

# Evaluation of Greenhouse Gas Emission from Municipal Solid Waste Leachate by two-stage Sequencing Batch Reactor

Nararatchporn Nuansawan<sup>1</sup>, Kwannate Sombatsompop<sup>1</sup> and Chayanid Witthayaphirom<sup>2</sup>

<sup>1</sup>Department of Civil and Environmental Engineering Technology, College of Industrial Technology, King Mongkut's University of Technology North Bangkok 1518 Pracharat 1 Rd., Bangsue, Bangkok, Thailand, 10800

<sup>2</sup>Department of Environmental Engineering, Faculty of Engineering, Kasetsart University 50 Ngam Wong Wan Road, Chatuchak, Bangkok, Thailand, 10900

## ABSTRACT

This work investigated greenhouse gas emission during the treatment of municipal solid waste leachate by two-stage Sequencing Batch Reactor (SBR). The SBR was carried out by anaerobic reactor followed by aerobic reactor. The system was operated at hydraulic retention times (HRT) of 4 and 2 days during 130 operating days. At steady state, the organic removal efficiencies were found to be 67.0% and 62.7% for 4 and 2 days of HRT. The organic carbon and nitrogen were mainly removed in aerobic reactor. The surface emission rates of methane in anaerobic reactor were 0.181 and 0.292 g/m<sup>2</sup>.d under HRT of 4 and 2 days, respectively. The emission factors of CH<sub>4</sub> at HRT 4 and 2 days were 0.297 and 0.238 gCH<sub>4</sub>/gCOD, respectively while those of N<sub>2</sub>O were 0.25 and 0.19 gN<sub>2</sub>O/gN for HRT of 4 and 2 days, respectively. The PCR technique confirmed that the microbial group was methanogenic bacteria which corresponded to the emission of greenhouse gas.

**Type of Paper:** Empirical

**Keywords:** Greenhouse gas; leachate; methane emission; microbial communities; sequencing batch reactor

## 1. Introduction

Leachate pollution from solid waste disposal is receiving more attention as the increase in the amount of solid waste collected from urban areas is dumped into landfills or open dumpsites, especially in developing countries. More stringent regulations on leachate control have been put forward for a better management of solid waste disposal sites. Municipal solid waste leachate contains other compounds, including organic substances and toxic substances.

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\* Corresponding author: Nararatchporn Nuansawan

E-mail: Nararatchporn.n@cit.kmutnb.ac.th

Affiliation: College of Industrial Technology, King Mongkut's University of Technology North Bangkok

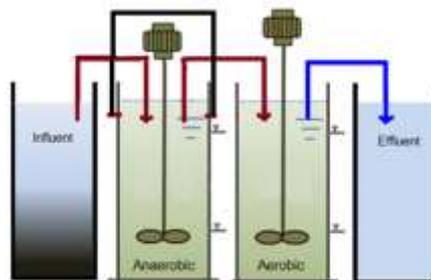
In order to meet the standards set for the release of leachate into the natural environment, an integrated treatment method, such as a combination of chemical physical and biological procedures (Wiszniewski et al., 2006) is required.

A study on the treatment of waste has developed rapidly (Kurniawan et al., 2006; Abbas et al., 2009; Gao et al., 2015; Nuansawan et al., 2018) including sequencing batch reactor (SBR), which is widely used for biological nutrient removal (BNR) in municipal and industrial wastewaters (Wang et al., 2014). Sequencing batch reactor (SBR) has become a global researcher's focus to optimize operational flexibility, space saving, and operating costs (Chamchoi and Nitorisavut, 2007; Jaramillo et al., 2018).

