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# Purification ability and carbon dioxide flux from surface flow constructed wetlands treating sewage treatment plant effluent

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## Abstract

In this study, a two-year experiment was carried out to investigate variation of carbon dioxide (CO<sub>2</sub>) flux from free water surface constructed wetlands (FWS CW) systems treating sewage treatment plant effluent, and treatment performance was also evaluated. The better 74.6–76.6% COD, 92.7–94.4% NH<sub>4</sub>-N, 60.1–84.7% TN and 49.3–70.7% TP removal efficiencies were achieved in planted CW systems compared with unplanted systems. The planted CW was a net CO<sub>2</sub> sink, while the unplanted CW was a net CO<sub>2</sub> source in the entire study period. An obvious annual and seasonal variability of CO<sub>2</sub> fluxes from different wetland systems was also presented with the average CO<sub>2</sub> flux ranging from -592.83 mg m<sup>-2</sup> h<sup>-1</sup> to 553.91 mg m<sup>-2</sup> h<sup>-1</sup> during 2012–2013. In addition, the net exchange of CO<sub>2</sub> between CW systems and the atmosphere was significantly affected by air temperature, and the presence of plants also had the significant effect on total CO<sub>2</sub> emissions.

**Keywords:** Constructed wetlands; Carbon dioxide; Wetland plants; Greenhouse gas emissions; Pollutant removal

## 1. Introduction

In past several decades, a rapidly expanding economy world-wide especially in developing countries has been accompanied by a great raise of severe environmental issues such as river pollution and water blooms occurred in lakes. However, conventional

wastewater treatment systems are still difficult to remove excess nutrients effectively from wastewater in an economical way (Li et al., 2014). Constructed wetlands (CWs) have proven to a low-cost and sustainable alternative for conventional wastewater treatment technologies, and are effective for treating a wide range of wastewaters such as domestic sewage, industrial drainage, agricultural and urban runoff wastewaters, polluted river water (Maltais-Landry et al., 2009; Wu et al., 2015a). Numerous studies have focused on the design and application of CWs, and have

shown that CWs could be efficient for removing various pollutants (organics, nutrients, heavy metals, etc.) through a variety of physical and biochemical mechanisms (Ju et al., 2014; Wu et al., 2016). In addition, great efforts have been made for improving the sustainable operation and application of these techniques (Wu et al., 2015b). Nevertheless, as an ecological wastewater treatment system which could be a supplement to the existing pollution control measures, few studies focused on purifying sewage treatment plant effluent with the aim of improving the water quality of lakes and rivers. The effluent from sewage treatment plants was generally characterized by relatively low organic content and moderate nitrogen and phosphorous concentrations, which might still be too high for discharge to surface waters sensitive to eutrophication (Wu et al., 2011).

Moreover, as a bias of the water purification, CW wastewater treatments have been found to emit variable amounts of carbon dioxide (CO<sub>2</sub>), methane (CH<sub>4</sub>) and nitrous oxide (N<sub>2</sub>O) which are important greenhouse gases (GHG), and might be main contributors to the global climate change (Chiemchaisri et al., 2009; Mander et al., 2014; Barbera et al., 2014). Considering the aspect of the CW sustainability, the emission of GHG from CWs may considerably reduce the environmental and ecological benefits. From the current literature review, Mander et al. (2014) indicated that there were more measurements of CH<sub>4</sub> and N<sub>2</sub>O emission from CWs. Average values of CH<sub>4</sub> and N<sub>2</sub>O emissions in various types of CWs were 97–142 mg m<sup>-2</sup> h<sup>-1</sup> and 2.2–3.1 mg m<sup>-2</sup> h<sup>-1</sup>, and can also be influenced by various environmental conditions and operating parameters such as dissolved oxygen (DO), hydraulic retention time (HRT), water depth, available organic carbon, influent C/N ratio, water temperature, season and vegetation (Wu et al., 2009; Mander et al., 2014). On the other hand, long-term measurements of CO<sub>2</sub> exchange from natural wetland ecosystems showed the great potential of many wetlands to sequester C and compensated the climate-warming impact of CH<sub>4</sub> and N<sub>2</sub>O emissions, which have improved our understanding of the responses of carbon cycles to climate change (Mitsch et al., 2013). However, only few works have been measured the CO<sub>2</sub> emission in CWs. Recently, Mander et al. (2014) pointed out that CO<sub>2</sub> emissions were significantly lower in free water surface (FWS) CWs than in subsurface flow (SSF) CWs (average rate of 2208–4440 mg m<sup>-2</sup> h<sup>-1</sup>), but there were obviously variations. Hence, a reliable and continuous quantification of the CO<sub>2</sub> emission in treatment processes from CWs is necessary for calculating the potential of CO<sub>2</sub> mitigation and evaluating the CW sustainability.

