Desalination and Water Treatment
www.deswater.com

doi: 10.1080/19443994.2016.1187089

57 (2016) 28743–28748
December

Taylor & Francis
Taylor & Francis Group

Effects of temperature on N_2O production in the process of nitrogen removal by micro-expansion aerobic granular sludge

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Received 30 January 2016; Accepted 3 May 2016

ABSTRACT

An SBR reactor was used to study the effect of temperature on N_2O production in the process of nitrogen removal by micro-expansion aerobic granular sludge, as well as the removal efficiency of COD and ammonia nitrogen. The experiments were conducted at 21 ± 0.5°C, 26 ± 0.5°C, and 31 ± 0.5°C with low dissolved oxygen. The results showed that simultaneous nitrification and denitrification occurred in this system. The temperature had a significant influence on N_2O production and N_2O release rate; both increased with temperature going up. The ratios of N_2O production to the influent nitrogen loading rate at 31 ± 0.5°C, 26 ± 0.5°C, and 21 ± 0.5°C were 8.07, 5.37, and 3.93%, respectively, which are higher than aerobic granular sludge. N_2O release was 0.96 mg/m^3 at 31 ± 0.5°C, which was 1.43 and 1.37 times the production at 21 ± 0.5°C and 26 ± 0.5°C, respectively, and dissolved N_2O production was 1.46 ± 0.05 mg/m^3 at 31 ± 0.5°C, which was 2.86 and 1.60 times the production at 21 ± 0.5°C and 26 ± 0.5°C, respectively. As a result, 26°C is recommended as a suitable temperature for reducing N_2O emission and maintaining nitrogen removal efficiency in wastewater treatment.

Keywords: Aerobic granular sludge; N_2O emission; Simultaneously nitrification and denitrification (SND); Biological nitrogen removal; Wastewater treatment

1. Introduction

Nitrous oxide (N_2O) is a greenhouse gas and participates in the destruction of stratospheric ozone [1]. Atmospheric N_2O concentrations increased from 270 μg/kg in 1,750 to 314 μg/kg in 1998 [2], and it is very stable in the atmosphere, with a lifetime of more than 120 years. Currently, N_2O accounts for 5% of the total global production of anthropogenic greenhouse gases (GHG) [3]. One of the most important sources of N_2O is denitrification, and the processes of nitrification and denitrification in traditional wastewater treatment are usually accompanied by N_2O emission to the atmosphere [4]. Studies on N_2O emission from the wastewater treatment process have shown that the fraction of nitrogen that is emitted as N_2O at the lab scale is 0–95% of the nitrogen load, and this fraction is 0–14.6% in full-scale studies [5].

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Many studies have examined the factors that affect N_2O production in the process of biological nitrogen removal, such as dissolved oxygen (DO) [6], nitrogen load [7], C/N [8], and aeration rate [9]. Temperature is another factor with an important influence on nitrogen removal efficiency and N_2O production, where the temperature varies greatly with season. However, little information is available about the effect of temperature on N_2O production by micro-expansion aerobic granular sludge, which has the same nitrogen removal effect as granular sludge but lower energy consumption [10]. Research on N_2O production from micro-expansion aerobic granular sludge systems is necessary and meaningful to the economic operation of practical engineering.

The purpose of this study was to investigate the effects of temperature on the performance and N_2O production of biological nitrogen removal process in an SBR reactor with micro-expansion aerobic granular sludge. The study will provide a new insight into the release of N_2O during biological nitrogen removal process.

