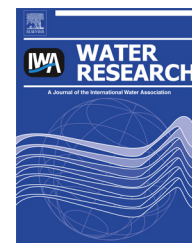


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Source identification of nitrous oxide on autotrophic partial nitrification in a granular sludge reactor

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ABSTRACT

Emission of nitrous oxide (N₂O) during biological wastewater treatment is of growing concern since N₂O is a major stratospheric ozone-depleting substance and an important greenhouse gas. The emission of N₂O from a lab-scale granular sequencing batch reactor (SBR) for partial nitrification (PN) treating synthetic wastewater without organic carbon was therefore determined in this study, because PN process is known to produce more N₂O than conventional nitrification processes. The average N₂O emission rate from the SBR was 0.32 ± 0.17 mg-N L⁻¹ h⁻¹, corresponding to the average emission of N₂O of $0.8 \pm 0.4\%$ of the incoming nitrogen load ($1.5 \pm 0.8\%$ of the converted NH₄⁺). Analysis of dynamic concentration profiles during one cycle of the SBR operation demonstrated that N₂O concentration in off-gas was the highest just after starting aeration whereas N₂O concentration in effluent was gradually increased in the initial 40 min of the aeration period and was decreased thereafter. Isotopomer analysis was conducted to identify the main N₂O production pathway in the reactor during one cycle. The hydroxylamine (NH₂OH) oxidation pathway accounted for 65% of the total N₂O production in the initial phase during one cycle, whereas contribution of the NO₂⁻ reduction pathway to N₂O production was comparable with that of the NH₂OH oxidation pathway in the latter phase. In addition, spatial distributions of bacteria and their activities in single microbial granules taken from the reactor were determined with microsensors and by in situ hybridization. Partial nitrification occurred mainly in the oxic surface layer of the granules and ammonia-oxidizing bacteria were abundant in this layer. N₂O production was also found mainly in the oxic surface layer. Based on these results, although N₂O was produced mainly via NH₂OH oxidation pathway in the autotrophic partial nitrification reactor, N₂O production mechanisms were complex and could involve multiple N₂O production pathways.

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1. Introduction

Nitrous oxide (N_2O) emissions draw attention since N_2O is expected to be a major stratospheric ozone-depleting substance in the future (Ravishankara et al., 2009) and is an important greenhouse gas with a global warming potential of about 300 times higher than CO_2 (Desloover et al., 2012; IPCC, 2007). It is generally accepted that nitrogen removal processes in a wastewater treatment system are an anthropogenic source of N_2O (Desloover et al., 2012). Conventionally, biological nitrogen removal is achieved by a combination of nitrification and denitrification processes. In contrast, an alternative and innovative approach is the use of a partial nitrification (PN) process followed by an anaerobic ammonium oxidation (anammox) process (PN-anammox process), which has several advantages, such as no need for external carbon addition, less energy and oxygen requirement, and less sludge production (van Dongen et al., 2001; Kartal et al., 2010). The PN-anammox process is applicable to reject water (Desloover et al., 2011; Kampschreur et al., 2008; 2009a; Joss et al., 2009; Okabe et al., 2011), landfill leachate (Wang et al., 2010), and wastewater from semiconductor factory (Tokutomi et al., 2011). N_2O emission from PN-anammox processes, especially from the PN process, has been reported (Desloover et al., 2011; Kampschreur et al., 2008; Okabe et al., 2011). Especially, a granular sludge reactor for PN process draws attention because of high specific nitrification rate, efficient biomass retention and excellent settleability.

