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# An innovative biochar-amended substrate vertical flow constructed wetland for low C/N wastewater treatment: Impact of influent strengths



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## GRAPHICAL ABSTRACT



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### ABSTRACT

Application of biochar can be an effective and low cost technique for wastewater treatment while reducing greenhouse gas emissions. In this study, biochar was used as substrates in Vertical flow constructed wetlands (VFCWs) for enhancing the removal of contaminant from low C/N wastewaters with different influent strengths. The removal of organic matter and nitrogen in biochar-added and non-biochar-added VFCWs with different low C/N influent strengths were evaluated systematically. The results demonstrated that combining VFCWs and biochar addition could be an appropriate strategy as compared to conventional VFCWs with average removal of organic pollutants (85%), NH4<sup>+</sup>-N (39%) and TN (39%) especially at high influent strengths. Meanwhile, N<sub>2</sub>O emission was also significantly lower in biochar-added VFCWs (138–1008  $\mu$ g m<sup>-2</sup> h<sup>-1</sup>) than that in non-biochar-added VFCWs (164–1304  $\mu$ g m<sup>-2</sup> h<sup>-1</sup>) under different influent strengths. We believe that VFCWs by adding biochar can be a useful technology for treating low C/N wastewaters.

#### 1. Introduction

With the rapid development of urbanization and economy, China has been facing many serious environmental issues. One of the typical problems is the water pollution triggered due to decentralized domestic wastewaters in vast rural areas is discharged directly into water bodies (Wu et al., 2011; Zhang et al., 2005; Ye and Li, 2009). Constructed

wetlands (CWs) are recognized as a promising technique for wastewater treatment with low energy consumption and easy management (Wu et al., 2015; Vymazal, 2010). According to the differences of water distribution and flow shape, CWs can be divided into free water surface (SFS) CWs, horizontal flow (HF) CWs and vertical flow (VF) CWs (Saeed and Sun, 2012; Wu et al., 2016). Among these CWs types, VFCWs have been widely applied for wastewater treatment owing to a higher oxygen

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transfer rate and small footprint (Saeed and Sun, 2012; Zhang et al., 2009).

Substrate, as one of the key elements of CWs in wastewater treatment, can be used to intercept significant contaminant in wastewater through sedimentation, filtration and adsorption (Dordio et al., 2007). Then microorganism living in substrate can remove pollutants ultimately through nitrification and denitrification, which are the main pathways for nitrogen and organic carbon removal in CWs (Wang et al., 2016; Wu et al., 2015). At present, typical and common fillers include zeolite, gravel, limestone, coal ash and some industrial byproducts (Lu et al., 2016). Some previous studies have demonstrated that different substrates have different abilities for effective water purification (Lu et al., 2016). Meanwhile, Vymazal (2007) indicated that high removal performances of organics such as chemical oxygen demand (COD) could be achieved in these conventional VFCWs treating low-strength wastewaters. However, the effects of above-mentioned fillers to treat highstrength wastewater are not ideal, and they can restrain the removal efficiencies of total nitrogen (TN) and phosphorus (P). Simultaneously, wastewater treatment in traditional CWs may release large nitrous oxide (N<sub>2</sub>O) emissions causing air pollution (Li et al., 2017). Therefore, finding a suitable and low cost filler to enhance wastewater treatment in CWs is a critical issue.

Recently, studies have increasingly paid close attention to the use of biochar to enhance CWs performance in wastewater treatment. Biochar is a product of pyrolysis of agricultural biomass waste and has been recognized as a multifunctional material for environmental applications (Lehmann, 2007). Biochar with characteristics such as large specific surface area and highly porous structure has been shown to be effective in the adsorption and thereby immobilizes pollutants in contaminated water (Beesley et al., 2011; Mohan et al., 2014). Additionally, biochar is a carbon rich material, indicating that it can be a potential carbon source for facilitating denitrification of low C/N wastewater (Liang et al., 2006; Liu and Zhang, 2009). A few research studies have detected that removal of COD, ammonia nitrogen ( $NH_4^+$ -N), nitrate nitrogen (NO3<sup>-</sup>-N) and TN in CWs with biochar addition were more efficient than those in conventional CWs (Gupta et al., 2015; De Rozari et al., 2015). Zhou et al. (2017) also revealed that biochar could reduce N<sub>2</sub>O production in CWs, which could lead to the reduction in atmospheric greenhouse gases.

Notwithstanding, biochar is conductive to environmental remediation and has been investigated widely. Nevertheless, the performance of CWs systems with the application of biochar when treating highstrength wastewater has not been systematically surveyed to any significant extent. Therefore, the primary objectives of this study were twofold: (a) to evaluate the effect of influent strength on the removal efficiency of COD,  $NH_4^+$ -N, TN and N<sub>2</sub>O emission in VFCWs combined with biochar addition for treating low C/N sewage; and (b) to elucidate the transformations of organics and nitrogen and variation of N<sub>2</sub>O emission under different influent strengths.

