.3 g/m<sup>2</sup> d due to no measurement. In dry period, the rapid oxygen exchange makes DO in the beds to maintain saturation level. During phase I, higher DO concentration (about 7 mg/L) was maintained in TF regardless of the value of OLR. Once DO meter merged, DO concentration declined rapidly and remained stable in a short time. A balance between oxygen transfer rate and oxygen uptake rate existed. Due to the increased OLR, the stable DO concentrations were 1.6, 1.3, 0.8, 0.6 mg/L under OLR of 300.9, 451.2, 602.4, and 751.2 g/m<sup>2</sup> d respectively. Although DO was not measured in OLR 166.3 g/m<sup>2</sup> d, it can be inferred that it must be higher than 1.6 mg/L. Although the DO concentrations appeared to exceed that required for anoxic condition, anoxic conditions could still exist due to the stratification of biofilms [24]. The DO in phase III presented a trend of decreasing first and then increasing immediately. Before DO meter exposed out, the DO dropped to the lowest because of constant oxygen consumption. As soon as the meter came out, the DO rise to a higher level until the next circle.

The profile of DO in VF (Fig. 3) showed significantly different compared with that in TF. The tested time (660 min) lasted from one hour before aeration (phase I) to one hour after the termination of aeration (phase III). Phase II was the period of aeration. Oxygen transfer rate into VF reported by Cooper et al. [25] was in the range of 50–90 g/m<sup>2</sup> d, which was far from the oxygen demand of treating strong wastewaters.

Table 1  
The running mode of constructed wetlands

Parameter	TF	VF
Influent/ Aeration	8:00–17:00	8:00–17:00
Full saturation	14:30–17:00	8:00–17:00
Effluent	14:30–23:00	8:00–17:00
Rest	23:00–8:00 (next day)	17:00–8:00 (next day)

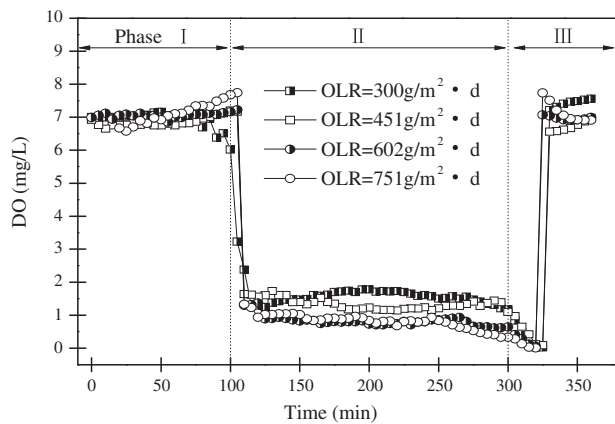


Fig. 2. The profile of DO in TF under various OLRs.

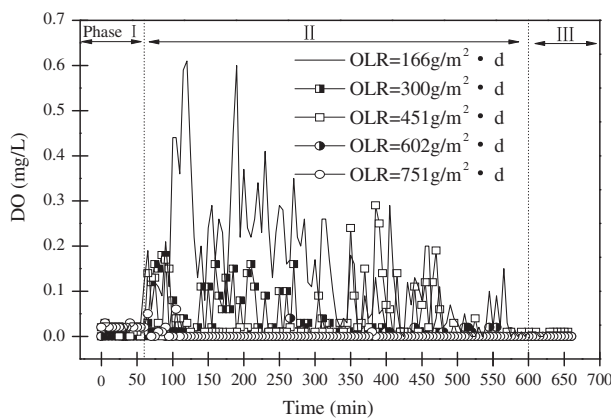


Fig. 3. The profile of DO in VF under various OLRs.

Whatever the OLR was, the DO concentration was below 0.5 mg/L during feeding, which was not beneficial for organic matter degradation and nitrification.

