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Achieving nitritation by treating sludge with free nitrous acid: The effect of starvation

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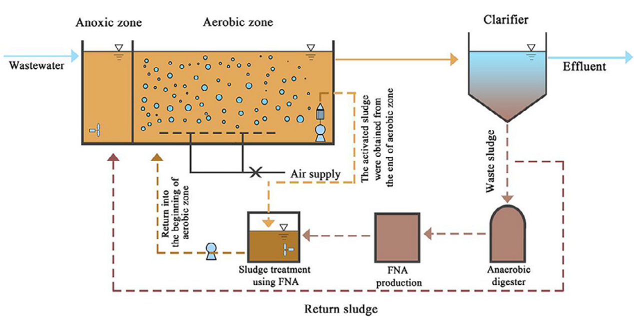
GRAPHICAL ABSTRACT



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ABSTRACT

Side-stream sludge treatment using free nitrous acid (FNA) is a novel strategy to achieve nitritation in mainstream of wastewater treatment plants (WWTPs). To optimize nitritation, the effect of starvation on this strategy was investigated in this study. The results showed that pre-starvation, which is the starvation before FNA treatment, enhanced the resistance of sludge to FNA. This led to a decrease in the nitrite accumulation rate (NAR), which dropped from 70% to 27% after aerobic pre-starvation. This was further confirmed in the FNA treatment using the sludge collected from the secondary settling tank (anoxic pre-starvation) and the aerobic zone (without starvation) of an Anaerobic-Anoxic-Oxic system. The post-starvation, which was the starvation after FNA treatment, decreased NAR from 63% to 14%. To obtain a higher NAR, the sludge used for FNA treatment should be collected from aerobic zone, and be returned to aerobic zone after treatment to avoid pre-starvation and post-starvation.

1. Introduction

Nitrogen removal from wastewater is critical to prevent eutrophication in receiving waters. The conventional biological nitrogen removal process includes of nitrification ($NH_4^+ \rightarrow NO_2^- \rightarrow NO_3^-$) and denitrification ($NO_3^- \rightarrow NO_2^- \rightarrow N_2$). Nitrification requires oxygen, which consumes energy. Denitrification requires organic matter, leading to a reduction in organic matter used to produce methane. This increases operating costs for carbon source dosing. One promising technology to reduce oxygen and organic matter demand is the nitritation-anammox process. Approximately 50% of ammonium is oxidized to nitrite instead of nitrate ($NH_4^+ \rightarrow NO_2^-$, i.e., nitritation); the

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remaining ammonium is oxidized to nitrogen gas using nitrite as electron acceptor ($NH_4^+ + NO_2^- \rightarrow N_2$, i.e., anammox). Nitritation-anammox process can reduce oxygen requirement by 60% and decrease organic matter demand by 100%. If this technology could be applied in the mainstream of wastewater treatment plants (WWTPs), WWTPs could become energy-neutral or even energy-positive (Ma et al., 2016).

Achieving nitritation is the basis for applying nitritation-anammox in mainstream treatment. The key to achieving nitritation is to create conditions under which nitrite oxidizing bacteria (NOB; $NO_2^- \rightarrow NO_3^-$) are eliminated from the system, while ammonia oxidizing bacteria (AOB; $NH_4^+ \rightarrow NO_2^-$) are retained. NOB and AOB are both inhibited by free nitrous acid (FNA) during aerobic nitrification. However, AOB tolerate FNA more than NOB. This results in a higher ammonium oxidation rate than the nitrite oxidation rate under high FNA conditions (Ma et al., 2017; Tan et al., 2008; Vadivelu et al., 2006a; Vadivelu et al., 2006b). For example, one study found that 0.023 mg HNO_2-N/L completely inhibited NOB (Vadivelu et al., 2006b), whereas 0.40–2.81 mg HNO_2-N/L reduced AOB activity by only 50% (Tan et al., 2008; Vadivelu et al., 2006a) . This inhibition has been used to achieve nitritation in N-rich wastewater treatment systems, including treating anaerobic digester effluent and landfill leachates (Kim et al., 2012; Sun et al., 2013).

However, in the mainstream of WWTPs, it is difficult to use FNA inhibition on NOB to achieve nitritation. This is because the ammonium concentration in sewage is typically lower than 60 mg/L. This means there is a lower cumulative nitrite concentration which could seldom reach the threshold FNA concentration for inhibiting NOB. Recently, Wang et al. (2014) found that FNA is more biocidal to NOB than to AOB under anoxic condition. After being exposed to 0.24 mg HNO_2-N/L for 24 h, NOB activity completely ceased; however, AOB activity continued at approximately 70% of the original value (Wang et al., 2014). Based on this finding, Wang et al. (2014) proposed a novel strategy to achieve nitritation in the mainstream treatment, which involved a side-stream sludge treatment using FNA. A nitrite accumulation ratio (NAR) above 80% was obtained in the mainstream treatment reactor (Wang et al., 2014).

Importantly, FNA is a green and renewable biocidal agent, because it can be produced on site from anaerobic digestion liquor by nitritation in a WWTP, and can be rapidly removed through denitrification (Law et al., 2015). Therefore, achieving nitritation by treating sludge with FNA is economically attractive and a promising technology. The FNA treatment conditions have been optimized to improve the stability of nitritation. These treatment conditions include the FNA concentration, FNA treatment frequencies, FNA treatment time, and oxygen control (Duan et al., 2018; Ma et al., 2017; Wang et al., 2016a). However, the effect of starvation on this strategy remains unclear, although it is widespread in WWTPs.