The aim of this research was to evaluate the treatment performance of FWS CW systems supplied with the effluent of sewage treatment plant. In addition, variations of CO<sub>2</sub> flux from the FWS CW treating effluent was also assessed using transparent chamber method for a better understanding of CW sustainability.

## 2. Material and methods

### 2.1. Study site and experimental system

This research was conducted in Baihua Park in Jinan, northern China (36°40′36″N, 117°03′42″E) where is characterized by a warm-temperature monsoonal climate. The experimental treatment system consisted of the FWS CW with a surface area of approximately 0.13 m<sup>2</sup> (50 cm in depth and 40 cm in diameter), and with an outlet at the bottom (as shown in Fig. S1). Each system was filled with washed river sand (particle size <2 mm, 0.39 porosity) as the substrate with a depth of 25 cm. Three macrophyte species were planted in each system (W1: *Phragmites australis*, W2: *Cyperus rotundus*, W3: *Zizania caduciflora*, W4: control; n = 3) at a density of 12, 20 and 20 rhizomes per system for W1, W2 and

W3, respectively. The water table was constant throughout the study, at approximately 10 cm from the sand surface, and each system held 20 L water when filled.

### 2.2. System operation

After an acclimation period (about one month), the systems were fed with synthetic wastewater to start the experiment, and were operated from April 2012 to December 2013 (except for January and February). The synthetic sewage treatment plant effluent used in this study was prepared from tap water, and composed of sucrose, (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub>, KH<sub>2</sub>PO<sub>4</sub> and KNO<sub>3</sub> based on Grade I treatment standard of municipal sewage treatment plants in China (Wu et al., 2011). All systems were operated in a sequencing fill-and-draw batch mode with the hydraulic retention time (HRT) of 10 d from April to November. When temperature was low in November and March, HRT was 15 d.

### 2.3. Sampling and analysis

#### 2.3.1. Water sampling and analysis

Influent and effluent of the wetland systems were sampled to evaluate their treatment performance. Water samples were collected using a syringe tube (100 mL) every ten days. The following water physicochemical parameters were analyzed according to APHA (2005): chemical oxygen demand (COD; HACH DR 2008™ Spectrophotometer, USA), ammonia nitrogen (NH<sub>4</sub><sup>+</sup>-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). The air temperature (oC) was recorded by a weather station that was close to the experimental site.

#### 2.3.2. Gas sampling and analysis

CO<sub>2</sub> flux across surface areas representing the net ecosystem exchange (NEE) from the FWS CWs has been estimated using the static-stationary chamber technique. Gas sampling was carried out using a transparent chamber system every two days during the whole experimental period. Four gas samples were collected in 150 mL nylon syringe at 20 min interval over 60 min, and the details of chamber systems and collecting steps of gas samples were according to the method described in the previous studies (Wu et al., 2009). The CO<sub>2</sub> 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 oC reformer temperature, 40 oC oven temperature and 200 oC detector temperature. The carrier gas was ultra-high purity N<sub>2</sub> (30 mL min<sup>-1</sup>). CO<sub>2</sub> flux (mg m<sup>-2</sup> h<sup>-1</sup>) was determined from the increase in concentration in the chambers over time with linear regression analysis as described by Wu et al. (2009).

### 2.4. Statistical analysis

Statistical analyses were performed through the software SPSS 11.0 (SPSS Inc., Chicago, USA). The normality of gas flux data obtained from the chamber measurements was checked using the Kolmogorov–Smirnov, Lilliefors’ and Shapiro–Wilk’s tests. Two-sample t-tests were used to evaluate the significance of differences between means obtained in this study. In all tests, differences and correlations were considered statistically significant only if P < 0.05.

