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Enhanced removal mechanism of iron carbon micro-electrolysis constructed wetland on C, N, and P in salty permitted effluent of wastewater treatment plant

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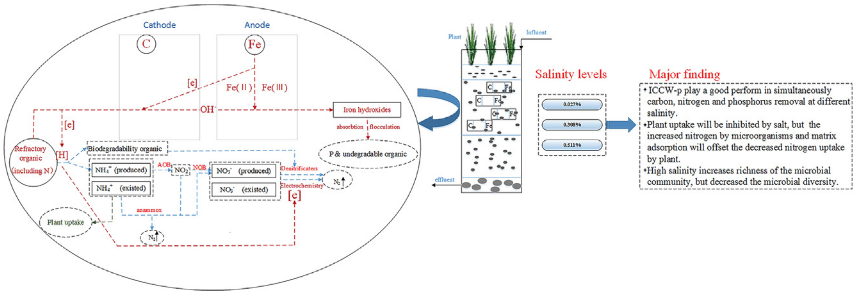
HIGHLIGHTS

- A new iron-carbon micro-electrolysis constructed wetland system, the combination of CW with iron-carbon system, was proposed.
- The simultaneous removal of contaminants in iron carbon micro-electrolysis constructed wetland was investigated.
- The enhanced removal mechanism of iron-carbon micro-electrolysis CWs was revealed in detail.

GRAPHICAL ABSTRACT



- ICCW-p play a good perform in simultaneously carbon, nitrogen and phosphorus removal at different salinity.
- Plant uptake will be inhibited by salt, but the increased nitrogen by microorganisms and matrix adsorption will offset the decreased nitrogen uptake by plant.
- High salinity increases richness of the microbial community, but decreased the microbial diversity.

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ABSTRACT

In this study, the combination of a constructed wetland (CW) with iron-carbon (Fe-C) system was used to enhance the simultaneous removal of carbon, nitrogen and phosphorus in salty permitted effluent of wastewater treatment plant (SPE-WTP). The removal mechanism of Fe-C micro-electrolysis CWs with different salinity (0.027, 0.308, and 0.511%) for treating SPE-WTP was investigated, including chemical oxygen demand (COD), phosphorus and nitrogen removal, the mass balance, as well as the changes in the microbial community structure. The results showed the salinity has a certain influence on the contaminant removals, and can enhance nitrogen removal under certain conditions. When the salinity increased from 0.308% to 0.511%, the removal of COD decreased from 68.20% to 62.69%, whereas the removal of total nitrogen (TN) increased from 72.02% to 81.21% in the ICCW-p system (including *P. australis* as the plant and gravel doped with 3% iron-carbon as the matrix). Microbial degradation, including the electrochemical effect (the degradation by iron-carbon micro-electrolysis) was the main N removal pathway in the ICCW-p system. The ICCW-p system always achieved higher removal rates (such as 81.21% TN and 62.69% COD removals at 0.511% salinity) than that in ICCW-n system (without plants and gravel doped with 3% iron-carbon as the matrix, 63.76% TN and 56.31% COD removals, respectively) and CW-n (without plants and gravel as the matrix, 14.90% TN and 22.39% COD removals, respectively). In addition, high-throughput sequencing analysis revealed that high salinity increased the abundance of N-removing bacteria in the ICCW-p system. Furthermore, with the introduction of iron-carbon in CWs, the removal methods in ICCW-p were diverse, which has enough ability to resist the impact of salinity. Fe electrolysis produced different valence states that acted as carriers for electron transport and accelerated the efficiency of biological and chemical reactions, which enhanced the simultaneous removal of carbon, nitrogen and phosphorus.

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1. Introduction

With the rapid development of the global economy and population growth, various serious problems caused by water pollution, such as shortage of freshwater resources, public health threats and other environmental issues, are attracting increasing attention (Shannon et al., 2008). In industry, agriculture and aquaculture, various types and large amounts of salts are produced, or occur naturally. Most of the saline wastewater contains not only inorganic salts but also other pollutants based upon specific sources (Liang et al., 2017a, 2017b). For example, saline wastewater usually contains salts, organic substances, nitrate, heavy metals and other pollutants such as sulfides (Quan et al., 2016; Tan et al., 2017a, 2017b). In the typical type of salty permitted effluent of wastewater treatment plant (SPE-WTP), which receives wastewater from various sources, nutrients (especially C, N and P in this study) of different chemical species are the most common potential pollutants (Liang et al., 2017a, 2017b). Nutrient enrichment or eutrophication of aquatic environments can increase algal growth and decrease dissolved oxygen, which further destroy the balance of an ecological structure (Hayes et al., 2017). Apart from the nutrients, salts (refers to inorganic salts in this study) also can inhibit the growth of plants, interfere with animal cellular activities and microbial metabolic processes, and even alter the community structure within an ecosystem (Chen et al., 2010). Therefore, high salinity has caused new problems that hinder the simultaneous removal of carbon, nitrogen and phosphorus in SPE-WTP, and these problems have posed a new challenge to the traditional technologies used to treat this wastewater.