### 3.2. The overall performance of wetlands

OLRs of 166.3, 300.9, 451.2, 602.4, and 751.2  $\text{g}/\text{m}^2 \text{d}$  were adopted, which were shown in detail in Table 2. The effects of various OLRs on the effluent  $\text{NO}_3^-$ -N and  $\text{NO}_2^-$ -N, and removal efficiencies for COD,  $\text{NH}_4^+$ -N, TN, TP were presented in Table 3. Treatment performances differed significantly between TF and VF.

Although organic matter can be decomposed via both aerobic and anaerobic microbial processes in wetlands, aerobic degradation is usually more important [26]. Therefore, the removal performance for COD in wetlands is positively correlated with the DO condition. According to Table 3, in the presence of aeration, better COD removal efficiencies (over 85%) were obtained in two wetlands, regardless of the value of OLR. Jia et al. [23] adopted intermittent operation strategy to create more oxidizing conditions in VF and the removal efficiency for COD was more than 90%. However, if the oxygen supply is not limited due to aeration, aerobic degradation rate will be elevated and governed by the amount of active organic matter available to the organisms. So with the OLR increased from 166.3 to 602.4  $\text{g}/\text{m}^2 \text{d}$ , the COD removal efficiencies in two wetlands were enhanced gradually. Increased DO concentration through aeration could not only enhance microbial activity but also accelerate the diffusion of pollutants [27].

In general, oxygen can be introduced into conventional wetlands through (1) atmospheric diffusion, (2) plant root release, and (3) influent. Prochaska et al. [28] demonstrated that conventional VF could just receive high COD removal in low OLRs (20–40  $\text{g}/\text{m}^2 \text{d}$ ) depending on these DO source. DO deficit often occurs inside the VF, especially with excessive influent organics [29]. Oxygen transfer is significantly promoted with tidal operation through multiple filling and drain cycles. In drained period, a rapid exchange of oxygen between the atmosphere and substrate pore spaces is generated. Average oxygen supply of 350  $\text{g}/\text{m}^2 \text{d}$  via tidal operation was much higher than that in conventional VF (<100  $\text{g}/\text{m}^2 \text{d}$ ) [30]. This principle could be well reflected on this study. As the OLR was increased from 602.4 to 751.2  $\text{g}/\text{m}^2 \text{d}$ , the COD removal efficiency was still elevated in TF (95.1–95.7%), which decreased instead in VF (93.9–89.6%). The oxygen supply through just constant aeration could not meet the DO demand for excess organic matter. The effluent COD concentration in VF could be as high as 104.6 mg/L compared with 42.6 mg/L in TF.

Fig. 4 showed the mass removal rates of COD in two wetlands. With the increase in OLR in the tested range of 166.3–751.2  $\text{g}/\text{m}^2 \text{d}$ , the COD mass removal rates were enhanced in two wetlands. VF presented

Table 2  
The values of OLR (mean  $\pm$  standard deviation)

Parameter	I	II	II	IV	V
COD (mg/L)	221.7 $\pm$ 18.8	401.2 $\pm$ 9.5	601.6 $\pm$ 19.0	803.2 $\pm$ 13.1	1,001.6 $\pm$ 21.9
OLR ( $\text{g}/\text{m}^2 \text{d}$ )	166.3 $\pm$ 14.1	300.9 $\pm$ 7.1	451.2 $\pm$ 14.3	602.4 $\pm$ 9.8	751.2 $\pm$ 16.4

