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Methanotrophic activities in tropical landfill cover soils: effects of temperature, moisture content and methane concentration

The methane oxidizing capacity of landfill cover soils was investigated through column and batch experiments by simulating conditions that are usually encountered in tropical climates. The rate of oxidation was monitored at different temperatures and moisture contents. It was observed that a low moisture content of 6% produced negligible oxidation, whereas oxidation rates were at a maximum at moisture contents between 15 and 20%. Temperature was found to be a dominant parameter which controlled the oxidation rates. The optimum temperature was between 30 and 36°C. In the column tests, the temperature influenced the methane oxidation capacity indirectly by causing the topsoil surface to become totally dry, resulting in almost zero oxidation in spite of aerobic conditions. Although some increase in oxidation rate was observed, a higher concentration of methane could not produce a corresponding increase in oxidation rates, indicating the limiting capacity of the soil to oxidize methane. A depth profile of the gas in the column system indicated that the depth of maximum oxidation was around 15 to 40 cm under normal test conditions. Experimental results indicated that the topsoil, if maintained at an optimum moisture content, could also produce a higher oxidation capacity. The results of this experimental program indicate the possibility of maximum methane oxidation in a tropical climate if the correct moisture content is maintained at the top surface.

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Introduction

The higher rate of increase in atmospheric temperature observed since the Industrial Revolution is currently being attributed to increased emissions of greenhouse gases. Carbon dioxide, the main greenhouse gas, is mainly produced by burning fossil fuels. Steps to reduce the emission of carbon dioxide could augment the contribution of another greenhouse gas, namely methane. Although atmospheric concentrations are relatively low, methane could be the major greenhouse gas of the future due to its high residence time and higher potential to cause global warming. Almost 80% of the methane emitted into the atmosphere is from the

biological degradation of organic matter. Its contribution to the greenhouse effect is estimated to be 19% (Foley 1991), with a global warming potential approximately 20 times greater than carbon dioxide. Methane is also a flammable gas, with a 5 to 15% V/V concentration in air capable of causing an explosion when accumulated in a confined space. Global methane production is 550 ± 105 Tg, with paddy fields, wetlands and landfills contributing 100 to 200, 25 to 170 and 20 to 70 Tg, respectively (Mendelsohn & Roseberg 1994). Thailand's contribution to the total methane emissions into the atmosphere is 6.2 Tg (TEI 1996). The annual production of methane from landfills in Asian countries is 11.8 Tg and Thailand's contribution is 0.28 Tg (TEI 1996).

Microbial activities within landfills are complex and interrelated. The various microbial transformations of materials and byproducts include the carbon and nitrogen cycle, together with the anaerobic decomposition of organic waste materials, the production of methane and carbon dioxide, and methane oxidation in landfill cover soils producing water and carbon dioxide. Different types of volatile organic compounds are also produced and transformed into different forms during their movement inside the waste and through the landfill cover.

The production of gases builds up pressure inside the landfill, which forces the gas to escape through the easiest route. Depending on the characteristics of the surrounding soil, the gas may migrate laterally hundreds of meters from the landfill. Normal practices for the disposal of gas include direct venting into the atmosphere, burning in flares, or utilization for energy production depending on the characteristics of the gas, the concentration of methane and the age of the landfill. The long-term collection and flaring of landfill gas is expensive, requiring high staffing levels and energy to recover the landfill gas during its active stage. An increase in the imperviousness of the landfill cover, on the other hand, causes the gas to migrate laterally, posing a threat to the surroundings.

The ever-increasing interest in global warming has forced researchers to investigate natural treatment methods for landfill gas. Few studies have been conducted in this field to determine the occurrence and behavior of the microorganisms responsible for oxidizing methane. Almost all of these studies are from temperate and cold climates; no study has been reported from tropical climates. The warm and moist conditions and high organic waste materials in landfills in most tropical climates require careful consideration.

This paper presents the initial results from a long-term research program to study methanotrophy in tropical climates and how this knowledge can be utilized in designing landfill cover systems to minimize the escape of methane. The results of an experimental study conducted to determine the methane oxidation capacity of soil are presented. The study was conducted to assess the methane oxidation capacity of landfill cover soils and its effectiveness in regulating methane emissions from tropical landfills.

