Bioresource Technology 179 (2015) 510–517

Contents lists available at ScienceDirect

Bioresource Technology

journal homepage: www.elsevier.com/locate/biortech

Minimization of nitrous oxide emission in a pilot-scale oxidation ditch: Generation, spatial variation and microbial interpretation

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HIGHLIGHTS

- Distinct spatial variations and low emission factor were found in oxidation ditch.
- Nitrifier denitrification contributed to 89.78% of the total N_2O generation.
- Denitrifiers performing N_2O reducing capabilities dominated microbial community.
- NOB abundance played a key role in the low nitrite and N_2O accumulations.
- N_2O emissions significantly increased when encountered ammonia or aeration shock.

ARTICLE INFO

Article history:
Received 25 October 2014
Received in revised form 8 December 2014
Accepted 9 December 2014
Available online 29 December 2014

Keywords:
Nitrous oxide
Oxidation ditch
Spatial variation
Nitrifier denitrification
Microbial community

ABSTRACT

Nitrous oxide (N_2O) emission from wastewater treatment plants (WWTPs) has received increasing attention. This paper presented how N_2O emission was significantly reduced in a pilot-scale Carrousel oxidation ditch under reasonable nitrification and denitrification. N_2O emission from the reactor was found as low as 0.027% of influent nitrogen, which was much less than that from other processes. Further measurements on spatial variation of N_2O emission in the alternative aerobic/anoxic zones with help of a series of batch experiments demonstrated that about 90% of the emission was contributed by nitrifier denitrification (ND). Moreover, the taxonomic analysis based on high through-put 16S rRNA gene sequencing revealed that the high abundance of denitrifying bacteria and nitrite-oxidizing bacteria (NOB) was responsible for low nitrite accumulations and consequent low N_2O emissions. However, N_2O generation would be greatly increased upon the normal operation being shocked by either ammonia overload or aeration failure of the oxidation ditch system.

1. Introduction

Nitrous oxide (N_2O) is the third largest greenhouse gas in the atmosphere after carbon dioxide and methane (IPCC, 2013). Apart from its significant global warming potential of 296 times (100-year period) as strong as that of carbon dioxide, N_2O is currently the single most important destructive source resulting in stratospheric ozone depletion (Ravishankara et al., 2009). Atmospheric N_2O concentration has increased continuously at an average rate of 0.73 ± 0.03 ppb per year during the past three decades, and anthropogenic sources were supposed to be responsible for this historic increase, as N_2O emissions from natural sources were quite stable (IPCC, 2013).

N_2O emission from wastewater treatment plants (WWTPs) has received increasing attentions since it was confirmed to be one significant atmospheric N_2O source (Aboobakar et al., 2013; Foley et al., 2010). A survey revealed that domestic wastewater treatment contributed to 14% of the overall N_2O emissions in U.S. (Ahn et al., 2010). To date, many efforts have been made to quantify N_2O emissions from WWTPs but a wide range of emission factors (0.03–28.2%) have been reported (Table S1). Generally, N_2O emissions measured in full-scale WWTPs was lower compared with those in laboratory studies, which was probably due to low microbial diversity facilitated by synthetic wastewater consisting of single carbon source.

N_2O generation mainly occurs in biological nitrogen removal (BNR) in WWTPs and commonly comprises three pathways, the contributions of which were still controversial: (1) nitrifier denitrification (ND): N_2O produced as a by-product when ammonia-oxidizing bacteria (AOB) use nitrite as an alternate electron acceptor instead of O_2 (Kampschreur et al., 2008; Yu et al.,

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http://dx.doi.org/10.1016/j.biortech.2014.12.027
0960-8524/

2010); (2) heterotrophic denitrification (HD): N_2O accumulated during nitrate or nitrite reduction by heterotrophic denitrifiers due to imbalanced activities of nitrogen-reducing enzymes (Hanaki et al., 1992; Lu and Chandran, 2010); (3) Chemical reactions: N_2O produced from oxidizing NH_2OH by nitrite or O_2 and decomposition from NOH or $N_2O_2H_2$, but this process is considered to occur rarely due to insufficient concentrations of these nitrification intermediates (Wunderlin et al., 2012).

Process design and operational parameters of WWTPs dominate the magnitude and variability of N_2O emissions. For instance, reactors achieving partial nitrification or simultaneous nitrification and denitrification (SND) always produce considerable amount of N_2O, while reactors with high recycle ratio emit low N_2O (Ahn et al., 2010; Foley et al., 2010). Several factors were generally accepted playing major roles in N_2O generation such as nitrite accumulation (Foley et al., 2010; Kampschreur et al., 2008; Zhou et al., 2008), dissolved oxygen limitation (Tallec et al., 2006; Lu and Chandran, 2010), insufficient organic carbon source (Hanaki et al., 1992; Lu and Chandran, 2010), short sludge retention time (SRT, Lotito et al., 2012), etc.

