

## DESIGN OF LANDFILL COVER SYSTEMS INCORPORATING SOIL METHANOTROPHY FOR METHANE EMISSION MITIGATION

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### Abstract:

Methane (CH<sub>4</sub>) has 21 times the global warming potential (GWP) of carbon dioxide (CO<sub>2</sub>) over a 100-year time horizon, and has a shorter atmospheric residence time than CO<sub>2</sub>. Therefore, control of CH<sub>4</sub> from anthropogenic sources, including landfills, could produce substantially more global warming mitigation in the short term than controlling CO<sub>2</sub>. Extraction, with or without energy recovery, is the most common method currently used to control gas emissions from landfills. However, this method is not adaptable in most landfills around the world. Even in landfills with gas extraction, conventional techniques are capable of capturing only 40 to 90% of produced gas. The rest escapes into the atmosphere via the landfill cover. Therefore, there is a need to identify and develop techniques to control CH<sub>4</sub> emissions across covers in landfills. A modified design approach, based on utilizing the CH<sub>4</sub> oxidation capability of soils, is proposed. The modified cover design aims for maximum CH<sub>4</sub> oxidation without compromising the leachate generation control function of a cover system. Preliminary results from on-going controlled experiments from two parallel studies currently being undertaken in Alberta, Canada, and in Bangkok, Thailand, and information from other studies, show a high degree of methane oxidation in landfill cover soils. Our results show that methane oxidation could be as high as 65% in cover soils used in tropical landfills.

### INTRODUCTION

Methane (CH<sub>4</sub>) is one of the key greenhouse gases produced within a landfill accepting biodegradable organic waste materials. Emissions from landfills around the world are estimated to account for almost 20% of the worldwide anthropogenic CH<sub>4</sub> emissions into the atmosphere (Nozhevnikova et al., 1993). Methane has 21 times the global warming potential (GWP) of CO<sub>2</sub> over a 100-year time horizon, and has a shorter atmospheric residence time than CO<sub>2</sub>. Therefore, control of CH<sub>4</sub> from anthropogenic emissions could produce substantially more global warming mitigation in the short term than controlling CO<sub>2</sub>. Controlling CH<sub>4</sub> emissions from anthropogenic sources such as rice paddies, intestines of ruminants, wastewater and sludge treatment processes are usually difficult. On the other hand, control of emissions from landfills is easier. Considering these facts, there is renewed interest in controlling CH<sub>4</sub> emissions from landfills.

Extraction, with or without energy recovery, is the most common method currently

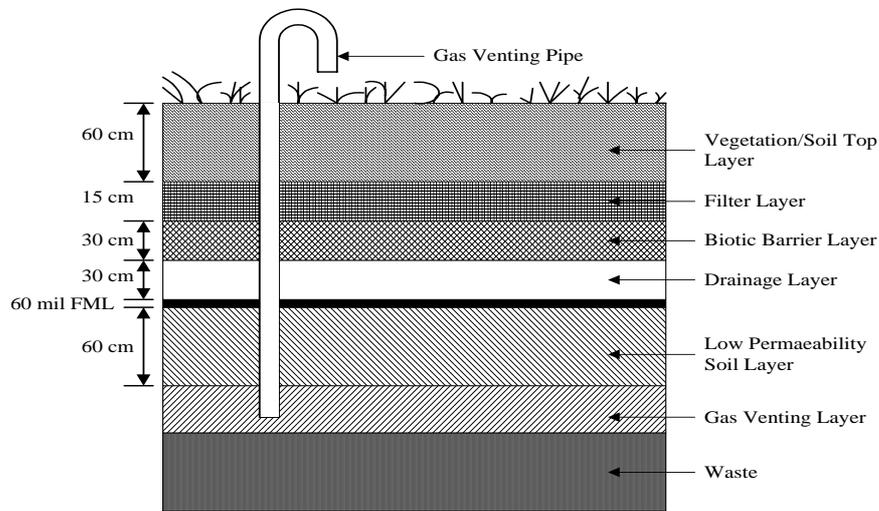
being used to control gas emissions from landfills. However, this method is not adaptable in most landfills in developed and developing countries. The US EPA recommends considering only the landfills containing more than 900,000 tonnes of solid waste for gas extraction and energy recovery (Gibbs and Bashki, 1996). Even in landfills with gas extraction, conventional techniques are capable of capturing only 40 to 90% of the produced gas (Augenstein and Pacey, 1996). The rest escapes into the atmosphere via the landfill cover. Therefore, there is a need to identify and develop techniques to control CH<sub>4</sub> emissions across final covers in landfills with or without gas extraction. This paper proposes a new design approach. The objective is to maximize the CH<sub>4</sub> oxidation capability of soils in a landfill cover. The approach is based on an evaluation of the current information on soil methanotrophy. Results from studies currently being undertaken at the University of Calgary, in Alberta, Canada, and at the Asian Institute of Technology, in Bangkok, Thailand to develop applicable design parameters are also presented.