Experiments were conducted at both column and batch levels at different moisture contents, temperatures and different methane flow-rates to determine the variation in oxidation rates and to study the influence of these parameters on methane oxidation rates. From these results, relations between natural and experimental conditions were developed. Furthermore, the depth at which maximum oxidation occurs and the parameters responsible for variations in maximum oxidation with depth were investigated. The depth of maximum oxidation could be one of the main factors to be considered in designing a landfill cover to attain maximum methane oxidation. The focus of this paper is on the factors controlling methane oxidation in tropical climates—temperature, moisture content and methane flow-rates—because landfill cover soils in the tropics are exposed to high temperatures, varying moisture contents and high methane flow-rates.

Materials and methods

Soil material

The experiments were conducted using three columns. Two main columns were filled with two different mixtures of soil and purged with a methane—carbon dioxide gas mixture (60 : 40%). The third column was used as a control, filled with both soils alternately and purged with nitrogen. The soil used for the experiments was a mixture of soils collected from different places. The soils were ground, mixed in different proportions and mixed with water to obtain an initial moisture content of 11%. The soil in column I contained 70% sand, 15% silt and 15% clay, whereas that in column II contained 70% sand, 5% silt and 25% clay. The experiment was carried out at a laboratory in the Asian Institute of Technology, Bangkok, Thailand.

Column system

The columns used for the experiment were acrylic tubes 20 cm in diameter and 120 cm in length (Fig. 1). Four gas sampling ports and four soil sampling ports were fixed on the

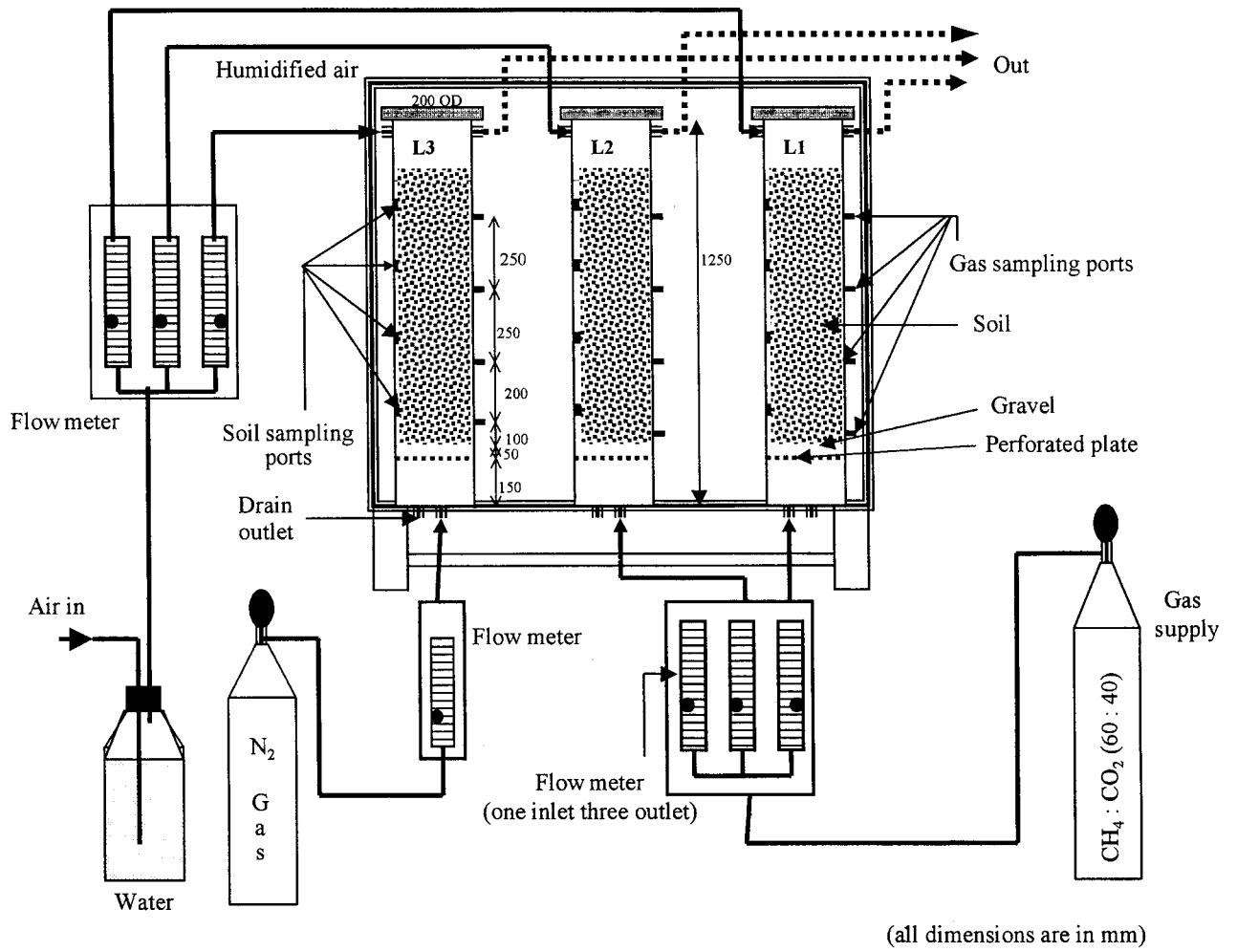

Fig. 1. Schematic diagram of experimental set-up.

