N2O EMISSION FROM A NITRITATION DOUBLE-SLUDGE SYSTEM TREATING WASTEWATER WITH A HIGH CONCENTRATION OF AMMONIUM

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ABSTRACT

Wastewaters such as landfill leachate and anaerobically digester effluent are usually contain high concentrations of ammonium (NH4-N) and low concentrations of biodegradable organic matters. A high efficient nitrite (NO2-N) based double-sludge system combining sequential denitrification and nitritation reactors has been developed to remove nitrogen from such types of wastewater. Denitrification process could fully utilize the limited low carbon source, and nitrite accumulation was achieved in the nitritation reactor. By this means, enhanced biological nitrogen removal could be realized. However, nitrous oxide (N2O), a potent greenhouse gas, can be produced and emitted during biological nitrogen removal. In this study, nitrite accumulation was achieved under low dissolved oxygen (DO) concentrations, high free ammonia (FA) concentrations and high NH4-N/DO ratios in the nitritation reactor. The nitritation efficiency (the ratio of NO2-N to the oxidized nitrogen, NOX-N) reached to 99.8% at the ammonium loading rate of 1.47 g/(L·d). N2O emission was measured under different aeration rates (0.24 L/min, 0.36 L/min and 0.48 L/min, respectively). With increasing the aeration rate, ammonium oxidizing rate increased, indicating that the activity of ammonia oxidizing bacteria was enhanced with increasing the aeration rate. However, N2O emission decreased with increasing the aeration rate. In addition, the effect of initial NO2-N concentrations on N2O emission under different aeration rates (0.24 L/min, 0.36 L/min and 0.48 L/min, respectively) was also examined. N2O emission was high with the addition of NO2-N and a high initial NO2-N concentrations led to a high emission of N2O. The aeration rate had a high effect on N2O emission under high initial NO2-N concentrations, with the N2O emission factor ranged from 0.61% to 1.82%. While the effect of aeration rate on N2O emission was not obvious without the addition of NO2-N, with the emission factor of around 0.24%.

Keywords: High Ammonium Wastewater, Nitrous Oxide, Nitritation, Aeration Rate, Nitrite.

1. Introduction

Wastewaters such as landfill leachate and anaerobically digester effluent are usually contain high concentrations of ammonium (NH4-N) and low concentrations of biodegradable organic matters. A high efficient nitrite (NO2-N) based double-sludge system combining sequential denitrification and nitritation reactors has been developed to remove nitrogen from such types of wastewater (Wu et al., 2014). In this process, ammonia oxidizing bacteria oxidize NH4-N to NO2-N by the alternative anoxic and aerobic operating mode and activities of nitrite oxidizing bacteria (NOB) are inhibited. However, for this NO2-N based system, nitrous oxide (N2O) can be produced and emitted both in denitrification and nitrification process during biological nitrogen removal, with the main contribution from the nitrification process. Ammonia oxidizing bacteria (AOB) have been identified as the major contributor to N2O production during nitrification (Ahn, 2010; Law, 2012; Gong et al., 2012), with three possible pathways of nitrifier denitrification, and biological and chemical oxidation of NH2OH.

Nitrite accumulation can be achieved in the nitritation reactor, but nitrite may induce a high N2O production. Weissenbacher et al. (2011) discovered that NO2-N was the main factor affecting N2O emission in the DEMON process. Xiao et al. (2013) demonstrated that N2O emission through AOB denitrification had a positive correlation with the NO2-N concentration, and the N2O

emission factor ranged from 0.41% to 7.25%. Law et al. (2013) found that N2O production reached the highest level when the NO2-N concentration was less than 50 mg N/L, decreased with the increasing NO2-N concentrations from 50 mg N/L to 500 mg N/L, and then decreased to the least around 0.20 mg/h/g VSS with the nitrite concentration ranged from 500 mg N/L to 1000 mg N/L.

In this study, nitrite accumulation was achieved in a double-sludge system, and the factors affecting nitrite accumulation and N2O production were furthered analyzed.

