METHANE BIOFILTRATION: A COST-EFFECTIVE APPROACH TO ELIMINATE FUGITIVE METHANE EMISSIONS

J. Patrick. A. HETTIARATCHI (1)

(1) Professor, Center for Environmental Engineering Research and Education(CEERE), University of Calgary, 2500 University Drive NW, Calgary, T2N 1N4, Calgary,Canada
Corresponding Author: Tel: 403-2205503, E-mail: jhettiar@ucalgary.ca

Abstract

Methane is estimated to be responsible for 15% of the excess warming of the earth’s atmosphere that has occurred since pre-industrial times. Methane emissions from landfills around the world are estimated to contribute at least 10 to 15% of the total atmospheric budget from anthropogenic sources. Waste landfills in many countries are the largest source of anthropogenic emissions of methane, a key greenhouse gas. Because of the need to minimize emission of greenhouse gases (GHGs) to the atmosphere, it is prudent to search for cost-effective ways of minimizing landfill methane emissions. Recent research has shown that microbiologically mediated oxidation of CH₄, or methanotrophy, may serve as an inexpensive technique for reducing CH₄ emissions from landfills. In this paper, a low-cost technique available to eliminate fugitive methane emissions from landfills and other sources is presented. This innovative technique, methane oxidation using biologically mediated methanotrophy, could be applied to control point sources of methane emissions as well as area sources. At oil and gas industrial facilities, such as oil wells and batteries, both engineered and fugitive emissions could be controlled using methanotrophy. This paper presents laboratory results as well as results from field application at various facilities.

Keywords: Biofiltration; Landfill gas; Fugitive; Oxidation; Methanotrophy

1. INTRODUCTION

The potentially negative impacts of climate change have resulted to search for techniques to reduce anthropogenic methane (CH₄) emissions. Methane is a greenhouse gas with a short atmospheric lifetime of only 12 years. The global warming potential (GWP) of CH₄ is 25 times that of carbon dioxide (CO₂) over a 100-year horizon (IPCC, 2007). Therefore, control of CH₄ emissions is desirable to minimize its impact on climate change. Landfill gas (LFG) emissions and engineered or fugitive emissions from oil and gas industry are significant point sources of anthropogenic methane emissions. Such point sources of methane emissions can be either collected for energy generation or can be treated on site by various technologies. Methanotrophic biofiltration (MBF) is one of the gas treatment technologies (Stein et al.,
In biofiltration, CH$_4$ emissions from landfills are oxidized into CO$_2$ in landfill cover systems before it escapes to the atmosphere. Methane oxidation can be achieved through the use of naturally occurring microorganisms i.e., methanotrophs, which are thought to be present in soil environment exposed to high CH$_4$ concentrations.

Although, a considerable amount of research on biofiltration has been carried out but relatively a few can be found on MBFs. Information is lacking on CH$_4$ oxidation rates for various filter media operating under variable moisture content, pH, temperature, nutrient concentration and other environmental factors, and also in the presence of inhibitors. Therefore, such data is necessary to develop configurations of MBFs to maximize the CH$_4$ oxidation efficiency. Methanotrophic biofiltration does not produce a marketable byproduct like incineration of wastes for recovery of energy. Hence, there is a need to construct and operate MBFs in an inexpensive but efficient manner.

Results of laboratory experiment conducted worldwide by different researchers have identified media properties e.g., particle size, media thickness, presence of nutrients, moisture content, temperature, pH as key parameters affecting CH$_4$ oxidation (Bogner et al., 1997; Stein and Hettiaratchi, 2001). The reported optimum temperature ranges between 20-30, optimum moisture contents are around 13-18% w/w for soil and more than 80% for compost (Stein and Hettiaratchi, 2001). Furthermore, recent research has shown that methanotrophs may prefer compost media to soil as a growth medium (Humer and Lechner, 1999).

Laboratory and field experimental results on CH$_4$ oxidation in landfill cover systems are presented in this paper. Laboratory incubation experiment was conducted to determine CH$_4$ oxidation and kinetic parameters of different materials viz., Soil, compost and soil-compost mix.

2. METHANE OXIDATION BED MEDIUM

Biofiltration is a biological air pollution control technology that has been proven to be effective for control of odour and removal of volatile organic compounds (VOCs) and other compounds produced by stationary sources. The principles governing biofiltration are similar to those of common biofilm processes. First, the substrate goes through a gas/liquid interface from the pore to the biofilm, which is supported by a solid particle and then the substrate diffuses through the biofilm to a consortium of microorganisms. These microorganisms, in presence of nutrients, such as nitrogen and phosphorus, obtain energy from the oxidation of the substrate and in some specific cases they co-metabolize some compounds via non-specific enzymes (Chandrakanthi et al., 2005; Philopoulos et al., 2009).