Table 3  
Inflow water quality, outflow water quality, and water temperature

Parameter	Unit	Standard deviation			Parameter	Standard deviation			Removal (%)	Removal (%)	Standard deviation	Removal (%)
		Mean	Standard deviation	VF (outflow)		Mean	Standard deviation	VF (outflow)				
<b>I Inflow</b>												
COD	mg/L	221.7	18.80	TF (outflow)	33.0	3.11	COD	32.1	6.19	85.3	85.3	
NH <sub>4</sub> <sup>+</sup> -N	mg/L	25.7	0.79	COD	3.6	0.29	NH <sub>4</sub> <sup>+</sup> -N	5.2	1.91	79.6	79.6	
NO <sub>3</sub> <sup>-</sup> -N	mg/L	0	0	NH <sub>4</sub> <sup>+</sup> -N	22.6	1.57	NO <sub>3</sub> <sup>-</sup> -N	16.3	3.33	-	-	
NO <sub>2</sub> <sup>-</sup> -N	mg/L	0	0	NO <sub>3</sub> <sup>-</sup> -N	0.1	0.02	NO <sub>2</sub> <sup>-</sup> -N	0.1	0.04	-	-	
TN	mg/L	29.8	0.93	NO <sub>2</sub> <sup>-</sup> -N	27.5	1.79	TN	22.6	2.42	24.2	24.2	
TP	mg/L	5.0	0.30	TN	3.9	0.27	TP	4.1	0.22	17.3	17.3	
Temperature	°C	17.1	0.98	TP								
<b>II</b>												
COD	mg/L	401.2	9.52	COD	34.5	5.52	COD	33.5	3.79	91.6	91.6	
NH <sub>4</sub> <sup>+</sup> -N	mg/L	27.0	1.33	NH <sub>4</sub> <sup>+</sup> -N	2.6	0.73	NH <sub>4</sub> <sup>+</sup> -N	11.3	1.48	58.3	58.3	
NO <sub>3</sub> <sup>-</sup> -N	mg/L	0	0	NO <sub>3</sub> <sup>-</sup> -N	18.9	1.38	NO <sub>3</sub> <sup>-</sup> -N	6.4	1.57	-	-	
NO <sub>2</sub> <sup>-</sup> -N	mg/L	0	0	NO <sub>2</sub> <sup>-</sup> -N	0.3	0.15	NO <sub>2</sub> <sup>-</sup> -N	0.1	0.05	-	-	
TN	mg/L	31.6	1.53	TN	22.6	1.28	TN	18.7	0.88	40.9	40.9	
TP	mg/L	5.0	0.46	TP	3.6	0.35	TP	4.1	0.50	18.4	18.4	
Temperature	°C	15.5	1.05	TP								
<b>III</b>												
COD	mg/L	601.6	18.96	COD	33.7	4.04	COD	40.9	3.80	93.2	93.2	
NH <sub>4</sub> <sup>+</sup> -N	mg/L	26.5	2.41	NH <sub>4</sub> <sup>+</sup> -N	1.9	0.27	NH <sub>4</sub> <sup>+</sup> -N	11.6	0.85	55.6	55.6	
NO <sub>3</sub> <sup>-</sup> -N	mg/L	0	0	NO <sub>3</sub> <sup>-</sup> -N	15.6	1.42	NO <sub>3</sub> <sup>-</sup> -N	3.3	0.77	-	-	
NO <sub>2</sub> <sup>-</sup> -N	mg/L	0	0	NO <sub>2</sub> <sup>-</sup> -N	0.3	0.07	NO <sub>2</sub> <sup>-</sup> -N	0.2	0.07	-	-	
TN	mg/L	30.3	2.02	TN	18.3	1.33	TN	17.1	2.37	43.4	43.4	
TP	mg/L	4.8	0.54	TP	3.0	0.19	TP	3.2	0.24	32.2	32.2	
Temperature	°C	17.7	0.99	TP								
<b>IV</b>												
COD	mg/L	803.2	13.10	COD	36.1	4.08	COD	48.3	10.80	93.9	93.9	
NH <sub>4</sub> <sup>+</sup> -N	mg/L	26.3	1.93	NH <sub>4</sub> <sup>+</sup> -N	1.8	0.41	NH <sub>4</sub> <sup>+</sup> -N	12.9	1.94	50.9	50.9	
NO <sub>3</sub> <sup>-</sup> -N	mg/L	0	0	NO <sub>3</sub> <sup>-</sup> -N	6.5	3.44	NO <sub>3</sub> <sup>-</sup> -N	1.4	1.34	-	-	
NO <sub>2</sub> <sup>-</sup> -N	mg/L	0	0	NO <sub>2</sub> <sup>-</sup> -N	0.3	0.15	NO <sub>2</sub> <sup>-</sup> -N	0.2	0.09	-	-	
TN	mg/L	30.0	2.64	TN	9.2	3.52	TN	15.0	2.19	49.8	49.8	
TP	mg/L	4.7	0.43	TP	2.1	0.32	TP	3.1	0.40	33.2	33.2	
Temperature	°C	16.0	0.56	TP								
<b>V</b>												
COD	mg/L	1001.6	21.90	COD	42.6	4.29	COD	104.6	24.23	89.6	89.6	
NH <sub>4</sub> <sup>+</sup> -N	mg/L	25.5	2.27	NH <sub>4</sub> <sup>+</sup> -N	0.7	0.64	NH <sub>4</sub> <sup>+</sup> -N	15.2	1.05	40.0	40.0	
NO <sub>3</sub> <sup>-</sup> -N	mg/L	0	0	NO <sub>3</sub> <sup>-</sup> -N	3.5	1.40	NO <sub>3</sub> <sup>-</sup> -N	0.1	0.06	-	-	
NO <sub>2</sub> <sup>-</sup> -N	mg/L	0	0	NO <sub>2</sub> <sup>-</sup> -N	0.2	0.07	NO <sub>2</sub> <sup>-</sup> -N	0.1	0.08	-	-	
TN	mg/L	29.6	3.75	TN	6.7	2.79	TN	20.5	2.84	30.8	30.8	
TP	mg/L	4.8	0.33	TP	2.2	0.27	TP	3.1	0.29	35.0	35.0	
Temperature	°C	16.9	0.97	TP								

