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A novel partial-denitrification strategy for post-anammox to effectively remove nitrogen from landfill leachate

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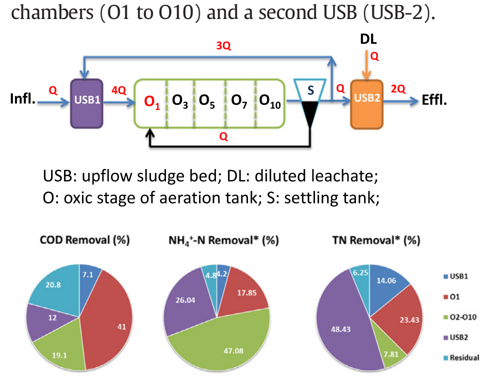
HIGHLIGHTS

- A partial nitrification-anammox and partial-denitrification-anammox (NADA) was developed to treat landfill leachate.
- NADA was efficient and economical to remove TN in leachate with 95% of removal rate.
- Diluted leachate was used as a carbon source and to convert excess NO_3^-–N to NO_2^-–N.
- A high population of anammox bacteria was enriched, accounting for 9.66% of the total bacteria.

GRAPHICAL ABSTRACT

The process consists of an upflow sludge bed reactor (USB-1), a multistage aeration tank with 10 separated chambers (O1 to O10) and a second USB (USB-2).



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ABSTRACT

Anaerobic ammonia oxidation (anammox) has shown great promise for nitrogen removal in low C/N wastewater such as landfill leachate. However, 11% of NO_3^--N is stoichiometrically produced, which decreases the total nitrogen (TN) removal efficiency. In this study, a strategy for post anammox was developed to effectively remove TN from leachate. A tandem conversion of NO_3^--N to NO_2^--N (partial denitrification) was accomplished by supplying substrate for subsequent anammox, by supplementing the electron donor deficient condition. This process greatly improved NH_4^+-N and TN removal from leachate, reaching a 95% efficiency. Quantitative real-time polymerase chain reaction results showed that a high abundance of anammox bacteria, with a titer of 10^9 copy numbers/L, was enriched, accounting for 9.66% of the total bacterial community, and indicating the success of this novel strategy for the TN removal in leachate.

1. Introduction

Leachate, generated principally by precipitation percolating through waste deposited in a landfill, is a notorious hazardous material. It contains large amounts of dissolved organic matter, inorganic macro components, including sulfate, chloride, and ammonia, heavy metals, and

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toxic xenobiotic organic compounds (Kjeldsen et al., 2002). If not properly treated, leachate causes severe environmental damage. The properties of leachate make it extremely difficult to treat, and biological treatment is currently the most commonly used technique (Renou et al., 2008). However, there are obstacles to total nitrogen (TN) removal because of the low chemical oxygen demand (COD)/N ratio for leachate. Conventional nitrification/denitrification processes normally require the addition of large amounts of external carbon sources to reach satisfactory TN removal efficiency, which diminishes the economic merit of the biological approach (Miao et al., 2015; Liang and Liu, 2008).

Anaerobic ammonium oxidation (anammox) has gained attention as a novel self-sustaining biological nitrogen removal process (Mulder et al., 1995; Kartal et al., 2010; Hu et al., 2013; Azari et al., 2017). This process is accomplished by the reactions of NO_2^- and NH_4^+ and does not require additional external carbon sources for denitrification. Compared with conventional nitrogen removal processes, partial nitrification-anammox offers advantages such as saving 62.5% of the aeration rate, large amounts of external carbon source, and low sludge production (Kartal et al., 2010). Previous studies have shown large nitrogen removal from leachate through partial nitrification-anammox processes (Miao et al., 2015; Wu et al., 2015; Wang et al., 2010). Although promising, single-stage anammox cannot obtain satisfactory TN removal, because around 11% of NO_3^--N is produced stoichiometrically as the main by-product (Zhang et al., 2017), sometimes causing the effluent TN to exceed stringent standards. Therefore, NO_3^--N removal after anammox is required, meaning that additional carbon source amendment is necessary, which is not ideal.

During heterotrophic denitrification, denitrifying bacteria carrying diverse denitrifying functional genes specific for nitrate, nitrite, nitric oxide and nitrous oxide reduction are present in activated sludge. Recently, to save the carbon source necessary for denitrification, partial denitrification of NO_3^--N to accumulate NO_2^--N has been advised for biological nitrogen removal processes. The applicability of this process is summarized as follows: (1) NO_2^--N can be autotrophically removed through anammox, thus saving the carbon source for complete heterotrophic denitrification; (2) Since denitrifying bacteria prefer NO_3^--N rather than NO_2^--N as an electron acceptor, the specific reduction rate of nitrate is much higher than that of nitrite (Du et al., 2016), which often results in nitrite accumulation during denitrification; and (3) Compared with partial nitritation of ammonia to nitrite, partial denitrification of nitrate is easier to control. Furthermore, ammonia-oxidizing bacteria (AOB) and nitrite-oxidizing bacteria (NOB) share similar ecological conditions. Suppressing the growth of NOB to selectively enrich AOB in one system is relatively difficult to control and needs stringent environmental conditions, and sludge age control. However, partial denitrification for NO_2^--N accumulation is easily obtained by regulating the appropriate COD/N ratio and the availability of the carbon source (Gong et al., 2013). It has been reported that when the ratio of readily biodegradable chemical oxygen demand (COD) to nitrate was <3.5, high nitrite accumulation could be obtained when the exogenous carbon source was depleted (Gong et al., 2013).

