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Dynamics of organic matter, nitrogen and phosphorus removal and their interactions in a tidal operated constructed wetland

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ABSTRACT

This paper demonstrates the potential of tidal flow operated constructed wetland application for the removal dynamics of organic matter, nitrogen and phosphorus. Near-complete removal of organic matter was achieved with a constant removal efficiency of 95%, irrespective of TOC influent loadings ranged from 10 $g/m^2 \cdot d$ to 700 $g/m^2 \cdot d$. High $NH_4^+ - N$ removal at 95% efficiency under influent loading of 17 $g/m^2 \cdot d$, was stably obtained and was not negatively influenced by increasing influent organic carbon loading rate. Increased influent TOC loading (350 $g/m^2 \cdot d$ to 700 $g/m^2 \cdot d$) significantly enhanced denitrification capacity and increased TN removal from 30% to 95%. Under tidal flow operation, a higher carbon supply ($C/N = 20$) for complete TN removal was demonstrated as comparing to that observed in traditional CWs approaches. In addition, the removal of phosphorus was strongly influenced by organic loadings. However, further investigations are needed to elucidate the detailed mechanism that would explain the role of organic loading in phosphorus removal.

1. Introduction

Constructed wetlands (CWs) have became increasingly and extensively used around the world for purifying wastewater due to their advantages, such as low operating cost and versatile pollutant removal performance (Knight et al., 2000; Bezbaruah and Zhang, 2003; Brisson and Chazarenc, 2009). The application of CWs as an effective, economical, and environment-friendly treatment alternative has already been successfully demonstrated in the treatment of various wastewaters (Scholes et al., 1999; Tanaka et al., 2006), including municipal (Hadad et al., 2006) and domestic sewage (Brix and Arias, 2005), industrial (Mitsch and Wise, 1998) and agricultural effluent (Sun et al., 1999), urban runoff (Cooper et al., 1996), mine drainage (Perkins and Hunter, 2000), and landfill leachate.

Horizontal subsurface flow CWs (HSSF CWs) and vertical flow CWs are two traditional types of wetlands that have been widely used in the past five decades. In these subsurface flow CWs, the pollutants are removed by multi-functioning substrate, microorganisms, and wetland plants as the wastewater flows through the wetland beds. Oxygen transfer and distribution in wetlands have been shown to play an important role in the performance of CWs. Moreover, the degradation of organic matter and ammonium is closely related to oxygen availability in the wetland system. The oxygen required for the degradation of pollutants in conventional HSSF CWs mainly comes from plant root secretion and atmospheric diffusion. However, the amount of oxygen secreted by plant roots and diffused from the atmosphere is extremely limited in a long-term saturated condition (Bezbaruah and Zhang, 2005; Wu et al., 2001). Despite that HSSF CWs are the most commonly applied CWs in Europe, the limited oxygen transfer capacity significantly constrains the quality of HSSF CWs performance in the reduction of organic matter and nitrogen in wastewater (Brix, 1994; Sun et al., 1999; Luederitz et al., 2001).

Compared with HSSF CWs, vertical flow CWs have a higher oxygen transfer rate of 17 $g/m^2 \cdot d$ to 25 $g/m^2 \cdot d$ due to the unsaturated water flow in the wetland beds (Wallace, 2013). However, such oxygen transfer capacity often fails to fully meet the microbial need for carrying out organic matter oxidation and nitrification. This is particularly important for the treatment of high-strength effluents, such as wastewater from pig farms (Zhao et al., 2004), and wine distillery effluent (Bernet et al., 1996). Therefore, investigations of new configurations and/or operation strategies that are modified based on traditional wetlands are essential to further

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improve treatment performance of CWs (Wu et al., 2014).

Tidal flow CWs adopt an innovative oxygen-intensifying operation and have been reported as another effective approach to improve the performance of CWs (Green et al., 1997; Sun et al., 1999; Leonard et al., 2003; Zhao et al., 2004; Austin, 2006). The tidal flow CWs act as a passive pump that repels and draws air from the atmosphere into the matrices while wastewater is rhythmically filling and draining into the CWs (Zhao et al., 2004). In addition, tidal flow CWs require only half the energy and area used by aerated CWs in treating the same volume of wastewater (Austin and Nivala, 2009). A few studies have demonstrated the increased effectiveness of tidal flow CWs in organic matter removal and ammonium oxidation due to their high reoxygenation capacity. However, limited denitrification process has also been reported as the increased accumulation of nitrate in the effluent of wetlands. This accumulation of nitrate may be caused by the aerobic environment in tidal flow wetland beds that inhibits the growth and activity of denitrifiers. It could also be attributed to the inadequate amount of carbon supply for the denitrification process that is dependent on the degradation priority of organic matter. However, knowledge on the dynamics of nitrogen transformations as related to varying organic loading rates remains fairly limited.

This study aims to explore the effect of increasing organic loading rates on the dynamics of nitrogen and phosphorus transformations in tidal flow CWs. Organic matter removal and oxygen transfer and consumption in wetlands are directly examined and calculated. The development of biomass on the surface of substrates and the intensity of nitrification and denitrification are also evaluated to better understand the dynamics of nitrogen transformation processes and its relationship with various rates of organic loading.

