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# Leachate treatment and greenhouse gas emission in subsurface horizontal flow constructed wetland

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#### ARSTRACT

Organic and nitrogen removal efficiencies in subsurface horizontal flow wetland system (HSF) with cattail (*Typha augustifolia*) treating young and partially stabilized solid waste leachate were investigated. Hydraulic loading rate (HLR) in the system was varied at 0.01, 0.028 and 0.056  $\rm m^3/m^2$  d which is equivalent to hydraulic retention time (HRT) of 28, 10 and 5 d. Average BOD removals in the system were 98% and 71% when applied to young and partially stabilized leachate at HLR of 0.01  $\rm m^3/m^2$  d. In term of total kjeldahl nitrogen, average removal efficiencies were 43% and 46%. High nitrogen in the stabilized leachate adversely affected the treatment performance and vegetation in the system. Nitrogen transforming bacteria were found varied along the treatment pathway. Methane emission rate was found to be highest at the inlet zone during young leachate treatment at 79–712  $\rm mg/m^2$  d whereas CO<sub>2</sub> emission ranged from 26–3266  $\rm mg/m^2$  d. The emission of N<sub>2</sub>O was not detected.

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#### 1. Introduction

One of the difficulties in dealing with solid waste landfill leachate is its wide variation in both quantity and characteristics terms. Various treatment methods have been applied to purify this problematic wastewater ranging from natural to mechanical intensive treatment systems. When considering leachate characteristics, organic substances and nitrogen are the major pollutants which need to be removed. However, high attention has been recently paid on nitrogen removal (Pelkonen et al., 1999; Kalyuzhnyi and Gladchenko, 2004) especially when dealing with leachate from old landfill sites. Advanced leachate treatment systems using biological and chemical treatment methods are recently adopted in developed countries but high investment and operating costs limited their application in most of the developing countries. Natural based treatment systems such as constructed wetland would to be more appropriate and practical for their treatment as the systems have significant merits of low cost and versatile removal mechanisms (Lee et al., 2004). In most of the cases, they are used as post-treatment or polishing systems. Direct application of constructed wetland to high strength wastewater especially leachate is still limited.

From the previous studies, it is well understood that nitrogen removal in subsurface horizontal flow constructed wetland is normally low because of limited oxygen availability for nitrification (Vymazal, 2007). This is true when the system is applied to the treatment of diluted organic wastewater and operated at relatively high hydraulic loading rate (Vymazal and Kröpfelová, 2008). During the treatment of landfill leachate, hydraulic loading in the system is usually kept at a low rate because it contains high organic concentration. Therefore, majority of biodegradable organics is possibly removed near the inlet zone and allowing nitrification to take place in the latter stage of the treatment when organic concentration become exhausted. This study is focusing on the removal of organic carbon and nitrogen with special attention on the existence of nitrogen transforming bacteria in this type of constructed wetland.

Despite of many advantages, constructed wetland emits considerable amount of methane ( $CH_4$ ), carbon dioxide ( $CO_2$ ), and nitrous oxide ( $N_2O$ ) gases that are formed under anoxic condition of inundated area.  $CH_4$  and  $N_2O$  have global warming potential (GWP) of 23 and 296 times in relative to  $CO_2$  over a 100 year time horizon. The gas fluxes from the wetland also have a strong seasonal and temporal variability resulting from variation in the environmental factors regulating the microbial processes (Liikanen et al., 2006).

This study deals with direct application of subsurface horizontal flow constructed wetlands to young and partially stabilized municipal solid waste leachate. The organic carbon and nitrogen removal efficiencies were evaluated at different hydraulic loading rates. The emission of target greenhouse gases, i.e.  $CH_4$ ,  $CO_2$  and  $N_2O$ , were measured during the treatment. Microbiological study targeting nitrifying bacteria in the systems was also conducted using fluorescent in situ hybridization (FISH) technique. This

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molecular biology method has been reported to be useful for the study of microbial population in constructed wetland system (Baptista et al., 2003; Shipin et al., 2005). The target microorganisms locating in soil and surrounding plant root along the treatment system are determined to correlate microbial populations with nitrogen transformation in the system.

#### 2. Methods

#### 2.1. Experimental system

Subsurface horizontal flow (HSF) constructed wetland system was used in this study. Four concrete ponds of 1 m wide 3 m long and 1 m depth were used. The inlet and outlet zones were filled with 30–60 mm gravel of 0.8 m depth and 1–2 mm sand was used in plantation zone. The water depth was 0.7 m, i.e. 0.1 m below the gravel surface level. The wastewater was fed into the system by a centrifugal pump through an inlet pipe (10 mm in size) with valve control. The wastewater moved downward and transported horizontally through the treatment (plantation) zone and discharged from the outlet zone through an outlet pipe (50 mm size). The sampling pipes (50 mm size inserted to 500 mm depth into the sand layer) were provided at three different locations, i.e. inlet, middle and outlet zone. Cattail (*Typha augustifolia*) was used as emergent in the system with initial planting density of 40 rhizomes/m².

