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Large-scale multi-stage constructed wetlands for secondary effluents treatment in northern China: Carbon dynamics $*$

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ABSTRACT

Multi-stage constructed wetlands (CWs) have been proved to be a cost-effective alternative in the treatment of various wastewaters for improving the treatment performance as compared with the conventional single-stage CWs. However, few long-term full-scale multi-stage CWs have been performed and evaluated for polishing effluents from domestic wastewater treatment plants (WWTP). This study investigated the seasonal and spatial dynamics of carbon and the effects of the key factors (input loading and temperature) in the large-scale seven-stage Wu River CW polishing domestic WWTP effluents in northern China. The results indicated a significant improvement in water quality. Significant seasonal and spatial variations of organics removal were observed in the Wu River CW with a higher COD removal efficiency of 64-66% in summer and fall. Obvious seasonal and spatial variations of CH₄ and CO₂ emissions were also found with the average CH₄ and CO₂ emission rates of 3.78–35.54 mg m⁻² d⁻¹ and 610.78–8992.71 mg m⁻² d⁻¹, respectively, while the higher CH₄ and CO₂ emission flux was obtained in spring and summer. Seasonal air temperatures and inflow COD loading rates significantly affected organics removal and CH₄ emission, but they appeared to have a weak influence on CO₂ emission. Overall, this study suggested that large-scale Wu River CW might be a potential source of GHG, but considering the sustainability of the multi-stage CW, the inflow COD loading rate of 1.8–2.0 g m⁻² d⁻¹ and temperature of $15-20$ °C may be the suitable condition for achieving the higher organics removal efficiency and lower greenhouse gases (GHG) emission in polishing the domestic WWTP effluent. The obtained knowledge of the carbon dynamics in large-scale Wu River CW will be helpful for understanding the carbon cycles, but also can provide useful field experience for the design, operation and management of multi-stage CW treatments.

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1. Introduction

The issue of water scarcity and deterioration has become a serious concern in the world, and this situation is becoming worse with the rapid urbanization, inadequate water/wastewater purification and management especially in developing countries in past decades [\(Greenway, 2005](#page--1-0)). Due to lack of convenient and effective wastewater treatments with lower cost (such as construction investment, operation costs and energy consumption), discharging

<https://doi.org/10.1016/j.envpol.2017.09.048> 0269-7491/© 2017 Elsevier Ltd. All rights reserved. the majority of untreated wastewater directly into urban rivers is a common practice in many cities and small towns, which leads to the serious pollution of the river basins [\(Wu et al., 2015a\)](#page--1-0). In addition, in recent years great concern has been growing regarding to the treated effluents, typically effluents from wastewater treatment plants (WWTP) ([Vivant et al., 2016; Wu et al., 2017](#page--1-0)). Such wastewaters contain excessive organic pollutants and nutrients, which are not properly/partially treated, are simply discharged into rivers and estuaries, which may impact aquatic ecosystem health and then pose potential risks to human health [\(Matamoros et al.,](#page--1-0) [2008; Wu et al., 2016a,b](#page--1-0)). In order to tackle this environmental concern, additional effective treatments could be employed to purify WWTP effluents and improve the urban river water quality.