### 3. Results and discussion

#### 3.1. Environmental parameters and water quality

Daily air temperature of the experimental site during the monitoring period was shown in Fig. 1a, and the average daily air temperatures in 2012 and 2013 were 10.5–19.1 °C and 10.8–19.6 °C, respectively. The maximum temperature was observed to reach above 30 °C from May to August, and the minimum temperature fell below 0 °C in January and February. The annual precipitation was recorded to be 665.7 mm, which was mainly in July and August.

The average influent concentrations of COD,  $\text{NH}_4^+\text{-N}$ , TN and TP in the present study were 72.71  $\text{mg L}^{-1}$ , 8.36  $\text{mg L}^{-1}$ , 21.14  $\text{mg L}^{-1}$  and 8.36  $\text{mg L}^{-1}$ , respectively. The average effluent concentrations and removal efficiencies of COD,  $\text{NH}_4^+\text{-N}$ , TN and TP for different FWS CW systems were given in Table 1. On the whole, systems with different plants achieved a satisfactory removal of COD,  $\text{NH}_4^+\text{-N}$ , TN and TP, when compared to unplanted systems. In addition, removal performance varied considerably among plant species. As summarized in Table 1, system W1, system W2 and system W3 achieved average COD removal efficiencies of 74.65–76.69%, while a lower COD removal (64.63%) was observed in the system W4. Average  $\text{NH}_4^+\text{-N}$  removal efficiencies in planted

Table 1

Characteristics of the effluent and respective removal efficiencies in wetland systems during the experimental period (mean  $\pm$  standard deviation).

Parameters	Experimental systems			
	W1	W2	W3	W4
COD (mg/L)	17.01 $\pm$ 1.87	18.31 $\pm$ 2.39	16.73 $\pm$ 2.48	24.65 $\pm$ 3.42
(%)	76.29 $\pm$ 2.56	74.65 $\pm$ 3.24	76.69 $\pm$ 3.41	64.63 $\pm$ 4.85
$\text{NH}_4^+\text{-N}$ (mg/L)	0.61 $\pm$ 0.12	0.49 $\pm$ 0.15	0.46 $\pm$ 0.13	1.35 $\pm$ 0.30
(%)	92.74 $\pm$ 1.44	94.13 $\pm$ 1.81	94.43 $\pm$ 1.56	83.62 $\pm$ 3.62
TN (mg/L)	7.71 $\pm$ 1.17	8.48 $\pm$ 0.83	3.22 $\pm$ 0.51	11.71 $\pm$ 1.14
(%)	63.53 $\pm$ 5.53	60.14 $\pm$ 3.93	84.77 $\pm$ 2.38	44.78 $\pm$ 5.36
TP (mg/L)	0.72 $\pm$ 0.07	0.89 $\pm$ 0.08	0.51 $\pm$ 0.09	1.11 $\pm$ 0.11
(%)	58.16 $\pm$ 4.54	49.36 $\pm$ 4.94	70.76 $\pm$ 5.42	36.65 $\pm$ 5.58
pH	7.78 $\pm$ 0.15	7.86 $\pm$ 0.12	7.54 $\pm$ 0.11	8.14 $\pm$ 0.24
DO (mg/L)	5.07 $\pm$ 0.16	5.02 $\pm$ 0.09	4.64 $\pm$ 0.11	6.14 $\pm$ 0.22

W1: *Phragmites australis*, W2: *Cyperus rotundus*, W3: *Zizania caduciflora*, W4: unplanted.

systems were all above 90%, and unplanted systems presented a slightly decrease of average  $\text{NH}_4^+\text{-N}$  removal efficiency (83.62%). Similarly, the efficiencies of TN and TP removal in planted systems were 60.14–84.77% and 49.36–70.76%, respectively, but an obvious decrease of TN and TP removal was appeared in the system W4 with the average efficiencies of 44.78% and 36.65%. The results of

