Evaluating the sustainability of free water surface flow constructed wetlands: methane and nitrous oxide emissions

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Abstract

Constructed wetlands (CWs) have been used as a green technology to treat various wastewaters for several decades, and greenhouse gases production in these systems attracted increasing attention considering the contributions of methane and nitrous oxide emissions to global warming. However, the detailed knowledge about the contribution of CWs to methane and nitrous oxide emissions in treating sewage treatment plant effluent are still limited in particular for a better understanding of the sustainability of CWs. The fluxes of methane (CH₄) and nitrous oxide (N₂O) from free water surface (FWS) CWs in northern China were measured continuously using the static-stationary chamber technique from 2012 to 2013. The results showed that CWs were the significant source of CH₄ and N₂O emissions. Average emission rates of CH₄ and N₂O ranged from -30.2 μg m⁻² h⁻¹ to 450.9 μg m⁻² h⁻¹, and -58.8 μg m⁻² h⁻¹ to 1251.8 μg m⁻² h⁻¹, respectively. Obvious annual and seasonal variations of CH₄ and N₂O emissions were observed over the 2-year period. In addition, temperatures and

plant species had an impact on CH₄ and N₂O emissions. The obtained results showed that FWS CWs, improving water quality but emitting lower CH₄ and N₂O, could be the alternative method for sewage treatment plant effluent.

Keywords: Constructed wetlands; Methane; Nitrous oxide; Wastewater treatment

1. Introduction

Over the last few decades, point and non-point pollution from agricultural, fishing, municipal and industrial drainage has become a worldwide environmental issue, especially in developing countries (Wu et al., 2015). On the one hand, untreated wastewater is directly discharged into continental surface waters because large scale municipal wastewater treatment plants (WWTPs) have not been constructed or fully operated due to large capital investments and operating costs in rural areas. Furthermore, considering the stringent discharge guidelines and standards, conventional wastewater treatment processes fail to remove large amount of nutrients efficiently, and are also not specifically designed to eliminate micropollutants (Luo et al., 2014; Kong et al., 2015; Lu et al., 2016; Wu et al., 2016). Consequently, untreated wastewater and sewage effluent which contain a variety of excessive organics and nutrients are discharged into rivers, estuaries and oceans, and may deteriorate the water environment quality and impact aquatic ecosystem health. Thus, the potential cost-effective treatment technologies of sewage/wastewater have been partially investigated in previous studies (Wu et al., 2011; Huang et al., 2013; Eveborn et al., 2014; Pan et al., 2016).

In recent years, constructed wetlands (CWs), as a green wastewater treatment technology by simulating natural wetlands, have been proven to be an effective alternative for conventional wastewater treatment technologies owing to their lower

cost, less operation and maintenance requirements, and little reliance on energy inputs (Vymazal, 2011; Wu et al., 2015). CWs are generally comprised of vegetation, substrates, soils, microorganisms and water, and have been found to be able to remove various pollutants (e.g., organics, nutrients and micropollutants) from wastewater by utilizing a variety of physical, chemical, and biological mechanisms (microbial degradation, plant uptake, sorption, sedimentation, filtration and precipitation etc.) (Vymazal, 2011; Saeed and Sun, 2012; Wu et al., 2015). Such natural-like systems can be mainly divided into free water surface (FWS) and subsurface flow (SSF) CWs, and are usually used to treat different wastewaters such as domestic sewage, industrial drainage, urban and agricultural, stormwater runoff, animal wastewaters, leachates, mine drainage and polluted river water (Rai et al., 2013; Li et al., 2014; Vymazal, 2014; Greenway, 2015; Saumya et al., 2015). In addition, CWs might be utilized as a supplement to the existing conventional WWTPs for reclaiming and reusing the sewage effluent (Greenway, 2005; Rai et al., 2013). However, with the aim of improving the water quality and conserving aquatic ecosystem, little attention has been paid to purification of WWTPs effluent which was characterized by relatively low organic content and moderate nitrogen and phosphorous concentrations. Meanwhile, as an artificial ecological system simulating natural wetlands, greenhouse gases (GHG) production in these systems attracted increasing attention considering the contributions of methane (CH_4) and nitrous oxide (N_2O) emissions to global warming (Kong et al., 2016). Compared to natural wetlands, heavy nutrient loading to CWs stimulates bacterial processing, resulting in higher fluxes of CH_4 and N_2O , and thus CWs might be significant sources of CH_4 and N_2O emissions. Many studies investigated the emission of CH_4 and N_2O in various types of CWs for treating various kinds of wastewaters (such as domestic wastewater, dairy

