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Preliminary Study of Methane Oxidation in Landfill Cover Soil: The Effect of Moisture Content

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Abstract: Methane oxidation is one of the important processes to mitigate the emission of methane gas at landfill to the environment. This study investigated the effect of moisture content in order to enhance the methane oxidation of final cover soil from Taman Medan Dumpsite. Three different PVC soil columns contained cover soils with moisture content of 7, 12 and 28% were used in this study. The columns were purged with the mixture of methane gas (60% by volume) and carbon dioxide gas (40% by volume) from the bottom of the soil columns. The mixture gas passing through the soil media was retrieved from the columns daily via sampling ports fabricated at every 10 cm of the column height. The soil cover was classified as sandy silt contained 56% of sand, 32% of silt and 12% of clay. It also had 12% of moisture content, bulk density of 0.229 g cm^{-3}, particle bulk density of 0.0304 g cm^{-3}, pH of 7.21, colour of dark yellowish brown, porosity of 86.72% and organic matter of 1.5%. Gas chromatography analysed the collected gas had showed that methane oxidation effectively occurred in the column with 12% of moisture content that the average methane oxidation efficiency of the 7 day experiment was 47.97%. However, the methane oxidation efficiency in the other two columns with 7 and 28% of moisture content was significantly lower with only 8.93 and 7.87% of efficiency. The analyses also showed highest methane oxidation efficiency by average, took place at the height of 80 cm of those columns.

Key words: Methane oxidation, landfill soil cover, CH_4 mitigation, bulk density, chromatigraphy

INTRODUCTION

Landfill Gas (LFG) is produced from the anaerobic degradation of organic compounds of the municipal solid waste is primarily constituted of 30-40% volume of Carbon Dioxide (CO_2) and 50-60% volume of Methane (CH_4) and trace elements (Abushammala *et al*., 2009). The flammable and harmful gas flows upward through the air space of the soil cover particles into the surface. It then accumulates as Green House Gases (GHG) in the environment and cause climate change. IPCC (2007) reported that methane is the second largest contributor of global warming after carbon dioxide. The Global Warming Potential (GWP_{100}) of methane was reported as 23 times higher than carbon dioxide in a 100 year horizon. It was also reported methane radiation forcing is the second-largest of the long-lived greenhouse gases. Due to this fact, a variety of field and laboratory studies have been carried out on the methane oxidation by microbes called methanotrophs present in the coversoil to mitigate the problem of undesirable methane emission from the landfill into the environment. However, there are several parameters should be taken into account to obtain the optimum condition for the methanotrophs to oxidise the methane, among them are moisture content, organic matter content, porosity and thickness of the soil cover. Hence, this preliminary study was conducted to find the optimum moisture content of the cover soil to enhance the microbial oxidation efficiency in which by the right amount of water could increase the gas molecular diffusion and microbial activities in the biofilter media (Yuan, 2006; Scheutz *et al*., 2009). Soil column experiments were performed for this purposeto determine methane oxidation efficiency of the coversoil at different percentages of moisture content.

MATERIALS AND METHODS

The cover soil used in this study was obtained from Taman Medan Dumpsite located in Petaling Jaya under administration of Majlis Bandaraya Petaling Jaya (Petaling Jaya City Council). Soil columns of height, inner diameter and thickness at 107, 15 and 0.5 cm, respectively were constructed from Polyvinyl Chloride (PVC) pipes to contain the cover soil medium in which the methane oxidation process occurred through it. The cylindrical columns were designed and modified according to the previous study by Kightley *et al.* (1995) and were equipped with 17 cm of gravel layer to allow gas distribution along the columns. The gas supplied was the

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mixture of methane (60%) and carbon dioxide (40%) to simulate the actual composition of landfill gas (Scheutz *et al*., 2009) and purged at flow rate of 5 mL min^{-1}. Further, the top of the columns were cap-sealed and embedded with inlet and outlet for air fed at 100 mL per min as reflection of real air movement at the surface of the soil cover. There were also 9 sampling ports along the soil columns at 10 cm intervals of the soil cover thickness where the samples of gas were retrieved daily. Gas Chromatography equipped with Flame Ionisation Detector (GC-FID) was used to analyse the gas samples.