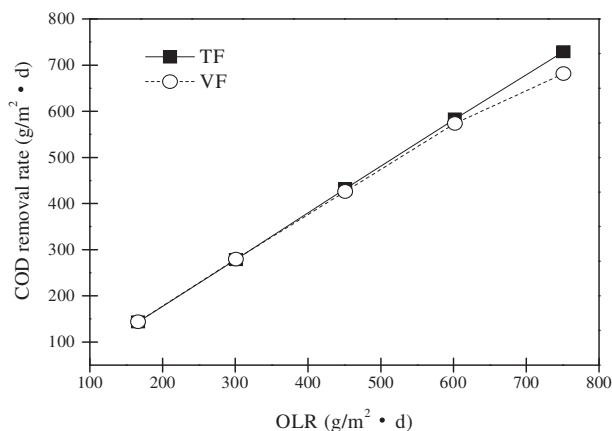


Fig. 4. Mass removal rates of COD with increased OLR in TF and VF.

pretty close to TF in mass removal rate of COD with OLR ranged from 166.3 to 451.2 g/m<sup>2</sup> d. However, due to the lack of oxygen, VF obtained a little smaller increasing trend than TF in the range of 451.2–751.2 g/m<sup>2</sup> d.

NH<sub>4</sub><sup>+</sup>-N can be removed through nitrification, ammonia volatilization, anaerobic ammonia oxidation, and nutrient assimilation into biomass [10,31], among which nitrification plays the most important role in wetlands [32]. Nitrification is an aerobic chemoautotrophic microbial process. Table 3 showed that the opposite trends appeared in NH<sub>4</sub><sup>+</sup>-N removal efficiency between TF and VF with the increased OLR. With the increase in OLR, the removal rate for NH<sub>4</sub><sup>+</sup>-N in TF was enhanced from 86.2 to 97.2%, while it dropped from 79.6 to 40.0% in VF. It is generally accepted that the DO concentrations above 1.5 mg/L are essential for nitrification [33]. Meanwhile, the DO available in wetlands is primarily consumed for COD degradation. A higher OLR would favorably select heterotrophic bacteria against autotrophic ammonia oxidation bacteria through competition for substrates and oxygen, thus rapidly inhibiting NH<sub>4</sub><sup>+</sup>-N removal. Results indicated that the oxygen supply through constant aeration in VF was primarily consumed to decompose the excess organic matter. The increased organics consumed more available DO and thus further restrained the activity of autotrophic oxygen-dependent bacteria, thus led to the decrease in nitrification rate. Therefore, significant reduction of NH<sub>4</sub><sup>+</sup>-N removal could be observed with the increase in OLR.

