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Effect of PHB and oxygen uptake rate on nitrous oxide emission during simultaneous nitrification denitrification process

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ABSTRACT

Simultaneous nitrification denitrification (SND) process was achieved in a SBR system to evaluate the impacts of intracellular carbon source PHB and oxygen uptake rate (OUR) on nitrous oxide (N_2O) emission. Compared with the sequential nitrification and denitrification (SQND) process, SND process significantly improved the nitrogen removal. N_2O emission during SND process was much higher than the SQND process. The amount of N_2O emission was 26.85 mg N per cycle in SND process, which was almost four times higher than that in SQND process. About 7.05% of the removed nitrogen during SND process was converted to N_2O-N. N_2O emission had great relations with the OUR and the OUR could reflect the N_2O emission trend more exactly than the DO concentration. At the aerobic stage of SND, the simultaneous denitrification could carried out using PHB as the carbon source and N_2O emission increased because of the slow degradation of PHB.

1. Introduction

In order to protect the water from eutrophication, many countries enforce the removal of nutrients through biological treatment of wastewater. The effluent quality standards are more stringent. So many modifications and novel processes have been developed and implemented for nitrogen removal from wastewater (Hu et al., 2011a). In biological nitrogen removal, inorganic nitrogen in the form of ammonium is removed through aerobic, autotrophic nitrification followed by anoxic, heterotrophic denitrification (Meyer et al., 2005). However, some heterotrophic nitrifiers have been reported to denitrify nitrite (NO_2^-) and nitrate (NO_3^-) aerobically (Zart and Eberhard, 1998). Several literatures have illustrated that nitrification and denitrification can occur simultaneously at low oxygen level (Yoo et al., 1999; Li et al., 2007). This is often referred to as simultaneous nitrification and denitrification (SND) process. The SND process represents a significant advantage over the conventional separated nitrification and denitrification processes (Chiu et al., 2007).

It is considered that the biological treatment process of domestic wastewater is an important source of greenhouse gas, like CH_4 and N_2O. N_2O is an important greenhouse gas, having an atmospheric lifetime of about 114 years, a global warming potential of 298 relative to CO_2 over a 100 year time horizon, and is responsible for about 6% of anticipated global warming (IPCC, 2007). N_2O also contributes to the depletion of stratospheric ozone, because of the stratospheric reaction with atomic oxygen to nitric oxide (NO) (Mosier, 1998). Therefore, even low amounts of N_2O emission are unwanted. Nowadays, the concentration of atmospheric N_2O is estimated to be approximately 319 ppbv, which is approximately 16% higher than that during the preindustrial era, and it is increasing at a rate of 0.3% $year^{-1}$ (IPCC, 2007). During the microbial transformations of nitrogenous compounds, N_2O can be produced during nitrification, denitrification, dissimilatory reduction of NO_3^-—NH_4^+ and chemo-denitrification (Wu et al., 2009a). Studies show that the heterotrophic nitrifying bacteria are often able to denitrify under aerobic conditions and N_2O is produced as an intermediate in this process. Heterotrophic nitrifiers produce much more N_2O per cell than autotrophic nitrifiers, and it might produce significant amount of N_2O under certain sets of circumstances (Wrage et al., 2001).

N_2O emission during the biological treatment is affected by many factors, such as COD/N ratio (Hanaki et al., 1992; Wu et al., 2009a), pH (Thoern and Soerensson, 1996), carbon content (Wu et al., 2009b), nitrite concentration (Tallec et al., 2006), dissolved oxygen (Tallec et al., 2006) and so on. The dissolved oxygen (DO) concentration is considered as a very important parameter controlling N_2O emission. In oxygen limiting conditions, autotrophic ammonia oxidizers use nitrite as the terminal electron acceptor to save oxygen for the oxygenation reaction (Hu et al., 2011b). Usually, the SND process occurred at a DO concentration lower

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than 0.5 mg/L (Chiu et al., 2007). This suggests that some heterotrophic nitrifiers have the ability to denitrify under low oxygen conditions to affect a SND process, and the N_2O emission during the SND process may be significant. Previous literatures studied the effect of DO concentration on N_2O emission (Tallec et al., 2006; Kampschreur et al., 2008; Hu et al., 2009). However, they only studied the trend of N_2O emission with DO concentration. Dissolved oxygen cannot describe the microbial condition of the sludge accurately and directly, especially at low oxygen level. The oxygen uptake rate (OUR) is a parameter that can be used to evaluate the rate at which metabolic processes take place in active sludge treatment processes. So according to the OUR, we could study the relation of N_2O emission and microbial activity. Another possible factor influencing the N_2O emission is the consumption of intracellular storage compounds, e.g. poly-hydroxybutyrate (PHB) and glycogen (Kampschreur et al., 2009). Glycogen accumulating organisms (GAO) and phosphate accumulating organisms (PAO) in the SND system both employ a special mechanism to store organic carbon during anaerobic periods, involving storage compounds, which are finally degraded via their internal PHB pool. The denitrification need carbon source to proceed when treating low C/N ratio wastewater and the microbes can carry out denitrification using their stored carbon compounds. PHB plays an essential ecological role in several wastewater treatment processes, so it is a general factor related to N_2O emission (Kampschreur et al., 2009). However, few studies focus on this point, and the relation between the growth of PHB and N_2O emission have not been well investigated.

In this study, the SND process was achieved using the SBR system. The contaminant removal performance and N_2O emission were evaluated, as well as OUR and PHB content. The aim of this paper was to (1) investigate the N_2O emission rate and amount during the SND process; (2) evaluate the impact of PHB consumption on N_2O emission and, (3) study the relation between OUR and N_2O emission during SND process.