The properties of the packing materials used are key factors influencing the performance of a biofilter. The following criterion outlines the desirable characteristics of a good filtering medium (Delhomenie et al., 2005):

- Presence and availability of intrinsic nutrients;
- High specific surface area for biofilm attachment, sorption capacity and gas/biofilm exchange;
- A high porosity to maintain a homogeneous distribution of gases and high retention times;
- Structural integrity to avoid medium compaction and low bulk density to reduce pressure drop;
Good moisture retention to avoid medium desiccation and to maintain microbiological activity. However, it is important to consider that excess water will fill biofilter’s void spaces and will slow the transfer of substrate, O2 and CO2 through the biofilm. Since compost exhibits the above-mentioned characteristics, it has been used as a granular filtering material in a number of research and pilot studies (Philopoulos et al., 2009). However, compost has two drawbacks. First, the feedstock or source material will define the physical, chemical and biological characteristics of the finished compost. Therefore, the performance of compost as a granular medium to support methanotrophy will vary with the feedstock raw material used to produce the compost. Second, compost is a biologically unstable material that tends to break down and become compacted over time. This compaction results in time dependent changes to diffusion within the filter bed and can lead to the inhibition of biological processes, such as biological oxidation of CH4 and its related co-metabolic processes (Chandrakanthi et al., 2005). In order to avoid performance problems due to compaction of granular bed, some researchers have employed bulking materials including wood chips, bark, perlite, sand, and vermiculite (Delhomenie et al., 2005; Philopoulos et al., 2009).

3. EXPERIMENTAL PROGRAM

3.1 Laboratory Experiment
Different soil-compost mix ratios (70/30, 50/50 and 30/70 by volume) were used for the laboratory incubation and column experiment. Soil used in the experiment was the same as used in landfill cover systems in Calgary. The compost was collected from City of Calgary Leaf Compost facility and considered as Grade-compost. The materials were sieved using 2.5 mm size sieve and soil, compost and soil-compost mix (70/30, 50-50 by volume) were used in the experiment. After sieving, soil-compost mixes (70/30, 50/50 and 30/70) were brought to different moisture contents. A specific quantity of material was placed in 250 ml serum bottles. The sample sizes selected were different for different soil-compost mixes and moisture contents. The optimum sample sizes were determined by conducting preliminary incubation experiment with different sample sizes. The incubation experiment was similar to the other incubation experiment, but for each incubation experiment, a number of samples with different weight were selected. The maximum oxidation capacity per g dry weight was calculated for each sample size and optimum sample sizes were fixed for each soil, compost and soil-compost mix samples at different moisture contents.

After filling the bottles with soil or compost materials, the bottles were sealed with caps fitted with teflon septa. Methane at different concentrations (approximately from 0.5% to 6% in air v/v) was injected into the bottles using a syringe. Initial gas concentrations were measured immediately after injecting CH4 into the bottles using Gas Chromatography. At least, two more gas concentration measurements were made with the samples at two different time intervals. The actual duration of gas sampling was determined by conducting a pre-incubation study. The incubation experiment was conducted to calculate the kinetic parameters (Vmax and Km).

3.2 Pilot Scale Landfill Test Cell
A pilot-scale landfill test cell (or Test Cell) of the dimensions shown on Figure 1 was constructed specifically to test the performance of selected media in relation to methane
oxidation. The Test Cell was filled with solid waste in two stages. First, the cell was filled with “as collected” waste to a maximum depth in the middle of 3.75 m, without manually breaking the garbage bags, to a compaction density of about 415 kg/m³. The water content of waste at the time of deposition ranged from 20% to 33% (Perera et al., 2002). Second, another layer of 3 m thick “shredded” waste was added (Figure 1) and compacted to a density of about 530 kg/m³. A layer of approximately 60 cm thick soil/compost mix (70:30) was placed as the final cover. This layer was lightly compacted to a (wet) density of about 1200 kg/m³. The 70:30 soil/compost mix was selected based on laboratory experimentation using a variety of soil/compost mixtures.

Methane surface flux from the Test Cell was monitored on a regular basis using the closed flux chamber technique before and after placement of the 70:30 soil/compost cover. Gas sampling tubes and thermocouples were placed at three different depths (10, 30, 40 cms from the surface) to measure the concentration profiles; pressure and temperature in cover system (Figure 2). Initial surface flux and gas concentrations at different depths were measured for two weeks.