side of each column at different levels. Gas inlet points were located at the bottom of each column. Inlet and outlet points were fixed at the top for the air supply. A perforated plate was placed at 15 cm height from the bottom of the tube. A 5 cm gravel layer was placed over the perforated plate to facilitate a uniform distribution of gas. The test columns were filled to a depth of 90 cm with the soil mixtures. In the two test columns, the gas supply was maintained at 5 ml min^{-1} at the initial stages and was increased to 9 ml min^{-1} to investigate the effect of different flow-rates on the methane oxidation rates. A control column was filled alternately with the two types of soil and was purged with nitrogen at the same supply rate as the test columns. The air supply from the top was maintained at 300 ml min^{-1} after humidifying by passing through a water column. Gas samples collected at the different sampling ports and the headspace gas were analyzed using a gas chromatograph (Shimadzu GC-15A system) connected to an integrator system (CR-4A). Helium was used as the carrier gas with a flow-rate of 50 ml min^{-1}. The temperatures were as follows: column, 55°C; injection block, 55°C; and TCD 110°C. The current was 130 mA and the column material SS. The column length was 10 ft, the outer diameter 1/8 in, the inner diameter 2 μm and the mesh range 60/80.

The moisture content in the column system was increased by spraying water from the top after determining the moisture content at different depths of the column. The moisture content was determined by oven-drying the soil sample removed from various depths of the column. The headspace gas concentration and the concentration of gases at different sampling points were determined at three different average moisture contents (11, 15 and 18%) and at different gas supply rates. The effect of temperature on the methane oxidation rate was monitored by taking headspace gas samples at different times of the day at different ambient temperatures (soil temperatures). Soil samples from different

depths were removed for sampling and analyzed for chemical (Page *et al.* 1982) and bacteriological parameters (Mancinelli *et al.* 1981) at the end of each experiment.

Batch experiments

The batch experiments consisted of the incubation of fresh and old column soils in 100 ml serum bottles at different incubation temperatures and with different moisture contents under various initial headspace methane concentrations. The soil was incubated with 5, 10, 15, 20 and 25% moisture content and at 5, 20, 30, 36 and 45°C. Headspace concentrations of various gases at different time intervals were also measured. Oxidation rates at different moisture contents and different incubation temperatures were determined and the kinetic parameters calculated. The oxidation rates at different initial headspace concentrations were also determined by incubating the soil samples with different initial headspace methane concentrations.

Soil samples collected from different depths of the column systems were incubated at 30°C with injected methane. Headspace concentrations were measured at different time intervals. From the decreasing headspace concentration, the oxidation capacity of each soil was determined. Microorganisms were enriched through growing cultures under a methanol atmosphere and indicator species for methanotrophic populations were identified (Mancinelli *et al.* 1981). The location of the maximum number of methanotrophic bacteria, and hence the depth at which maximum oxidation occurs, was identified from the oxidation rates obtained from batch experiments.

Data analysis

The gas flow-rates in all three columns were monitored and gas concentrations at different sampling ports were determined and compared with the physical, chemical and bacteriological properties of the soil. Gas concentration data were used to determine the oxidation rates. The mass flux method was used to obtain preliminary estimates of the oxidation rates assuming that the total inflow and outflow of the gas were the same, although some of the carbon was utilized by the microorganisms. An average temperature of 32°C for the column system was used to calculate the methane oxidation rates (this temperature was based on the measured temperatures between 28 and 36°C). The oxidation rates in batch experiments were calculated from the measured decrease in headspace methane concentration. A first-order kinetic reaction was assumed when calculating the methane oxidation kinetics parameters.

