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Treatment of anaerobic digestate supernatant in microbial fuel cell coupled constructed wetlands: Evaluation of nitrogen removal, electricity generation, and bacterial community response

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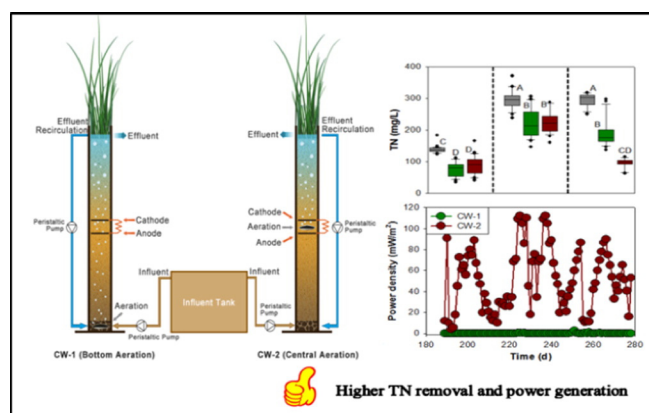
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HIGHLIGHTS

- The performance of central aerated CWs with effluent recirculation was assessed.
- The new configuration CWs can efficiently treat anaerobic digestate supernatant.
- The removal of NH_4^+-N , TN and COD was achieved at 92%, 69% and 69%, respectively.
- Higher electricity generation (112 mW/m^2) was found in the new configuration CWs.
- Higher abundance of denitrifiers and anammox was found in the new configuration CWs.

GRAPHICAL ABSTRACT



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ABSTRACT

The objective of this study was to assess whether the improved configuration of vertical upflow constructed wetlands (CWs) coupled with aeration in the centre part and effluent recirculation can strengthen the treatment performance of high strength anaerobic digestate supernatant. Moreover, electricity generation and bacterial community characteristics were also examined. The results indicated that intermittent aeration in vertical upflow CWs significantly enhanced organic matter ($>69\%$, $214\text{--}401 \text{ g/m}^2 \text{ d}$) and ammonium ($>92\%$, $62\text{--}138 \text{ g/m}^2 \text{ d}$) removal, regardless of aeration position. However, the removal efficiency of total nitrogen (TN) was limited to 24%–40%. Effluent recirculation was examined to enhance TN removal up to 69% in the central aerated CW, as compared to 44% observed in the bottom aerated CW. Accordingly, significantly higher abundances of denitrifiers (*nirK* and *nirS*) and anaerobic ammonium oxidation bacteria (anammox) were found in the bottom layer of the central aerated CW. In addition, the central aerated CW coupled with effluent recirculation generated significantly higher electricity (maximum power density of 112 mW/m^2) than traditional bottom aerated CWs when integrated with a microbial fuel cell (MFC). Results confirmed the application potential of this new configuration of upflow CW integrated with central aeration and effluent recirculation.

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1. Introduction

The rapid improvement in Chinese living standards has led to an increased demand for higher dietary levels and, subsequently, the number of livestock farms continues to grow. These farms have in turn generated large quantities of livestock manure and related organic wastewater (Huaitalla et al., 2013). Presently, most farms use anaerobic digestion to treat livestock wastewater because it can produce renewable energy (CH₄) as well as reduce greenhouse gas emissions (Deng et al., 2006; Nasir et al., 2012). During this process, most biodegradable organic matter is converted to CO₂ and CH₄ (Massé et al., 2007). However, the concentration of nutrients in the effluent from anaerobic reactors remains high, especially in the case of NH₄⁺-N, which is not efficiently removed by anaerobic digestion, and might at times actually increase.

Several techniques employing biological, physical, chemical, or a combination of these methods have been reported to recover nutrients and treat anaerobic digestate supernatant, such as struvite formation (Song et al., 2011b), ammonia stripping (Degermenci et al., 2012), and biological membrane reactors (Song et al., 2011a), as well as a sequential combination of aerobic and anaerobic batch reactors (Frison et al., 2013). Although these techniques have been reported to be effective, they are still limited by high costs. Considering the low-profit nature of Chinese livestock breeding farms, finding a more economical technique for post-treatment of anaerobic digestate supernatant is crucial.

Constructed wetlands (CWs) are often recognized as a low-cost and operationally simple technology for wastewater treatment (Vymazal, 2010). Recently, CWs have been used to treat livestock wastewater with high NH₄⁺-N levels (Harrington and Scholz, 2010; Liu et al., 2014). However, given the limited oxygen transfer rate of traditional CWs and the high content of residual organic matter and ammonium in these wastewaters, the treatment efficiency of traditional wetlands is often quite low and requires a large land area (Liu et al., 2014).