Owed to its unique running mode, the total oxygen available in TF could not only be applied to deal with excess organic matter but also guarantee efficient nitrification. After filling, the DO concentration

remained approximately 1.6, 1.3, 0.8, 0.6 mg/L under OLR of 300.9, 451.2, 602.4, and 751.2 g/m<sup>2</sup> d, respectively. Moreover, microbial assimilation occurred to help to remove NH<sub>4</sub><sup>+</sup>-N due to the increase in OLR. The amount of NH<sub>4</sub><sup>+</sup>-N immobilized by biomass assimilation is up to 0.074 g for each gram of biochemical oxygen demand removal [34]. However, nutrient assimilation into micro-organisms is highly insignificant compared to influent nitrogen loads.

Complete TN elimination is firstly dependent on efficient nitrification and then good denitrification. After nitrification, in the presence of organic carbons as the electron donors, facultative bacteria use nitrates as the terminal electron acceptor in anoxic condition and then transform them into innocuous fundamental nitrogen gas [35,36]. Denitrification is reported to account for a proportion of typically 60–95% of TN permanently removed from wetlands [37]. The profiles of removal performance for TN in two wetlands illustrated in Table 3 differed slightly with the increase in OLR. TF presented an increasing trend, while VF showed a trend of first increasing and then decreasing.

Denitrification is facilitated with DO level <0.5 mg/L. In the OLR range of 166.3–451.2 g/m<sup>2</sup> d, VF performed better TN removal than TF, although the former showed a relatively lower nitrification rate. Plenty of oxygen transfer in TF destroyed the anoxic conditions, which was not beneficial for denitrification. However, even though prevailing anoxic and anaerobic conditions in VF offered suitable environment for denitrification, the supply of organic matter was limited, thus inhibiting denitrification process. NO<sub>3</sub><sup>-</sup>-N and NO<sub>2</sub><sup>-</sup>-N as electron acceptors could not be removed permanently unless sufficient organic carbon was supplied as electron donor in anoxic condition. Intermittent operation adopted by Jia et al. [23] significantly enhanced NH<sub>4</sub><sup>+</sup>-N removal, which could be 93.9% while low TN removal (46.9%) was obtained because of low carbon source supply. In this OLR range, the TN removal efficiencies of TF and VF increased from 7.7 to 39.4%, 24.2 to 43.4%, respectively.

Sufficient denitrification will occur with rich nitrates and carbons. When OLR elevated to 602.4 and 751.2 g/m<sup>2</sup> d, the TN removal efficiencies in TF were up to 68.9 and 77.5%. A higher OLR could not only provide enough electron donors but also produce more anoxic regions for denitrification. However, denitrification in VF was limited by the lack of electron acceptor (nitrate) under higher OLR. The effluent NO<sub>3</sub><sup>-</sup>-N dropped to 1.4, 0.1 mg/L under OLR of 602.4, 751.2 g/m<sup>2</sup> d, respectively. So strategy of greater aeration rate could be adopted for nitrification enhancement by increasing the DO concentrations [38,39].

The primary phosphorus removal mechanisms in wetland systems are microbial assimilation, plant uptake, adsorption, complexation, and precipitation [40]. Because of the wetlands in this study unplanted, the contribution of plant uptake to TP removal could be eliminated. With the increase in OLR, the removals for TP were elevated in two wetlands, which increased from 21.2 to 54.2%, 17.3 to 35.0%, respectively.