Results and discussion

Effect of moisture content on methane oxidation

Batch experiments with fresh soil samples

As shown in Fig. 2, the oxidation of methane increased with increasing unsaturated moisture content up to 20% *V/V*. Higher moisture contents (more than 20%) in the batch experiments produced a lower oxidation capacity, indicating the negative effects of a higher moisture content on the methane oxidation capacity in soils. The highest oxidation rate of 1.22×10^{-6} g CH_4 (g dry soil)$^{-1}$ h^{-1} was observed in fresh soil with a 15% moisture content when incubated at 30°C. Moisture contents of 11 and 25% showed similar oxidation rates of 0.75×10^{-6} g CH_4 (g soil)$^{-1}$ h^{-1}. No significant methane oxidation was observed at 6% moisture content. A relatively high oxidation rate was observed in batch experiments at 20% moisture content, which is similar to the results reported by Schnell & King (1996). The optimum moisture content reported by Boeckx & Van Cleemput (1996) was 15%, with an oxidation rate of 2.36×10^{-9} g CH_4 (g soil)$^{-1}$ h^{-1}.

Column experiments

A decrease in moisture content in the column system with time was noticed, with the top surface of the column becoming almost dry and the moisture content decreasing to less than 5%. The moisture content increased with the depth of the column with an approximately constant moisture content of around 12% at 30 to 90 cm depth. Spraying with water from the top increased the moisture content at the top surface, but did not maintain a uniform moisture content with depth. The average moisture content of the column system was taken as the average at a depth of 15 to 60 cm. A comparison of oxidation rates (Fig. 3a) at different average moisture contents in the columns indicated a maximum

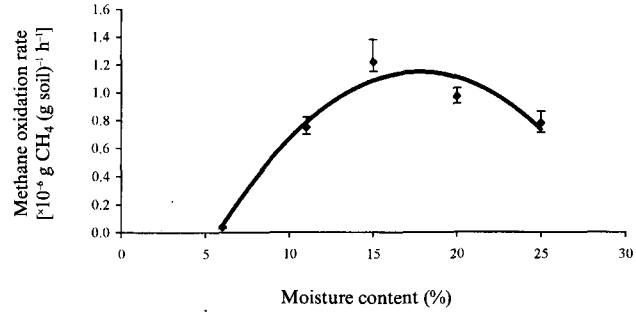

Fig. 2. Batch experiment on the methane oxidation capacity of a fresh soil sample incubated at 30°C under 3 to 4% initial headspace methane concentration.

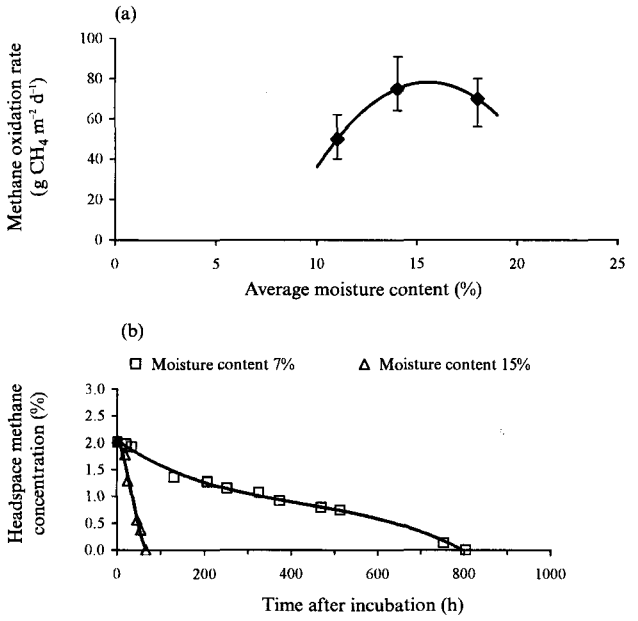

Fig. 3. (a) Average methane oxidation rates in test column I at different moisture contents with a gas supply rate of 5 ml min^{-1}. (b) Headspace methane concentration measured by incubating 10 g of column I topsoil with two different moisture contents in a batch experiment.

oxidation rate at an average moisture content of about 14% (or 75 g m^{-2} d^{-1}). Whalen *et al.* (1990) reported an optimum moisture content of 11% (45 g m^{-2} d^{-1}) and Boeckx & Van Cleemput (1996) reported a range of optimum moisture content from 10 to 20%, similar to our results. The different proportions of silt and clay used in the two test columns did not show any significant difference in oxidation capacity.