farm wastewater, municipal wastewater and mining runoff) based on the lab-scale and full-scale experiments (Tanner et al., 1997; Mander et al., 2008; Van der Zaag et al., 2010; Mander et al., 2014). From the current literature review by Mander et al. (2014), it indicated that average values of CH₄ and N₂O emissions in various types of CWs are 97-142 mg m⁻² h⁻¹ and 2.2-3.1 mg m⁻² h⁻¹, and can be influenced by various physical, hydrological and operational factors such as dissolved oxygen (DO), hydraulic retention time (HRT), water depth, inflow loading, influent C/N ratio, climate and vegetation. Therefore, in order to comprehensively evaluate the environmental benefit of using CWs as a sustainable wastewater treatment technology, a continuous measurement of CH₄ and N₂O emissions in treatment processes from CWs is absolutely necessary. Moreover, the detailed knowledge about the contribution of CWs to CH₄ and N₂O emissions in treating WWTPs effluent would be required in particular for the potential of GHG mitigation.

The aim of this work was to quantify the long-term CH₄ and N₂O emissions from FWS CWs for treating sewage treatment plant effluent. Annual and seasonal variations of CH₄ and N₂O emissions were analyzed over an approximate 2-year period. The CH₄ and N₂O fluxes and their global warming potential were further comparatively compared with common CW treatments and current WWTPs.

2. Material and methods

2.1 Experimental system and operation

Experimental FWS CW systems which were built in Baihua Park in Jinan, northern China (36°40'36"N, 117°03'42"E) were designed to treat the effluent of sewage treatment plant (Figure 1). The climate of the area is characterized by a warm-temperature monsoonal climate. The experimental treatment system consisted

of twelve FWS CW systems with a surface area of approximately 0.13 m² (50 cm in depth and 40 cm in diameter), and each system had an outlet at the bottom. All CW system were filled with washed river sand (particle size <2 mm, 0.39 porosity) as the substrate with a depth of 25 cm. Nine of CW systems were planted with three macrophyte species (W1: *Phragmites australis*, W2: *Cyperus rotundus*, W3: *Zizania caduciflora*) with three duplicates, and three of CW systems were not planted (U4: control). The density of plants was 12, 20 and 20 rhizomes per system for W1, W2 and W3, respectively. Each system held 20 L water when filled. The water depth of each system was approximately 10 cm from the sand surface.

All experimental CW systems were operated for a period of approximate two years (from April 2012 to December 2013). The synthetic sewage treatment plant effluent was used as influent in each wetland in this study, The synthetic wastewater was prepared from tap water and mainly composed of sucrose, (NH₄)₂SO₄, KH₂PO₄ and KNO₃ based on Grade I treatment standard of municipal sewage treatment plants in China (Wu et al., 2011). Specially, the characteristics of the influents in the present study were COD 72.71 mg L⁻¹, NH₄⁺-N 8.36 mg L⁻¹, TN 21.14 mg L⁻¹ and TP 8.36 mg L⁻¹, respectively. Sequencing fills-and-draw batch mode was applied to influent in the whole experimental period. The HRT was 10 d from April to November and 15 d in November and March when temperature was low.

2.2 Sampling and analysis

2.2.1 Environmental parameters

The following environmental parameters and climatic data in the experimental site were recorded: air temperature (°C) and relative humidity (%).