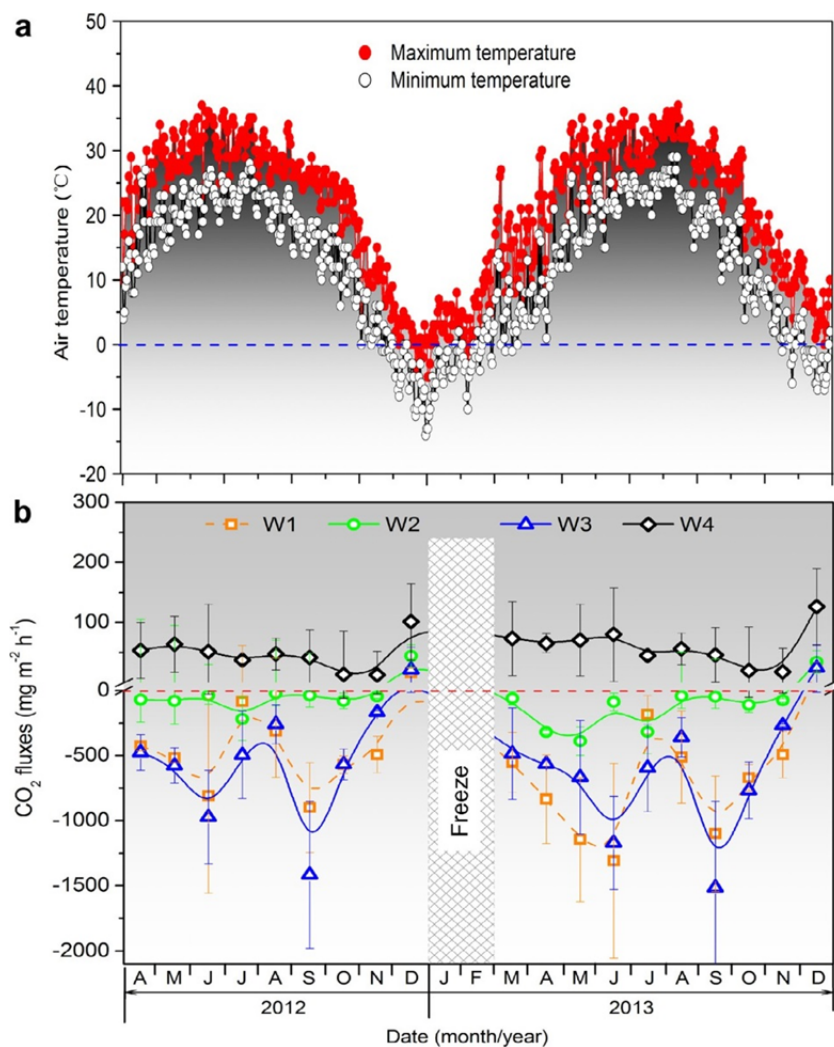


Fig. 1. The variation of air temperature (a) and  $\text{CO}_2$  fluxes (b) from different wetland systems (W1: *Phragmites australis*, W2: *Cyperus rotundus*, W3: *Zizania caduciflora*, W4: unplanted) during the experimental period.

the higher treatment performance in planted systems could be explained by that plants not only directly assimilated nutrient for the growth and reproduction, but also released oxygen and provided enough medium for purification reactions, which would be beneficial to a variety of removal processes for pollutants in CWs (Wu et al., 2011, 2015a).

