



Research article

Biological denitrification from mature landfill leachate using a food-waste-derived carbon source

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ABSTRACT

The mature landfill leachate containing high ammonia concentration (>1000 mg/L) is a serious threat to environment; however, the low COD to TN ratio (C/N, <3) strongly inhibits the denitrification process and poses a severe obstacle for efficient treatment. Herein, two kinds of acidogenic liquids, fermented from oil-removed food waste and oil-added food waste, were first applied as external carbon sources for the biological nitrogen removal from mature landfill leachate in an aerobic/anoxic membrane bioreactor. "Acidogenic liquid b" served quite better than commercial sodium acetate, considering the higher denitrification efficiency and the slightly rapider denitrification rate. The effect of C/N and temperature were investigated under hydraulic retention time (HRT) of 7 d, which showed that C/N ≥ 7 (25 °C) was enough to meet the general discharge standards of NH₄⁺-N, TN and COD in China. Even for some special areas of China, the more stringent discharge standards (NH₄⁺-N ≤ 8 mg/L, TN ≤ 20 mg/L) could also be achieved under longer HRT of 14 d and C/N ≥ 6. Notably, the COD concentration in effluent could also be well reduced to 50–55 mg/L, without further physical-chemical treatment. This proposed strategy, involving the high-value utilization of food waste, is thus promising for efficient nitrogen removal from mature landfill leachate.

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1. Introduction

Municipal solid waste (MSW) has been explosively increasing around the world during the past decades (Ye et al., 2011), and its annual generation in China increased to 180 million tonnes by 2014 (Hong et al., 2017). In spite of the rising in composting and incineration, sanitary landfill is still the principal technology that predominates over 65% of MSW treatment in China, due to convenience and cost-effectiveness (Zhou et al., 2017). However, the landfill leachate generated from landfills, which contains high concentration of organics, ammonia, inorganic salts and heavy metals (Yong et al., 2018), becomes a serious threat to environment and poses a severe obstacle for efficient treatment (Chen et al., 2016). In terms of landfill age, landfill leachate can be classified as young landfill leachate (<5 years), middle-age landfill leachate (5–10 years), and mature landfill leachate (>10 years) (Brennan

et al., 2017). As the increasing of landfill age, mature landfill leachate generally contains the higher ammonia concentration (>1000 mg/L), the lower chemical oxygen demand (COD, <3000 mg/L), and the lower ratio of biochemical oxygen demand (BOD₅) to COD (<0.1) (Miao et al., 2014). These characteristics are significantly adverse to the removal of organic matter and ammonia nitrogen (NH₄⁺-N), and mature landfill leachate is thus extremely difficult to treat.

Biological treatment have been identified as one of the most favorable nitrogen removal techniques for mature landfill leachate, which involves of the nitrification in an aerobic reactor and the denitrification in an anoxic reactor (Miao et al., 2015). However, the low ratio of COD to TN (C/N, <3) strongly inhibits the denitrification process, due to the serious shortage of organic carbon source as an electron donor (Wu et al., 2015). Thus, the addition of external carbon source is often considered as necessary for the effective denitrification completion (Remmas et al., 2016). Traditionally, the easily biodegradable organics, such as methanol, ethanol (Sun et al., 2009), acetate, propionate, glucose (Miao et al., 2016), glycerine (Kulikowska and Bernat, 2013), sucrose, and meat peptone (Santos et al., 2016), were used as

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external carbon sources, however, the expensive cost restricts their viability in full-scale application. At present, various organic wastes have been investigated as cheaper alternative carbon sources for efficient denitrification, which is also benefit for the disposal of these organic wastes. Solid carbon sources, including fresh garden refuse, composted garden refuse (Plüg et al., 2015), pine bark (Frank et al., 2015), and straws of ornamental flowers (Chang et al., 2016), were proposed as one kind of slow-release alternatives, yet the slow denitrification rates were disappointed. Organic wastewaters, including bio-refractory organic (Wu et al., 2016), fermentation broth of plant (Fu et al., 2017), industrial organic wastewater (Fernández-Nava et al., 2010), mecellulose wasted liquor (Park et al., 2005), were regarded as another kind of sustainable and efficient alternatives; nevertheless, the application range is restricted since these wastewaters could only be generated from some special industry. Therefore, cost-effective and widely available alternative carbon sources still need to be identified to ensure the economic and efficient nitrogen removal from mature landfill leachate.

Acidogenic liquid fermented from activated sludge (Wang et al., 2016) or food waste (Jiang et al., 2013), which mainly contains volatile fatty acids (VFAs), ethanol, and soluble protein, is considered as promising and abundant carbon source for its direct role in bacterial metabolism. Recently, Guo et al. (2017) and Liu et al. (2017a) reported the improvement of biological nitrogen removal from municipal wastewater using an activated-sludge-derived carbon source. Although much effort has been made to improve the bioproduction of short-chain fatty acids from activated sludge (Li et al., 2011), the COD contents in activated-sludge-derived carbon sources (<5 g/L) were much less than those in food-waste-derived carbon sources (~80 g/L) (Zhang et al., 2016a). Thus, food waste, whose annual generation in China reached up to 96 million tonnes by 2013 (Clercq et al., 2017), is more promising for the production of acidogenic liquid. Kim et al. (2016) optimized both hydraulic retention time (HRT) and pH for the production of VFAs from food waste and examined their potential for denitrification. Zhang et al. (2016b) applied the food-waste-derived carbon source for treating both summer and winter sewage and verified its feasibility in denitrification process. Zhang et al. (2016c) and Tang et al. (2017) paid attention to the effect of food-waste-derived carbon source on the characteristics of nitrogen removal, microbial community, and membrane fouling. Kim et al. (2017) further clarified the relationship between bacterial populations and different carbon sources, based on the treatment of sewage wastewater in full-scale wastewater treatment plants (WWTP). However, there are few studies focusing on the feasibility of food-waste-derived carbon source for the treatment of mature landfill leachate, which contains much higher $\text{NH}_4^+\text{-N}$ concentration (>1000 mg/L) than sewage wastewater (~50 mg/L).