Many studies on salts and removal pathways of specific contaminants in SPE-WTP, which is an urgent task in many countries, have been conducted. Conventional treatment plants struggle to find feasible methods for treating SPE-WTP. The most commonly reported methods for treating SPE-WTP include coagulation-filtration, membrane separation, electrochemical removal and advanced oxidation methods (Kim and Logan, 2013; Moussavi et al., 2010). However, most of these methods have disadvantages that include large economic investment in construction and large operation costs. Constructed wetland system (CWs), an environmentally friendly technology, has been shown to outperform conventional techniques (owing to its low cost and good performance) in treating permitted effluent from wastewater treatment plants (PE-WTP) (Wu et al., 2015). However, CWs have some disadvantages that limit their use for treating these effluents, such as blockages, inadequate carbon for nitrogen removal, inadequate oxygen, and low operation efficiency at low temperatures (Zhao et al., 2016). More research is necessary regarding the design, operation, and optimization of CWs for simultaneous carbon, nitrogen and phosphorus removal from SPE-WTP.

Plants have been proven to be an important component in constructed wetland treatment (Gao et al., 2014; Galletti et al., 2010). Some halophytic plants (e.g. *Spartina maritima*, *Juncus maritimus*, *Phragmites australis* and *Digitaria bicornis*, etc.) can take up excessive salts and enhance the removal of nutrients from wastewater, thereby playing an important role in reducing salt toxicity toward other functional system within CWs (De Lange et al., 2013; Jesus et al., 2017). Furthermore, iron has been widely applied in biological wastewater treatment to improve the removal efficiency of nutrients because its changeable chemical valence can induce various physico-biochemical processes (Ma et al., 2014). Iron-carbon micro-electrolysis, as one of several widely used wastewater treatment technologies in advanced oxidation processes (AOPs), has proven to be a high-efficiency and low-cost method for the treatment of various wastewaters (Yang et al., 2017). Fe (II) can be easily oxidized into Fe (III) under aerobic conditions, whereas Fe (III) can be easily reduced into Fe (II) under reductive conditions (Stefanakis and Tsihrintzis, 2009). Especially in CWs, various oxidation-reduction zones were found (Zhao et al., 2017), which provide the potential for combining a CW system with an iron-carbon system. However, previous studies have rarely focused on the simultaneous removal of carbon, nitrogen and phosphorus from SPE-WTP in a combined iron-carbon micro-electrolysis constructed wetland system. Furthermore, little is known about the influence of salinity on carbon, nitrogen and phosphorus removal in iron-carbon micro-electrolysis constructed wetland system.

Therefore, the major objectives of this study were: (a) to assess chemical oxygen demand (COD), phosphorus and nitrogen removals in CWs with different salinity levels; (b) to examine the effect of salinity on the nitrogen mass balance in the CWs; and (c) to evaluate the variations in microbial richness and diversity in the iron-carbon micro-electrolysis constructed wetland systems and identify the simultaneous removal mechanism of carbon, nitrogen and phosphorus for iron-carbon micro-electrolysis CWs. The information obtained from this study will provide both practical references for using CWs to remove nutrients from SPE-WTP, and an academic reference for understanding the interactions between salts and nutrients and their respective accumulations when iron-carbon micro-electrolysis is used in CWs.

2. Materials and methods

2.1. Experimental design

Experiment was conducted in a greenhouse laboratory (27 ± 2 °C) at the College of Environment, Hohai University, Nanjing, China. For all experiments, two-month old seedlings (about 20 cm in height) of the wetland plant species, *Phragmites australis* (*P. australis*) were purchased from a supplier in Huaian, Jiangsu Province in April 2016. The seedlings were cultivated in a 1/12 Hoagland solution until required. Previously, *P. australis* was shown to grow well under moderate salinity conditions (e.g., electrical conductivity (EC) 7 mS/cm) (Shang et al., 2014), and was therefore considered to be a potential species for use in CWs to treat saline wastewaters. The experiments conducted in CWs made from methyl methacrylate with an inner diameter of 200 mm, the depth of 700 mm. All the systems were covered with foil to avoid direct exposure to sunlight (thereby minimizing algal growth).

2.2. CWs establishment and operation

Three CW systems were established: CW-n (without plants and using gravel as the matrix), ICCW-n (without plants and gravel doped with 3% iron-carbon as the matrix), and ICCW-p (including *P. australis* as the plants and gravel doped with 3% iron-carbon as the matrix). The working thickness of the CW matrix was 50 cm, and was comprised of a 5 cm top cover layer, a 40 cm middle filtration layer, and a 5 cm bottom supporting layer. The three systems were continuously operated as downflow vertical CWs (Fig. 1). Because the ratio and dopant amount of Fe-C could influence the effect of micro-electrolysis, the optimal ratio (Fe:C = 5:1) and dopant amount of iron-carbon (3%) has been obtained according to our previous studies. In addition, five sampling points were established in the matrix layer (at 10, 20, 30, 40, and 50 cm from the bottom) in each CWs. The hydraulic retention time (HRT) of each CWs was 2 days while the surface hydraulic load was 0.20 $m^3/(m^2 \cdot d)$.

2.3. Experimental operation

A synthetic wastewater was used as influent to the CWs. In a previous study, the salt content of actual SPE-WTP was found to be 0.099–0.428%, so the experiment was divided into three periods: Period I (duration 40 days, 0.027% salinity), Period II (duration 40 days, 0.308% salinity) and Period III (duration 40 days, 0.511% salinity). Sodium chloride (NaCl), ammonium chloride (NH_4Cl), Potassium nitrate (KNO_3) and monopotassium phosphate (KH_2PO_4) were added to adjust salinity, N and P in the influent. The synthetic wastewater was composed of the following constituents (all measured in mg/L): COD 60 (from a mixture of glucose, sodium humate, sodium alginate and peptone), NH_4^+-N 8 (NH_4Cl), PO_4^{3-}-P 1 (KH_2PO_4), and NO_3^--N 12 (KNO_3). A trace elements