### 3.2. Variation of CO<sub>2</sub> flux from surface flow constructed wetlands

Although the mineralisation of organic matter to CO<sub>2</sub> is the objective of wastewater treatment processes, emission (sequestration) of CO<sub>2</sub> in CWs involves plant photosynthesis, soil respiration and plant autotrophic respiration (Mitsch et al., 2013). Therefore, depending on meteorological, environmental and operating conditions, CWs can be sources or sinks of carbon (Mitsch et al., 2013). CO<sub>2</sub> exchange between CW systems and the atmosphere in this study was directly determined, and the variation of CO<sub>2</sub> fluxes from different wetlands during the experimental period is shown in Fig. 1b. An obvious annual variability of CO<sub>2</sub> fluxes from the planted and unplanted wetlands was presented during the whole experimental period. Specially, the average CO<sub>2</sub> flux from the different wetland systems during 2012–2013 ranged from -592.83 mg m<sup>-2</sup> h<sup>-1</sup> to 553.91 mg m<sup>-2</sup> h<sup>-1</sup>. Moreover, a tendency to increase was found with time. The average CO<sub>2</sub> flux from CW systems was -676.35 to 60.12 mg m<sup>-2</sup> h<sup>-1</sup> in the second year, and significantly higher than that (-544.74 to 47.08 mg m<sup>-2</sup> h<sup>-1</sup>) obtained in the first year. Since the majority of organic pollutant removal in the FWS wetland was largely attributed to microorganisms attached to the plant biomass, the substantially increased plant biomass was shown in the second year. On the other hand, plant autotrophic respiration and plant photosynthesis were enhanced with high biomass production, which directly resulting a higher net exchange of CO<sub>2</sub> between CWs and the atmosphere (Mander et al., 2014). The properties of CO<sub>2</sub> fluxes from the different wetlands also showed significant differences with seasonal fluctuations. On the whole, a W-shaped trend of CO<sub>2</sub> fluxes was found in planted wetland systems, while CO<sub>2</sub> fluxes from unplanted systems varied slightly. This also showed that the vegetated (W1, W2 and W3) and unvegetated (W4) wetland systems expressed significant differences in the CO<sub>2</sub> flux that was emitted in the atmosphere. In addition, the rate of CO<sub>2</sub> fluxes was higher in summer than that in other seasons because of the plants withering and microorganism activity decreasing in fall and winter. These results are also consistent with other research which reports that we can assume that vegetation in CWs could significantly reduce emissions of CO<sub>2</sub> by photosynthesis of plants (Mander and Teiter, 2005). The peak exchange of CO<sub>2</sub> appeared in growth seasons (June and September) with the CO<sub>2</sub> flux of -1308.36 mg m<sup>-2</sup> h<sup>-1</sup>, and can be a sink of carbon. However, CWs might be a source of carbon in December when the CO<sub>2</sub> emission rate was highest (126.49 mg m<sup>-2</sup> h<sup>-1</sup>). This indicated that the CO<sub>2</sub> flux from the wetlands might be affected by air temperature, which could be explained by the line regression relationship between CO<sub>2</sub> fluxes and air temperature shown in Fig. 2a. During the 2-year monitoring period, the rate of CO<sub>2</sub> flux from planted wetlands was significantly associated with seasonal air temperatures, and it seemed that generally CO<sub>2</sub> flux increased with the temperature rising. The possible reason may be that microbial activity in CWs generally increases at a proper climatic condition (such as temperature) (Wu et al., 2011; Mander et al., 2014). In addition, temperature affects directly plant photosynthesis and respiration but also evaporation, which influences directly CO<sub>2</sub> emission (Hao et al., 2011; Mitsch et al., 2013).

As shown in Fig. 2b, the planted wetlands had higher exchange rate of CO<sub>2</sub> than the unplanted wetlands. There were also differences in mean CO<sub>2</sub> fluxes among the different plant species sys-

