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Research article

Optimization of high-rate TN removal in a novel constructed wetland integrated with microelectrolysis system treating high-strength digestate supernatant

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ABSTRACT

The potential of high-rate TN removal in three aerated horizontal subsurface-flow constructed wetlands to treat high-strength anaerobic digestate supernatant was evaluated. Different strategies of intermittent aeration and effluent recirculation were applied to compare their effect on nitrogen depuration performance. Additional glucose supply and iron-activated carbon based post-treatment systems were established and examined, respectively, to further remove nitrate that accumulated in the effluents from aerated wetlands. The results showed that intermittent aeration (1 h on:1 h off) significantly improved nitrification with ammonium removal efficiency of 90% (18.1 g/(m^2·d)), but limited TN removal efficiency (53%). Even though effluent recirculation (a ratio of 1:1) increased TN removal from 53% to 71%, the effluent nitrate concentration was still high. Additional glucose was used as a post-treatment option and further increased the TN removal to 82%; however, this implementation caused additional organic pollution. Furthermore, the iron-activated carbon system stimulated with a microelectrolysis process achieved greater than 85% effluent nitrate removal and resulted in 86% TN removal. Considering the high TN removal rate, aerated constructed wetlands integrated with a microelectrolysis-driven system show great potential for treating high-strength digestate supernatant.

1. Introduction

With the increasing demand of meat consumption and modifications in the pig-breeding industry, large-scale intensive pig farms have developed very fast in China (Jiang et al., 2011). The manure produced from these pig farms significantly increased by up to 243 million tons in the year of 2010 (Zhong et al., 2013). The technology of anaerobic digestion has been strongly promoted worldwide owing to its double benefits of waste treatment for environmental protection and renewable energy production. For example, the number of manure-based biogas plants in rural regions of China has reached up to 91, 600 by the end of 2012, which increased by almost 3 times that in 2007. In particular, more than 80% of these biogas plants was within medium to large scale and located in coastal and southwest areas (Hu et al., 2015a).

With this considerable construction of biogas plants, large volumes of anaerobic digestate are also simultaneously produced. Owing to the process of hydrolysis and subsequent methanogenesis, most organic compounds are converted to methane and the concentration of ammonium nitrogen generally increased in the anaerobic digestate (Möller and Müller, 2012; Tambone et al., 2009). Given the typical composition of the anaerobic digestate, there are two mainly possible solutions for its application or disposal. Traditionally, it would be considered as bio fertilizers for use in farmlands, but in special cases when the quantity of these anaerobic digestate exceeds the land consumption capacity. This current situation consequently leads to the pollution of environment and causes epidemic diseases, which has to be considered as wastewater that requires treatment (de la Fuente et al., 2013; Feng et al., 2011). Therefore, proper post-treatment techniques for economical discharge of this high-strength anaerobic digestate supernatant into water bodies are desirable.

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Compared with other energy-intensive treatment technologies such as biological membrane reactors (Song et al., 2011) and ammonia stripping (Değermenci et al., 2012), constructed wetlands (CWs) are more potential techniques for reducing pollutants in the supernatant, owing to its low cost of construction and easy maintenance (Wu et al., 2014). In particular, sub-surface, horizontal flow CWs as one category of treatment wetlands have been proven to be a pathogenically safe and aesthetic treatment technique, and they are widely used in European countries (Ayaz, 2008). However, given the limited oxygen supply, treatment efficiency of conventional horizontal subsurface flow CWs for treating high-strength wastewaters such as digestate supernatant was far from satisfactory and requires further improvement (Wu et al., 2015a).

In order to improve the dissolved oxygen in horizontal subsurface flow CWs, additional artificially aeration appears to be the most effective alternative. In particular, when dealing with high strength wastewater, artificial aeration seems to be very important for achieving complete nitrification. Thus far, artificial aeration in most CWs studies have been performed in continuous mode (24 h per day) (Maltais-Landry et al., 2009; Ong et al., 2010), resulting in significant energy consumption. Nonetheless, energy requirements of these aerated CWs (0.16–0.49 kWh/m^3) are still lower than conventional wastewater treatments such as activated sludge (0.76–0.88 kWh/m^3) by a large extent (Austin and Nivala, 2009; Kadlec and Wallace, 2009). However, this arrangement always leads to contradiction between the removal of ammonium nitrogen and total nitrogen because alternate aerobic and anaerobic conditions for nitrification and denitrification are unavailable (Saeed and Sun, 2011), and the influent carbon source is depleted (Wu et al., 2014).