2. Materials and methods

2.1. System operation

Two lab-scale sequencing batch reactors (SBRs), one for denitrification and the other for nitrification, each with volume of 6 L, were operated in the laboratory at 27oC. The two reactors were seeded with sludge acclimated in the laboratory, and the operation cycles were both 4 h. One cycle in the denitrifying reactor consisted 180 min anoxic stirring, 20 min aeration, 30 min settling, and 10 min decanting and idle periods. During each cycle, 3 L of mixed wastewater (2 L synthetic wastewater and 1 L recycled nitrified wastewater from the nitrification reactor) was fed to the reactor, and 3 L was discharged, resulting in a hydraulic retention time (HRT) of 8 h. While one cycle in the nitrifying reactor consisted of 80 min aeration-I, 40 min anoxic stirring, 80 min aeration-II, 30 min settlement, and 10 min decanting and idle periods, and the HRT was also 8 h. The effluent from the denitrifying reactor was fed to the nitrifying reactor. The solids retention time (SRT) of the denitrifying reactor was kept at 20 days, and sludge was not wasted in the nitrifying reactor under steady state. In each cycle, 2 L of synthetic wastewater was fed to the denitrifying and nitrifying reactors, and 2 L of treated wastewater was decanted. Therefore, the HRT of the combined system was 1 d. The synthetic wastewater comprised: 3.06 g/L of NH4Cl (0.8 g/L NH4+-N), 1.00 g/L of NaAc, 7.00 g/L of NaHCO3, 0.10 g/L of Na2HPO4, 0.056 g/L of CaCl2, 0.36 g/L of MgSO4, 0.01 g/L of yeast extract and 0.4 mL/L of a trace element stock solution.

2.2. Batch experiments

Batch experiments were carried out to study the effect of different aeration rates and initial NO2-N concentrations on N2O emission for activated sludge acclimated in the nitrifying reactor.

For the effect of different aeration rates on N2O emission at a high initial NO2-N concentrations, 1.2 L mixed liquor was taken from the nitrifying reactor after filling and divided into three batch reactors. The mixed liquor was then aerated under the flow rate of 0.24, 0.36 and 0.48 L/min, respectively. The effect of different aeration rates on N2O emission was also examined without a high initial NO2-N concentration. 1.2 L mixed liquor was taken from the nitrifying reactor before the end of the aeration phase. After centrifugation, supernatant was discarded and the residual sludge was re-suspended with synthetic wastewater but without the organic carbon and the initial NH4-N concentration was controlled at around 165 mg/L. The mixed liquor was then divided into three batch reactors and aerated at the flow rate of 0.24, 0.36 and 0.48 L/min, respectively. During all batch experiments, samples (both liquid and gas samples) were taken at intervals of 10 min, and simultaneously DO and pH were measured. Mixed liquor sample was also taken from the batch reactor to determine the mixed liquor suspended solids (MLSS) and its volatile fraction (MLVSS) concentrations.

2.3. Analytical methods

NH4-N,NO2-N and nitrate nitrogen (NO3-N) were analyzed according to standard methods. DO and pH were determined by a DO meter (oxi 315i, WTW, Germany) and a pH meter (pH3110, WTW, Germany), respectively. FA and free nitrous acid (FNA) were calculated according to Anthonisen et al. (1976). N2O concentration in gas phase was measured with a gas chromatograph (Agilent 6820, Agilent Technologies, USA). To compared conveniently, the produced N2O in gas phase was represented as mg/L, which was mg N2O (gas) produced from the unit volume (litre) of mixed activated sludge liquor.

3. Results and discussion

The denitrifying reactor reached to steady state after operation for 46 d. The average concentrations of NH4-N, NO2-N and NO3-N in effluent were 489.8 mg/L, 2.2 mg/L and 0.4 mg/L, respectively. Figure 1 shows that the dynamics of pH, DO, NH4-N and NO2-N concentrations during a typical cycle under steady state in the nitrifying reactor. During the first aeration period, DO concentration was around 1.0 mg/L, and the nitritation efficiency kept at high steady state, indicating that nitritation could be achieved stably under a low DO condition.

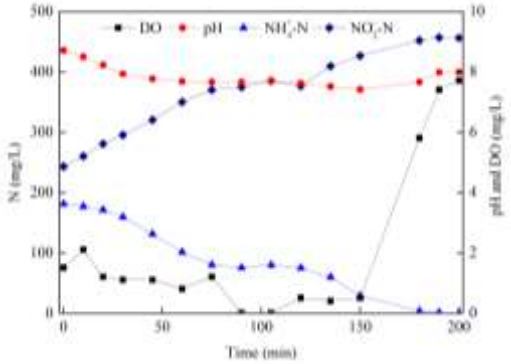

Figure 1. Dynamics of parameters during a typical SBR cycle.