This study aims to develop a two-step SBR process. However, greenhouse gases (GHGs) can be produced significantly from biological activity during treatment because methane (CH<sub>4</sub>) can be produced under anaerobic conditions in the initial stages of treatment (Yan et al., 2014). Significant CH<sub>4</sub> emissions can occur at non-oxygenated areas of the leachate system under high loading (Nuansawan et al., 2016). In addition, the appearance of high levels of nitrogen in the leachate can cause the emission of N<sub>2</sub>O significantly soon after the raw leachate is aerated (Lin et al., 2008). N<sub>2</sub>O is released during nitrogen removal since N<sub>2</sub>O is produced by autotrophic nitrifying bacteria, most of which are ammonia oxidation bacteria (Kampschreur et al., 2008) during the nitrification, but most of them would be produced during denitrification (Aboobakar et al., 2013). Oxygen profiles are important variables influencing the release of CH<sub>4</sub> and N<sub>2</sub>O during wastewater, depending on aerobic and anaerobic conditions and leachate treatment (Yan et al., 2014). Among the factors affecting CH<sub>4</sub> and N<sub>2</sub>O emissions during leachate treatment, there is still very limited information on the impact of hydraulic retention time (HRT) on SBR emissions being investigated. Therefore, this study was carried out to investigate CH<sub>4</sub> and N<sub>2</sub>O gas emission characteristics from the two-stage SBR system incorporating anaerobic and aerobic conditions during the treatment of leachate under different HRT during operation of more than 130 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, confirmed microbial group using molecular biology technique in the two-stage SBR operated under different conditions was carried out for better understanding of the dynamic of GHG produced in the system.

## 2. Methodology

Lab-scale SBR unit with capacity of 1.5 and 3 l/d was used in this study. The schematic diagram of the experimental system is shown in Figure 1. The system consisted of two treatment steps. The anaerobic and aerobic reactors have 0.005 m<sup>3</sup> working volume. The aeration was continuously supplied to the aerobic reactor which maintained DO level of 4-5 mg/l. Hydraulic retention time (HRT) in both tanks was kept at 4 and 2 days. The study was conducted in two experimental systems. Systems 1(HRT 4 days) and 2(HRT 2 days), which last for 130 days, are operated continuously from anaerobic to aerobic reactors. The two systems have been used to provide stable conditions in terms of



water quality and emissions.

Figure 1. Schematic of two-stage SBR system

## 2.1 Leachate preparation and water quality analyses

Raw leachate was obtained from solid waste collection trucks into waste disposal area. Wastewater samples were kept in a glass container and stored at 4°C. Before analyzing, the wastewater samples were filtered through a glass microfibre filter (GF/C). All leachate analysis was performed according to the standards for water and wastewater (APHA, 2005). The water parameters used in the analysis include pH, DO, BOD, COD, TOC, SS, NH<sub>3</sub>, TKN, NO<sub>2</sub> and NO<sub>3</sub>, while greenhouse gases include CH<sub>4</sub> and N<sub>2</sub>O. Chemical characteristics of leachate are shown in Table 1. The leachate used exhibited high organic concentrations in terms of BOD, COD and TOC and was acidic in nature. SBR was prepared by mixing fresh leachate and tap water at a ratio of 1: 3 v/v to maintain a constant COD concentration in leachate feed. The average concentration was 7,760 mg COD/L and was to be consistent in feed water.

Table 1. Chemical characteristics of raw and feed leachate

Parameters	Raw leachate		Diluted leachate used in the experiment	
	Range	Average(SD)	Range (System I & II)	Average (System I & II)
pH	3.1-4.3	3.80(0.41)	4.7-5.7	5.14(0.42)
BOD(mg/l)	52,350-68,700	61,030(5,218)	4,950-5,570	5,130(170)
COD(mg/l)	71,550-85,300	76,230(4593)	7,300-8,350	7,760(369)
TOC(mg/l)	17,250-22,460	19,690(2,011)	2,470-3,290	2,960(309)
SS(mg/l)	320-1,350	690.5(21)	78-450	225(115)
NH <sub>3</sub> -N(mg/l)	2,420-3,260	2,802(291)	720-840	780(47)
TKN(mg/l)	2,610-3,540	3,150(308)	810-1,020	897(70)
NO <sub>2</sub> -N(mg/l)	0.3-0.8	0.6(0.20)	0.3-0.5	0.4(0.01)
NO <sub>3</sub> -N(mg/l)	0.8-2.7	1.9(0.6)	0.3-2.1	1.4(0.7)

Note: The numbers show avg. (SD) values

## 2.2 Determination of greenhouse gas emission

During the operation, a closed-flux chamber was occasionally placed on top of anaerobic and aerobic reactors to determine greenhouse gas emission from the system. Close flux chamber is a chamber made of plastic plate with 150-mm in diameter and 100-mm in height. During the measurement, special care was taken to make sure that there were not any gas leakages. The reactor surface area which covered the chamber was 0.018 m<sup>2</sup>. 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 minutes) up to 120 minutes. Then, gas composition in a vial was analyzed by using a gas chromatograph (GC). For CH<sub>4</sub> and N<sub>2</sub>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.

