



# Effect of hydraulic retention time and sludge recirculation on greenhouse gas emission and related microbial communities in two-stage membrane bioreactor treating solid waste leachate



Nararatchporn Nuansawan, Jarungwit Boonnorat, Wilai Chiemchaisri, Chart Chiemchaisri\*

Department of Environmental Engineering, Faculty of Engineering, Kasetsart University, 50 Ngam Wong Wan Road, Chatuchak, Bangkok 10900, Thailand

## HIGHLIGHTS

- Long term (>500 days) of CH<sub>4</sub> and N<sub>2</sub>O emissions from two-stage MBR was investigated.
- More than 90% of CH<sub>4</sub> emissions were contributed from first stage anaerobic reactor.
- N<sub>2</sub>O emission were at the same level from anaerobic and aerobic reactors.
- Effect of HRT and sludge recirculation was more pronounced for CH<sub>4</sub> than N<sub>2</sub>O.
- Microbial diversity and abundance were less when hydraulic loading was increased.

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## ABSTRACT

Methane (CH<sub>4</sub>) and nitrous oxide (N<sub>2</sub>O) emissions and responsible microorganisms during the treatment of municipal solid waste leachate in two-stage membrane bioreactor (MBR) was investigated. The MBR system, consisting of anaerobic and aerobic stages, were operated at hydraulic retention time (HRT) of 5 and 2.5 days in each reactor under the presence and absence of sludge recirculation. Organic and nitrogen removals were more than 80% under all operating conditions during which CH<sub>4</sub> emission were found highest under no sludge recirculation condition at HRT of 5 days. An increase in hydraulic loading resulted in a reduction in CH<sub>4</sub> emission from anaerobic reactor but an increase from the aerobic reactor. N<sub>2</sub>O emission rates were found relatively constant from anaerobic and aerobic reactors under different operating conditions. Diversity of CH<sub>4</sub> and N<sub>2</sub>O producing microorganisms were found decreasing when hydraulic loading rate to the reactors was increased.

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## 1. Introduction

Disposal of municipal solid wastes in sanitary landfills lead to the formation of landfill leachate, a highly polluted wastewater. Municipal solid waste landfill leachate generally contains high concentrations of dissolved organic matter and nutrients (Kjeldsen et al., 2002). In order to meet strict quality standards for direct discharge of leachate into natural water environment, integrated treatment methods, i.e. combination of chemical, physical and biological steps are required (Wisniewski et al., 2006). Membrane bioreactor (MBR) technology which combined biological wastewater treatment process and advanced physical separation has been developed as a promising technology for treating various types of wastewater including landfill leachate. The

membrane separation in MBR allows complete retention of biomass and maintaining high biomass concentration, resulting in an efficient biological digestion system (Ahmed and Lan, 2012). The advantages of MBRs over conventional biological processes are well-known including improvement of effluent quality, good process stability, reduce reactor size by retaining higher biomass or mixed liquor suspended solids (MLSS) concentrations, and lower sludge production (Van Dijk and Roncken, 1997). In order to apply MBR under high organic loading of wastewater, a novel type MBR utilizing inclined-plate separator in first stage anoxic reactor followed by second stage aerobic submerged MBR has been developed (Xing et al., 2006). The unique feature of the system was its capacity to storage majority of biomass in the first stage reactor while maintaining relatively constant biomass concentration in the second reactor for membrane fouling control.

Two-stage MBR has also been applied to the treatment of partially stabilized leachate yielding satisfactory results

\* Corresponding author.

E-mail address: [fengccc@ku.ac.th](mailto:fengccc@ku.ac.th) (C. Chiemchaisri).

(Chiemchaisri et al., 2011). Nevertheless, greenhouse gases (GHGs) can be significantly produced from biological activities during the treatment as methane ( $\text{CH}_4$ ) gas could be produced under anaerobic condition during the initial step of treatment (Yan et al., 2014) and recirculation of sludge between the anaerobic and aerobic reactors yield alternate oxygen absence and presence condition which is favorable for nitrous oxide ( $\text{N}_2\text{O}$ ) production (Aboobakar et al., 2013). Significant emission of  $\text{CH}_4$  could occur at the anaerobic zone of leachate treatment system under high organic loading (Chiemchaisri et al., 2009). Moreover, high nitrogenous characteristics of leachate could lead to significant  $\text{N}_2\text{O}$  emissions soon after raw leachate was aerated (Lin et al., 2008). The production of  $\text{CH}_4$  and  $\text{N}_2\text{O}$  during leachate treatment depends on aerobic and anaerobic conditions in the treatment system and it varied from one treatment technology to the others. There were only few studies conducted on  $\text{N}_2\text{O}$  emissions during landfill leachate treatment even though high emissions are anticipated during the treatment of this nitrogen-rich leachate in the past years (Barton and Atwater, 2002). Only recent researches have been reported that several factors could influence the emission of  $\text{CH}_4$  and  $\text{N}_2\text{O}$  during wastewater and leachate treatment. They include the configuration of wastewater treatment process (Yan et al., 2014) with oxygen profile as an important parameter (Aboobakar et al., 2013), wastewater characteristics (Wang et al., 2014; Liu et al., 2014). Nevertheless, the acquired knowledge is still far from complete understanding of GHG emission during wastewater treatment processes as they could be operated under wide range of environmental conditions. For instance, the  $\text{N}_2\text{O}$  emission were greatly influenced by dynamic conditions and variation of influencing parameters including oxygen, nitrite and ammonium concentrations during nitrification and denitrification reactions (Kampschreur et al., 2008). Among the factors affecting  $\text{CH}_4$  and  $\text{N}_2\text{O}$  emission during leachate treatment being reviewed, there was still very limited information on the effect of hydraulic retention time (HRT) and sludge recirculation on their emissions especially from the treatment systems operated under high biomass concentration such as MBR. Therefore, this study was carried out to investigate  $\text{CH}_4$  and  $\text{N}_2\text{O}$  gas emission characteristics from the two-stage MBR system incorporating anaerobic and aerobic conditions during the treatment of leachate under different HRT and sludge recirculation during long term operation (>500 days) in order to determine their emission rate during steady operating condition and understand the effect of hydraulic condition on their emissions from the treatment system. Furthermore, detailed characterization of microbial community using molecular biology technique in the two-stage MBR operated under different conditions was carried out for better understanding of the dynamic of GHG producing microorganisms in the system.

