METHANE OXIDATION EVALUATION OF THE LANDFILL COVERS

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Abstract

Municipal solid waste landfills, industrial woodwaste landfills and sludge ponds contribute significant emissions of methane and other greenhouse gases to the atmosphere. Only 33 landfills in Canada have some form of landfill gas collection systems. There are numerous smaller community landfills and other waste piles which have no gas collection systems, nor top cover and produce methane from anaerobic degradation. There is a need to develop a passive greenhouse gas reduction method for small and old landfills and as an auxiliary method to augment landfill gas collection. Laboratory methane oxidation tests show to provide a methane emission reduction potential of 10 to 90% of the methane produced. Several different materials were tested under different levels of moisture content and temperature. The soil that provides the highest methane oxidation potential will be used for additional in-situ testing.

Key words: Methane oxidation, landfill cover, greenhouse gas reduction.

Research Problem Statement and Objectives

With about 40 – 70 Mt of methane emitted each year worldwide, landfills are the largest antropogenic source of this greenhouse gas in North America. Landfill gas collection reduces methane emission by 50 – 60 %. The remaining 40 – 50% of landfill gas is emitted to the atmosphere. Furthermore, there is significant number of small, old, and remote landfills, as well as operating areas of landfills before the closure where the gas collection is not either possible or economically feasible. To achieve greenhouse gas reduction at these facilities, an engineered passive methane emission reduction method is a suitable option.

Methane oxidation, the biochemical oxidation of methane into carbon dioxide and water in the top cover soil layer has been shown to provide a methane emission reduction potential of 10 to 70% of the methane produced in landfills, or 30 to 50% of the methane emitted through the surface. Thus, methane oxidation appears to provide a significant potential for greenhouse gas reduction if the engineering design and operation of the landfill cover has to be optimized in order to achieve a minimum reliable reduction of methane emissions.

The objectives for this study were as follows:

- Investigate different type of materials as adequate media for methane oxidation;
- Vary key variables – temperature, moisture content and porosity; and
- Suggest cover design approaches to improve methane oxidation rates.

Methane Oxidation Cover Design Scenario

Oxidation of methane, which intends to escape from anaerobic areas inside landfills to the atmosphere, occurs primarily as part of aerobic metabolism in methanotropic bacteria. The net reaction of methane oxidation under aerobic conditions can be described as:

$$CH_4 + 2O_2 \Rightarrow CO_2 + 2H_2O$$

Methane as a nutrient and oxygen are essential for the reaction. Oxygen availability strongly depends upon the soil diffusivity, which in turn affects the utilization of methane. The longer the reactants are mixed, the higher the reaction rate. Figure 1 shows the optimal concentration profiles for the gases that take place in methane oxidation. A good design of the soil cover provides deep oxygen penetration although its concentration decreases with the depth as a result of the reaction consumption. If oxygen is not limiting factor methane concentration constantly decreases going up to the surface because of the reaction consumption and air dilution.