#### 2.2. Experimental condition and water quality analyses

Two different types of wastewater were used, i.e. representing young and partially stabilized waste leachate. Their chemical characteristics are shown in Table 1. Fresh leachate was collected from a solid waste transfer station in Bangkok. It was diluted with tap water to obtain final COD of 5000–10,000 mg/L. For partially stabilized waste leachate, collected wastewater from leachate storage pond in a closed landfill was used. Influent COD was controlled in range of 3000–5000 mg/L in the first experiment (Run I). It was diluted by rainwater in subsequent experiments (Runs II and III) in order to control nitrogen loading into the system.

The treatment efficiencies of the system were examined at different hydraulic loading rates (HLR) of  $0.01~\text{m}^3/\text{m}^2$  d (Run I),  $0.028~\text{m}^3/\text{m}^2$  d (Run II) and  $0.056~\text{m}^3/\text{m}^2$  d (Run III). They were equivalent to the feeding rates of 20, 56 and 112 L/d and hydraulic retention times (HRT) of 28, 10 and 5 d respectively. Water quality analyses included temperature, pH, BOD, COD, SS, NH<sub>3</sub>–N, TKN, NO $_2^-$ , NO $_3^-$ , PO $_3^4$ , Cl $^-$  and electrical conductivity (EC). Oxidation reduction potential (ORP) of underlying soil at the inlet, middle and outlet zones of constructed wetland unit was also monitored. All the analyses were performed according to standard methods for the examination of water and wastewater (APHA, 1989)

**Table 1**Chemical characteristics of young and partially stabilized leachate.

pH - 4.3-6.5 8.2-8.5 7.9-9.2 BOD mg/L 3150-7400 209-278 15-68 COD mg/L 5850-12,820 1613-4506 414-2184 SS mg/L 320-825 124-223 10-158	achate
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ND. not detected.

#### 2.3. Nitrogen transformation bacteria population

Relative comparison of nitrogen transforming bacteria populations in soil and surrounding plant root at 0.40 m depth of gravel bed (or 0.2-0.3 m from the water surface) along the treatment pathway was determined. This sampling depth was selected based on the penetration depth of plant root above which the aerobic condition prevailed. Fluorescent in situ hybridization (FISH) technique was used to study 3 targeting bacteria groups in the system, i.e. ammonium oxidizing bacteria (AOB), nitrite oxidizing bacteria (NOB) and ANNAMOX bacteria (AMX). Specific 16 S rRNA-targeted oligonucleotide probes, i.e. Nsm156 (Nitrosomonas spp.) representing AOB, NIT3 (Nitrobacter spp.) representing NOB and Amx820 (AMX) were used. The probes were labeled with fluoroscein isothiocyanate (FITC). The fluorescent area obtained from each specific probe is determined as percentage of the targeting bacteria to the total microorganisms (stained with 4'.6-diamidino-2-phenylindole or DAPI).

#### 2.4. Greenhouse gas emission measurement

Close flux chamber technique was used for the measurement of greenhouse gas emission. The chamber was made of acrylic plate with 0.3 m diameter and 0.3 m height. It is covered by acrylic plate that having gas sampling and temperature measurement ports. The chamber base was made of stainless steel with 0.3 m diameter and 0.125 m height. The base part was inserted in to the soil one day before the gas measurement at the inlet, middle and outlet zones of experimental unit. Gas samples were collected into a 5 ml vial by a gas-tight syringe from the chamber at 15 min interval for 6 h. The gas samples were then analyzed for their CH<sub>4</sub>, CO<sub>2</sub> and N<sub>2</sub>O concentrations using gas chromatograph (HP6890, Agilent) equipped with Alltech-CRT and Heyesep Q, 80/100 columns, and a thermal conductivity detector, using helium as the carrier gas. The gas flux was then determined from its concentration increase in the chamber as described in the following equation.

$$F = \frac{V}{A} \frac{dC}{dt} \tag{1}$$

where F, is the gas flux (mg/m<sup>2</sup> d), V is the chamber volume (m<sup>3</sup>), A is the area enclosed by the chamber, (m<sup>2</sup>) and dC/dt is the gas concentration gradient (mg/m<sup>3</sup> d).

#### 3. Results and discussion

#### 3.1. Organic matter and nitrogen removal efficiencies

Table 2 shows the effluent qualities from the constructed wetland unit treating young and partially stabilized leachate at different hydraulic loading rates (HLR). In case of young waste leachate treatment, high organic removal of 98% and 94% in terms of BOD and COD was achieved during steady state conditions when the system was operated at HLR of 0.01 m<sup>3</sup>/m<sup>2</sup> d. Subsequent increase in HLR to 0.028 and 0.056 m<sup>3</sup>/m<sup>2</sup> d did not deteriorate organic removal efficiencies. They were between 94% and 99% resulting in average effluent BOD and COD concentrations of 32-136 and 364–757 mg/L, respectively. High BOD and COD removal occurred simultaneously with moderate suspended solid removal efficiencies (71-88%) as the major treatment mechanisms were sedimentation and filtration of suspended solids in gravel bed, plant uptake and biological degradation of organic substances by attached growth microorganisms under aerobic, facultative and anaerobic conditions in the top (rhizosphere), middle and bottom zones respectively (Stottmeister et al., 2003). Organic concentration profiles along constructed wetland unit (Fig. 1a) suggested that most

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