2.2.2 Water sampling and analysis

Water samples of influent and effluent were taken to evaluate their treatment performance. According to standard methods (APHA, 2005), and all samples were transferred immediately to the lab and analyzed immediately for the following water physicochemical parameters: chemical oxygen demand (COD; HACH DR 2008™ Spectrophotometer, USA), ammonia nitrogen ($\text{NH}_4^+\text{-N}$), total nitrogen (TN) and total phosphorus (TP). Dissolved oxygen (DO) and pH were measured in situ by a DO meter (HQ 30d 53LED™ HACH, USA) and a glass pH meter (SG2-T SevenGo pro™ MTD, Switzerland).

2.2.3 Gas sampling and analysis

CH_4 and N_2O fluxes from the FWS CWs have been investigated in this study. Gas sampling was done using the static-stationary chamber every two days during the whole experimental period. The transparent chamber system ($50\text{ cm} \times 50\text{ cm} \times 50\text{ cm}$) was made of polymethyl methacrylate, and the details of collecting steps of gas samples were according to the method described in the previous studies (Wu et al., 2009). The N_2O concentration was determined using the gas chromatography (SP-3410, China) with an electron capture detector (ECD) and a Poropak Q column, using 30 mL/min high-purity nitrogen as the carrier gas. The temperature of the detector and column were set at 36 °C and 50 °C, respectively. The CH_4 concentration was determined using the gas chromatography (SP-6890, China) equipped with a flame ionization detector (GC-FID) and stainless steel packed columns (GDX502). The operating conditions for the GC were: 375 °C reformer temperature, 40 °C oven temperature and 200 °C detector temperature. The carrier gas was ultra-high purity N_2 (30 mL min⁻¹). CH_4 and N_2O fluxes ($\mu\text{g m}^{-2}\text{ h}^{-1}$) were

determined from the increase in concentration in the chambers over time with linear regression analysis according to the method described in the previous studies (Wu et al., 2009).

2.3 Statistical analysis

Statistical analyses were performed through the software SPSS 11.0 (SPSS Inc., Chicago, USA). A two independent samples t-test was conducted to determine the significance of differences between means. In all tests, differences and correlations were considered statistically significant when $P < 0.05$.

3. Results and discussion

3.1 Environmental variables and water characteristics

As shown in Figure 2, monthly mean air temperature during the whole monitoring period ranged from 2.1 °C to 29.5 °C, and the annual average air temperature in 2012 was 21.4 °C, which was slightly higher than that in 2013 (19.1 °C). Moreover, the maximum temperature was observed to appear from May to August, and the minimum temperature was recorded in January and February. The average relative humidity during the study period was 57.4%, with the higher value in the first year (59.6%) and the lower value in the second year (55.6%). The average effluent concentrations of COD, $\text{NH}_4^+\text{-N}$, TN and TP in different FWS CW systems in the present study were 16.7-24.6 mg L⁻¹, 0.5-4.4 mg L⁻¹, 3.2-11.7 mg L⁻¹ and 0.5-1.1 mg L⁻¹, respectively. These results indicated a significant improvement in water quality of sewage treatment plant effluent by treatment through FWS CWs. However, the average removal performance of the planted FWS CW systems was higher than that of unplanted CW systems, which suggested that there was a positive correlation

among water purification and plant growth and establishment. On the whole, our results are consistent with other research which reported that reduction of pollutants was found to increase with growth and establishment of the plants (Rai et al., 2013).

3.2 Variation of CH₄ emission

The variation of CH₄ emission from different FWS CW systems in 2012-2013 is shown in Figure 3a. Average CH₄ fluxes had obvious annual and seasonal variations in different FWS CWs, and ranged from -30.2 $\mu\text{g m}^{-2} \text{h}^{-1}$ to 450.9 $\mu\text{g m}^{-2} \text{h}^{-1}$. Specially, the average CH₄ flux in CW systems in the second year (138.6 $\mu\text{g m}^{-2} \text{h}^{-1}$) was significantly higher than that (88.6 $\mu\text{g m}^{-2} \text{h}^{-1}$) measured in the first year. The higher flux of CH₄ was observed in summer compared to spring and fall, and a general seasonal peak occurred at the end of the summer/beginning of fall. However, it should be noted that the weak absorption (the sink) of CH₄ was found in cold season (November and December) compared with other period when wetland became a source of CH₄. These results suggested that seasons might have a significant effect on CH₄ emissions in FWS CW systems. These results can also be illustrated by the line regression relationship between CH₄ fluxes and air temperature. As shown in Figure 3b, during the 2-year monitoring period, the rate of CH₄ emission in CW systems was significantly associated with air temperatures, and CH₄ flux generally increased with the temperature rising. However, clear difference was found between vegetation and non-vegetation systems. The possible reason may be that microbial activity in CWs would increase with the increasing of temperature at a certain climatic condition (Wu et al., 2011; Mander et al., 2014). On the other hand, it is well recognized that temperature affects plant photosynthesis and plant biomass directly, and thus increases organic matter and gas transportation, which would give a positive