### **CURRENT APPROACHES TO COVER SYSTEM DESIGN AND LANDFILL GAS EMISSION CONTROL**

The current landfill final cover system design approach is based on the desire to minimize post-closure leachate generation arising from percolation of rainfall and melting snow (US EPA, 1993). In modern landfills, cover systems convert precipitation into surface runoff or evapotranspiration, without eroding the cover. The US EPA requires at least an infiltration barrier layer overlain by an erosion layer. However, some of the alternate designs incorporate a gas emission control function into cover systems. Typically, such designs include a permeable layer for gas collection and a venting system, low permeability layer, drainage layer, biotic barrier layer, filter layer, and a vegetative layer (see Figure 1). The drainage layer removes percolating water that has infiltrated through the erosion layer. Entry of deep plant roots or burrowing animals is prevented by the biotic barrier layer. The filter layer prevents top soil from migrating into the biotic barrier.

The primary concern addressed by such a design is the gas pressure build-up in a landfill, which increases the lateral migration potential of landfill gas. Emission of landfill gas into the atmosphere, and related global warming impact, is not addressed. Flaring of collected gas is a possible solution to this problem. Although the conversion of CH<sub>4</sub> into CO<sub>2</sub> reduces the global warming impacts, partial combustion of landfill gas in a flare is known to produce undesirable by-products of human health concern. A more efficient system available for landfill gas control is a combination of an active gas collection system and power generation. High temperature burning under a controlled environment reduces the potential for formation of undesirable by-products.

The efficiency of an active gas collection system depends on the design and age of the landfill unit, soil characteristics, hydrogeologic, and hydraulic conditions of the facility and surrounding environment. Usually, the collection efficiency is believed to be in the range 40 to 90%. However, current concern on global warming effect of methane requires much more effective emission control. The potential of incorporating methanotrophy in landfill cover designs is important in this regard.



**Figure 1: Cross Section of a Landfill Cover System with Gas Venting (US E.P.A., 1993)**

**MIGRATION OF GAS ACROSS LANDFILL COVER SYSTEMS;  
Mathematical Modelling**

The design of cover systems to accommodate landfill gas control requires an understanding of the physical, chemical and biological processes governing gas migration. A comprehensive mathematical model to describe gas migration across landfill cover systems is described herein. Flow through a landfill cover is essentially multiphase, involving a water phase and a gas phase. The gas phase may include many gases, such as CH<sub>4</sub>, CO<sub>2</sub>, oxygen (O<sub>2</sub>), nitrogen (N<sub>2</sub>), and water vapour. The effect of moisture infiltration into a landfill cover has an impact on gas migration, but considering the time scales, such effects can be neglected. However, the effect of moisture saturation on gas permeability should be taken into account.