$$F_{AN} = \frac{V\Delta C(298)}{A\Delta t(273+T)} \quad (1)$$

Where  $F_{AN}$  = Mass of gas emitted from anaerobic (g/m<sup>2</sup>.d) at 25°C ;  $V$  =volume of chamber (m<sup>3</sup>);  $\Delta C/\Delta t$  = gas concentration gradient (g/m<sup>3</sup>.d);  $T$  = temperature measured in degree: Celsius (°C). The gas emission was measured from anaerobic reactor at different times along the operation period. For the determination of gas emission from aerobic reactor, gas samples were collected from the cover chamber equipped with gas outlet port. The size of cover chamber was identical to that used in anaerobic reactor. The gas emission was determined from supplied air flow rate and measured outflow gas concentration using the following equation.

$$F_{AE} = Q_{air} C/A \quad (2)$$

Where  $F_{AE}$  = Flux of gas emitted from aerobic reactor (g/m<sup>2</sup>.d),  $Q_{air}$ = supplied air flow rate (m<sup>3</sup>/d),  $C$  = outflow gas concentration (g/m<sup>3</sup>) and  $A$ = area of the cover chamber (m<sup>2</sup>).

### 2.3 Determination of Biochemical Methane Potential (BMP)

The method used to analyze the BMP is a modified version as described by Luna-delRisco et al., 2011. In this study, 3.0 g of sludge in anaerobic wastewater treatment tank was added to a 118 ml of each sample. Next, 60mL of leachate in each tank was added. The growth medium was seeded with the sludge to promote the growth of anaerobic microbes. After doing so, they were sealed and mixed by shaking and then placed in an oven at 37°C. The total gas was recorded as the headspace in the bottle added to the volume of gas collected in the sampling tubes. Each sample was then run through a Gas Chromatograph 6890 (TCD). Using blanks, the overall methane potential was reported as litres of methane (STP) per gram of COD removed (L CH<sub>4</sub>/g COD removed).

### 2.4 Confirm group of microbes that produce methane.

The PCR was performed using 338GC-F and 518R primers for most bacteria 344GC-F and Univ522R for methanogen via Toptag Master Mix Kit, Quiagen by Swift™ MaxPro thermal cyclers (Esco Healthcare Pte. Ltd). The combination of these primers generated a PCR fragment about 190-200 bp. The PCR amplification of genes was then performed according to the following conditions; 25 µl of PCR reaction containing 5 µl of genomic DNA template, 0.25 µl of each primer (8µmol), 7 µl of H<sub>2</sub>O and 12.5 µl of taq DNA polymerase (Qiagen, Germany). The PCR was conducted in a Perkin-Elmer GeneAmp PCR System 9700 (Applied Biosystems, USA). The thermal program of PCR was set as follows; initial denaturation at 100°C for 3 minutes, followed by 26 cycles of denaturation at 100°C for 30 seconds, annealing at 58°C for 30 seconds and extension at 72°C for 1 minute. Finally, the extension step was performed at 72°C for 10 minutes. The size of PCR products were visualized on a 1.2% (w/v) agarose gel electrophoresis staining with ethidium bromide.

## 3. Results

### 3.1 Treatment performance of SBR

During the 1st system (HRT 4 day), the BOD and COD efficiencies in the SBR system were 63.9% and 52.8%, while the Anaerobic reactor was removed 25.0% and 12.2%, respectively. NH<sub>3</sub> and TKN removals were also higher than 50% as shown in Table 2. Most of nitrified nitrogen was denitrified resulting in low concentrations of oxidized nitrogen. In the 2nd system (HRT 2 day), slightly higher COD and TKN removal efficiencies of 32.2% and 33.7% were obtained but the increased removal efficiencies were not significant. At these lowered HRT conditions, much higher biodegradable organic (BOD) concentrations were detected in the effluent of aerobic reactor even though the effluent

from first stage anaerobic reactor were only moderately elevated. These results suggested that long HRT had insignificant effect on the SBR performance on organic and nitrogen removals. Biomass concentration in aerobic reactors is regularly monitored. At the start of the MLSS, the aerobic reactor gradually increased from 5,370 to 8,110 mg/l (HRT 4 d) and 5,490 to 7,090 mg/d (HRT 2 d). It was found that HRT and long-term aerobic conditions influenced the concentration of biomass in aerobic reactors. It does not affect the overall performance of SBR system.