![Figure 1: Details of Field Test Cell](image)

Soil samples were collected from different depths and surface, and brought to laboratory to determine CH₄ oxidation capacity.
4. RESULTS AND DISCUSSION

4.1 Sample Size Determination

Figure 3 shows the results of the experiment conducted to determine the sample size for incubation experiment with different soil-compost mixes. The results indicates the effects of sample size on types of materials and moisture contents. Higher the moisture contents, smaller is the sample sizes and higher the compost fraction, smaller is the sample sizes. The deviation in sample sizes with moisture is significant in samples with low soil fractions, whereas an increase in the soil fraction reduces the variation in sample size with respect to moisture contents (Figure 3). The results indicated that selection of sample sizes plays a significant role in determining the effectiveness of the experiment to determine kinetic parameters.

Figure 3: Effect of Compost Fraction on Sample Size for Incubation Experiment
4.2 Effect of Moisture Content

Figure 4 shows that the increase in compost fraction increases Vmax significantly. The optimum moisture content to attain the particular Vmax also increases with the increase in compost fraction. The results showed that the optimum moisture contents for maximum CH₄ oxidation for the soil-compost mixes of 70/30 and 50/50 are 26% and 34%, respectively.

Theoretically, a landfill final cover material containing a high compost fraction appears to be the best option to optimize methane oxidation rates. For example, if the 44% moisture content is achievable under field conditions for a soil-compost mixture of 30/70, an oxidation rate of more than 0.3 mg/g DW/h can be achieved. But, if the field moisture content is lower than optimum the moisture content, a medium with lower compost fraction could exhibit higher CH₄ oxidation. Therefore, field moisture content has to be considered while selecting the cover material, because the optimum moisture might not be achievable under field conditions. Another important factor that needs to be considered is the flux rate. If the flux rate is very low, it is better to consider the lower compost fraction with lower Vmax values because lower ranges of moisture contents are achievable under field conditions.

![Figure 4: Vmax of Different Soil Compost Mixes at Different Moisture Contents](image)

Although ideal for high-rate methane oxidation, maintaining compost (alone) at its optimum moisture content of 85% (i.e. optimum moisture content) is difficult under field conditions. If the landfills are not emitting high fluxes of methane, as in the case with the Calgary Test Cell, a soil-compost mix with lower methane oxidation potential is sufficient. The 30:70 compost-soil mixture was selected because of: high porosity promoting higher diffusion of oxygen into the medium, availability of nutrients when amended with compost,
the lower optimum moisture content (of about 26%) which can be maintained easily under field conditions.

4.3 Comparison of Performance of Soil/Compost Cover and Soil Final Cover

Samples collected from different depths of Test Cell cover during different periods of operation were incubated in laboratory to study methanotrophic activities using batch experiments. The Vmax values were determined using a procedure outlined in Stein et al (2001). The moisture contents and Vmax values of samples collected at two different periods are presented in Figure 5. Figure 5 shows that at soil compost mix ratio of 70/30, the field moisture content at depths (20-30 cm) are higher than soil alone, resulting in higher Vmax values. Although, the field moisture content (~21-22%) of 70/30 cover is lower than the optimum moisture content (27%) for maximum CH$_4$ oxidation, the difference is not significant. The observed surface emission of CH$_4$ was zero indicating the presence of methanotrophic activity. The design of biocover (i.e., soil-compost mix ratio) depends on the surface flux, which is the function of waste volume and organic fraction. For larger landfills, to achieve higher oxidation rate, the biocover can be designed with a relatively higher compost fraction, if the moisture content can be maintained at or near optimum level.

The moisture content of the cover material decreases with time as shown in Figure 5. Figure 5 also shows the concentration profile of biocover system. The moisture content of biocover is reduced from approximately 20% to 15% (at depths below 15cm), which is not the optimum moisture content for CH$_4$ oxidation. However, when compared with the Figure 4 results, this material still has a CH$_4$ oxidation potential of half of all time maximum CH$_4$ oxidation potential (Vmax). For a landfill situated in a semi-arid climate, the landfill gas emission rate is very low and therefore, 100% CH$_4$ oxidation can still be achieved at low moisture content, provided the moisture content does not fall far below the optimum.

![Figure 5. Comparison of Kinetic Parameters of Cover Material (soil/compost) Samples](image-url)
For example, a biocover made of 50/50 (soil/compost) at a moisture content of 25 % is less effective than a biocover made of 70/30 soil/compost at 15% moisture content. Therefore, the selection of a biocover depends not only one Vmax value at optimum moisture content, but also field moisture content and the distribution of Vmax at different moisture contents.

5. CONCLUSIONS
This paper presents results from a comprehensive research program involving laboratory studies, a field-scale monitoring and quantification of CH$_4$ emission and oxidation in a landfill test cell. The results indicated that the establishment of a bio-cap made of 70:30 soil-compost mixtures eliminates CH$_4$ emissions from small-scale landfills in semi arid regions. The 70:30 soil-compost combinations was selected based on laboratory batch experiment performed to determine the optimum moisture contents required to achieve maximum CH$_4$ oxidation levels in a variety of soil-compost combinations.

REFERENCES