In a landfill cover, flow occurs both due to pressure and concentration gradients. For simplicity, only flow in the vertical direction is considered, assuming no lateral flow.

$$\phi \frac{\partial C_i}{\partial t} = \frac{\partial^2 (D_z C_i)}{\partial z^2} - u_z \frac{\partial C_i}{\partial z} \mp S \dots\dots\dots (1)$$

Where, C<sub>i</sub> = concentration of gas i (in volume percent); i = CH<sub>4</sub>, CO<sub>2</sub>, O<sub>2</sub>; D<sub>z</sub> = coefficient of dispersion in vertical direction (m<sup>2</sup>/s); u<sub>z</sub> = gas velocity in z direction (m/s); φ = gas porosity; S = source, sink, decay, or production rate (s<sup>-1</sup>)

Using Darcy's law;

$$u_z = - \frac{k}{\mu} \frac{\partial P}{\partial z} \dots\dots\dots (2)$$

Where, k = gas permeability (m<sup>2</sup>/s); μ = gas mixture viscosity (N.s/m) and P = pressure (Pa)

From the equation of state for gases;

$$P = R_1 C_1 + R_2 C_2 + R_3 C_3 \dots\dots\dots (3)$$

Where, R<sub>i</sub> = R/M<sub>i</sub>; R = universal gas coefficient, M<sub>i</sub> = molecular weight of gas i, T = absolute temperature (°K) and 1, 2, 3 represent the gases CH<sub>4</sub>, CO<sub>2</sub>, and O<sub>2</sub>

There are no sources or sinks within the cover itself. The only decay reaction of importance is CH<sub>4</sub> oxidation. Assuming Monod kinetics, the oxidation rate (..) is given by;

$$\beta = \frac{KXC_{CH_4}}{K_s + C_{CH_4}} \dots\dots\dots (4)$$

Where, K = maximum rate of CH<sub>4</sub> utilization per unit mass of micro-organisms, X = concentration of organisms; K<sub>s</sub> = half saturation constant and C<sub>CH<sub>4</sub></sub> = Concentration of CH<sub>4</sub>

At high CH<sub>4</sub> concentrations, the reaction rate follows zero order kinetics (assuming microbial concentration is constant). At low concentrations it is a first order reaction. The complete equation, which incorporates the physical and biological reactions, is as follows;

$$\phi \frac{\partial C_i}{\partial t} = D_z \frac{\partial^2 C_i}{\partial z^2} + \frac{k}{\mu} \frac{\partial}{\partial z} \left[ C_i \frac{\partial}{\partial z} (R_1 C_1 + R_2 C_2 + R_3 C_3) T \right] + \mp \beta \dots(5)$$

As evident from this equation, gas migration is controlled by the source strength of landfill gas, permeability of gas across cover soil, and factors that control soil methanotrophy. Although indirect control may be possible, direct control of gas source strength is not possible. However, gas permeability across cover soil and the factors affecting soil methanotrophy can be controlled by selecting appropriate design parameters.

**OXIDATION OF METHANE IN SOILS: Soil Methanotrophy**

Methanotrophy is the process of CH<sub>4</sub> oxidation by a specific type of bacteria residing in soils, known as “methanotrophs”. Methanotrophs possess the specific enzyme methane mono-oxygenase that enables them to utilise methane as a sole source of energy and as a major carbon source (Haber et al., 1983). They oxidise methane through methanol to formaldehyde, which they then either assimilate for the synthesis of cell material or further oxidise to carbon dioxide. All methanotrophic bacteria isolated and characterised to date have been gram negative, obligately aerobic, and have possessed intra-cytoplasmic membranes (Topp and Hanson, 1991).

Soil methanotrophy has been reported most commonly in agricultural soils, forest soils, tundra, and bogs (Topp and Hanson, 1991). More recently, researchers have isolated methanotrophic bacteria in landfill cover soils. According to their characteristics, methanotrophs can be categorised into two types: low oxidation capacity methanotrophs and high oxidation capacity methanotrophs. As the gases emitted by landfills contain a high amount of CH<sub>4</sub>,

landfill methanotrophy is concerned only with the latter type of methanotrophic bacteria.