2. Methods

2.1. Synthetic wastewater

Synthetic wastewater was used in this study. The wastewater contained, per liter: 260.2 mg $C_6H_{12}O_6{\cdot}H_2O$; 260.2 mg $CH_3COONa{\cdot}3H_2O$; 191 mg NH_4Cl; 200 mg $NaHCO_3$; 11 mg KH_2PO_4; 18 mg $K_2HPO_4{\cdot}3H_2O$; 10 mg $MgSO_4{\cdot}7H_2O$; 10 mg $FeSO_4{\cdot}7H_2O$; 10 mg $CaCl_2{\cdot}2H_2O$ and 1 mL nutrient solution. One liter of nutrient solution contained: 0.15 g H_3BO_3; 0.03 g $CuSO_4{\cdot}5H_2O$; 0.18 g KI; 0.12 g $MnCl_2{\cdot}4H_2O$; 0.06 g $Na_2MoO_4{\cdot}2H_2O$; 0.12 g $ZnSO_4{\cdot}7H_2O$; 0.15 g $CoCl_2{\cdot}6H_2O$ and 10 g ethylene diamine tetraacetic acid (Zeng et al., 2003). The influent characters were shown in Table 1.

Table 1
Mean contaminant concentrations with standard deviations (in brackets) and removal efficiencies (%) in each system.

	Influent	SND	SQND
COD (mg/L)	342.24 (36.32)	28.68 (7.08)	29.58 (12.74)
COD removal (%)		91.62	91.36
TP (mg/L)	3.14 (0.51)	0.28 (0.20)	0.44 (0.29)
TP removal (%)		91.08	85.99
NH_4^+ (mg/L)	49.31 (5.71)	0.97 (0.49)	1.05 (1.04)
NH_4^+ removal (%)		98.03	97.87
TN (mg/L)	51.08 (5.99)	7.63 (1.96)	17.22 (3.64)
TN removal (%)		85.06	66.29
NO_3^-	1.39 (1.08)	6.74 (1.63)	16.46 (1.22)
NO_2^-	ND	0.06 (0.10)	0.03 (0.01)
pH	7.21 (0.14)	7.15 (0.23)	7.30(0.32)

ND: not detected.

2.2. Reactor setup and operation

The experiments were conducted in two gastight sequencing batch reactors (SBRs), constructed using transparent, rigid plexiglas cylinders, with an effective volume of 15 L each. The schematic diagram was illustrated in Fig. 1. Biomass was enriched in the SBRs seeding with sludge from the Second Wastewater Treatment Plant of Everbright Water (Jinan) Ltd. (Jinan, China). Both the two SBRs were operated with a cycle time of 6 h consisting of a 6 min feeding, 90 min anaerobic reaction and 180 min aerated period, followed by 70 min settling and 14 min decant. The SBRs were operated at 25 ± 2 °C. At the feeding period 7.5 L of synthetic wastewater was feed into each reactor using peristaltic pump. The electric agitator with a rectangular paddle was used to keep the suspension of the sludge at the anaerobic stage. At the aeration stage, air pump was used to supply air through the diffuser located at the bottom of the reactor. The difference between the two reactors was the aeration rate. In one SBR, the DO concentration at the aeration stage was maintained 0.35–0.80 mg/L by on/off control of air pump to achieve SND process. As a contrast, another SBR was operated with an aeration rate of 7.5 $L_{air}/(L_{reactor}\ h)$ at the aeration stage to mimic the actual wastewater treatment plant to achieve the sequential nitrification and denitrification (SQND) process. After settling, 7.5 L of supernatant was removed, resulting in a hydraulic retention time (HRT) of 12 h. The mixed liquor suspended solid (MLSS) was maintained at approximately 3000 mg/L and certain amount of excess sludge was disposed at the end of aerobic phase to control the SRT at approximately 15 days.

The effluent was analyzed every 5 days to evaluate the operation conditions of the reactors at first. After running for about 4 months, the effluent of the two reactors was stable and the SND process was achieved. Then the COD and nutrients removal performances were evaluated every 2 days. Meanwhile on day 130, the N_2O emission during one cycle was also measured by collecting the off-gases at intervals of 15 min. At the same time, liquid phase and sludge samples were taken to measure the water quality and PHB content.

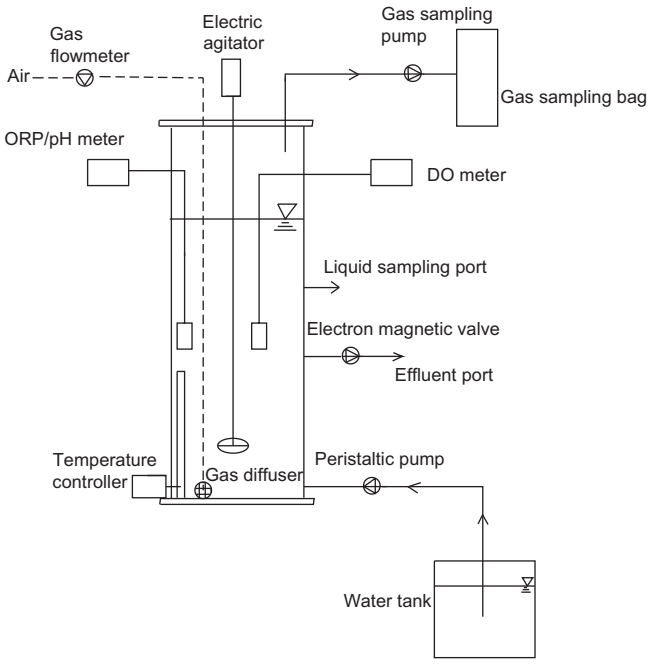

Fig. 1. Schematic description of the experiment system.