Literature review

Biofilter/Biocover: Several studies have documented biological methane (CH_4) removal in landfill cover soils. It has been concluded that biological oxidation of methane in engineered systems is possible by providing optimum conditions for methanotrophic bacterial growth. As a result, there have been developments related to systems for biological removal of methane like biocovers, biofilters and biowindows. However, additional research is required before these alternatives can be commercially used (Stein and Hettiaratchi, 2001; Humer and Lechner, 2001; Scheutz *et al*., 2009).

Methane removal in biofilter/column: Gebert and Groengroeft (2009) performed diffusion tests and column studies to investigate the effect of air-filled porosity and degree of compaction on diffusivity and methane oxidation efficiency. The filter media was collected from closed landfill and filled into three PVC columns at different degree compaction which were 75, 85 and 95% of Proctor density. At the bottom, the columns were continuously charged with moisturised gas composed of methane gas (60% by volume) and carbon dioxide gas (40% by volume) reflecting the typical composition of landfill gas (Scheutz *et al*., 2009).

At the highest degree of compaction (95% of the Proctor density), the methane oxidation efficiency declined strongly with increasing inlet flux. In the experiment, the average of 74% efficiency was maintained at inlet rate of 1.47 gm^{-2} h^{-1}. However, the methane oxidation removal process dropped to an average of 32% after the inlet flux was increased to 2.4 gm^{-2} h^{-1}. The shrinking value of methane oxidation efficiency continued to decrease toonly 20% as the inlet rate was raised to 3.53 $gm^{-2}h^{-1}$. At 85% of the Proctor density, the increase of flux from 1.47-2.4 $gm^{-2}h^{-1}$did not reduce the oxidation efficiency that fluctuated around 94%. Performanceonly decreased to an average 62% after increasing the rate of charged gas to the highest level of 3.53 g CH_4 $m^{-2}h^{-1}$. The least compacted sample (75% of the Proctor density) maintained very high methane oxidation efficiencies of close to 100% over the entire flux range.

Barasarathi and Agamuthu (2011) carried out a batch experiment to investigate the different methane oxidation rate which was influenced by incubation time and moisture content by using compost as biocover. The compost moisture content was 62.17% and the pH was 6.33. In this study, it was found that composts should be able to retain water which was important to sustain the microbial population for the methane oxidation. Hilger and Humer (2003) stated that compost can offer a good water holding capacity to optimise methane oxidation. Furthermore, pH of the compost should be neutral to slightly acidic to optimize the methane oxidation (Moldes *et al*., 2007). Chanton and Liplay (2000) mentioned that the methane oxidation is higher in organic rich soils. The C:N ratio of the compost was 17. In addition, according to Boeckx *et al*. (1996), a high maturity of the compost material is crucial for an efficient methane consumption.

Humer and Lechner (2001) also indicated that at 35°C, the methanotrophic activity was the highest compared to the other incubation temperature studied ranging from 15-35°C. Temperature affected the methane solubility in water which eventually changed the methane oxidation rate by the change of the methane uptake rate (Barlaz *et al*., 2004).

Moisture content also plays role in determining the rate of methane oxidation in landfill cover soil by providing optimum condition within the medium for microorganism to maintain their activity of transporting nutrient supply and also removing the residual metabolic compounds (Scheutz *et al*., 2009; Graf and Gomez, 2007). Yuan (2006) concluded in his studies that there were three essential roles of moisture content in methane oxidation process. First, optimum moisture content provided optimum environment for methanotrophic bacteria to carry out their activity. Second, the moisture content significantly influenced the diffusion of oxygen into then soils, whereby, the oxygen was the main activator in methane oxidation process. Third, moisture content was one of the controlling factors of soil porosity that influenced gas transportation throughout the soil.

Study by Boeckx *et al*. (1996) suggested that the decreasing of methane oxidation rate at high moisture contents in the soilwas caused by a shift of gas-phase molecular diffusion to aqueous-phase molecular diffusion which is 10^4 times less rapid. It was also reported that the rate of methane oxidation decreased when there was insufficient moisture content in landfill cover soil due to response to water stress which would reduce microbial activities (Cao and Staszewska, 2011; Scheutz *et al*., 2009).