### **THE APPROACH FOR LANDFILL COVER SYSTEM DESIGN INCORPORATING SOIL METHANOTRPHY**

A landfill final cover is a layered system, consisting of layers with specific functions. If methanotrophy is to be incorporated, it will be necessary to modify an existing layer or introduce a new layer. But, this should be done without compromising the other objectives.

Since methanotrophy requires O<sub>2</sub>, the layer responsible for CH<sub>4</sub> oxidation should be located at or near the surface. Nozhevnikova et al. (1993) reported penetration of O<sub>2</sub> up to a depth of about 60 cm. However, the top most layer in a landfill cover could encounter very low temperatures (eg. in Northern climates) and high moisture contents. Both these factors are known to substantially reduce the methanotrophic activity. The selection of an appropriate vegetative layer as the surface erosion layer could reduce potential negative impacts. For example, the use of a vegetation type with a high evapotranspiration capability would decrease the soil moisture. In addition, an extensive root system could make diffusivity of O<sub>2</sub> much easier.

Considering the above requirements, the parameters identified below should be specified in the new design approach which incorporates soil methanotrophy:

- Soil structure; as defined by grain size distribution and pore space distribution,
- Thickness of the soil methanotrophy/erosion layer
- Nutrient and organic content, and type of vegetation
- Gas flow rates for optimum methanotrophy.

Literature contains reports from several research studies which shed some light on the effect of these parameters on soil methanotrophy. These findings are discussed below, together with preliminary results from our own studies, in Canada and Thailand, to define the design parameters for landfill cover systems.

### **EFFECT OF VARIOUS PARAMETERS ON GAS EMISSION AND SOIL METHANOTROPHY; Results from Field Studies and Laboratory Experiments**

A summary of methane flux rates observed at various landfills are presented in Table 1. All field investigations, except our investigation in Thailand, have been conducted in temperate climates. Reported surface gas emission rates vary widely, ranging from a low of 0.003 g/m<sup>2</sup>.day to a high of 1000 g/m<sup>2</sup>.day. The maximum rate is reported at a landfill in Illinois by Bogner et al. (1995). Although one would expect higher methane flux rates in tropical climates, preliminary field investigations at a landfill in Thailand found methane flux rates about two orders of magnitude higher than the lower limit, but well below the highest reported rate. Several factors may have contributed to this observation. First, the numbers of measurements taken at the site are relatively small, which may have missed the "hot spots". Second, it is possible a high percentage of methane gas is being oxidised by increased methanotrophic activity resulting from high temperatures (about 30 °C). The field measurements were carried out at the Kampaeng Saen landfill located about 110 km from Bangkok. This landfill occupies an area of about 12.8 ha. It received waste from Bangkok over the last eight years. The total amount of waste deposited was about 2.5 million tonnes. The average depth of the landfill was

about 10m. Although it is a relatively new landfill, high methanotrophic activity explains the low surface emission rates observed.

**Table 1: Methane Flux Rates Across Landfill Covers: Results From Field Studies**

Landfill Information			Landfill Cover Material	Observed CH <sub>4</sub> Flux Across Cover (g/m <sup>2</sup> .d)*	Source of Data
Location	Landfill Depth (m)	Landfill Cover Thickness (m)			
New York	14	0.3	clay cover	192.8	Lu and Kunz, 1981
Moscow, Russia	15 - 20	unknown	sandy clay cover	0 - 31.2	Nozhevnikova et al., 1993
California	30	0.5	granular soil	315.4- 1000	Bogner and Spokas, 1993
Illinois	40	1.5	silty clay	0.002 - 19.7	Bogner et al., 1995
California	30	1	sandy silt	0.003 - 1000	Bogner et al., 1995
New Hampshire	unknown	1 - 2	sandy-clay loam	0 - 1495	Czepiel et al., 1996a
Hagby, Sweden	unknown	0.2 - 1.0	sandy loam	0 - 334.8	Börjesson & Svensson, 1997
Bangkok, Thailand	10	0 - 1.0	silty clay	0.17 - 0.4	This study

\* observed CH<sub>4</sub> flux is after oxidation, at the surface of the landfill.