Table 2 Effluent qualities from SBR during steady operation

Parameters	HRT 4 day			HRT 2 day		
	Eff.	Eff.	%	Eff.	Eff.	%
	(An-SBR)	(Ae-SBR)	Removal	(An-SBR)	(Ae-SBR)	Removal
pH	7.2(0.3)	8.9(0.3)	-	6.4(0.2)	7.9(0.2)	-
DO	0.0(0.02)	3.7(0.40)	-	0.3(0.1)	2.5(0.2)	-
BOD	3,854(139)	1,854(136)	63.9	4,597(185)	2,764(173)	46.2
COD	6,801(93)	3,652(143)	52.8	7,493(401)	5,250(302)	32.2
TOC	1,910(139)	972(109)	67	2,170(139)	1,094(142)	62.7
NH+3-N	602(44)	387(31)	50.2	666(31)	391(31)	49.8
TKN	653(32)	410(16)	54.5	723(27)	597(27)	33.7
NO-2-N	0.08(0.07)	0.3(0.07)	47.4	0.19(0.10)	0.3(0.09)	46.8
NO-3-N	0.71(0.16)	0.6(0.08)	57.8	0.94(0.2)	0.73(0.11)	54.2

Note: The numbers show avg. (SD) values

### 3.2 Measurements of greenhouse gas emissions from SBR

Table 3 presents surface emission rates of CH<sub>4</sub> and N<sub>2</sub>O from anaerobic and aerobic reactors during leachate treatment. During the 1st System (HRT4 days), average CH<sub>4</sub> emission rate from anaerobic and aerobic reactors was 0.181 and 0.012 g/m<sup>2</sup>.d. This is equivalent to CH<sub>4</sub> mass of 0.003 and 0.0003 g/d. Meanwhile, N<sub>2</sub>O emission rate was 0.017 and 0.002 g/m<sup>2</sup>.d. These results show that both greenhouse gases were mainly emitted from the anaerobic reactor. When HRT 2 day operated in the 2nd system, CH<sub>4</sub> emission rates were found increasing in both reactors. Meanwhile, N<sub>2</sub>O emission rates were found at lower level of 0.0003g/d only for aerobic reactor. These results show that both GHG were mainly emitted from the anaerobic reactor. Similar observation on the CH<sub>4</sub> emission trend along the treatment process was reported in Wang et al. (2014). The major source of CH<sub>4</sub> emission came from the first reactor which was favorable for methanogens. In this study, comparable N<sub>2</sub>O emission found in aerobic reactors was found to increase. Previous studies have reported much higher N<sub>2</sub>O emission from aerobic reactor mainly due to air stripping mechanism (Yan et al., 2014; Wang et al., 2014). Meanwhile, Anaerobic reactor N<sub>2</sub>O production could take place where DO was maintained at about 0.5 mg/l. In previous research, it was reported that high N<sub>2</sub>O production was observed under a DO level less than 2 mg/l (Nuansawan et al., 2016; Kampschreur et al., 2008) as N<sub>2</sub>O was produced from denitrification instead of N<sub>2</sub> in low oxygen condition (Itokawa et al., 2001).

Table 3 CH<sub>4</sub> and N<sub>2</sub>O emission from anaerobic and aerobic reactors of SBR system

Conditions	GHGs	Anaerobic(g/m <sup>2</sup> .d)		Aerobic(g/m <sup>2</sup> .d)	
		Range	Avg	Range	Avg