Oxidation ditches are widely employed in WWTPs due to their advantages in processing efficiency and operational cost (Liu et al., 2010). They have distinct features combining continuous stirring tank reactor (CSTR) and plug flow reactor (PFR), which have significant impact on magnitude and variation of N_2O emission. CSTR-like characteristic is demonstrated with the large internal recycle ratios of oxidation ditch (dozens to hundreds of times influent flow rate). Therefore, the high influent nitrogen concentration gets sufficient dilution and thereby the pressure on N_2O generation gets released (Foley et al., 2010; Liu et al., 2010). On the other hand, oxidation ditch demonstrates a PFR-like spatial characteristic during one cycle that the activated sludge experiences alternative aerobic and anoxic regions for nitrification and denitrification. Accordingly, N_2O emission presents a distinct spatial variation within the reactor, which also increases difficulties of precise quantification of the total emission amounts (Aboobakar et al., 2013).

To date, researches on N_2O emissions from the distinct wastewater treatment process oxidation ditch were scarcely reported. During this study, the magnitude and spatial variations of N_2O emissions were systematically monitored within a pilot-scale oxidation ditch. Furthermore, N_2O generation sources were explored with a series of batch experiments at various oxygenation levels and further interpreted by microbial community analysis based on high-throughput 16S rRNA gene sequencing. Finally, responses of the oxidation ditch to abnormal operations like nitrogen overload shock and aeration failure shock were investigated.

2. Methods

2.1. Set-up and operation of the pilot-scale oxidation ditch

This study was conducted in a pilot-scale Carrousel oxidation ditch which was fabricated with plexiglass with working volume of 1.4 m^3 (Fig. 1). The reactor comprised four channels with 1.15 m long, 0.35 m wide and 0.5 m working depth. Two surface impellers and four submerged stirrers drove the circulating flow in the channels at an average rate of 0.01 m s^{-1}. A bioselector with working volume of 50 L was setup to avoid sludge bulking. A clarifier of 0.15 m^3 was used for the separation of the activated sludge which was recycled back to the bioselector at 100% of the influent flow by a magnetic pump.

For the initial start-up, the reactor was seeded with activated sludge taken from a full-scale municipal WWTP in Langfang, China. Synthetic wastewater was pumped into the bioselector at a flow rate of 0.1 m^3 h^{-1} to maintain a hydraulic retention time (HRT) of 14 h. The synthetic wastewater was stored in a tank with a volume of 1.8 m^3 and the composition in 1 m^3 tap water was as follows: 250.0 g sugar, 191.1 g NH_4Cl, 36.8 g $K_2HPO_4{\cdot}3H_2O$, 12.0 g $MgSO_4{\cdot}7H_2O$, 10.0 g $CaCl_2$, 3.0 g $FeSO_4{\cdot}7H_2O$, and 50 mL trace elements solution (Zheng et al., 2014). This influent gave concentrations of chemical oxygen demand (COD) and NH_4^+-N as 250.0 mg L^{-1} and 50 mg L^{-1}, respectively.

Aeration was controlled at the rate of 1.8 m^3 h^{-1} (an optimal value for efficient nitrogen removal) by a flowmeter and well distributed into the reactor with four 1 m long microporous aerated pipes installed at the base of the second and third channels. SRT was maintained at approximately 25 d by discharging 60 L mixed liquor daily from the fourth channel. The temperature was maintained at 22 ± 2 °C with help of heating rods and pH was around 7.8 without any extra control and adjustment. Influent and effluent samples were taken daily for analysis of COD, NH_4^+-N, NO_2^--N and NO_3^--N.

After two-month steady-state operation, two kinds of stress tests were carried out including "ammonium overload shock" and "aeration failure shock" experiments. In the former experiment, synthetic wastewater containing NH_4^+-N concentration of 250 mg L^{-1} (other compositions not changed) was pumped to the reactor at 0 h and turned back to the normal influent after 1 h. In the latter experiment, aeration rate was reduced to 0.9 m^3 h^{-1} at time 0 h and switched back to normal 1.8 m^3 h^{-1} at 2 h.

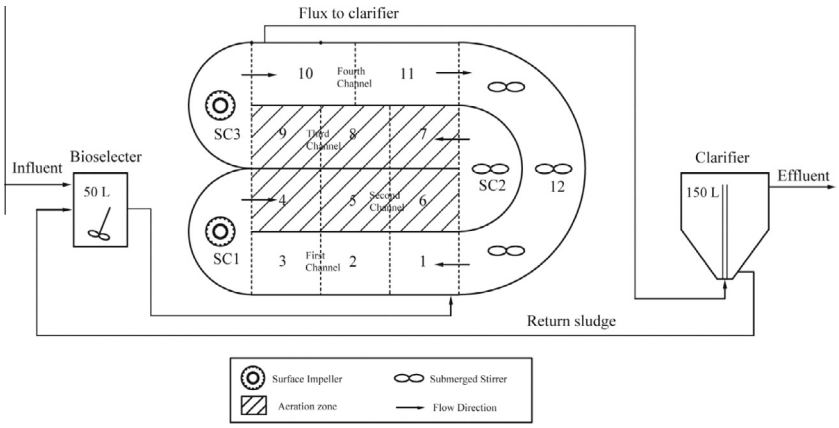

Fig. 1. The sketch of the pilot-scale Carrousel oxidation ditch and sampling zones.