Zeiss (2006) stated that optimum moisture content in the soil for methane oxidation rate to occur effectively was affected by soil type and porosity. It was also supported by Yuan (2006) that the optimum moisture content in landfill soil would differ from various types of soil and depends on other environmental factors such as temperature. Graf and Gomez (2007) also found that the lowest optimal moisture content was available for coarse sand (~10% weight ratio (w/w)) while the highest one was found in yard waste compost at >30% (w/w). Studies by Boeckx *et al.* (1996), it was found that the optimum moisture content for methane oxidation rate in a soil cover from a landfill in Schoten, Belgium ranged between 15.6 and 18.8% in which similarly found by Stein and Hettiarachi (2001) for soil cover of a landfill in Calgary, Canada in 2001. In other studies, the optimum moisture content for maximum methane oxidation efficiency was obtained by different researchers. Czepiel *et al.* (1996), Whalen *et al.* (1990) and Visvanathan *et al.* (1999) found the value of optimum moisture content for methane oxidation occurred most efficiently was at 15.7, 11 and 15-20%, respectively.

RESULTS AND DISCUSSION

Properties of landfill coversoil: The properties of cover soil used for this study are shown in Table 1. The soil colour was determined as dark yellowish brown. Via soil textural analysis, it was found that the cover soil of Taman Medan Dumpsite to be sandy silt which contained 56% of sand, 32% of silt and 12% of clay. This soil had pH of 2.61, initial moisture content of 12% and organic matter content of 50.4%. Further, it had bulk density of 0.0304 g cm^{-3}, particle density of 0.229 g cm^{-3} and porosity of 86.72%.

Table 1: Physical and chemical properties of soil from Taman Medan Dumpsite

Properties	Characteristic/Value
Colour	Dark yellowish brown
Texture	Sand silt
Sand (%)	14.3
Silt (%)	15.2
Clay (%)	70.2
pH	7.21
Moisture content (%)	12
Organic matter content (%)	1.5
Bulk density (g cm^{-3})	0.0304
Particle density (g cm^{-3})	0.229
Porosity (%)	86.72

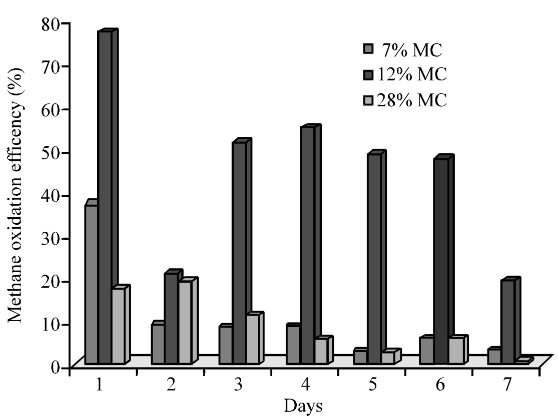

Fig. 1: Methane oxidation efficiency at different moisture content levels over the period of the experiment

Moisture content response experiments: The efficiency of methane oxidation in the soil with different moisture content was studied and compared. Figure 1 shows soil with 12% of moisture content has the highest total methane oxidation efficiency where of 21-77% of the methane was oxidised in the soil column daily. It followed by soil column contained 7% of moisture content where the methane oxidation efficiency was reduced to 3-37%. The third soil column with moisture content of 28%, on the other hand indicated only 1-19% of methane was oxidised over the period of experiment. The results obtained agrees with the studies by Cao and Stazewski (2011) that the optimum percentage of moisture content of landfill cover soil to boost the methanotrophic activities was in the range of 11-20%. This is essential since the methanotrophs, by using the methane monooxygenase enzyme, consumed methane as their carbon source (Scheutz and Kjeldsen, 2004).

However, the inadequate water content in the soil cover occurred in the soil cowith 7% moisture content in which the methane oxidation efficiency was reduced as a result of water stress that hindered the microbial activities (Cao and Stazewski, 2011). It was also reported by Wang *et al.* (2011) that low moisture content caused the bacterial cells to lyse as the water was drained out of the cell body via osmotic pressure and finally led to the death of the microbes.