2. Materials and methods

2.1. Laboratory-scale CW system description

The laboratory-scale tidal flow CW used in this study was made of poly vinyl chloride (PVC) column measuring 1800 mm in height and 1200 mm in diameter. The PVC column was filled with gravel (φ10 mm to 50 mm) to a depth of 400 mm to create a bottom drain layer with an average porosity of 75%. It was also filled with coarse sand (φ5 mm to 10 mm) to a depth of 1000 mm to generate a main filter layer with an average porosity of 41% above the drain layer. The gravel drain layer at the bottom of the column was designed to enhance the negative pressure that would facilitate air intake into the matrix. The wetland was planted with *Juncus effusus* during the experimental period and then kept in an indoor area at the Bio-Energy Engineering and Low Carbon Technology Laboratory of China Agricultural University.

To better understand the microbial transformations of carbon, nitrogen, and phosphorus, artificial wastewater containing organic carbon, ammonium, and phosphate was used in this study. Influent artificial wastewater was prepared in a feed tank and fed into the wetland by a vacuum pump. To investigate the influence of organic loadings on the dynamics of microbial nitrogen transformation in tidal flow CWs and to explore the maximum capacity of organic matter removal, five experimental phases of different influent concentrations were conducted. The composition of wastewater used in this study during different experimental phases is listed in Table 1. The wastewater used for loading the wetland contains glucose as an organic compound, with total organic carbon (TOC) concentrations varying between 30 mg/L and 1200 mg/L during different experimental phases. Potassium dihydrogen phosphate and ammonium chloride were used in the preparation of the inorganic compounds. In all cases, a trace mineral solution (Wu et al., 2012) containing EDTA-Na (0.100 g/L), $FeSO_4 \cdot 7H_2O$ (0.100 g/L), $MnCl_2 \cdot 4H_2O$ (0.100 g/L), $CoCl_2 \cdot 5H_2O$ (0.170 g/L), $CaCl_2 \cdot 6H_2O$ (0.100 g/L), $ZnCl_2$ (0.100 g/L), $CuCl_2 \cdot 5H_2O$ (0.020 g/L), $NiCl_2 \cdot 6H_2O$ (0.030 g/L), H_3BO_3 (0.010 g/L), $Na_2MoO_4 \cdot 2H_2O$ (0.010 g/L), and H_2SeO_3 (0.001 g/L) was added to the artificial wastewater (1 mL/L).

Table 1
Operation parameters during experimental phases A to E (in mg/L).

Phases	TOC ($C_6H_{12}O_6$)	$NH_4^+ - N$ (NH_4Cl)	$PO_4^{3-} - P$ (KH_2PO_4)
A	20 (55)	10 (38.2)	5 (17.4)
B	100 (275)	30 (114.7)	5 (17.4)
C	300 (825)	30 (114.7)	5 (17.4)
D	600 (1650)	30 (114.7)	5 (17.4)
E	1200 (3300)	30 (114.7)	5 (17.4)

The flood period started when wastewater filled the wetland, whereas the drain period started when pore water of the wetland bed had been completely discharged. The flood and drain cycles were set to occur every 8 h, with each of the cycle lasting for 4 h. The pore volume of the bed is 200 L. The effluent was recycled as influent at a ratio of 3:1, which provided the system with a hydraulic loading rate of 0.53 $m^3/m^2 \cdot d$.

2.2. Sampling and analysis

To evaluate the pollutants removal performance of the tidal flow operated wetland, wastewater samples were collected in triplicate from the influent and effluent flow-through at 9 AM every two days. The samples were analyzed immediately for dissolved oxygen (DO), pH, oxidation redox potential (ORP), TOC, ammonium ($NH_4^+ - N$), nitrate ($NO_3^- - N$), nitrite ($NO_2^- - N$), and phosphate ($PO_4^{3-} - P$). To investigate the dynamics of carbon and nitrogen transformations across the profile of the wetland bed, 13 samples were collected along with 50 mL pore water at each vertical sampling point at an interval depth of 10 cm from the top to the bottom of the bed. The samples were stored without headspace at 4 °C and analyzed for the aforementioned parameters on the same day. To investigate the relationship between oxygen consumption and biodegradation of pollutants in the wetland bed, 13 air samples were collected using a portable oxygen/carbon dioxide detector at each vertical sampling point at an interval depth of 10 cm from the top to the bottom of the bed. The samples were analyzed immediately upon collection using the detector.

2.2.1. Water analysis

DO, pH, and ORP were measured using a portable Orion 5-Star DO/pH/ORP meter, a DO electrode (086030MD, Thermo, USA), a pH electrode (9172BNWP, Thermo, USA), and an ORP electrode (9172BNWP, Orion, USA). TOC was determined using a TOC analyzer (TOC-VPN, Shimadzu, Japan). $NH_4^+ - N$ (4500-NH_3 F. phenate method), $NO_2^- - N$ ($4500 - NO_2^-$ B. colorimetric method), and $PO_4^{3-} - P$ (4500-P E. ascorbic acid method) levels were measured using an ultraviolet–visible spectrophotometer (722S, China) according to the standard calibration and operation procedures. $NO_3^- - N$ content was analyzed using continuous flow colorimetry (SEAL Auto-Analyzer 3, British). Total nitrogen (TN) was calculated as the sum of the measurements for ammonium, nitrite, and nitrate.

2.2.2. Air analysis

The volume fraction of O_2 and CO_2 were monitored using a portable oxygen/carbon dioxide detector (HND880-O_2, China; and HND880-CO_2, China) at a resolution ratio of 0.1 vol% during each experimental phase.