Generally, substrate adsorption, complexation, and precipitation represent the main removal pathways. Phosphate retention in wetlands is dependent upon the type of media, as well as the calcium, aluminum, and iron contents of the substrates [41]. Moreover, the removal efficiency of TP depends strongly on input concentration. Influent TP concentration determines the percentage removed by adsorption or biomass [42]. When the input TP concentration is higher than 0.5 mg/L, adsorption is the main removal mechanism. At input concentrations lower than 0.25 mg/L, the adsorption is weak and biomass becomes more important. According to Table 3, the average influent TP concentration was 4.8 mg/L, which determined that adsorption was the main TP removal approach in this study. However, due to poor adsorption substrates used, the proportion of TP removal via adsorption was low. A study on VF equipped with different substrates in the runoff treatment by Chen et al. [43] demonstrated under the same condition, volcanics received just 13.3% TP removal, much lower than woodchip (38.8%), pot gravel (64.4%), and synthetic fiber (74.5%).

Biological phosphorus removal is based on the activity of phosphorus accumulating organisms (PAOs). In the anaerobic phase, PAOs take up organic carbons and store them as polyhydroxyalkanoates (PHAs), meanwhile followed by the release of orthophosphate ( $\text{PO}_4^{3-}\text{-P}$ ) to the bulk water. About 0.67–0.84 g COD will be taken to release 1 g  $\text{PO}_4^{3-}\text{-P}$  into the bulk liquid [44]. In the subsequent aerobic phase, PAOs use the PHAs for generating energy for growth, glycogen synthesis, and luxury phosphate uptake. Sufficient oxygen supply was in favor of excess phosphorus absorption by PAOs, which can well explain the phenomenon that TF performed better TP removal than VF, regardless of the value of OLR. The competition between phosphorus release and denitrification for the available carbon source was blamed to the slight drop of TP removal under OLR 751.2 g/m<sup>2</sup> d.

Microbial assimilation to micro-organisms also contributes to the TP removal. The amount of  $\text{PO}_4^{3-}\text{-P}$  by biomass assimilation is up to 0.02 g for removing 1 g COD [45]. However, the phosphorus in bacteria only accounts for less than 2% of the nutrients removed. Only when OLRs are very high, can the TP removed by micro-organisms reach a significant percentage [46].

#### 4. Conclusions

The total oxygen supply in TF matches the demand for COD degradation and nitrification. Although DO exceeds that required for anoxic condition, anoxic conditions could still exist due to the stratification of biofilms. Whatever the OLR was, the DO concentration was below 0.5 mg/L during feeding in VF.

Increased OLR promoted the performances of COD and  $\text{NH}_4^+\text{-N}$  degradation in TF. High removal rates of COD (95.7%) and  $\text{NH}_4^+\text{-N}$  (97.2%) were obtained simultaneously with OLR of 751.2 g/m<sup>2</sup> d. However, excess organics consumed more oxygen available in VF, thus inhibiting the  $\text{NH}_4^+\text{-N}$  removal through nitrification. The COD removal efficiency could be 89.6% while low  $\text{NH}_4^+\text{-N}$  removal (40%) was obtained under OLR 751.2 g/m<sup>2</sup> d.

The removal of TN was enhanced by the increase in OLR, which could not only provide enough electron donors but also produce more anoxic regions for denitrification. The best TN removal efficiencies occurred at OLR of 751.2 g/m<sup>2</sup> d in TF (77.5%) and at OLR of 602.4 g/m<sup>2</sup> d in VF (49.8%). A drop of TN removal in VF was attributed to the lack of  $\text{NO}_3^-\text{-N}$  when reaching OLR of 751.2 g/m<sup>2</sup> d.

The increase in OLR was beneficial to TP removal, which was 54.2 and 35% in TF and VF, respectively. Due to the introduction of volcanic, the proportion of TP removal through substrates adsorption was low, which was blamed to the overall lower TP removal performance.

It is feasible that PV intermittent aeration system without batteries replaces conventional aeration and it can reduce the dependence of wastewater treatment plants on the grid.

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