Wu et al. (2014) reviewed various expanded CWs with innovative designs, configurations, and technology combinations. Among these, integrating aeration in traditional horizontal and/or saturated vertical flow CWs showed the highest potential, but this step was mostly incorporated at the bottom of CWs. This design can result in an aerobic condition in the entire CW and may not be efficient enough to conserve energy and drive sequential nitrification under oxidative conditions and denitrification under anaerobic conditions (Dong et al., 2012). Changing the aeration position to the central layer of a CW may create an anaerobic section that benefits denitrification activity in the unaerated bottom portion of the CW (Wang et al., 2015). In addition, recirculation of effluent back to the inflow zone of CWs can further fuel the process of denitrification by providing nitrate from the effluent. However, the performance of this improved configuration with central aeration and effluent recirculation for the treatment of high strength wastewater, such as anaerobic digestate supernatant, should be evaluated. Moreover, the bacterial community response to the configuration changes should also be investigated to better understand the pollutant removal mechanisms.

A microbial fuel cell (MFC) coupled with a CW (CW-MFC) system is an expanded wetland that allows for simultaneous wastewater treatment and energy production (Doherty et al., 2015). An MFC produces bioelectricity through an external circuit by transferring electrons from the anode region to the cathode region (Logan, 2008). Various factors, such as system configuration and electrode material, will affect electricity/energy generation in a CW-MFC system. Doherty et al. (2015) reviewed that a CW-MFC can generate a power density of 1.84–44.63 mW/m² under influent COD concentrations of 135–8000 mg/L with COD removal efficiencies of 64%–95%. However, the energy recovery dynamics by electricity generation associated with high strength anaerobic digestate supernatant treatment in vertical upflow CWs with central aeration and effluent recirculation has not been sufficiently investigated.

In this study, two laboratory-scale aerated vertical upflow CWs were set up to treat effluent supernatant from the anaerobic digestion of pig slurry. To provide an improved configuration to enhance total nitrogen (TN) removal, the performances of these two vertical upflow CWs coupled with either central or bottom aeration, as well as effluent recirculation, were tested. An abundance of nitrogen-transforming bacteria were detected in both CWs through molecular biology analysis. Moreover, the capacity and dynamics of electricity generation from both CWs were reviewed.

2. Materials and methods

2.1. Aerated vertical upflow CWs setup

Two lab-scale aerated vertical upflow CWs (CW-1 and CW-2), with identical dimensions (inner diameter of 9 cm and height of 150 cm), were set up (Fig. 1). Both CW columns were filled with quartz sand (0.2–0.6 cm in diameter) and a height of 10 cm of gravel (1–3 cm in diameter) at the bottom to prevent clogging. The effective volumes of both CWs were 3.5 L. The inlets were positioned at the bottom of the CWs and were controlled by peristaltic pumps to allow the systems to operate in continuous upflow mode. The outlets were set at the top of the CWs. The intermittent aeration was set at the bottom of CW-1 and in the centre (70 cm in height) of CW-2. Both CWs were planted with 30–40 *Juncus effusus* stalks.

Both CWs were equipped with microbial fuel cells (MFCs) and cathode (above) and anode (below) graphite electrodes (8 cm diameter and 0.5 cm thickness) were buried in the middle portion of the system with an electrode space of about 12 cm (Fig. 1). The electrodes were porous plates to allow wastewater to flow through. Cathodes and anodes were connected by insulated titanium wire through an external circuit with an external resistance of about 950 Ω, which was selected from the literature on previous CW-MFC studies (Doherty et al., 2015). It should be noted that in CW-2, the aeration was set in the middle of the graphite plates, which intermittently aerate the cathode compartment.

The CWs were wrapped with black shading membranes to prevent microalgae formation and placed in the Laboratory of Bioenergy Engineering and Low Carbon Technology at China Agricultural University. The laboratory was operated under defined environmental conditions simulating an average summer day in moderate climate conditions. The temperature was adjusted to 22 °C from 6 a.m. to 9 p.m. and to 16 °C from 9 p.m. to 6 a.m. Lamps (Master SON-PIA 400 W; Philips, China) were switched on at daytime as an additional constructed light source when natural illumination was below 60 klx.

2.2. Experimental conditions

The anaerobic digestate supernatant (influent of the CWs) was collected from a mesophilic biogas plant (400 m³), which was used solely to digest piggery slurry with a digested effluent generating capacity of 10 m³/d. The hydraulic retention time of this biogas plant is about 40 d. The anaerobic biogas plant is located in Dong Hua Shan Village, Shunyi District, Beijing, China (N40°06'24.59", E116°54'30.68").

The experiment was conducted from April 2014 to January 2015 for approximately 300 days under three continuous phases, A, B, and C, based on different influent concentrations and operation conditions (Table 1). During phase A (130 days), the influent anaerobic digestate supernatant was diluted 10 times from the original collection with average chemical oxygen demand (COD) and NH₄⁺-N concentrations of 647 and 137 mg/L, respectively. To evaluate the higher treatment ability of the CWs, the influent in phase B was increased to 5 times dilution with average COD and NH₄⁺-N concentrations of 1111 and 297 mg/L, respectively, for 100 days. To intensify TN removal efficiency, effluent recirculation to the influent with the ratio of 1:1 was investigated in both CWs during phase C for 70 days. The average COD and NH₄⁺-N

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