Environmental conditions, such as soil temperature, moisture content, nutrient availability, O<sub>2</sub> and CH<sub>4</sub> levels in soil and soil type are critical for growth and survival of methanotrophs (Stein and Hettiaratchi, 1997).

#### **Effect of Temperature on Soil Methanotrophy**

Required optimum soil temperatures of between 20 to 30°C (Whalen et al., 1990) are hardly attainable in northern climates, but are common in most tropical countries year around. Soil methanotrophy is reported at 6°C, albeit at low CH<sub>4</sub> oxidation rates (Nozhevnikova et al., 1993). The survivability of methanotrophs between freezing and thawing cycles is not well understood.

#### **Effect of Gas Flow Conditions and Patterns on Soil Methanotrophy**

Once established, the methanotrophs survive under intermittent CH<sub>4</sub> flow conditions (Kightley et al., 1995). Intermittent flow conditions are common in landfills in northern climates. The methanotrophic activity is directly related to the detention time of CH<sub>4</sub> in the oxidation layer at the surface of the landfill cover. Source strength of landfill gas, as well as gas permeability will affect this factor. Although it is possible to decrease the permeability of this layer, CH<sub>4</sub> flow rate will be mainly governed by the gas permeability of the hydraulic barrier layer. This is because the effective permeability is determined by the lowest permeability in a layered structure.

#### **Effect of Soil Parameters on Soil Methanotrophy**

The soil moisture content (MC) affects both physics and biology of soil methanotrophy. High MCs usually decrease microbial activity and increase occurrence of drying cracks which promote by-pass flow (Gostomsky et al., 1997). Parameters such as soil type and structure,

hydraulic conductivity and MC are inter-related, and could be influenced by weather conditions. Field and laboratory observations show that, optimum MC for sandy-loam and sandy-clay varies between 10% and 20% (Czepiel et al., 1996a and Whalen et al., 1990). Freeze-thaw cycles are known to change soil micro-structure with resulting changes in hydraulic conductivity; which invariably impacts gas migration. Bender and Conrad (1994) have observed the greatest methanotrophic activity when particle diameters range between 0.5 and 2 mm. Kightley et al. (1995) also found that porous, coarse sand develop a greater methanotrophic capacity than fine sand or clay soil.

While sufficient information on factors influencing CH<sub>4</sub> oxidation in soils is available in literature, the interactions among variables are not well known.

### Lab-scale and Pilot-scale Methane Oxidation Studies

Two types of lab-scale studies are reported in literature; batch experiments and column experiments. A summary of CH<sub>4</sub> oxidation rates found in literature is presented in Table 2. Most lab studies have been carried out on soil samples taken from landfill cover soils.

**Table 2: Maximum CH<sub>4</sub> Oxidation Rates Observed Under Controlled Conditions**  
**a) Batch Experiments**

Soil Type	Oxidation Rate (nmol/g dry soil.hour)*	Source
Sand and clay	585	Whalen et al., 1990
Sandy clay	25000	Nozhevnikova et al., 1993
Sandy-clay loam	955	Czepiel et al., 1996b