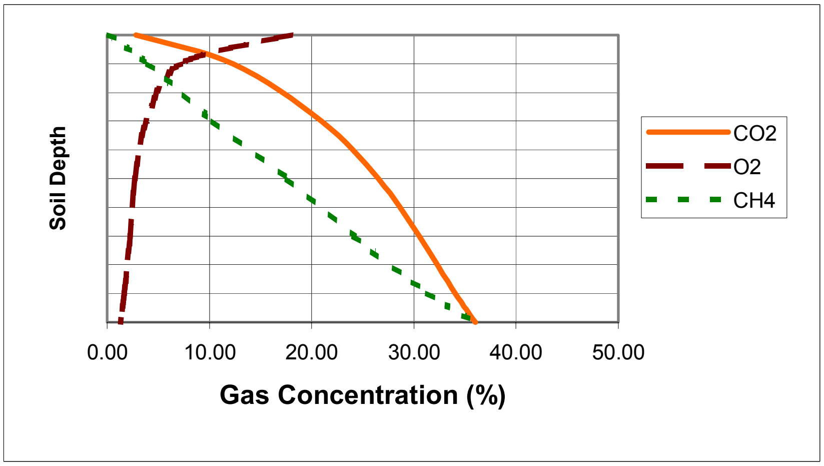

Figure 1. Optimal gas concentration profiles in landfill soil cover

Carbon dioxide concentration has similar trend as methane concentration but it is larger since carbon dioxide is a product of the reaction. Figure 2 shows the optimal profiles for moisture content and temperature. Depending of the seasons temperature profile changes but an optimal range is between 10 to 30°C. An optimal range for moisture in landfill covers is between 11 to 25% by wet wt. A large number of variables can affect methane oxidation rates but the objective of this research focuses on three most significant: porosity, moisture content, and temperature.

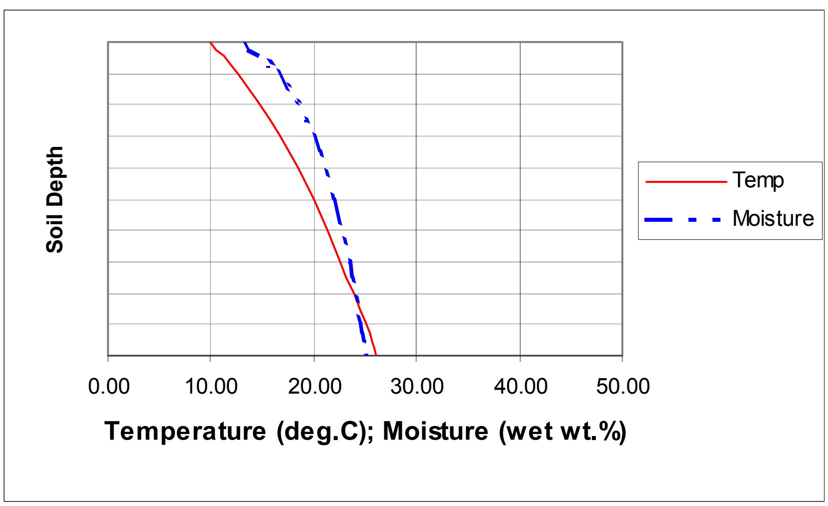

Figure 2. Optimal profiles for temperature and moisture content in landfill soil cover

Soil Porosity

Since the gas availability is dictated by soil porosity, it is one of the most important variables for methane oxidation. Soil porosity, if properly designed, can control moisture content of the soil cover and gas permeability throughout the soil cover. Furthermore, soil porosity is a function of material type, particle size, compaction, and moisture content. Porosity affects oxygen concentration inside the soil. Methane oxidation is an oxygen limited and methane saturated reaction where the rate constant depends of soil porosity.

Moisture Content

Moisture content of the soil regulates methane oxidation by affecting gas transport through soil cover and through the phyisiological requirement by methanotrophs. Methane oxidation decreases when the moisture content is over 40% by volume (close to level of saturation for subarctic soils) and under 10% by volume. The desirable moisture content ranges between 11 to 25% by wet wt. (Boeckx et al. 1996, and Whalen et al. 1990). High moisture content will reduce the advecitve flow of landfill gas entering the soil from below and will reduce the diffusion of oxygen from above, while low moisture content will reduce biological activity. The data obtained in the field and laboratory studies indicate that soil moisture is an important factor regulating methane oxidation. Unfortunately, none of the studies related to methane oxidation measured water content directly.

Temperature

Temperature also plays an important role in methane oxidation reaction. Boeckx et al. (1996) found that methane oxidation reaches a maximum between 25 and 30ºC with a consistently increased rate between 12 to 25ºC. When temperatures decreased to 5 to 7ºC methane oxidation rate decreased by 3 to 5 times (Nozhevnikova et al., 1993).