There are three main microbial pathways involved in N_2O production. During nitrification, it is produced from hydroxylamine (NH_2OH) as a side product of the oxidation of ammonium (NH_4^+) to nitrite (NO_2^-) (Poughon et al., 2001; Hooper and Terry, 1979). During denitrification, N_2O is produced as an intermediate during reduction of nitrate (NO_3^-) to N_2 by heterotrophic denitrifiers (Lu and Chandran, 2010; Schmidt et al., 2004). Some ammonia-oxidizing bacteria (AOB) reduce NO_2^- to N_2O or N_2 through a process called nitrifier denitrification (Tallec et al., 2006; Wrage et al., 2001; Colliver and Stephenson, 2000). Many studies have been conducted to estimate N_2O emission rate of PN processes (Kong et al., 2013a,b; Okabe et al., 2011; Law et al., 2011; Desloover et al., 2011; de Graaff et al., 2010; Kampschreur et al., 2008). In contrast, there are few studies on N_2O production pathways. Nitrifier denitrification was the key biological pathway of N_2O production in an intermittently aerated sequencing batch biofilm reactor for PN treating synthetic ammonium-rich wastewater (Kong et al., 2013b) while NH_2OH oxidation pathway was the main source of N_2O in a sequencing batch reactor (SBR) for PN (PN-SBR) (Law et al., 2011; Yang et al., 2009). To determine which pathway is responsible for N_2O production in a wastewater treatment process is still challenging, because a variety of operational parameters (concentrations and loading rates of nitrogenous compounds, dissolved oxygen (DO) and organic carbon, pH, a ratio of organic carbon and nitrogenous compounds (COD/N) and temperature) influence N_2O production in a PN process (Tallec et al., 2006; Kampschreur et al., 2009b; Desloover et al., 2012; Wunderlin et al., 2012; Law et al., 2011). Furthermore, their temporal changes also affect N_2O production.

Analyses of the intermolecular distributions of ^{15}N in N_2O (isotopomers) are regarded as useful parameters to infer the

predominant N_2O production pathway (Wunderlin et al., 2013; Sutka et al., 2006; Toyoda et al., 2005, 2011). Isotopomer ratios (site-specific N isotope ratios in asymmetric molecules of NNO) give us qualitative information on N_2O production and consumption pathways. Toyoda et al. (2011) and Wunderlin et al. (2013) conducted isotopomer analysis and distinguished N_2O produced during NH_2OH oxidation from N_2O produced during NO_2^- reduction in wastewater treatment processes. However, N_2O production pathways in a PN-SBR have not been investigated by isotopomer analysis. In a PN-SBR, temporal changes in the operational parameters (DO, NH_4^+ and NO_2^- concentrations and pH level) are more significant than conventional activated sludge processes, which likely play an important role in N_2O production pathways.

In this study, source of N_2O produced in an autotrophic granular PN-SBR was investigated. A lab-scale PN-SBR was operated and N_2O emission from the reactor was determined with an online monitoring system. Dissolved N_2O in the reactor was monitored with a microsensor for N_2O . N_2O , DO, pH, NH_4^+ , NO_2^- and NO_3^- concentrations in the PN-SBR for one cycle were continuously monitored. We measured temporal changes in intermolecular ^{15}N -site preference (SP) in N_2O in the PN-SBR for one cycle. In addition, the spatial distribution of N_2O , DO, pH, NH_4^+ , NO_2^- and NO_3^- in the PN granules was determined with the microsensors to estimate net production and consumption rates of N_2O , NH_4^+ and NO_2^- in single granules. The spatial distribution of AOB and other bacteria in the PN granules was determined by FISH. The combination of microsensor measurements and FISH analysis allows us to deduce function of AOB. Finally, these results were compared and we discussed the source of N_2O in the PN-SBR.

2. Materials and methods

2.1. Operation of a lab-scale autotrophic PN-SBR

A lab-scale autotrophic PN-SBR with working volume of 2.0 L was operated. The reactor was inoculated with 0.3 L of PN granules (3–5 mm in diameter), which was obtained from the PN reactor operated in our laboratory (Okabe et al., 2011). One cycle of the reactor operation was 4 h. It consisted of feeding of a synthetic wastewater (3 min), aeration (232 min), settling of the granules (3 min), and discharging of treated wastewater (2 min). The composition of a synthetic wastewater was as follows: $(\text{NH}_4)_2\text{SO}_4$ (1650 mg L^{-1}), KHCO_3 (3300 mg L^{-1}), $\text{CaCl}_2 \cdot 2\text{H}_2\text{O}$ (135 mg L^{-1}), $\text{MgSO}_4 \cdot 7\text{H}_2\text{O}$ (300 mg L^{-1}), and KH_2PO_4 (22 mg L^{-1}). Trace element solutions I and II were prepared and added as described by van de Graaff et al. (1996). The influent pH was adjusted to 7.7 ± 0.1 . The hydraulic retention time (HRT) of the PN reactor was fixed at 8 h. The airflow rate was changed according to reactor performance until the PN process became stable. After the PN process became stable, airflow rate was fixed at 0.2 L min^{-1} .

2.2. Water and gas analyses

The PN reactor performance was determined by collecting grab samples of influent and effluent at arbitrary time

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