Figure 2 show N2O emission with a high initial NO2-N concentrations under the aeration rates of 0.24 L/min, 0.36 L/min and 0.48 L/min, respectively. NH4-N were mostly oxidized to NO2-N, and the rate of NH4-N oxidizing increased from 10.42 mg/g·h to 16.35 mg/g·h with the increasing of aeration rates. While the NO3-N production rate kept low. It indicated that the activity of AOB could be enhanced by increasing aeration rates. N2O production rate and the N2O emission factor were both decreased with increasing of aeration rates, which was likely due to that AOB denitrification pathway was inhibited with increasing of aeration rates.

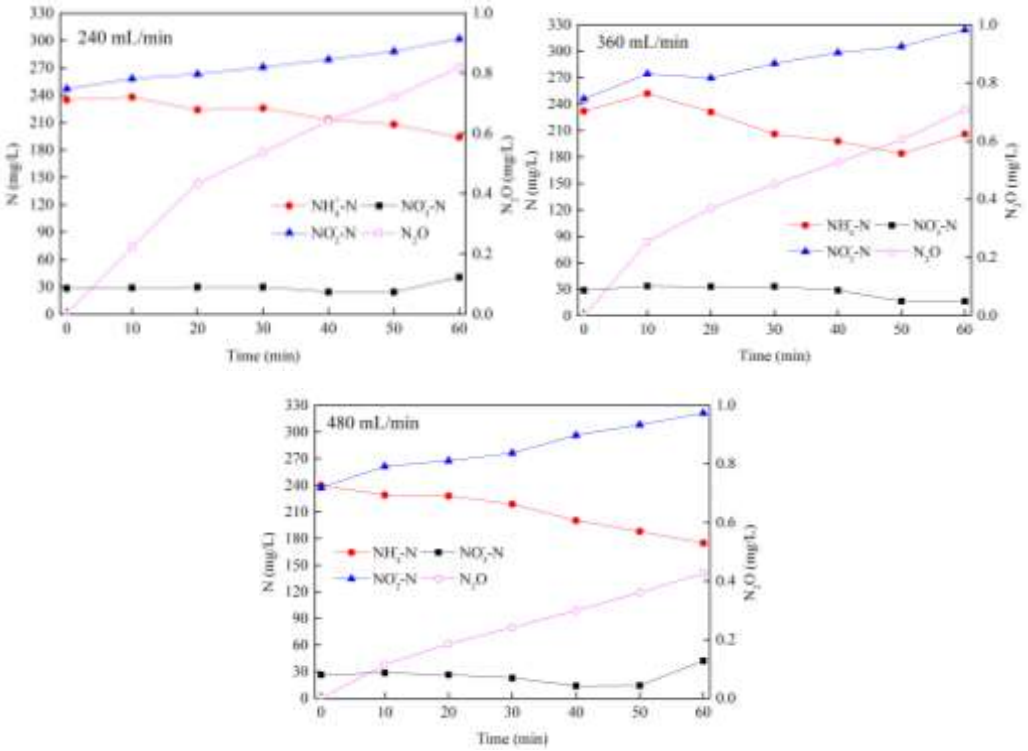

Figure 2. Dynamics of NH4-N, NO2-N, NO3-N concentrations and N2O emission under different aeration rates with different initial NO2-N concentrations during batch nitrification.

Both the N2O production rate and the N2O emission factor at high initial NO2-N concentrations were higher than those at a low initial NO2-N concentration at the same aeration rate, indicating that high initial NO2-N concentrations might lead to a high N2O emission. The reason could be due to that high NO2-N concentrations might stimulate AOB denitrification pathway by promoting the expression of the nirK gene.

4. Conclusions

In double-sludge system, a high ammonium loading rate and nitritation was achieved by optimizing the operation mode and reaction cycle. The nitritation efficiency reached to 99.8% at the ammonium loading rate of 1.47 g/(L·d). Low DO concentrations, high FA concentrations and high NH4-N/DO ratios are main factors contributing to nitrite accumulation. The aeration rate had a high effect on N2O emission under high initial NO2-N concentrations, and the N2O emission factor decreased with the increasing of aeration rates, ranging from 0.61% to 1.82%. While the effect of aeration rate on N2O emission was not obvious without the addition of NO2-N, with the emission factor of around 0.24%.

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