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The intensified constructed wetlands are promising for treatment of ammonia stripped effluent: Nitrogen transformations and removal pathways☆

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ABSTRACT

This study investigated the treatment performance and nitrogen removal mechanism of highly alkaline ammonia-stripped digestate effluent in horizontal subsurface flow constructed wetlands (CWs). A promising nitrogen removal performance (up to 91%) was observed in CWs coupled with intensified configurations, i.e., aeration and effluent recirculation. The results clearly supported that the higher aeration ratio and presence of effluent recirculation are important to improve the alkalinity and pollutant removal in CWs. The influent pH (>10) was significantly decreased to 8.2–8.8 under the volumetric hydraulic loading rates of 0.105 and 0.21 d^{-1} in the CWs. Simultaneously, up to 91% of NH_4^+-N removal was achieved under the operation of a higher aeration ratio and effluent recirculation. Biological nitrogen transformations accounted for 94% of the consumption of alkalinity in the CWs. The significant enrichment of $\delta^{15}N$-NH_4^+ in the effluent (47–58‰) strongly supports the occurrence of microbial transformations for NH_4^+-N removal. However, relatively lower enrichment factors of $\delta^{15}N$-NH_4^+ (−1.8‰ to −11.6‰) compared to the values reported in previous studies reflected the inhibition effect of the high pH alkaline environment on nitrifiers in these CWs.

1. Introduction

Anaerobic digestion (AD) technology has been widely used in livestock waste treatment as a result of the benefits to environmental protection and bioenergy generation (Mao et al., 2015; Sakar et al., 2009). The number of AD plants has dramatically increased in recent years. In Europe, the total number of AD plants increased by 10% in comparison to the previous year and reached 14,572 in 2013 (Torrijos, 2016). In China, the number of large scale anaerobic digestion plants with monomer volumes of more than 300 m^3 rapidly increased to 15,531 at the end of 2013; these plants can process approximately 42% of the 3.8 billion ton livestock manure (NDRC, 2014). The generated biogas from these intensive scaled AD plants can then be further purified and/or upgraded to cooking gas or converted to electricity (Mao et al., 2015). However, if the surrounded arable land is not sufficient to consume digestate as fertilizer, the largely generated digestate will cause potential negative impacts on the environment. Therefore, the adequate management or disposal of the surplus digestate has been recently discussed in order to avoid to the drawbacks of the sustainable development of the AD technology (Sheets et al., 2015).

Nitrogen (mainly in the form of ammonium) concentrations in the surplus digested effluent from the AD process of the animal slurry are generally far over the treatment capacity of the conventional biological treatment technology (Lei et al., 2007). Moreover, the relatively low chemical oxygen demand/total nitrogen (COD/TN) ratio (1–3) of the digested effluent is also insufficient to facilitate comprehensive TN removal through heterotrophic denitrification (Li et al., 2016). However, instead of being converted to dinitrogen gas, this high nitrogen content effluent could also be sustainably reused in agriculture (Kizito et al., 2015).

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For the recovery of nitrogen from waste effluents, several techniques have been suggested, including microbial fuel cells (Kuntke et al., 2012), reverse osmosis (Mondor et al., 2008), electrodialysis (Mondor et al., 2008), and adsorption (Tada et al., 2005). Ammonia stripping, which transfers the ammonia from the liquid into an air stream through pH adjustment and aeration, is recognized as an effective technology for ammonia recovery/removal from ammonia-rich wastewater due to its low cost, easy installation, and high ammonia removal ability (Yuan et al., 2016). For the treatment of digested effluents or for the recovery of nitrogen from digested effluents, the ammonia stripping technique has already been well studied by scientists (Jiang et al., 2014) and implemented by engineers (Zeng et al., 2006). However, the discharged effluent of this stripping process often has a high alkalinity with $pH > 10$, which may give rise to another environmental issue by adversely affecting aquatic ecosystems (Mayes et al., 2009). Serna-Maza et al., 2015) proposed using this alkaline effluent to upgrade methane content of biogas by consuming CO_2 from the biogas and OH^- from the alkaline effluent simultaneously while pumping the biogas through this solution. The pH of the effluent after this step is still normally higher than 9. The most conventional management option for high alkaline wastewater is confined to direct chemical neutralization as the priory treatment. However, this requires a sustained capital input that may not be suitable for low-income farmers and/or agricultural industries in developing countries.

Constructed wetlands (CWs) are man-made systems that have already been acknowledged in recent years for the treatment of polluted waters, owing to their cost-effectiveness, easy maintenance, and efficient performance (Wu et al., 2014). Some previous studies have preliminarily reported the potential of traditional horizontal subsurface flow CWs to buffer high alkaline ($pH > 10$) industry wastewaters, such as cement and lime drainages (Mayes et al., 2005), bauxite residue leachate (Higgins et al., 2017), and steel slag drainage (Mayes et al., 2009). Most of the published work, however, only focuses on the alkalinity buffering potential of these wetlands and ignores the removal performance of associated pollutants and degradation pathways (Buckley et al., 2016; Mayes et al., 2009). Only a few studies examined the precipitation of divalent metals, e.g., Co, Mn, Ni, and Zn, in these alkaline environments (Higgins et al., 2016, 2017). Nitrogen transformation dynamics and respective pathways could be strongly influenced by and interact with highly alkaline conditions, but knowledge in this field is still insufficient.