effect on CH₄ emission (Zhao et al., 2016). However, some other studies have reported negative effect of plant biomass on CH₄ emission, and these impacts on CH₄ emission from CWs vary significantly among plant species (Bhullar et al., 2014). Specifically, significant difference of CH₄ emission rate was found among different CWs with various plant species in this study. The wetlands (W1) vegetated with *Phragmites australis* emitted higher CH₄ (164.1 µg m⁻² h⁻¹) than wetlands (W3) planted with *Zizania caduciflora* (152.1 µg m⁻² h⁻¹), following by and the wetlands (W2) with *Cyperus rotundus* (104.5 µg m⁻² h⁻¹) throughout experimental period. Similarly, Zhao et al. (2016) studied the effects of plant diversity on CH₄ emission and nitrogen removal, and concluded that the best combination of low CH₄ emission and high N removal rates could be achieved in CW microcosms planting *P. arundinacea*. These results about difference among various plant species suggested that CH₄ emission could be affected by other various factors such as CW type, oxygen level, inflow loading and C/N ratio (Mander et al., 2014; Zhao et al., 2016).

On the whole, analysis of CH₄ emission in two years showed FWS CW systems were a CH₄ source, but the mean CH₄ emission rate measured in this study were lower than the values (0.2-36 mg m⁻² h⁻¹) in FWS CW treatment systems and the values (0.064-23 mg m⁻² h⁻¹) in SSF CWs reported in the literature (Mander et al., 2014), and as high as the values (0.003-6.2 mg m⁻² h⁻¹) in enhancing CW systems such as aerated CWs (Maltais-Landry et al., 2009). CH₄ emission was also found to be significantly lower than the results (0.61-9.7 mg m⁻² h⁻¹) from natural wetlands (Chen et al., 2013). Moreover, when compared with common WWTPs, the emission rate was greatly lower than the values (0.06-978 g m⁻² d⁻¹) obtained among different processing units in the typical conventional WWTPs (Ren et al. 2015).

3.3 Variation of N₂O emission

The variation of N₂O emission from FWS CW systems during the experimental period is illustrated in Figure 4a. It is shown that N₂O emission from the planted and unplanted wetlands varied annually and seasonally. The N₂O emission rate ranged from -58.8 $\mu\text{g m}^{-2} \text{h}^{-1}$ to 1251.8 $\mu\text{g m}^{-2} \text{h}^{-1}$, and specially, the average N₂O flux in CW systems in 2013 (381.8 $\mu\text{g m}^{-2} \text{h}^{-1}$) was significantly higher than that (311.1 $\mu\text{g m}^{-2} \text{h}^{-1}$) recorded in 2012. This result indicated that the wetlands plants were well developed in the second year, and flourishing plants and active microbial population beneficial to nitrification and denitrification would promote production and transport of N₂O as compared with in the initial first year. The N₂O emission was also observed to be higher in summer than in spring and fall, and there had a lower N₂O emission rate in winter due to the plants withering and microorganism activity decreasing. This result indicated that temperature had an important effect on the N₂O emission in CWs, which is in agreement with other reports (Zhang et al. 2005). Figure 4b presents the polynomial regression relationship between N₂O emission rates and air temperature during the 2-year operating period. It can be illustrated that the N₂O emission rate in CWs was increased with the rising temperatures, but the statistics was not significant. Mander et al. (2014) reported that the higher temperature of the environment slightly increased CH₄ emission in CWs, whereas in terms of N₂O emission the relationship was insignificant and unclear.