Inhibition effects on methane oxidation at low moisture contents have been reported by Whalen *et al.* (1990). Very low oxidation rates in topsoil at 7% moisture content and zero oxidation at 1.5% moisture content were observed in our experiments. A similar result of zero oxidation in air-dried soil samples was reported by Nesbit (1992). When moisture was introduced from the top, the high moisture content (15%) produced very high methane oxidation rates (Fig. 3b), indicating the re-establishment of a high oxidation capacity in topsoil with an optimum moisture content. This also indicates the capacity of methanotrophs to survive in the extreme condition of low moisture content. Lower oxidation rates at low moisture contents could be due to physiological water stress. Nesbit (1992) reported the reduction of oxidation rate by 56% of the maximum in a saturated soil. A lack of adequate diffusion of atmospheric oxygen into the column soil with a high moisture content could be the reason for the low oxidation rates in the column. The gas profiles for all three columns are shown in Fig. 8a.

Both batch and continuous experiments showed similar results with an optimum moisture content of around 15% in the column system and 15 to 20% in the batch experiments, whereas Boeckx & Van Cleemput (1996) reported a range of 15.6 to 18.8%.

Effect of temperature

Batch experiments

Fresh and column soil samples, when incubated at different temperatures with initial headspace methane concentrations between 3 and 4%, produced different rates of methane disappearance. An increase in oxidation rate was observed at temperatures up to 32°C, with an oxidation rate of 1.20×10^{-6} g CH_4 (g soil)$^{-1}$ h^{-1} being the maximum with fresh soil (Fig. 4). A rapid decrease in oxidation rate was observed at temperatures higher than 36°C, with complete inhibition at 45°C.

Column experiments

The natural diurnal variation of temperature also affected the oxidation rate. As evident from Fig. 5, low temperatures (about 30°C) during the night produced higher oxidation rates than high daytime temperatures (about 37 to 40°C).

Both direct and indirect effects of temperature on the methane oxidation rate were observed. The published optimum temperatures for methane oxidation usually fall within the range 30 to 36°C. Whalen *et al.* (1990) reported an optimum temperature of 31°C and William & Zobell (1949) 32°C. Boeckx & Van Cleemput (1996) and Nesbit (1992) reported optimum temperatures between 25 and 30°C and 20 and 30°C, respectively, a range lower than the experimental results reported here.

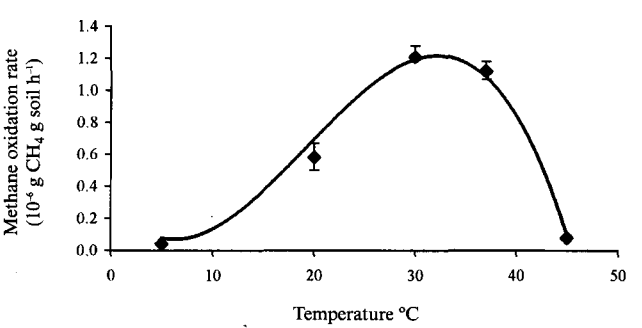

Fig. 4. Average methane oxidation capacity of fresh soil with 16% moisture content and incubated at different temperatures under 3 to 4% initial headspace methane concentration.

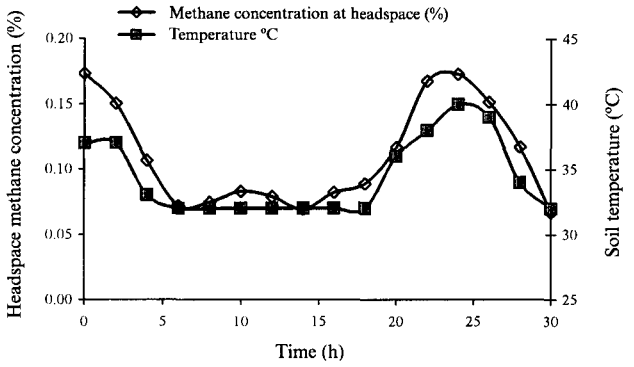

Fig. 5. Headspace methane concentration observed in test column I at different temperatures (0 h sampling time corresponds to 14:00 h).

Inhibition effects at low temperatures, reported by Nozhevnikova *et al.* (1993) and Whalen *et al.* (1990), were also observed, with only half the oxidation capacity at 20°C compared with that at 30°C (Fig. 4). Zero oxidation capacity at 5°C, as reported by Nesbit (1992), was also noticed in the experiment. The experimental results indicated faster inhibition effects at temperatures higher than optimum (45°C). Indirect effects of temperature on the methane oxidation rate were also observed, with high temperatures causing a reduction in topsoil moisture. Borjesson & Svensson (1997) reported a lower oxidation rate at night due to low temperatures in temperate climates. Sufficient moisture content and an appropriate temperature (about 20°C) could result in higher oxidation. Contrary to this, the extremely high atmospheric temperatures (about 40°C) in tropical regions could cause the topsoil to become totally dry, resulting in low oxidation (Fig. 5). This could force the methanotrophs to survive in starvation and/or migrate well below the top surface to where insufficient oxygen is available for them to operate at optimum capacity.