2. Materials and methods

2.1. SBR design and operation

The study was conducted with a laboratory-scale SBR made of plexiglass, with a working volume of 3.2 L [11]. The seeding sludge was a micro-expansion aerobic granular sludge with 3 mm in diameter, and the SBR system was fed with synthetic wastewater containing 400 mg/L COD and 30 mg/L NH_4^+-N. The composition of the synthetic wastewater included 1.32 g/L $C_6H_{12}O_6{\cdot}H_2O$, 0.37 g/L NH_4Cl, and 1 mL/L nutrient solution. One liter of nutrient solution contained: 1.5 g $FeCl_3{\cdot}6H_2O$; 0.15 g H_3BO_3; 0.03 g $CuSO_4{\cdot}5H_2O$; 0.18 g KI; 10 g EDTA; 0.06 g $MnSO_4{\cdot}H_2O$; 0.12 g $ZnSO_4{\cdot}7H_2O$; 0.18 g $CoSO_4{\cdot}7H_2O$. The mixed liquor suspended solids (MLSS) were maintained at 3,000 mg/L. The DO was maintained below 1.0 mg/L. The reactor was run in 1 min feeding, 180 min aeration, 30 min settling, and 20 min decant. The temperature was controlled by a heater. During the study, the reactor operated stably for 25 d each at 21 ± 0.5℃, 26 ± 0.5℃, and 31 ± 0.5℃.

2.2. Analytical methods

COD, NH_4^+-N, NO_2^--N, and NO_3^--N, MLSS were measured according to the national standard methods [12]. DO, pH, oxidation–reduction potential, and temperature were measured using a WTW Handheld Multi-parameter Instrument (Multi340i, Germany).

The value of N_2O production during the biological nitrogen removal process included gaseous N_2O and dissolved N_2O. The concentration of N_2O was determined using a gas chromatograph (GC) equipped with an electron-capture detector (GC-14B, Shimadzu, Japan) at 345℃, which was controlled by a Chromatpac (CR-7A, Shimadzu, Japan) and a program controller (PRG 12A, Shimadzu, Japan). The columns were packed with Porapac Q. The carrier gas was 5% methane in argon. Dissolved N_2O was sampled according to the following steps: 20 mL micro-expansion aerobic granular sludge mixture was drawn from SBR reactor by a 50 mL glass syringe, then air and 1 mL H_2SO_4 with concentration of 1 mol/L were successively drawn using the syringe until the full-scale range was reached (acid was added to inhibit microbial activity). The mixture was mixed and stood for 1 h at room temperature, and the gas was collected in the syringe for dissolved N_2O detection by gas chromatograph [13]. N_2O production analysis was performed in triplicate, and the average value was calculated.

2.3. Data analysis

To further evaluate the effect of temperature on the N_2O production during the process of nitrogen removal by micro-expansion aerobic granular sludge, the nitrification rate and N_2O release rate were calculated at different temperatures according to the following formulas:

$$Ka = \frac{\Delta\left(NH_4^+\text{-N}\right)}{X_v \Delta t} \tag{1}$$

$$Re = \frac{\Delta C_{N_2O\text{-N(emission)}}}{\Delta t} \tag{2}$$

$$\Delta\rho\text{N/COD} = \frac{\Delta\left(NH_4^+\text{-N}\right)}{\Delta\text{COD}} \tag{3}$$

where Ka is the nitrification rate (mg/g min), Re is the N_2O release rate (mg/m^3 min), $\Delta(NH_4^+$-N) is the consumed ammonia nitrogen concentration (mg/L), $\Delta C_{N_2O\text{-N(emissions)}}$ is emitted N_2O (mg/m^3), X_v is the sludge concentration in the reactor (mg/L), and Δt is the ammonia oxidation time (min). $\Delta\rho$N/COD indicates the efficiency of carbon source utilization, ΔCOD is the consumed COD concentration (mg/L).

3. Results and discussion

3.1. Effect of temperature on SBR performance

During the entire operation, the performance of the SBR with the micro-expansion aerobic granular sludge was stable at three test temperatures (Fig. 1). The NO_3^--N and NO_2^--N concentrations in the effluent were all below 1 mg/L. Furthermore, the temperature had no significant impact on COD removal efficiency, and the removal exceeded 90% at all three test temperature. COD removal is mainly completed by heterotrophic microbes, which are abundant in microbial communities and have bioactivity over a wide range of temperature.