HRT 4 day	CH <sub>4</sub>	0.142-0.297	0.181	0.008-0.018	0.012
	N <sub>2</sub> O	0.015-0.048	0.027	0.0017-0.0025	0.002
HRT 2 day	CH <sub>4</sub>	0.212-0.396	0.292	0.011-0.023	0.002
	N <sub>2</sub> O	0.028-0.049	0.042	0.0015-0.0018	0.0017

the experiment at steady condition, (At 1st system, HRT 4 day) COD and TN loading to the system was 11.64 and 1.34 mg/d. At 2nd system, COD and TN loading to the system was 23.27 and 2.70 mg/d. Therefore, emission factors for CH<sub>4</sub> and N<sub>2</sub>O of 0.297 gCH<sub>4</sub>/gCOD loading and 0.250 gN<sub>2</sub>O/gN loading. (At 2nd system, HRT 2 day) found that 0.238 gCH<sub>4</sub>/gCOD loading and 0.190 gN<sub>2</sub>O/gN loading for combining the emission from both reactors can be calculated. Frison et al., 2015 reported that low N<sub>2</sub>O emission (0.24% of influent nitrogen load) could be achieved during biological nitrogen removal from anaerobic effluents through the operation at sufficient DO level and lower volumetric nitrogen loading than nitrogen removal capacity of the system.

### 3.3 BMP of influent and effluent leachate

Methane producing potential of influent, treated leachate from effluent reactors from two-stage SBR were examined to determine the effect of treatment on GHG (methane) mitigation. The total volume of gas that was produced in the assay bottles was measured until day 55 when the experiment was ended due to low gas production rate from the anaerobic sludge. The experimental results are illustrated in Figure 2. During the BMP test, it was found that methane production became constant for most samples after 40 days of incubation among which aerobically treated samples reached maximum production after 10-20 days. The untreated leachate had the highest rate of methane production, but produced methane for a longer period of time than the treated leachate. Furthermore, lag period (10-20 days) for methane production was found for all influent samples indicating the requirement for hydrolysis reaction of organic substances presented in leachate prior to methane production. The BMP values obtained from this study were between 0.19-0.28 L CH<sub>4</sub>/gCOD<sub>removed</sub>. They were comparable to the previously published reported figures, i.e. 0.114-0.334 L CH<sub>4</sub>/gCOD<sub>removed</sub> (Elsayed et al., 2012; Gimenez et al., 2011; Harish et al., 2014; Huang et al., 2013; Yoo et al., 2012), However, BMP of samples depend heavily on the types of wastewater employed in the testing. Produced biogas through anaerobic fermentation process (digestion) should ensure a favorable environment for the development and optimal metabolic activity of microorganisms involved in the process. The fermentation medium must fulfill several important conditions such as availability of biodegradable organic matter; favorable condition, e.g. pH between 6.8 – 7.3 and C/N ratio between 15 and 25.

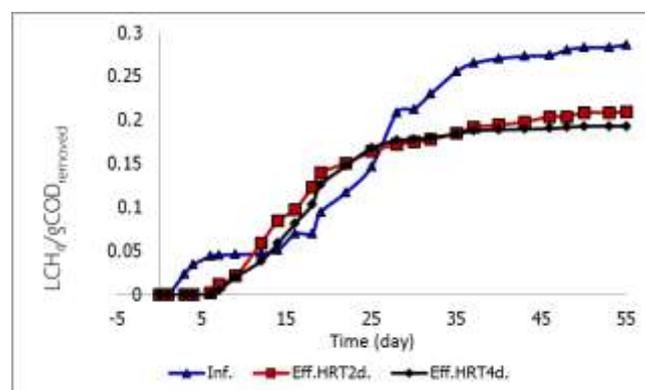


Figure 2. BMP of treated and untreated samples for all experimental.

#### 4. Conclusion and discussion

An experimental study on greenhouse gas emission from two-stage SBR treating highly concentrated leachate suggested CH<sub>4</sub> gas were mainly emitted from first anaerobic stage at an average rate of 0.18 and 0.29 g/m<sup>2</sup>.d at HRT 4 and 2 day. Meanwhile, the emissions from second aerobic reactor were 0.012 and 0.017 g/m<sup>2</sup>.d, respectively. These emissions accounted 0.086 and 0.139 g/d of CO<sub>2</sub> equivalent of 0.06% and 0.05 of C at HRT 4 and 2 day, respectively. Increases in hydraulic loading through shortened HRT increased CH<sub>4</sub> emission by 37%. Based on this conclusion, it is recommended to run the system at a very high storage capacity, which will improve the efficiency of wastewater treatment and reduce greenhouse gas emissions.

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