Cover Design Approaches

Concentration Control with Porosity

Gas concentrations can be controlled if top cover is properly designed. This means that an engineered design should be applied where top cover would contain different types of materials layered according to its porosity. Bottom layer with its low porosity should minimize landfill gas emission allowing (if applied) regular gas collection, restrict air intrusion and keep anaerobic condition inside a landfill. This nonporous, like clay, material has a function to keep moisture content near an optimal level inside the cover. Further layering should be performed so that porosity slightly increases finishing with a highly porous material (e.g., sand or gravel), which allows deeper intrusion of oxygen and collects enough moisture.

Moisture Control and Measurements

To keep moisture level inside the desirable ranges layering based on porosity may help but is not a guarantee, especially in the areas where precipitation is not equally distributed per seasons. Possible solution for this problem would be an additional layer inside the cover soil. That layer needs to distribute moisture in both directions, to drain surplus of water during the wet seasons or to distribute the moisture during the dry seasons. An automated moisture measurement system (Zeiss and Li, 1999), which includes TDR (time domain reflectrometers) would be suitable solution. Once installed

TDR probes allow continuously moisture content measurements with relatively high precision of ±5% by volume.

Temperature Control

According to USEPA yearly averaged methane oxidation rate is 10% of methane produced. This was calculated assuming that methane oxidation rate is 20% of methane produced during the warm season and zero percent during the cold season. This means that temperature is responsible for additional methane oxidation potential of 10% for which methane is not reduced. Previous studies including this one show that temperature inside landfills ranges from 25°C to 30°C two meters from surface. To keep temperature over the freezing point the first solution would be deep access of oxygen to move the reaction deeper inside the cover. Additional protection, especially for the areas with extremely cold winters, would be an insulating layer over the surface based on porous material such as, compost soil or shredder fluff.

Soil Column Results

A test series of four column reactors was conducted to simulate methane oxidation in landfill cover soil. Each column was constructed from 20 cm diameter PVC pipe containing a 50 cm thick layer of soil. The columns were filled with 1) clay – bottom half and landfill soil (mixed clay, silt, and sand), 2) soil and sand, 3) soil and compost, and 4) soil. The first three columns were run at room temperature (22°C) while the fourth column was run at low temperature (4°). The columns were fed from below by synthetic landfill gas, composed of a 45:45% mixture of methane and carbon dioxide, and 10% neon (used as a tracer gas). A gas flow flux was $2.34 \cdot 10^{-7}$g CH_4 $cm^{-2}d^{-1}$, which was lower midrange of reported landfill methane fluxes. Atmospheric conditions were maintained at the top of the soil where air inflow was 300 mL min^{-1}. Fresh soil was collected from the Clover Bar Landfill, an active municipal landfill in Edmonton, Canada. All materials were air-dried prior to filling of the columns. All columns were compacted on the same way (ASTM D 1557-78). The initial soil moisture was around 5% by wet wt. Later moisture contents were increased up to 12% by wet wt. The gas concentrations were measured by Gas Chromatography (GC). 1mL gas samples were automatically withdrawn and analyzed by Varian Micro GC (Model CP – 2003P)

Figures 3 to 6 show concentration profiles for methane, carbon dioxide, and oxygen of each column under optimal moisture content. Concentration profiles for oxygen indicate that the reaction occur as deep as oxygen penetrates inside soils. In Figure 3 the methane oxidation rate is the highest at depths of 5 to 15 cm from the surface and slightly decreases with depth. This occurs because of an insufficient amount of oxygen, which was limited by the porosity of the soil. However, in column 2 (Fig. 4), despite the high presence of oxygen, the reaction is "layered" as well as the material components. The reaction is very low in the soil and increases slightly in the sand where the highest reaction rate occurred at 15 to 25 cm of depth. Since water was been recently added, moisture controls the reaction level. Close to the surface, the moisture content is low because of low field capacity of sand. In the same time the lower part of sand layer is saturated because of the low porosity inside the top soil layer. High moisture content in the contact area restricts the intrusion of oxygen. However, a mixture of soil (bottom half) and compost (Column 3 in Figure 5) provides an ideal media for methane oxidation.