Ammonia nitrogen removal efficiency increased with the increase in temperature (Fig. 1). At 21 ± 0.5°C, the ammonia nitrogen removal efficiency was 42.97 ± 6.29% as well as the ammonia nitrogen concentration in effluent was 16.6 mg/L and when the temperature increased to 26 ± 0.5°C and 31 ± 0.5°C, the ammonia nitrogen removal efficiencies raised to 50.14 ± 6.95% and 68.29 ± 2.61%, respectively.

3.2. Effect of temperature on N_2O production

3.2.1. N_2O production

N_2O emissions did not differ notably at constant temperature during the operation of the SBR which means the SBR system operated under a stable state (Fig. 2(a)). The N_2O generation rate increased with temperature going up, which led to an increase in N_2O emissions (0.67 ± 0.004 mg/m^3 at 21 ± 0.5°C, 0.7 ± 0.004 mg/m^3 at 26 ± 0.5°C, and 0.96 ± 0.005 mg/m^3 at 31 ± 0.5°C). The concentration of N_2O in the atmosphere is 0.3 mg/m^3 [14], and the N_2O emissions in this study are greater than the amount in the atmosphere, suggesting that biological nitrogen removal processes including nitrification may have caused the increase in N_2O concentration in the environment [15]. The further research is needed to study other factors

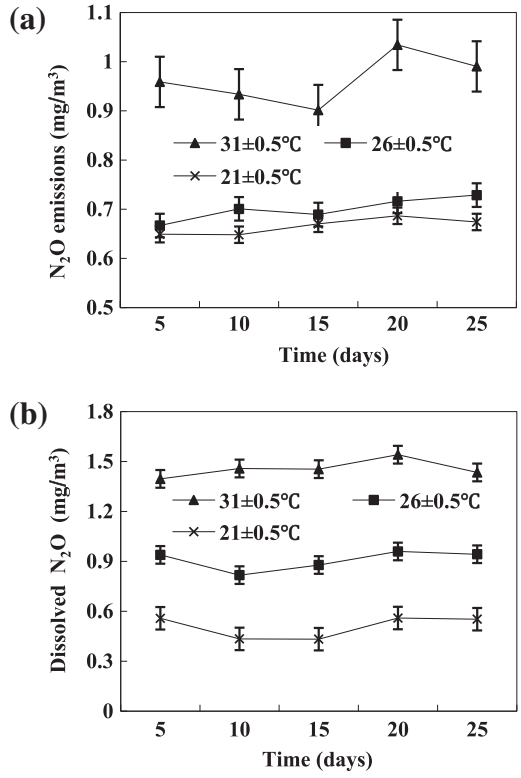

Fig. 2. N_2O production at different temperatures: (a) N_2O emission and (b) dissolved N_2O.