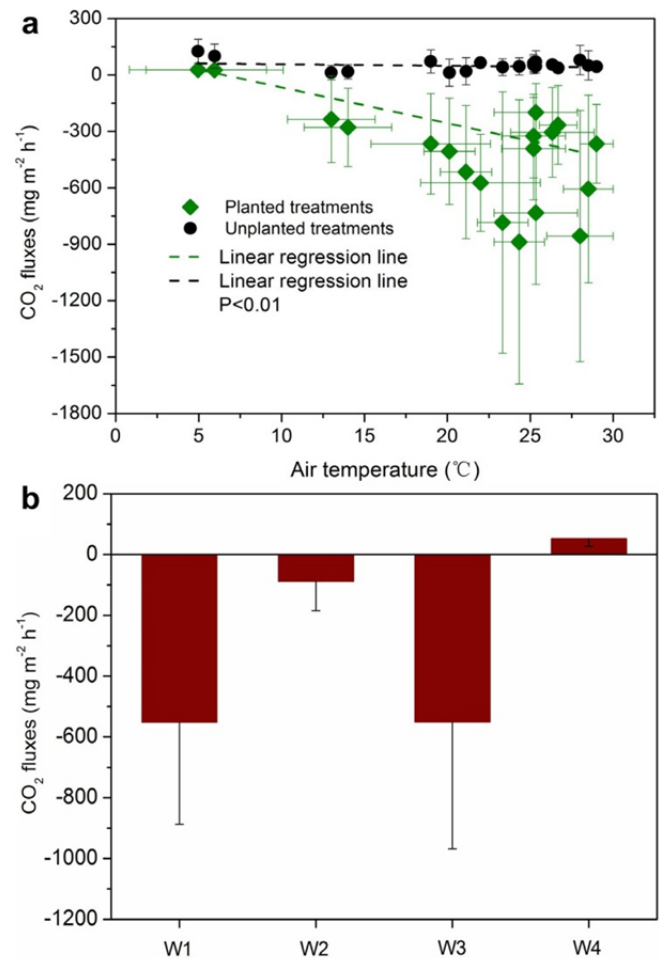


Fig. 2. CO<sub>2</sub> fluxes response to air temperature (a), and the effect of plants on the CO<sub>2</sub> fluxes from wetland systems (W1: *Phragmites australis*, W2: *Cyperus rotundus*, W3: *Zizania caduciflora*, W4: unplanted).

tems, ranging from -552.13 mg m<sup>-2</sup> h<sup>-1</sup> to -88.26 mg m<sup>-2</sup> h<sup>-1</sup>, while mean CO<sub>2</sub> fluxes from unplanted systems was observed to be 52.76 mg m<sup>-2</sup> h<sup>-1</sup>. This indicated that the plant species had an impact on CO<sub>2</sub> flux but the trend was not statistically significant. Variation of CO<sub>2</sub> flux from planted wetlands among plant species was due to the relative differences in intrinsic species, possible ecotype and growth characteristics. On the whole, wetlands (W3) vegetated with *Zizania caduciflora* had highest net CO<sub>2</sub> fluxes following by the wetlands (W1) with *Phragmites australis* and the wetlands (W2) with *Cyperus rotundus* throughout experimental period. These results are in agreement with previous studies. Maltais-Landry et al. (2009) reported the significant effect of the plant presence on CO<sub>2</sub> emissions, with higher fluxes in planted systems compared with unplanted systems. Barbera et al. (2014) also found the cumulative CO<sub>2</sub> efflux in vegetated sites was higher than that in unvegetated areas, which demonstrated that the existing plants were important for total ecosystem respiration. Overall, analysis of CO<sub>2</sub> fluxes in two years showed the planted CWs was a net CO<sub>2</sub> sink, but the mean net CO<sub>2</sub> fluxes measured in this study were lower than the values (0.54 mg m<sup>-2</sup> s<sup>-1</sup>) in natural wetland ecosystems reported by Hao et al. (2011). However, the unplanted CWs was a net CO<sub>2</sub> source, but the mean CO<sub>2</sub> fluxes was lower than the values (117 mg m<sup>-2</sup> h<sup>-1</sup>) from artificially aerated wetland for treating wastewater reported by Maltais-Landry et al. (2009), and greatly lower than the values (24.8 g d<sup>-1</sup>) measured in conventional sewage treatment systems (Ren et al., 2015). Based on the

results obtained in this study, the strategy of selecting the wetland plants with large biomass would be beneficial for reducing CO<sub>2</sub> emission and increasing carbon storage in CW wastewater treatments, and be also helpful for management of the plant in CWs.

#### 4. Conclusions

The FWS CW systems were efficient for purifying the sewage treatment plant effluent with the better removal efficiencies of COD (74.6–76.6%), NH<sub>4</sub><sup>+</sup>-N (92.7–94.4%), TN (60.1–84.7%) and TP (49.3–70.7%). The average CO<sub>2</sub> flux from CW systems in the second year (-676.35 to 60.12 mg m<sup>-2</sup> h<sup>-1</sup>) was significantly higher than that (-544.74 to 47.08 mg m<sup>-2</sup> h<sup>-1</sup>) in the first year. The CO<sub>2</sub> fluxes from CW systems varied seasonally, and the mean CO<sub>2</sub> fluxes were higher in summer than that in other seasons. Furthermore, the net exchange of CO<sub>2</sub> between CW systems and the atmosphere was significantly influenced by air temperature and wetland plants.

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#### Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at <http://dx.doi.org/10.1016/j.biortech.2016.08.030>.

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