The average N₂O emission rate from the different wetlands in the whole experiment also varied each other. The planted wetlands had higher N₂O emission rate than the unplanted wetlands, which indicated that the plant species had an impact on N₂O emission. N₂O emission rates from the planted wetlands also varied among plant

species because of the relative differences in intrinsic species, possible ecotype and growth characteristics. On the whole, wetlands (W1) vegetated with *Phragmites australis* had highest N₂O fluxes (mean value 514.2 $\mu\text{g m}^{-2} \text{h}^{-1}$) following by wetlands (W3) planted with *Zizania caduciflora* (mean value 334.9 $\mu\text{g m}^{-2} \text{h}^{-1}$) and the wetlands (W2) with *Cyperus rotundus* (mean value 328.2 $\mu\text{g m}^{-2} \text{h}^{-1}$). The results suggested that FWS CW systems were a source of N₂O throughout experimental period when treating sewage treatment plant effluent. The maximum N₂O emission rates measured in this study were higher than the values (50 $\mu\text{g m}^{-2} \text{h}^{-1}$) in natural ecosystems reported by Sagggar et al. (2007), but lower than the values measured in CWs treating wastewater (2145 $\mu\text{g m}^{-2} \text{h}^{-1}$) reported by Wu et al. (2009), and still greatly lower than the values ($2 \times 10^3 \text{ mg m}^{-2} \text{h}^{-1}$) measured in sewage treatment plants (Benckiser et al. 1996).

4. Conclusions

In this study, CH₄ and N₂O emissions from FWS CWs treating sewage treatment plant effluent, ranging from -30.2 $\mu\text{g m}^{-2} \text{h}^{-1}$ to 450.9 $\mu\text{g m}^{-2} \text{h}^{-1}$, and -58.8 $\mu\text{g m}^{-2} \text{h}^{-1}$ to 1251.8 $\mu\text{g m}^{-2} \text{h}^{-1}$, respectively, were significantly low compared with traditional WWTPs, but obvious temporal variations were found in different FWS CWs. The average emission rates of CH₄ and N₂O in CWs in the second year (138.6 $\mu\text{g m}^{-2} \text{h}^{-1}$ and 381.8 $\mu\text{g m}^{-2} \text{h}^{-1}$) were significantly higher than that in the first year (88.6 $\mu\text{g m}^{-2} \text{h}^{-1}$ and 311.1 $\mu\text{g m}^{-2} \text{h}^{-1}$), and the higher fluxes of CH₄ and N₂O were observed in summer compared to spring and fall. The rate of CH₄ and N₂O emission was increased as the temperature rising, and the planted wetlands had higher CH₄ and N₂O emission than the unplanted wetlands. These results showed that FWS CWs, achieving the better treatment performance and lower GHG emission, could be an

alternative method for sewage treatment plant effluent, which would be beneficial for the sustainable operation and successful application of CW systems.

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Figure Captions:

Figure 1 Figure 1 Profile of the laboratory-scale constructed wetland (a) and photograph of the experimental constructed wetland systems (b)

Figure 2 The variation of air temperature and relative humidity during the experimental period.

Figure 3 The variation of CH₄ emissions from different wetland systems (W1: *Phragmites australis*, W2: *Cyperus rotundus*, W3: *Zizania caduciflora*, W4: unplanted) during the experimental period (a), and linear regression between air temperature and CH₄ emission rates (b).

Figure 4 The variation of N₂O emissions from different wetland systems (W1: *Phragmites australis*, W2: *Cyperus rotundus*, W3: *Zizania caduciflora*, W4: unplanted) during the experimental period (a), and polynomial regression between air temperature and N₂O emission rates (b).

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Figure 4 The variation of N_2O emissions from different wetland systems (W1: *Phragmites australis*, W2: *Cyperus rotundus*, W3: *Zizania caduciflora*, W4: unplanted) during the experimental period (a), and polynomial regression between air temperature and N_2O emission rates (b).