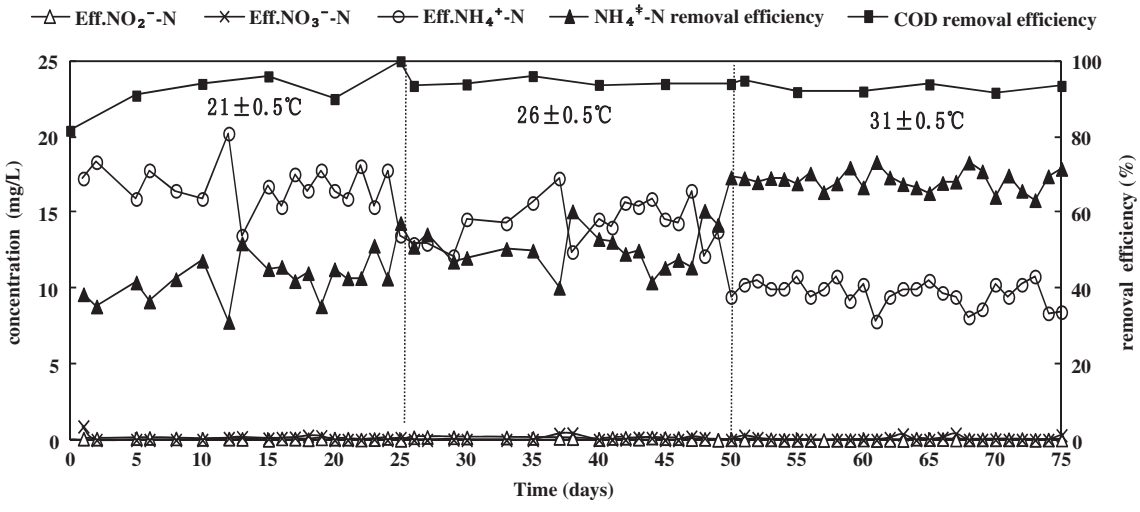

Fig. 1. Performance of SBR at different temperatures.

that may affect N_2O emission in the process of nitrogen removal by micro-expansion aerobic granular sludge during the actual wastewater treatment engineering process.

The amount of dissolved N_2O varied greatly with changing temperature, and it decreased at lower temperatures (Fig. 2(b)). With the decrease in temperature, the activity of N_2O reductase and synthetase both decreased [16], which could be responsible for the downward trend of dissolved N_2O and released N_2O with temperature dropping down. However, the change in the trend of dissolved N_2O at lower temperatures was more obvious than the N_2O emissions, indicating that temperature more strongly influenced the dissolved N_2O.

3.2.2. N_2O generation characteristics in a typical SBR cycle

The performance of the SBR with the micro-expansion aerobic sludge was analyzed during a typical SBR cycle (Fig. 3). The ammonia nitrogen removal efficiencies were above 40% at all temperatures. NO_3^--N and NO_2^--N did not changed significantly with the temperature, and the concentrations of NO_3^--N and NO_2^--N in the effluent remained below 1 mg/L at all temperatures, which means that the ammonia nitrogen removal mechanism did not change with varying temperature but always occurred by simultaneous nitrification and denitrification (SND) in the micro-expansion aerobic granular sludge system. DO is an important factor in the wastewater treatment process, the SBR reactor with the micro-expansion aerobic granular sludge operated under low DO conditions (1 mg/L) with the same nitrification type (SND).

The production of N_2O is the result of both nitrification and denitrification processes in the SND system [17]. During the steady operation periods, the N_2O production decreased with the temperature dropping down, with values of approximately 0.115 mg/g NH_4^+-N, 0.109 mg/g NH_4^+-N, and 0.097 mg/g NH_4^+-N at 31 ± 0.5, 26 ± 0.5, and 21 ± 0.5℃. In general, 8.07, 5.37, and 3.93% of the total nitrogen load of the SBR at 31 ± 0.5℃, 26 ± 0.5℃ and 21 ± 0.5℃ was emitted as N_2O (Table 1). However, only 5.41% of the total nitrogen load was converted into N_2O in the oxic–anoxic granular sludge SBR at 31 ± 0.5℃ (12), and 0.60 ± 0.17% in an aerobic granulation sequencing batch airlift reactor (SBAR) at 18 ± 3℃ [18].

3.3. Dynamic analysis

Temperature had a remarkable effect on the removal of ammonia nitrogen by micro-expansion aerobic granular sludge (Table 2). With the decrease in temperature, the removed amount of ammonia nitrogen decreased, and the nitrification rate slowed at the same time. Temperature also had a significant influence on N_2O emission, which decreased with temperature; this trend might be explained by the great impact of temperature on the entire process of the nitrification and denitrification reaction. $\Delta\rho$N/COD indicates the efficiency of carbon source utilization [19]. This value did not reach that of the optimal nitrogen removal efficiency at 26 ± 0.5℃, but it complied with the peak of $\Delta\rho$N/COD, 0.054 mg N/mg COD. Considering the actual working conditions and the N_2O generation characteristics with temperature, 26℃ is suitable for reducing N_2O emission and maintaining nitrogen removal efficiency in wastewater treatment.