Excessive water content present in the soil cover with 28% of moisture content whereby the methane oxidation efficiency was found to be reduced in the soil column. This was explained by Wang *et al.* (2011) that too moist condition limited the oxygen and methane diffusion to the microbial community along the soil profile. It was also stated in the report of Graf and Gomez (2007) and Cabral *et al.* (2004) that very wet soil cover caused the movements of those gases to be limited due to the fact that liquid diffusion is as much as 10^4 times slower than gaseous diffusion.

The 12% of optimum percentage of moisture content was also by comparison different with a few previous studies. Methane oxidation experiment using the mixture of soil of paddy field and earthworm cast by Moon *et al.* (2010) found that the engineered filter media was capable to oxidise methane at rate between 57-82% with moisture

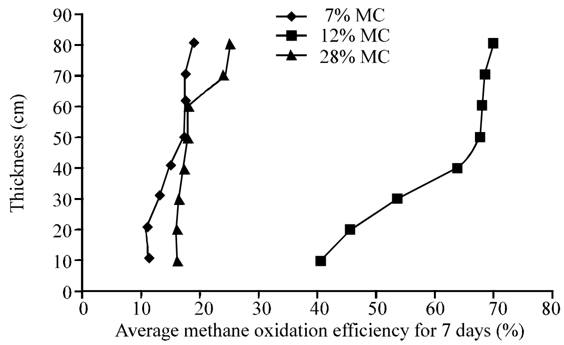

Fig. 2: Methane oxidation efficiency at different moisture content levels over the period of the experiment by profiles

content range of 15-40%. In addition, the studies by Park *et al*. (2002) showed that the addition of fertiliser containing nitrogen, phosphorous and potassium to loamy sand type collected from C horizon was best in oxidising methane at moisture content 3 and 13%. The varying ranges of optimum moisture content in the filter media to abate the methane concentration was hypothesised to be influenced by the physical and chemical properties of the filter media (Park *et al*., 2002).

According to Yuan (2006), there are 3 crucial roles of moisture levels in methane oxidation which are to provide optimum environment for methanotrophic bacteria to allow penetration of oxygen into the soils and to permit gas transportation throughout the soil. These 3 points reflect the lower methane oxidation efficiency in the soil column with 28% moisture content which was slightly greater than the range of the optimum values obtained for the literature.

Methane oxidation efficiency was also determined at different thicknesses of the three soil columns as shown in Fig. 2. At 12% moisture content, the methane oxidation efficiency was >40% at the first 10 cm of the soil layer and continually increase up to almost 70%. This indicated that there was sufficient oxygen diffusion at the thickness level supplied to the methanotrophs to oxidise methane (Park *et al*., 2002). Meanwhile, at 7 and 28% moisture content, the methane oxidation process took place gradually in the soil column with 11-18 and 16-25% of efficiency.

The study also recorded that the methane oxidation was more efficient along the soil column thickness in the three columns containing different percentages of moisture content, in which, it was deducted there was no significant loss of moisture at any level of thickness of the column. Although, Kightley *et al*. (1995) found that <3% of water loss columns at thickness of 85-90 cm due to air movements at the surface of the top soil column, the methane oxidation was still in incremental pattern to the highest thickness in their soil columns. Hence, the fact can explain of this study that the methane oxidation efficiency was still at a good level at 90 cm thickness. Further, it rejects the idea that the top layer of soil cover in the soil columns to reach permanent wilting point that limited the microbial activities as the moisture content dropped below 13% (Bender and Conrad, 1992).

CONCLUSION

This study showed that cover soil is essential in the abatement of methane emission into the environment and moisture content is one of the most important parameters to promote methane oxidation to take place. This is important in order to provide optimum condition for the growth of methanotrophic microorganisms that is responsible to convert methane to carbon dioxide. Besides, the percentage of moisture content should be approximately in the range of saturation and permanent wilting point to allow the methane oxidation process to occur efficiently (Park *et al*., 2002).

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