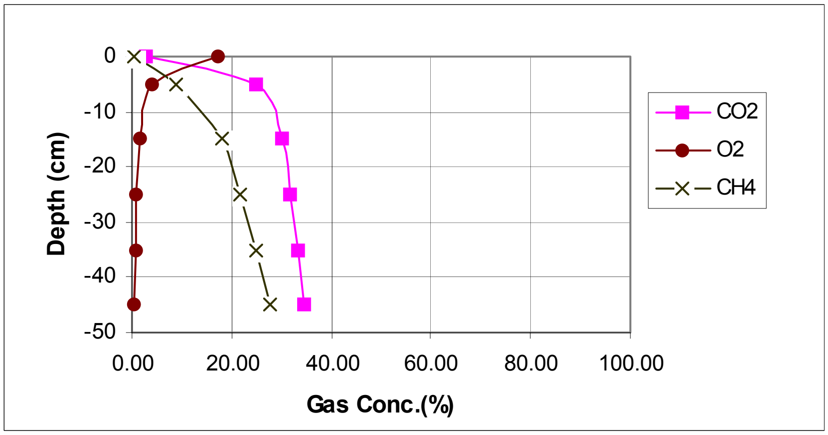

Figure 3. Gas concentration profile in clay and soil.

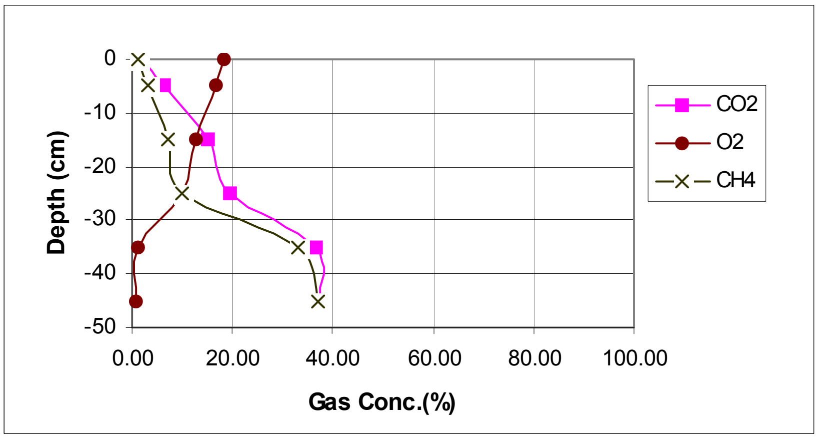

Figure 4. Gas concentration profile in soil and sand

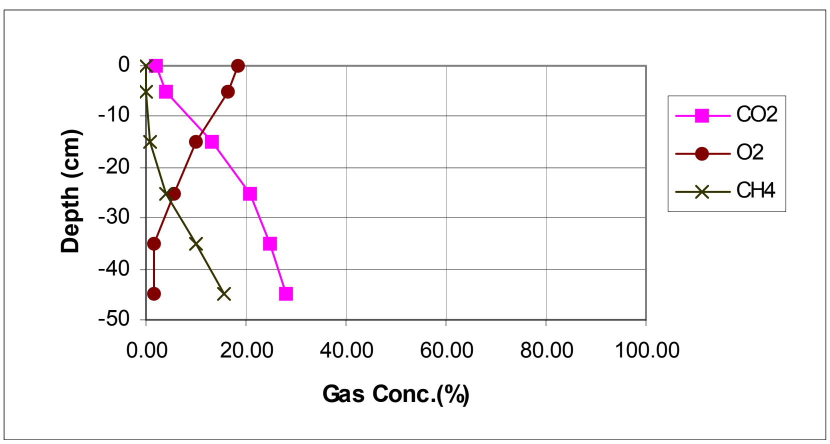

Figure 5. Gas concentration profile in soil and compost

The high porosity of compost allows deep penetration of oxygen into the soil which results that the reaction starts as soon as methane enters the column. High field capacity of the compost provides distribution of moisture through the whole depth of the column. As a result an equal and intensive reaction occurs between 45 and 15 cm. Above 15 cm the reaction decreases as the methane concentration becomes the limiting factor. Finally, the reaction rate in this column is over 80% of the methane entering the column.