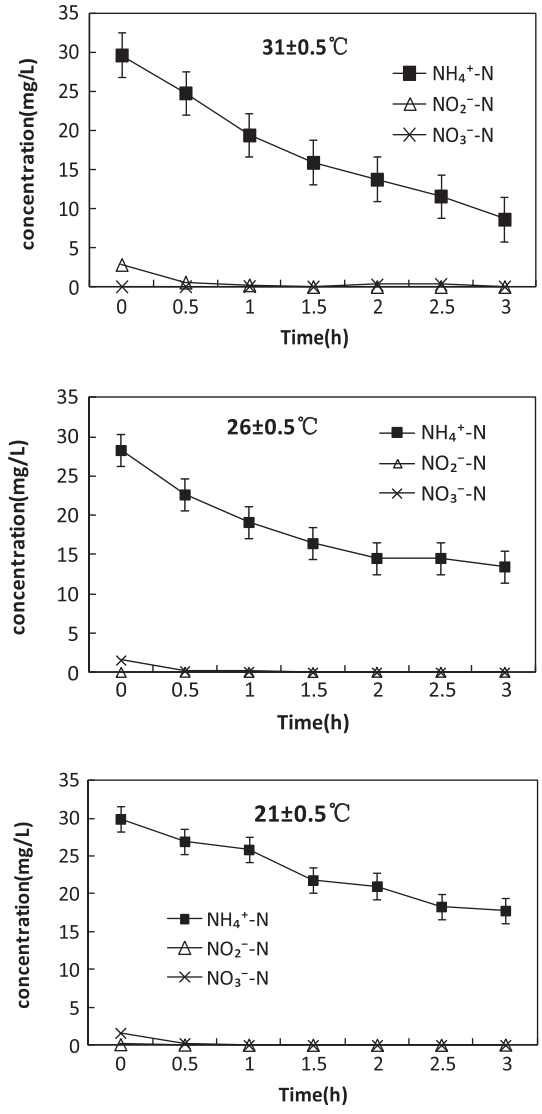

Fig. 3. Nitrogen concentration at different temperatures.

Table 1
N_2O generation characteristics in a typical SBR cycle

T (℃)	NH_4^+-N removal efficiency (%)	N_2O production (mg/m^3)		N_2O production (mg/g NH_4^+-N)	N_2O production (% of N load)
		N_2O emission	Dissolved N_2O		
31	68.29	0.96	1.46	0.115	8.07
26	50.14	0.70	0.91	0.109	5.37
21	42.97	0.67	0.51	0.097	3.93

Table 2
Pollutant removal characteristics at different temperatures

Temperature (℃)	31	26	21
COD degradation rate (mg COD/(g SS) h)	48.17	30.77	42.26
NH_4^+-N removal (mg/L)	21.03	14.83	12.13
Specific nitrification rate (mg N/(g SS) min)	38.95×10^{-3}	27.47×10^{-3}	22.47×10^{-3}
N_2O emission rate (mg N_2O/(g SS) min)	3.20×10^{-4}	2.33×10^{-4}	2.23×10^{-4}
$\Delta\rho$N/COD (mg N/mg COD)	0.048	0.054	0.031

4. Conclusions

(1) The effect of temperature on N_2O production by micro-expansion aerobic granular sludge was remarkable, and the proportions of N_2O production to the influent nitrogen loading at 31 ± 0.5℃, 26 ± 0.5℃, and 21 ± 0.5℃ were 8.07, 5.37, and 3.93%, respectively, which are higher than aerobic granular sludge. Decreasing the temperature reduced the release rate of N_2O, resulting in a decrease in N_2O production.
(2) Temperature also had a remarkable effect on the removal of ammonia nitrogen with the micro-expansion aerobic granular sludge. Based on the calculation of ammonia nitrogen removal efficiency, $\Delta\rho$N/COD, and N_2O production, 26℃ is recommended as a suitable temperature for reducing N_2O emission and maintaining nitrogen removal efficiency in wastewater treatment.

Acknowledgements

This work was supported by National Natural Science Foundation of China (No. 21177033) and Natural Science Foundation of Heilongjiang Province (No. ZD201412).

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