\* nmol of CH<sub>4</sub> oxidised per 1 gram of dry soil per hour

### b) Column Experiments

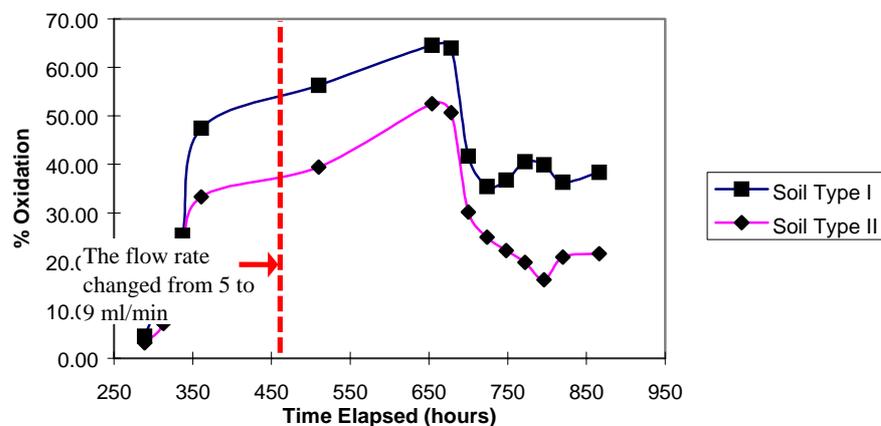
Soil Type	CH <sub>4</sub> Flow Rate (ml/min)	Oxidation Rate (g/m <sup>2</sup> .day)	Source
Coarse sand	5	166.4	Kightley et al., 1995
Clay top soil	5	108.8	Kightley et al., 1995
Fine sand	5	110	Kightley et al., 1995
Silty sand	3 - 5.4	61.7-120.6	Our study
Clayey silty sand	3 - 5.4	50.7-87.3	Our study

The current joint research program at the University of Calgary and the Asian Institute of Technology includes laboratory-scale column experiments, a pilot-scale field lysimeter study and full-scale landfill studies. The use of soil columns, instead of batch experiments performed by most researchers, provides a more realistic simulation of soil methanotrophy as encountered in landfills. Whalen et al. (1990) concluded that 50% of the landfill CH<sub>4</sub> in the US is oxidised on the basis of data collected from batch experiments. However, batch experiments do not account for the reduction in the areal extent of O<sub>2</sub> penetration that might be caused by its advective displacement by CH<sub>4</sub>.

A four metre high, 2.4 m diameter test lysimeter has been installed in the University of

Calgary premises. It contains about 4 tonnes of commercial and residential solid waste. The cover system of the lysimeter consists of three layers; a silty soil layer above the solid waste, a compacted clay layer as the hydraulic barrier and vegetative layer of silty soil and compost. Thermocouples were installed to obtain the temperature profile in the waste and the cover. Moisture probes were installed in the cover. Gas emissions through the cover will be measured using flux chambers. Data from this lysimeter will be available in late spring and summer.

Details of our laboratory set-up for column studies are described elsewhere (Stein and Hettiaratchi, 1997). Preliminary column study results from Thailand are presented in Table 3, and Figures 2 and 3. Table 3 contains methane oxidation rates at a temperature of 30-35°C as a function of time for two consecutive flow rates, 5 and 9 mL/min. Percentage oxidation as a result of soil methanotrophy is presented in Figure 2. When simulated landfill gas, containing 60% CH<sub>4</sub> and 40% CO<sub>2</sub>, at a volumetric rate of 5 mL/min was passed through a Type I soil (consisting of 70% sand, 15% clay and 15% silt), about 65% methane oxidation was observed at steady state. When the flow rate was increased to 9 mL/min, the immediate result was a decrease in percentage oxidation (to about 35%). However, methane oxidation rate increased from 61.74 g/m<sup>2</sup>.day to 120.64 g/m<sup>2</sup>.day (a 95% increase). A similar trend was observed with the Type II soil (consisting of 70% sand, 25% clay and 5% silt). The change in flow rate decreased the % oxidation from a high of about 51% to about 20%, whereas the oxidation rate increased from 48.88 to 87.29 g/m<sup>2</sup>.day (a 44% increase). These results show the pronounced impact of the composition of soil on oxidation. An ideal soil layer should contain mostly sand and silt and small amounts of clay.