## 2. Methodology

### 2.1. Experimental system

A laboratory scale MBR unit consisting of anaerobic and aerobic reactors with 30 L working volume each was used. The schematic diagram of the experimental system is shown in Fig. 1. In the anaerobic reactor, an inclined tube module (2.5 cm opening) was installed for separating sludge from the mixed liquor so that sludge can be stored inside the system while allowing low mixed liquor suspended solids (MLSS) overflow into the aerobic tank. Anaerobic condition were kept in major part of the reactor especially beneath the inclined tube module. Meanwhile, periodical aeration was supplied at the surface of the reactor above the inclined tube module to maintain dissolved oxygen (DO) at about 0.5 mg/l for odor control purpose. In aerobic tank, a hollow fiber membrane module

(Mitsubishi Rayon, PVDF, 0.4  $\mu\text{m}$  pore size, 0.077  $\text{m}^2$  surface area) was used for solid–liquid separation. Intermittent suction (5 min on and 1 min off) was performed for withdrawal of filtrate water through the membrane module. The aeration was continuously supplied to the aerobic reactor by maintaining DO level at 3–4 mg/l. HRT in each reactor was set at 5 days during 1st and 2nd experiment run and 2.5 days during 3rd and 4th experimental run. Consequently, average membrane permeate flux was 0.1 and 0.2  $\text{m}^3/\text{m}^2 \text{d}$  at HRT of 5 and 2.5 days respectively.

The study was carried out in four experimental runs. During the 1st run (day 1–275) and 3rd run (day 410–496), the system was operated without sludge re-circulation. During the 2nd run (day 276–409) and 4th run (day 497–575), the system was operated with sludge re-circulation from aerobic reactor back to anaerobic reactor at 100% of influent feed flow rate. Under each operating condition, the system was operated until steady condition in terms of water qualities and gas emission rate have been reached.

### 2.2. Leachate preparation and water quality analyses

Raw leachate was obtained from solid waste collection trucks entering a solid waste disposal site in Thailand. The wastewater samples were kept inside glass containers and stored at a temperature of 4 °C. Prior to analysis, the waste water samples were filtered through the glass microfiber filter (GF/C). All leachate sample analysis was performed according to Standard Methods for the Examination of Water and Wastewater (APHA, 2005). The analytical parameters included pH, DO, BOD, COD, TOC, SS,  $\text{NH}_3$ , TKN,  $\text{NO}_2^-$  and  $\text{NO}_3^-$ . In the reactors, MLSS concentrations were monitored.

Chemical characteristics of leachate used in this study are shown in Table 1. The leachate used exhibited high organic concentrations in terms of BOD, COD and TOC and acidic in nature. Feeding leachate used in the laboratory scale MBR was prepared by mixing fresh leachate and tap water at about 1:1.5 v/v ratio to maintain BOD and COD concentrations in the feed leachate at about 20,000 and 40,000 mg/l to ensure steady operation and greenhouse gas production along the experimental period.

### 2.3. Determination of greenhouse gas emission

GHG emission was determined on regular basis from the anaerobic and aerobic reactors during the MBR operation. For the anaerobic reactor, a closed-flux chamber was occasionally placed on top of anaerobic reactor to determine greenhouse gas emission from the system. Close flux chamber is made of acrylic plate with 250-mm width, 300-mm in length and 100-mm in height. During the measurement, special care was taken to make sure that there are no gas leakage. The coverage area of the chamber was 0.075  $\text{m}^2$ . In order to determine the emission rate, gas samples from the closed-flux chamber were collected into a 9-ml vial by a gas-tight syringe at different time intervals (e.g. every 30 min) up to 120 min. Then, gas composition in a vial was analyzed by using a gas chromatograph (GC). For  $\text{CH}_4$  analysis, GC (Agilent 6890) with thermal conductivity (TCD) and Alltech-CRT column was used. For  $\text{N}_2\text{O}$  analysis, GC (Shimadzu Clarus 580) with thermal conductivity (ECD) installed with Heyesep D column was used. Closed flux chamber operated by allowing upward diffusive gas to accumulate in the chamber. As, the area of flux chamber and reactor was equal, the increasing rate of gas in the chamber was used to determine the mass of emitting gas as follows.

$$F_{\text{AN}} = \rho V \Delta C / \Delta t \quad (1)$$

where  $F_{\text{AN}}$  = mass of gas emitted from anaerobic reactor;  $\rho$  = density of gas;  $V$  = volume of chamber;  $\Delta C / \Delta t$  = gas concentration gradient.

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