AOB and NOB are under starvation at the anaerobic, anoxic or settling stage of the mainstream of WWTPs, because they are aerobic bacteria and have no oxygen or other substrate to use. Under these starvation conditions, AOB and NOB can initiate programmed cell death and adjust their metabolic processes to reduce their maintenance energy requirement. This decreases the specific bacteria activity (Elawwad et al., 2013; Liu et al., 2017a; Liu et al., 2017b; Salem et al., 2006). Du et al. (2015) found that starved cells had a higher tolerance than normal cells during disinfection. The E. coli cells in a nutrient-limited environment develop a resistance to different stress conditions (Matin, 1991). Therefore, if sludge undergoes a starvation phase before FNA treatment (pre-starvation, e.g., collecting sludge from secondary settling tank), does it develop a tolerance to FNA? If sludge treated by FNA undergoes starvation (post-starvation, e.g., returning sludge treated by FNA to anaerobic zone), does NAR decrease? This knowledge is important for achieving stable nitritation in the mainstream of WWTPs by treating sludge with FNA.

In this context, this study investigated the effect of starvation on sludge treatment using FNA, to achieve nitritation in the mainstream. To study the pre-starvation effect, the sludge first underwent anaerobic, anoxic and aerobic starvation. It was then treated by FNA and evaluated in term of AOB and NOB activities. To confirm the research discussed above, the sludge samples used for FNA treatment were collected from the aerobic zone (without starvation) and the secondary settling tank (under anoxic pre-starvation) of an Anaerobic-Anoxic-Oxic (A^2O) biological nitrogen and phosphorus removal system. Additionally, to study the effect of post-starvation, sludge was first treated by FNA and then underwent a starvation phase. It was evaluated in term of AOB and NOB activities. Finally, the paper discusses the applications of this finding for WWTPs.

2. Materials and methods

2.1. Sludge treatment batch tests using FNA

Batch tests were conducted to evaluate the effect of starvation on sludge treatment using FNA. Before the sludge treatment, the activated sludge was washed three times with deionized water using a centrifuge (r = 4000 rev/min, t = 10 min) to remove organic matter, which controlled nitrite reduction during the sludge treatment using FNA. The mixed liquid volatile suspended solids (MLVSS) concentration was adjusted to 4000 mg/L. The batch tests were conducted in reactors with a working volume of 0.5 L. The reactor was sparged with nitrogen gas to remove any residual oxygen, and sealed with a rubber stopper to maintain anoxic condition. These reactors were placed in a digital circulating water bath with a steady temperature of 22 ± 0.5 °C under strict anoxic condition for 6 h at a pH of 6.0 ± 0.5 (Ma et al., 2017). The pH value was adjusted by adding 0.1 mol/L HCl or 0.1 mol/L NaOH. The FNA concentration was calculated based on the nitrite concentration, pH, and temperature (Anthonisen et al., 1976).

To measure the AOB and NOB activities after treating sludge with FNA, the sludge was washed three times to remove the nitrite. No nitrite was detected after washing. After that, the ammonia (30 g·L^{-1} NH_4^+-N) and nitrite (30 g·L^{-1} NO_2^--N) stock solution were added into the batch reactor to achieve initial NH_4^+-N and NO_2^--N concentrations of 30 mg·L^{-1} and 15 mg·L^{-1}, respectively. Then, the sludge was aerated for 3 h and the DO was controlled at above 5.0 mg/L (Wang et al., 2014). The batch reactor was operated in a temperature-controlled room at a temperature of 22.0 ± 0.5 °C. Mixed-liquor samples were collected at 30 min intervals to measure NH_4^+-N, NO_2^--N and NO_3^--N concentrations. When the batch test was finished, a mixed-liquor sample was collected to analyze the MLVSS concentration. The concentration degradation rates of ammonia and nitrite were characterized as $CC_{NH_4^+}$ and $CC_{NO_2^-}$ (Qian et al., 2017). The ammonia and nitrite degradation curve fitted well with the experimental data with a good linear relationship (R^2 > 0.9). The AOB and NOB activities were represented by specific ammonia ($r_{NH_4^+}$) and nitrite oxidation rate ($r_{NO_2^-}$) respectively. $r_{NH+\ 4}$ and $r_{NO-\ 2}$ were calculated by dividing the corresponding degradation rate by the MLVSS concentration. The ammonia oxidized by AOB was only from influent water, while the nitrite oxidized by NOB was from both influent water and the ammonia oxidation process. Thus, above relationship can be derived in following Eqs. (1) and (2). The nitrite accumulation rate (NAR) in the reactor was calculated using Eq. (3).

$$r_{NH_4^+} = CC_{NH_4^+}/MLVSS \tag{1}$$

$$r_{NO_2^-} = (CC_{NH_4^+} + CC_{NO_2^-})/MLVSS \tag{2}$$

$$\mathrm{NAR} = (r_{NH_4^+} - r_{NO_2^-})/\text{-}\ r^{+}_{NH_4^+} \times 100\% \tag{3}$$

2.2. Effect of pre-starvation on sludge treatment using FNA

The activated sludge was obtained from Gaobeidian WWTP in Beijing, China, and was divided into four segments (Fig. 1A). The first