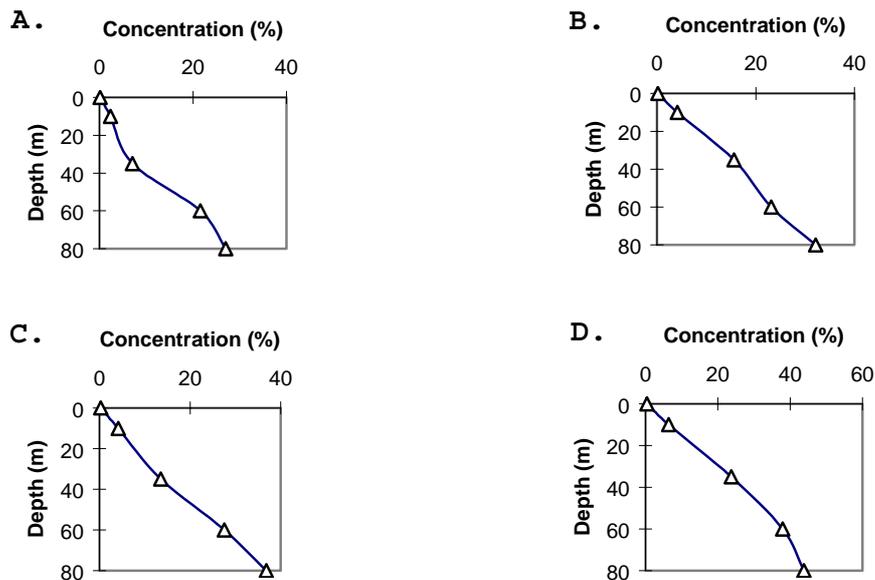


**Figure 2: Variation of Percentage Methane Oxidation with Time and Gas Flow Rate**

The results in Figure 2 and Table 3 are for the entire soil columns, which are each 90 cm deep. The variations in methane concentration throughout each soil column at steady state are shown in Figure 3. Similar curves are obtained for all four situations. Evidently, methane reductions occur throughout the soil columns. These reductions are due to two reasons; dilution effects and the oxidation effects. High reductions in methane (indicating continuous oxidation) seem to occur at the bottom 10 cm of the soil column when the flow rates are low.

**Table 3: Methane Oxidation Rates as a Function of Time (for two flow rates; 5 and 9 mL/min)**

Gas Flow Rate = 5 ml/min			Gas Flow Rate = 9 ml/min		
Time Elapsed (hours)	CH <sub>4</sub> Oxidation Rate (g/m <sup>2</sup> .day)		Time Elapsed (hours)	CH <sub>4</sub> Oxidation Rate (g/m <sup>2</sup> .day)	
	Soil Type I	Soil Type II		Soil Type I	Soil Type II
289	4.37	3.09	700	120.64	87.29
313	13.63	6.95	724	102.54	72.29
337	24.44	19.29	748	106.42	64.28
361	45.79	32.15	772	117.28	57.3
408	43.73	26.24	796	115.47	46.96
457	40.39	33.96	820	105.13	60.4
510	54.28	38.07			
533	44.5	39.61			
603	49.13	38.84			
628	56.34	37.04			
654	62.25	50.68			
678	61.74	48.88			



**Figure 3: Methane Concentration Profiles: A- soil type I with a flow rate of 5 mL/min, B-soil type II with a flow rate of 5 mL/min, C- soil type I with a flow rate of 9 mL/min, D- Soil type II with a flow of 9 mL/min**

In conclusion, our preliminary data show that very high methane reductions could be achieved in

tropical landfill covers if appropriate soils are selected for a “methanotrophic” layer and a barrier layer is designed to maintain the gas flow rates at optimum levels. Further research is required (and currently being undertaken) to define design parameters for cover systems world-wide for maximum methane oxidation.

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