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Comparing subsurface flow constructed wetlands with mangrove plants and freshwater wetland plants for removing nutrients and toxic pollutants

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ABSTRACT

Constructed wetlands (CWs) have been increasingly used to remove nutrients from wastewater, but their effectiveness to treat toxic wastewater remains largely unexplored. This study compared the treatment efficiency of CWs using mangrove plants (*Aegiceras corniculatum* and *Bruguiera gymnorrhiza*) and non-mangrove plants (*Acorus calamus*, *Canna indica* and *Phragmites australis*) in different cultural arrangements (mono-culture vs. mixed-culture) to remove nutrients (TOC, TKN, TP, NH_4^+ and NO_3^-) and toxic pollutants (heavy metals, PAHs and phenol) from wastewater. Additionally, the effect of tidal flushing on the treatment efficiency of the mangrove CWs was examined. The effectiveness of CWs was evaluated based on the health status of plants after 6-month irrigation with toxic wastewater, and the removal percentage of nutrients and pollutants. Following the experimental period, the mangrove plants remained healthy, while the non-mangrove plants were impaired by the toxic wastewater (e.g. chlorosis and wilting). In both mangrove and non-mangrove CWs, the presence of plants slightly enhanced the removal of nitrogenous compounds, while the pollutants were mostly adsorbed onto the sediment. The mangrove CWs generally had higher removal percentage of both nutrients and pollutants than the non-mangrove CWs. In the mangrove CWs, however, tidal flushing was necessary not only to facilitate the removal of nutrients, but also to prevent the production of NO_3^-. Cultural arrangement had no significant effect on the treatment efficiency. We conclude that the mangrove CWs, especially planted with *A. corniculatum*, have higher application values than the non-mangrove CWs to treat toxic wastewater on condition that tidal flushing is provided.

1. Introduction

Owing to the rapid urbanization and industrialization, water quality has been deteriorating in many developing countries over the last few decades (e.g. Singh et al., 2002; Wu et al., 2016). Given the high operating and maintenance cost of conventional wastewater treatment facilities, discharge of untreated wastewater into water bodies is commonly observed. In recent years, therefore, constructed wetlands (CWs) have been increasingly used to treat wastewater in view of cost-effectiveness and environmental friendliness (Kivaisi, 2001).

Constructed wetlands are composed of wetland plants, substrates and the associated microbial assemblages to treat wastewater through various physical and biological processes. While pollutants are primarily removed by adsorption onto substrates (Brix, 1997), absorption by plants, especially nutrients, can be substantial (Thullen et al., 2005; Yang et al., 2007; Yadav et al., 2012; Li et al., 2013; Sehar et al., 2015). Besides, roots provide suitable habitats by releasing oxygen or organic exudates for microbes to degrade nutrients (Stottmeister et al., 2003; Vymazal, 2007; Peng et al., 2014). Nevertheless, the influence of plants is species-specific, depending on their growth rate and nutrient uptake rate (Yang et al., 2007). Cultural arrangement may be associated with treatment efficiency. Mixed-culture, for example, is shown to have higher removal percentage of nutrients than mono-culture by enhancing the effective distribution of roots and diversity of microbial community (Zhang et al., 2010, 2012; Abou-Elela and Hellal, 2012). To maximize the treatment efficiency of CWs, using the

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optimal planting arrangement (i.e. optimal plants and cultural arrangement) is monumental.

Freshwater wetland plants, such as *Phragmites australis* and *Acorus calamus*, have been widely used in CWs, but it is noteworthy that most of the previous studies focused on the removal of nutrients with low level of toxic pollutants (e.g. Li et al., 2014; Zhang et al., 2014). In reality, high concentrations of salts, nutrients and toxic pollutants are often found in wastewater, especially in the developing countries with less stringent discharge standards. As such, freshwater wetland plants are probably not versatile enough for CWs because they may not be able to tolerate the stress due to salinity and toxic pollutants. For example, toxic pollutants can directly damage the plants or microbial communities (Schützendübel and Polle, 2002; Alkio et al., 2005; Wang et al., 2007; Park et al., 2012), thereby reducing the treatment efficiency of CWs. In this regard, mangrove plants may be competent for CWs because of their strong tolerance to pollutants as well as their special adaptations to survive in the highly fluctuating environment (e.g. salinity, temperature and anoxic substratum) (Wu et al., 2008). Since mangrove plants are distributed in the intertidal environment and subject to periodic flooding, tidal flushing should also be considered in the design of CWs when mangrove plants are used.

The present study aimed to examine the effectiveness of subsurface flow constructed wetlands to treat toxic wastewater using *Aegiceras corniculatum*, *Bruguiera gymnorrhiza*, *Acorus calamus*, *Canna indica* and *Phragmites australis* in different cultural arrangements (mono- and mixed-cultures). The former two are dominant mangrove plants, while the rest are common wetland plants which have good ability to treat wastewater (Konnerup et al., 2009; Xu et al., 2010; Li et al., 2014). These wetland plants are collectively denoted as ‘non-mangrove’ in this paper. In addition, the effect of tidal flushing on treatment efficiency of the mangrove CWs was evaluated. The chemical compositions of the wastewater used in this study stimulated those in Shenzhen River, South China, which suffers from severe water pollution due to discharge of industrial sewage. The effectiveness and suitability of CWs to treat toxic wastewater were evaluated by the removal percentage of nutrients and pollutants, and the health status of plants after long-term irrigation with toxic wastewater. The findings could provide a more realistic evaluation of CWs for wastewater treatment and shed light on the significance of planting arrangement for optimization of CWs.