The gas concentration profiles shown in Figure 6 represent conditions from a column filled with soil and run at a low temperature of 4°C. Although most of the other factors are very similar to first column very low consumption rate for methane is the result of the low temperature. Despite the sufficient concentration of oxygen at 15 to 25 cm, the reaction is close to zero deeper than 15 cm. These conditions differ from those encountered in the field because the temperature of the supplied gas is the same as the temperature of surrounding air.

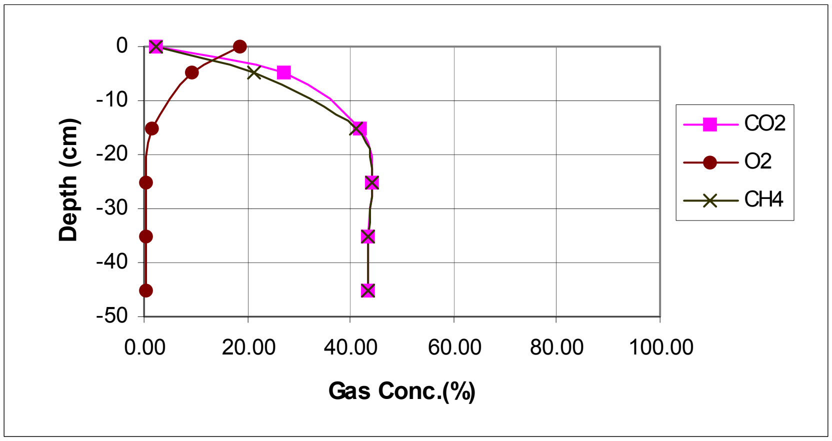

Figure 6 Gas concentration profile in soil at low temperature (4°C)

The results of the media used in column three (soil and compost) provide a good base for further testing in the field under real atmospheric condition.

Research Direction and Issues

Additional studies need to approve all the assumptions made above about layered soil porosity design of landfill covers. How sustainable this cover will be under the atmospheric conditions. Waste settlement and surface erosion often promote significant change in landfill cover, so their influence under these approaches has to be investigated. Furthermore, precipitation, evaporation, and lateral flow may affect pore size and pore distribution so that they should be considered in future studies.

Decreasing pore size with depth will enable air intrusion, but its influence on the temperature profile of landfill covers is still an issue. Further research will explain whether an additional insulation layer can keep methane oxidation rate at positive level, even if the low temperature causes movement of the methane oxidation zone deeper into the soil. The possible movement of methane oxidation zone could bring an additional benefit forcing methanotrops from self-produced extracellular polymeric substances (Hilger et al., 1999). An additional issue is whether this engineered approach needs to involve whole area or some certain areas to which the gas would be driven.

Finally, the measurement and verification methods to determine the amount of methane emissions reduced by oxidation have be determined and structured into a protocol so that landfill owners can determine the reduction in tonnes of CO_2 equivalents for their greenhouse gas reduction reporting and for greenhouse gas emission reduction trading.

In the next planned phase of research, field scale tests with the best soil profile from the column tests, will be performed, based on the findings of the lab tests. These tests will be performed under actual atmospheric conditions over a 12-month period. Test plots will be constructed on an area with previously determined methane emission. The soil layers will be constructed and compacted to achieve decreasing pore size with depth to enable air intrusion while limiting the landfill gas flow upwards. The significance of these tests is to develop an engineered cover layer that will passively reduce greenhouse gas emissions. This approach is applicable as a stand-alone method at smaller, remote and older landfills (where gas generation is low, or gas use infeasible), or in conjunction with landfill gas extraction at large landfills to reduce the part of methane that can not be collected.

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