

## Greenhouse gases (GHGs) emission from aerobic methanotrophic denitrification (AeOM-D) in sequencing batch reactor (SBR)

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**Abstract.** This study presents the effect of hydraulic retention time (HRT) on the characteristics of emission of three major greenhouse gases (GHGs) including CH<sub>4</sub>, CO<sub>2</sub> and N<sub>2</sub>O during operation of a sequencing batch reactor for AeOM-D (AeOM-D SBR). Maximum specific denitrification rate (SDNR) increased as the HRT increased from 0.25 to 1.0 d. However, the highest overall SDNR during the entire reaction time occurred at the medium HRT of 0.5 d since that of the longer HRT tended to be lowered due to deficiency of residual nitrate. Dissolved N<sub>2</sub>O concentration increased and then leveled-off or slightly decreased. Concentration of the dissolved N<sub>2</sub>O was higher at the shorter HRT, which was highly associated with the lowered C/N ratio. A longer HRT leads to a higher C/N ratio with a sufficient carbon source produced by methanotrophs via methane oxidation, which provided a favorable condition for reducing N<sub>2</sub>O formation. Similar to the dissolved N<sub>2</sub>O formation, N<sub>2</sub>O emission rate was higher at a shorter HRT condition. Opposite to the N<sub>2</sub>O emission, emission rates of CH<sub>4</sub> and CO<sub>2</sub> were higher at a longer HRT. Longer HRT resulted in the greater total GHGs emission as CO<sub>2</sub> equivalent due to the higher emission rate of CH<sub>4</sub>. It was doubled when the HRT increased from 0.5d to 1.0 d. Contribution of CH<sub>4</sub> onto the total GHGs emission was most dominant accounting for 98 - 99% compared to that of N<sub>2</sub>O (< 2%).

**Keywords:** Aerobic oxidation of methane with denitrification (AeOM-D), hydraulic retention time (HRT), methane (CH<sub>4</sub>), carbon dioxide (CO<sub>2</sub>), nitrous oxide (N<sub>2</sub>O), greenhouse gases (GHGs)

### 1. Introduction

Denitrification in wastewater treatment is significant since discharge of nitrate at a high concentration to water environment can potentially cause eutrophication which eventually deteriorates quality of water resources (Kim et al., 2005). In addition to damages on water environmental quality, nitrate is known to be forming carcinogenic compounds such as nitrosamines and nitrosamides (Ono et al., 2000; Forman et al., 1985). A typical method for

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enhancing denitrification is to add external carbon sources such as methanol and acetate into either biological nutrient removal (BNR) process or post-denitrification process as a tertiary step (Costa et al., 2000). However, the excess addition of the external carbon sources often generates concerns such as increase in operating cost, excess biological growth and deterioration of effluent quality due to the residual organic carbon (Lee et al., 2014).

As an alternative, denitrification using methane (or hydrogen) has been recently attempted at a laboratory level study aiming water and wastewater treatment (Daelman et al., 2014; Cuba et al., 2011; Modin et al., 2010). It can be attractive due to its some benefits with less potential of biomass production and organic pollutants remaining in the effluent (Sun et al., 2013). The denitrification using methane as a sole carbon source under aerobic condition is known as aerobic oxidation of methane with denitrification (AeOM-D). Despite some benefits in AeOM-D, its applicability is still limited due to low mass transfer rate of methane gas into water and utilization rate of the dissolved methane by the relevant microorganisms. As for the latter, methane in AeOM-D is firstly metabolized by methanotrophs with methane monooxygenase (MMO) oxidizing methane to methanol. The rate of MMO has not been clearly explained in AeOM-D for water treatment system but the denitrification rate is strongly subject to the enzymatic reaction rate (Houbroun et al., 1999). This means that methane oxidation is the rate-limiting step and AeOM-D might have a higher potential of emission of unutilized  $\text{CH}_4$  into the atmosphere. Waki et al. (2005) pointed out that the risk of explosion during operation if  $\text{CH}_4$  is not sufficiently transformed or utilized and its concentration in the off-gas may exceed 5-14%.

In addition to  $\text{CH}_4$ , there should be concerns for emission of the other greenhouse gases (GHGs) including  $\text{CO}_2$  and  $\text{N}_2\text{O}$  from AeOM-D. Global warming impacts of  $\text{CH}_4$  and  $\text{N}_2\text{O}$  are 28 and 265 times as strong as carbon dioxide ( $\text{CO}_2$ ), respectively (IPCC, 2013). Similar to common denitrification process, nitrous oxide ( $\text{N}_2\text{O}$ ) could be also produced from AeOM-D (Kits et al., 2015). In spite of low production potential of  $\text{N}_2\text{O}$  compared to  $\text{CH}_4$  and  $\text{CO}_2$ , it is very seriously considered due to its higher global warming impact. Mechanisms for  $\text{N}_2\text{O}$  production are still under study but in many studies,  $\text{N}_2\text{O}$  could be formed through various biological reaction related to nitrogen transformation: denitrification, autotrophic and heterotrophic nitrification, nitrifier-denitrification (Hu et al., 2011; Wrage et al., 2001).  $\text{N}_2\text{O}$  production in denitrification is more significant when nitrate is highly loaded (Hu et al., 2012; Wunderlin et al., 2012). It is important to point out that the most possible conditions for greater  $\text{N}_2\text{O}$  emission were associated with unfavorable conditions to nitrifiers and denitrifiers such as low DO, solids retention time (SRT), C/N ratio, and temperature (Zheng et al., 1994; Her and Huang, 1995; Thoern and Soerensson, 1996; Noda et al., 2003; Tallec et al., 2008; Hu et al., 2013; Paudel et al., 2015). The  $\text{N}_2\text{O}$  production in the full-scale conventional wastewater treatment plant is estimated from 0 to 14.6% of input nitrogen (Kampschreur et al., 2009) accounting for 26% of total GHGs emission from wastewater plant (Frijns et al., 2008). However, to our knowledge,  $\text{N}_2\text{O}$  emission from biological denitrification using methane oxidation has not been intensively studied yet. Some relevant studies attained so far are more likely related to anammox and denitrifying anaerobic methane oxidation (DAMO) in wastewater and soil area (Shi et al., 2013; Ding et al., 2014).

The overarching goal of this study was to characterize emission potential of GHGs from AeOM-D in a sequencing batch reactor (SBR) operated under different hydraulic retention time (HRT) conditions. Specifically, nitrogen transformation together with  $\text{N}_2\text{O}$  formation during AeOM-D was