2. Materials and methods

2.1. Setup of constructed wetlands

To optimize the performance of plants, sediments collected from the mangrove in Sai Keng (22°25′12″N, 114°16′06″E) and freshwater wetland in Long Valley (22°29′55″N, 114°06′59″E) were used to fill up the purpose-made tanks (66 cm long × 25 cm wide × 37 cm tall, 20 cm sediment depth, Fig. 1) for the mangrove and non-mangrove CWs, respectively. The sediment in Sai Keng mangrove was sandy (Sand: 78.4%; Silt: 21.6% and Clay: 0%) with hydraulic retention time of 10.6 h, whereas the sediment in Long Valley wetland was silty (Sand: 30.0%; Silt: 67.4% and Clay: 2.6%) with hydraulic retention time of 12.0 h in the CWs. Individuals of *B. gymnorrhiza* (Bg) (ca. 20 cm tall) and *A. corniculatum* (Ac) (ca. 20 cm tall), collected from the mangrove in Sai Keng, were cultured in the mangrove CWs under greenhouse conditions (temperature: 24 ± 1 °C; relative humidity: 80%) for one month prior to experimentation. There were four planting arrangements: monoculture of Ac, monoculture of Bg, mixed culture of Ac and Bg (individual ratio = 1:1), and control (i.e. no plants). Four healthy individuals were evenly planted in each tank (*n* = 3 replicate tanks per planting arrangement). To investigate the effect of tidal flushing, artificial seawater (salinity: 15 ppt) was added to the CWs (5 cm above sediment surface) to stimulate high tide, while drained to stimulate low tide. The sediment was flooded daily from 6:00 p.m. to 10:00 a.m. (next day). For the non-mangrove CWs, *A. calamus* (Acc) (ca. 120 cm tall), *C. indica* (Ci) (ca. 80 cm tall) and *P. australis* (Pa) (ca. 70 cm tall) were used. Seedlings of Acc were grown from seeds under greenhouse conditions, while seedlings of Ci were purchased from a local gardening company. Seedlings of Pa were collected from the reed bed in Mai Po Nature Reserve (22°29′33″N, 114°02′08″E), Hong Kong. There were seven planting arrangements: three monocultures of Acc, Ci and Pa, three mixed cultures in crossed combination (individual ratio = 1:1), and control. Four healthy individuals were evenly planted in each tank (*n* = 3 replicate tanks per planting arrangement).

2.2. Maintenance of constructed wetlands and record of health status

Following the setup of CWs, the plants were daily irrigated with 1 L artificial wastewater and allowed to acclimate under greenhouse conditions for two months prior to experimentation. The chemical compositions of the artificial wastewater, which mimic those of the wastewater in Shenzhen River, are shown in Appendix I. After acclimation, the initial and final (six months after commencement) stem height and leaf number of each plant were recorded. The growth status of plants, such as sprouting, flowering and wilting, was also recorded. During the acclimation and experimental periods, the plants were cleaned regularly to prevent fungal or insect infestation.

2.3. Collection of effluent and sediment samples

Effluent samples from the CWs were collected bimonthly. The debris in the effluent was filtered and the filtrate was used for chemical analyses. Sediment samples were collected at the end of the experiment. To do so, each plant was carefully removed with the aid of a spade and only the sediment underneath was collected. The sediment samples were freeze-dried, ground into powder and passed through a 2 mm sieve prior to chemical analyses.

2.4. Analyses of effluent and sediment samples

The pH of sediment was measured using a pH meter (HI 9025, Hanna Instruments, USA) after mixing with deionized water (1:5, w/v). Redox potential of sediment was measured *in situ* by a handheld Conductivity-pH-Redox-Temperature Meter (WP81, Science Essentials, Australia). Total organic carbon (TOC) in the effluent was measured using a TOC analyzer (TOC-5000A, Shimazu, Japan). Flow injection analyzer (QuikChem 8000, Lachat Instruments, USA) was used to analyze the concentrations of ammonium (NH_4^+) and nitrate (NO_3^-) in the effluent and sediment. Potassium chloride (2 M) extraction was applied to extract NH_4^+ and NO_3^- in the sediment. Total Kjeldahl nitrogen (TKN) and total phosphorus (TP) in the effluent and sediment were also determined by flow injection analyzer, following Kjeldahl acid digestion. Heavy metals in the sediment were extracted using concentrated nitric acid, followed by microwave digestion. The concentrations of heavy metals in the extract and effluent, including cadmium (Cd), chromium (Cr), copper (Cu), iron (Fe), manganese (Mn), nickel (Ni), lead (Pb) and zinc (Zn), were analyzed by inductively couple plasma optical emission spectrometry (Optima 2100 DV, PerkinElmer, USA). A certified reference material (MESS-3, National Research Council, Canada) was used to estimate the recoveries of heavy metals, which ranged from 73.5 to 101.2% with relative standard deviation (RSD) less than 1.78%.