Previous studies have demonstrated successful partial denitrification in a batch system (Chen et al., 2009), but information on the continuous mode system remains scarce. In practice, partial denitrification in SBR is more easily controlled by step feeding of a carbon source to create electron donor deficient conditions. However, in continuous flow, except for providing carbon source deficient conditions, the loading of the carbon source, the hydraulic retention time (HRT), and the influent concentration of the feed should be considered, according to the NO_3^--N concentration. Only when the proper carbon source loading fits the needs, can optimal partial denitrification and subsequent anammox be realized. This study investigated solving the problem of anammox-induced nitrate accumulation through partial-denitrification, followed by anammox processes to improve TN removal efficiency in a continuous and complete mixed reactor. The entire process was divided into three steps in a three-sludge system, which consisted of an upflow sludge blanket (USB-1), a multi-stage aeration, and a second USB (USB-2). Firstly, raw influent (mixture of leachate and sewage), and reflux effluent from the sedimentation tank, entered the USB-1 reactor for organics removal and heterotrophic denitrification. Subsequently, the effluent from the USB-1 reactor entered the oxygen-limited aeration tank for further organics and partial ammonia oxidation into nitrite. The effluent from the settling tank of the second stage was finally injected into the USB-2 reactor for subsequent autotrophic denitrification through anammox. To minimize NO_3^--N produced during anammox, a small amount of diluted leachate was injected into the USB-2 reactor to bring about limited electron donor for partial denitrification of NO_3^--N, which produces the feed NO_2^--N for further anammox. This partial nitritation and anammox coupled with partial denitrification and anammox, provides a reliable and economic strategy for leachate treatment with lower carbon requirements.

2. Materials and methods

2.1. Characteristics of leachate

Landfill leachate was obtained from a mature landfill site (10 years old) in Beijing, China. Table 1 lists the characteristics of the leachate. The COD of the mature landfill leachate was 4000–5000 mg/L and the NH_4^+-N concentration was 1200–1300 mg/L, with a COD/N ratio of 3–4. In order to decrease the toxicity of high organic pollutants and increase biodegradability (Brennan et al., 2016), the leachate was combined with municipal sewage as influent in this study. After the leachate was diluted with municipal sewage at a ratio of 1:2, the mixed liquor had COD and NH_4^+-N concentrations of 1400–2000 and 460–500 mg/L, respectively, with a COD/N ratio of 3–4.

2.2. Experimental setup of the three-sludge A/O/A process

In this study, leachate diluted with municipal sewage was treated with a three-sludge A/O/A system for organics and NH_4^+-N removal, as shown in Fig. 1. A/O/A consists of an upflow sludge bed reactor (USB-1), a multi-stage aeration tank, and a second USB reactor (USB-2), with effective volumes of 8.25, 15 and 4.25 L respectively. The multi-stage aeration tank consisted of 10 identical compartments with the same volume. All the reactors were made of polymethyl methacrylate. Anaerobic sludge of the USB1 and USB2 was taken from a UASB reactor of the Beijing Drainage Group's research and development center, which anammox bacteria were 10^4 copy numbers/L of hzsB gene. The sludge of A/O was taken from the Liulitun landfill oxidation ditch in Beijing. The anammox bacteria of this sludge were low due to the high dissolved oxygen (DO) concentration. The USB reactors were controlled between 35 and 38 °C with a water bath, and the oxic stage was controlled between 28 and 35 °C using a heating rod. In the oxic stage, the first chamber was set with a mechanical stirrer, while each of the nine chambers was equipped with an aerator. DO was monitored and controlled between 0.3 and 1.5 mg/L. The mixed liquor suspended solid (MLSS) concentration was maintained at 3000–4000 mg/L. The mixed liquor volatile suspended solid (MLVSS)

Table 1
Characteristics of raw and sewage diluted leachate.

Parameters	Concentration[a] (mg/L)	Concentration[b] (mg/L)
COD	4000–5000	1400–2000
NH_4^+-N	1000–1300	460–500
TP	8–10	5–8
NO_x^--N	0.5–15	0.5–10
TN	1400–1600	480–520
pH	7.2–8.3	7–8

[a] Concentrations in raw leachate.
[b] Concentrations in sewage diluted leachate.