#### 2. Material and methods

#### 2.1. Apparatus setup and operation

The experiment was carried out in Northwest A & F University. Eight parallel microcosm VFCWs were set up for treating simulated domestic wastewater with the same configurations (height, 65 cm; diameter, 20 cm). The schematic diagram of the experimental VFCWs is shown in Fig. 1. The structure and multidimensional gradation of substrate of each system was described in detail by Zhou et al. (2017). In order to measure various physical and chemical parameters in situ, a vertical perforated PVC pipe was installed into the substrate. In this study, *Oenanthe Javanica* species were collected from water channels and were planted in all VFCWs at a density of 12 rhizomes per systems. Based on the previous study (Xu et al., 2012), biochar which was made from bamboo at a 500 °C under anaerobic environment was chose and

used in this experiment. The proportion of mixing biochar and fine gravel was 1:1 (volume ratio). The experimental VFCWs were acclimatized for a period of four weeks, for allowing the development of plants. At this time, supernatant liquid (300 ml) of activated sludge from the secondary sedimentation was added into each system for introducing microbes, and then the experiment started. During the whole experimental period, temperature was 22–27 °C.

#### 2.2. Operation of the experimental CWs

Synthetic wastewater was dosed manually in every CW system. Each system held 5.5 L wastewater and effluent was discharged from the outlets at the bottom of VFCWs. The influent strength was manipulated by changing the content of sucrose and (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub>. The characteristics of four different influent strengths (I, II III and IV) were shown in Table 1. The average concentrations of strength I was 200 mg  $L^{-1}$  COD, 40 mg L<sup>-1</sup> TN, strength II was 400 mg L<sup>-1</sup> COD, 80 mg L<sup>-1</sup> TN, strength III was 600 mg L<sup>-1</sup> COD, 120 mg L<sup>-1</sup> TN and strength IV was 800 mg L<sup>-1</sup> COD, 160 mg L<sup>-1</sup> TN. The eight VFCWs were divided into four groups, where each group was operated with different strengths of influent, and the two VFCWs in each group were fed with the same influent strength, one of which was biochar addition. Hydraulic retention time (HRT) was 72 h according to a previous study (Wu et al., 2016), which just a cycle in this study. At about 8: 00 am on the first day of each cycle, the effluent was supplied in batch mode into each VFCW within 15 min. In particular, in order to elucidate the transformation of pollutants in each system, one cycle was chose as a typical cycle in the 3th day of the operation period.

#### 2.3. Sampling and analysis

#### 2.3.1. Water sampling and analysis

Water samples of influent and effluent in eight systems were used to analyze the treatment performance of organics and nitrogen every three days. In the typical cycle, the water samples were collected 15 times at different time to analyze the transformation of organics and nitrogen. The collected samples were stored at -4 °C before testing. COD, NH<sub>4</sub><sup>+</sup>-N, NO<sub>3</sub><sup>-</sup>-N, nitrite nitrogen (NO<sub>2</sub><sup>-</sup>-N), and TN were analyzed according to standard methods (APHA, 2005). Dissolved oxygen (DO) was measured in situ at the midpoint of the water depth from the PVC pipe by a DO meter (HQ 30d 53LEDTM HACH USA) and the values of pH was measured by a pH meter (PHSJ-4F).

#### 2.3.2. Gas collection and measurement

Gas sampling was done every experimental cycle using the staticstationary chamber to determine N<sub>2</sub>O fluxes from eight systems. In the typical cycle, the gas samples were collected 10 times at different time to monitor the variation of gas release. Gas samples were collected at 0, 20, 40 and 60 min between 9:00 and 10:00 am of the sampling day. The steps of gas sampling were described in detail in the previous studies (Wu et al., 2009). The N<sub>2</sub>O concentration was measured using the gas chromatography (GC, Agilent Technologies 7890B) within 24 h. The detail calculating method of N<sub>2</sub>O emission fluxes ( $\mu g m^{-2} h^{-1}$ ) was also elucidated in the previous studies (Wu et al., 2009).

#### 2.4. Statistical analysis

All statistical analyses were carried out with software SPSS 11.0 (SPSS Inc., Chicago, USA), including analysis of variance (ANOVA), Bartlett's and Levine's test for homogeneity of variance and normality, and Duncan's multiple range tests for differences between means. Repeated measures analysis of variance was used to evaluate the significance of differences between the experimental eight CWs. In all tests, differences and correlations were considered statistically significant when P < 0.05.

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