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In fact, constructed wetlands (CWs), as an efficient and sustainable ecological treatment technology, have been proven to be an effective alternative to traditional wastewater treatment systems. The typical CW is generally comprised of vegetation, substrates, soils, microorganisms and water, and it can remove various pollutants (such as organics and nutrients) from wastewater by means of physical, chemical, and biological mechanisms (microbial degradation, plant uptake, sorption, sedimentation, filtration, precipitation and volatilization etc.) [\(Saeed and Sun, 2012; Wu et al.,](#page--1-0) [2015a](#page--1-0)). Particularly, microbial removal mechanism plays a key role in the removal of pollutant in CWs ([Mitsch et al., 2013; Meng](#page--1-0) [et al., 2014\)](#page--1-0). Those processes can be influenced by various environmental conditions and operating parameters such as dissolved oxygen (DO), hydraulic retention time (HRT), water depth, inflow loading, water temperature, season and vegetation ([Saeed and Sun,](#page--1-0) [2012](#page--1-0)). According to the hydrological condition, CWs can typically be divided into free water surface (FWS) and subsurface flow (SSF) wetlands with different technical characteristics. SSF CWs can be further classified into vertical flow (VF) and horizontal flow (HF) CWs. There is another classification for CWs based on the type of wetland plants used. In the past several years, CWs have been widely applied for removing various pollutants in domestic sewage, agricultural wastewater, industrial effluent, mine drainage, landfill leachate, urban runoff, and polluted river water ([Wu et al., 2014\)](#page--1-0). In addition, given their low cost and ease operation, CWs have been shown to be able to efficiently remove organic pollutants, nutrients and harmful bacteria from WWTP effluents with aim to improve the water quality and conserve ecological environment of the urban river ([Chen et al., 2014; Morvannou et al., 2015](#page--1-0)). But most of the previous studies were performed in microcosm-scale or pilot-scale CW systems with a small surface area [\(Vivant et al., 2016; Wu et al.,](#page--1-0) [2015b\)](#page--1-0). [Matamoros et al. \(2008\)](#page--1-0) investigated the removal of a variety of organic pollutants in a full-scale surface flow CW fed with secondary effluent from a conventional WWTP, and reported that CW was efficient for removing 12 organic micropollutants from WWTP effluent discharged into the River Besos of northeastern Spain. [Chen et al. \(2014\)](#page--1-0) studied the removal efficiencies and the kinetics of disinfection byproducts in SSF CWs treating secondary effluent, and a high removal efficiently removed of >90% was achieved in laboratory-scale SSF CWs. In a recent study using FWS CWs receiving secondary effluent from a France WWTP, [Vivant et al.](#page--1-0) [\(2016\)](#page--1-0) found that FWS CWs could be an efficient treatment for extended-spectrum beta-lactamase-producing E. coli disinfection of wastewater. However, it should be indicated that the treatment capacity of a single basic CW was still limited for polishing urban rivers receiving large inputs of WWTP effluent. Concurrently, the more application of multi-stage CWs by combining different types of CWs has increased particularly for achieving the greatest potential for wastewater polishing and more stable pollutant removal ([Wu et al., 2015b; Kato et al., 2013](#page--1-0)). This type multi-stage CW has been proved to substantially improve the treatment performance as compared with the conventional single-stage CWs ([Vymazal,](#page--1-0) [2013a](#page--1-0)). [Kato et al. \(2013\)](#page--1-0) designed the six multi-stage CWs for treating high-content wastewater in cold climates of northern Japan, and the satisfactory purification was obtained for organics, nitrogen and phosphorus. [Jia et al. \(2014\)](#page--1-0) constructed a four-stage wetland system for treating a heavily polluted river, and significant improvement in the water quality was observed. Moreover, several important studies on multi-stage CWs have been conducted to treat emerging organic contaminants (such as personal care products and pharmaceuticals), and demonstrated the great capacity of a multi-stage wetland system as a cost-effective alternative or supplementary to conventional WWTP [\(Avila et al., 2014a, 2014b\)](#page--1-0). Above previous studies have convincingly proved the benefits of multi-stage CWs in the treatment of various types of wastewater, such as dairy, tannery or domestic wastewater, however, further studies reporting their treatment performance on the treatment of effluent from WWTPs are still lacking. Additionally, few long-term full-scale multi-stage CW systems have been constructed and evaluated for organic pollutant purification and processes of WWTP effluents. What is more, considering the sustainability and the potential of greenhouse gases (GHG) mitigation, not enough data are currently available on the dynamics and influencing factors of GHG emissions in the large-scale multi-stage CWs for polishing WWTPs effluent.

Therefore, the aim of this study was to evaluate the carbon dynamics of a large-scale seven-stage FWS CW polishing secondary effluents from a conventional domestic WWTP in northern China. Specific objectives of this study are: (i) to assess the removal performance and variability of organics in the seven-stage FWS CW for polishing WWTPs effluent; (ii) to evaluate spatial and seasonal variation of methane (CH₄) and carbon dioxide (CO₂) emission in this multi-stage CW system; and (iii) to investigate the influence of the key factors (input loading and temperature) on organics removal and $CH₄$ and $CO₂$ emission. The results from this study would be helpful in designing multi-stage CW systems in polishing WWTP effluent for urban river water quality improvement as a sustainable wastewater treatment technology.

2. Material and methods

2.1. Study site description

Fig. S1 presents the site map of the studied CW (the Wu River large-scale CW), which located on the west side of the Yi River in Linyi, Shandong province, northern China at a latitude of $34^{\circ}51'$ -35°06 N and a longitude of 118°06'-118°28'E. Wu River large-scale CW as a treatment system for improving river water quality was directly constructed along the riverbed of the Wu River, and Wu River as the downstream of Yi River and Xianni River is currently the main urban drainage channel to receive the treated effluents from a municipal WWTP in Linyi. The climate condition of the study site is warm temperate continental monsoon climate with a mean annual temperature 14.1 \degree C and an average precipitation of 818.8 mm, respectively.

2.2. Large-scale system design and operation

The Wu River large-scale CW is designed as a multi-stage CW and comprises a rubber dam, a distribution ditch, a sequence of seven stages of FWS CWs which are interconnected by using 500 mm concrete pipes and an outlet ([Fig. 1](#page--1-0)). In order to enhancing reoxygenation or reaeration in CW system, several concrete overflow weirs were designed between the fourth wetland unit and the fifth wetland unit of the CW system. The secondary effluent from the municipal WWTP as the influent was directly held by the rubber dam, and flowed into the following FWS CW systems through the distribution ditch. Finally, the effluent from the CW system flowed into the Yi River through the outlet. The total land area of the system was about 8660000 $m²$ (over 15000 m long and 120–320 m wide) with the treatment capability of 380000 m^3 d⁻¹. A natural soil layer of the Wu River was used as substrates for the CW system, and the CW system had a $30-50$ cm depth of water above the substrate. The theoretical hydraulic retention time (HRT) was around 7 d, and average hydraulic load was 3.5 cm d $^{-1}$. The CW system was planted with a diversity of macrophyte types which was mainly the dominant native wetland plants included Phragmites australis (P. australis), Typha orientalis (T. orientalis), Zizania latifolia (Z. latifolia), Nelumbo nucifera (N. nucifera), Nymphaea tetragona (N. tetragona), Potamogeton crispus (P. crispus), Lemna minor

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