To improve the treatment performance and minimize the occupied land area, some intensive operational strategies with new configurations (e.g., bottom aeration and effluent recirculation) were gradually integrated into the traditional CW beds (Wu et al., 2014). In alkaline conditions, the artificial bottom aeration can improve the biological degradation of organics into CO_2 that can theoretically be easily dissolved to neutralize alkalinity. Moreover, the aeration by introducing air into the bed will also physically supply more CO_2 dissolved in water to neutralize alkalinity (Summerfelt et al., 2015). The aeration is also closely linked to nitrogen removal, by not only effectively intensifying the nitrification process with extra oxygen into the bed, but also benefiting simultaneous alkalinity consumption due to the generation of protons in nitrification (Li and Irvin, 2007). Another commonly used operation is the effluent recirculation, which could additionally increase the water/substrate contact time. The effluent recirculation was demonstrated to improve denitrification by utilizing the numerous NO_3^--N accumulated in the effluent (Wu et al., 2017). Due to the aforementioned nitrogen removal pathways through microbial activities, the contribution of nitrogen removal through NH_3 volatilization should be studied. The higher hydraulic loading rate (HLR), which induces more pollutants into CWs, may cause negative effect on the microbial community and plant growth and decrease the treatment performance. Associated with such various nitrogen transformations, the knowledge on how the abundance and functioning of the microbial community response to the alkaline environment and operational strategies should be investigated.

In this study, three lab-scale horizontal subsurface flow CWs have been established to 1) investigate the treatment feasibility of highly alkaline ammonia-stripped digestate effluent in CWs; 2) determine the influence of volumetric hydraulic loading rates (HLR_v) and intensifying strategies (bottom aeration and effluent recirculation) on pollutant removal under the highly alkaline conditions; and 3) discuss the nitrogen transformation dynamics and removal pathways by N-isotopic fractionation assessment and microbial community response.

2. Materials and methods

2.1. Experimental setup and operational conditions

Three laboratory-scale horizontal subsurface flow CWs (CW1, CW2, and CW3) were established in this study. They weremade of Plexiglass containers with a length, width, and height of 100 cm, 15 cm, and 50 cm, respectively. Each container was filled with a 45-cm layer of sand (Ø 0.2–0.6 cm, with average porosity of 35%) as the main CW media. The water height in each CW was approximately 40 cm. Two perforated plastic boards were placed at a distance of 3 cm in front of the influent and effluent openings to create small water distribution and collection zones, respectively. These zones were designed to ensure the equal distribution of horizontal-flow influent in the main CW bed and were filled with large gravel pieces with diameters of 2–4 cm. The CWs shared a 150-L influent tank, constantly mixed by a submerged centrifugal pump placed at the bottom. The pump was controlled by a timer and was switched on for 5 min in every 30 min with an air flow rate of 50 L/h. Three individual peristaltic pumps were used to pump the wastewater from the influent tank to the three CWs separately. All CWs were planted with *Juncus effuses*, at a density of 70–80 stalks per CW. Experimental CWs were placed in a greenhouse and operated under defined environmental conditions simulating an average summer day under moderate climatic conditions. The conditions were controlled at 22 °C during the day (from 6 a.m. to 9 p.m.) and 16 °C at night (from 9 p.m. to 6 a.m.). Lamps (Master SON-PIA 400 W; Phillips, Shanghai, China) were switched on during the day as an artificial light source when natural illumination was below 60 klx.

The experiment was conducted for 200 d under two continuous phases at different influent volumetric hydraulic loading rates (HLR_v) (Table 1). To evaluate the operational conditions of CWs treating the ammonia-stripped digestate effluent, the HLR_v was set at 0.105 and 0.21 d^{-1} for all CWs in phase I and II, respectively. To decrease the toxic effect of alkalinity on microbial communities and enhance NH_4^+-N removal under intensified operations, different artificial aeration ratios and effluent recirculation rates were set up in the CWs. An air bar connected to an air pump (ACO-818, Aiseng, Guangdong, China) intermittently supplied air to the bottom of the CWs with a flow rate of about 180 L/h. The pump was actuated via a timer and kept the aeration ratios (on: off) at 1 h:5 h, 1 h:1 h, and 1 h:5 h for CW1, CW2, and CW3, respectively. The effluent of CW2 and CW3 were recirculated to the influent collection area of the CWs controlled by a peristaltic pump (BT 100-2J, Longer Precision Pump Co. Ltd., Baoding, China) with effluent recirculation ratio of 1:1 and CW1, as the control, had no effluent recirculation.

All CWs were fed by ammonia-stripped digestate effluent. The original anaerobic digestate was collected from a mesophilic biogas