In this study, two kinds of acidogenic liquids were first fermented from oil-removed food waste and oil-added food waste to investigate the influence of oil on their biological properties, since Chinese food waste generally contains 15–35 g/L oil (Liu et al., 2017b). The real mature landfill leachate (C/N = ~1.5) were then nitrified and denitrified in a membrane bioreactor (MBR) with the assistant of the above acidogenic liquids as external carbon sources. The effect of C/N, temperature and HRT during the denitrification process was also investigated, focusing on the denitrification rate and the effluent quality indicators. Owing to the high biodegradable COD contents in acidogenic liquids, the fast denitrification rate, and superior effluent quality, this approach could be used for efficient nitrogen removal from mature landfill leachate as well as the high-value utilization of food waste.

2. Material and methods

2.1. Acidogenic liquids fermented from food waste

The oil-removed food waste was composed of 35% rice, 45% cabbage, 16% pork, and 4% tofu by weight, while additional 15 g/L oil (blended oil, Golden Dragon; 55% peanut, 22% sunflower, 12% linseed, 6% soybean, and 5% rapeseed) was added into the oil-added food waste to simulate the real Chinese food waste. Mesophilic anaerobic digested sludge was obtained from the anaerobic digester in Gaobeidian WWTP (Beijing, China). The fermentation was conducted following our previous study (Jiang et al., 2013), under the optimal conditions of pH = 6, temperature = 35 °C, volatile solids (VS) = 15% and HRT = 72 h. The acidogenic liquids were separated from the residues by centrifugation and filtration, which were designated as “A.L. a” and “A.L. b” for oil-removed food waste and oil-added food waste, respectively. Table 1 presented the characteristics of the resultant acidogenic liquids, which possessed different VFA compositions and different C/N of 156 and 51, respectively.

2.2. Mature landfill leachate and seed sludge

The mature landfill leachate was collected from the Asuwei MSW Sanitation Landfill (Beijing, China), which has been in operation since 1994. And it was pretreated by an anoxic/anaerobic up-flow anaerobic sludge bed (UASB) in the Sanitation Landfill and then was conserved in closed containers at 4 °C to prevent the natural degradation of organic. The major characteristics of the pretreated landfill leachate was as follows: COD = 3250 ± 100 mg/L, $\text{BOD}_5 = 100 \pm 20$ mg/L, $\text{NH}_4^+\text{-N} = 2120 \pm 100$ mg/L, nitrite nitrogen ($\text{NO}_2^-\text{-N}$) < 1 mg/L, nitrate nitrogen ($\text{NO}_3^-\text{-N}$) < 1 mg/L, and pH = 7.8–8.2. The active sludge for MBR was mixed of both aerobic sludge and anoxic sludge (ratio = 1:1), which were obtained from aerobic tank and anoxic tank in the Xiaojiahe WWTP (Beijing, China), respectively. After the mixed active sludge being inoculated, the activated sludge concentration in MBR was adjusted to 3500 ± 200 mgVSS/L.

2.3. MBR setup and operation

The lab-scale MBR (Fig. S1), with a working volume of 7 L, was operated for the nitrogen removal from mature landfill leachate. The membrane tank was equipped with a Polyvinylidene Fluoride (PVDF) hollow fiber membrane module (FP-AI, Motimo, Tianjin, China), with a surface area of 8 m² and a nominal pore size of 0.2 μm. The membrane module could be backwashed for particular needs using sodium hypochlorite (1000 mg/L) and citric acid (1%). The membrane tank was surrounded by water bath and could be controlled at a consistent temperature of 0–50 °C. The operational cycle (24 h) was in a multi-stage sequence, including 10-min filling of influent, 16-h aeration with stirring (100 rpm), 30-min settling, 10-min filling of external carbon source, 6.5-h anoxic with stirring (100 rpm), 30-min settling, and 10-min decanting. During the aeration stage, the dissolved oxygen (DO) concentration was maintained at 3–4 mg/L by adjusting the gas flow and was online monitored by a DO meter (InoLab 740, WTW, Germany). Excess sludge was discharged daily from the membrane tank to maintain the solids retention time (SRT) at 25 ± 5 days in the whole experimental period.

In each operational cycle, only 1-L supernatant was decanted after settling and equivalent volume (1 L) mature of landfill leachate was fed to the MBR, and thus the real HRT for MBR became 7 d. The total volume of sludge and treated water were always maintained at 7 L, except for the negligible volume (<30 mL) of

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