Further improvements in aerated CWs could be achieved by applying intermittent aeration, in which the level of aeration can be adjusted and controlled (Fan et al., 2013a). This is beneficial not only for saving energy, but also enables alternate aerobic and anaerobic conditions for simulating nitrification and denitrification, and further helps in achieving high total nitrogen removal (Boog et al., 2014; Labella et al., 2015; Wu et al., 2015b). The regime of intermittent aeration plays an important role in nitrogen removal: excessively long or short regimes both result in imbalances of nitrification and denitrification. For instance, higher aeration frequency can lead to more complete nitrification, whereas impose restrictions on denitrification, and further low total nitrogen removal. However, to the best of our knowledge, intermittent aeration in most CWs studies mainly focus on treating domestic sewage (Nivala et al., 2007; Vymazal, 2009; Zhang et al., 2010a), and little attention has been paid to treating high-strength anaerobic digestion supernatant.

Moreover, except for dissolved oxygen for limiting nitrification, organic carbon source is the major limiting factor as electron donor in traditional heterotrophic denitrification. However, considering that the degradation of organic matter is prior to ammonium oxidation, the low C/N ratio of high strength anaerobic digestion supernatant hinders classical denitrification metabolism in CWs (Wu et al., 2015a). Effluent recirculation has been proposed by Arias et al. (2005) and Wu et al. (2014) as an operational modification to promote nitrogen reduction in CWs, which can transfer a part of the effluent with sufficient nitrate back to the inflow with more organic matter content to promote denitrification. Even though several studies have been conducted on wastewater treatment through the integration of CWs with intermittent aeration and effluent recirculation (Chang et al., 2014), very few have focused on improving total nitrogen removal through synergistic intermittent aeration with effluent recirculation, especially for treating anaerobic digestion supernatant.

Considering that nitrate would accumulated in the effluents from aerated wetlands and total nitrogen removal was limited, post-treatment should be established. External carbon source addition could effectively provide electron donor for nitrate reduction, but may lead to secondary pollution with surplus organic matter in the effluent. While nitrate could be effectively removed by zero-valent iron, the resulting iron corrosion product(s) (e.g., iron oxides or oxide hydroxides that are usually coated on the surface of zero-valent iron) could significantly inhibit nitrate reduction reaction under environmental conditions (Huang et al., 2003). However, if the solution contained enough amount of Fe^{2+}, Fe^{2+} would trigger a rapid nitrate reduction with a large amount of precipitates (γ-FeOOH) formed as the iron corrosion product (Huang and Zhang, 2005). Numerous microscopic galvanic cells can be formed in the iron-activated carbon microelectrolysis system, and electrons are supplied from the galvanic corrosion of iron (anode) to form Fe^{2+} (Mehrabi et al., 2015). However, how this conceptual technology effective to reduce nitrate from wastewater as comparing to the traditional carbon source addition has not been significantly evaluated so far.

In this study, three laboratory-scale horizontal subsurface flow CWs integrated with different strategies of intermittent aeration and effluent recirculation were set up to evaluate and compare the nitrogen depuration performance in treating anaerobic digestion supernatant. Moreover, the feasibility of iron-activated carbon column and glucose addition column as post treatment in enhancing the total nitrogen removal was tested. During each operation condition, the gene copy number of ammonia-oxidizing bacteria (AOB) and nitrite reductase-S (nirS), the levels of nitrification and denitrification in three CWs, were analyzed to better demonstrate microbial response to changes in conditions.

2. Materials and methods

2.1. Laboratory-scale wetlands

Three experimental horizontal subsurface-flow constructed wetlands (CWs) were set up in plexiglass containers (length, 100 cm; width, 15 cm; height, 50 cm) CW-1, CW-2, and CW-3. These containers were uniformly filled with approximately 100 kg of gravel (diameter, 2 mm–6 mm; density, 1.67 g/cm^3; porosity, 35%) up to a height of 45 cm. The water level was adjusted to be 5 cm below the gravel bed surface. Sieves of perforated plastic were placed at a distance of 3 cm in front of the inflow and outflow of the gravel bed to create small inlet and outlet zones. The free liquid volume was specially designed to ensure an equal distribution of inflow and laminar (plug) liquid flow through the gravel bed.

Considering accumulated NO_3^--N in the effluents from aerated wetlands throughout the entire experimental phases, two post-treatment systems after wetlands were developed. One was set up by using iron and activated carbon to develop microelectrolysis process (Column I) and the other was operated by supplying additional glucose as electron donor (Column II). Both columns were set up in two up flow plexiglass containers (height, 30 cm; diameter, 12 cm). Column I was filled with iron chips (diameter, 1–3 mm) and activated carbon particles (diameter, 4–6 mm) with a volume ratio of 4:1. Column II was filled with activated carbon particles (diameter, 4–6 mm) and the glucose was supplied at the bottom of this column according to the effluent nitrate concentration with a ratio of $C_6H_{12}O_6$–C: NO_3^--N = 2:1.

The three horizontal subsurface-flow CWs were wrapped with black plastic cloth all around the sidewalls to prevent periphyton formation. *Juncus effusus*, a species native to Asia and other parts of the world, was planted in the three CWs at a density of 70–80 stalks per wetland. All the experimental CWs were placed in the Bioenergy and Environment Science & Technology Laboratory at