Average methane oxidation rates

Batch experiments

The average and maximum oxidation rates calculated for fresh soil samples were 1.78×10^{-6} and 3.00×10^{-6} g CH_4 (g soil)$^{-1}$ h^{-1}, respectively, when incubated at 30°C and 15% moisture content. The average methane oxidation capacity of the column soil was higher than the fresh soil. This could be due to the presence of relatively higher numbers of methanotrophs in the column soil exposed to methane for a longer period. The highest reported oxidation capacity is 6.4×10^{-3} g CH_4 (g soil)$^{-1}$ d^{-1} in soil samples collected from an old landfill (Nozhevnikova *et al.* 1993). A low oxidation capacity of 5.7×10^{-8} g CH_4 (g soil)$^{-1}$ d^{-1} was also reported by Boeckx & Van Cleemput (1996). Barratt (1995) reported a maximum oxidation capacity of 166×10^{-6} g (g soil)$^{-1}$ h^{-1} for a soil–vermiculite mix. However, when the same experiment was conducted with pure soil, the value reported was 6.25×10^{-6} g (g soil)$^{-1}$ h^{-1}. In the experimental runs reported here, the average and maximum oxidation capacity in column I soil samples (150 days) were 7.7×10^{-6} and 9.1×10^{-6} g CH_4 (g soil)$^{-1}$ h^{-1}, respectively, when incubated at 3 to 4% initial headspace methane concentration at 30°C. The relatively higher value could be due to the presence of a vibrant community of methanotrophs, as reported elsewhere. This result can be interpreted in terms of the methane oxidizing capacity of the landfill cover soil with the age of the landfill. The cover soil in old landfills with long-term exposure to methane could be the cause of the extremely high methane oxidation capacity of landfill soil reported by Nozhevnikova *et al.* (1993).

Column experiments

The headspace methane concentration decreased with time in both test columns, indicating the increase in methanotrophic activity with time. Low oxidation rates were observed on the ninth day of the experiment and the oxidation rates increased to 50 and 41 g CH_4 m^{-2} d^{-1} on the 20th day. The methane supply rates were increased from 5 to 9 ml min^{-1} on the 32nd day to investigate the effect of a rate increase on oxidation rates. As seen in Fig. 3a, an average oxidation rate of 63 g CH_4 m^{-2} d^{-1} was observed at a 5 ml min^{-1} flow-rate. Similarly, a decrease in the average oxidation rate at the same moisture content was noticed at lower supply rates (Fig. 6). The methane oxidation capacity determined by Borjesson & Svensson (1997) was 120 to 390 mmol CH_4 m^{-2} h^{-1} (about 46 and 149 g m^{-2} d^{-1}) in a

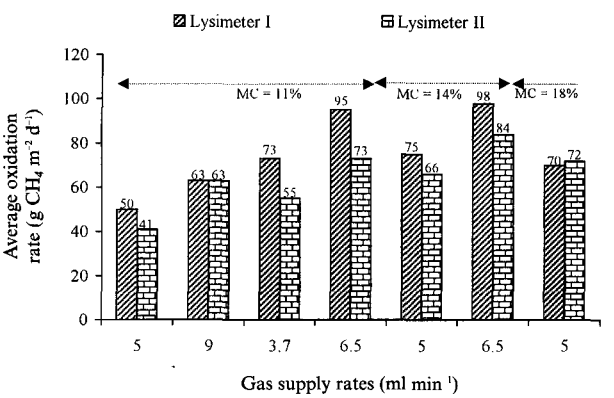

Fig. 6. Average methane oxidation rates at different methane supply rates and average moisture contents for test columns I and II.

Swedish landfill. The lower rate is less than the oxidation rate observed in these experiments.

Average methane oxidation rate at different initial headspace gas concentrations and methane supply rates

Initial headspace gas concentrations in batch experiments

As seen in Fig. 7, an increase in the initial headspace methane concentration in the batch experiments resulted in an increase in oxidation capacity. However, the increase in oxidation capacity was not proportional to the increase in initial headspace concentration. A higher rate of increase in oxidation capacity was observed at low initial headspace gas concentrations. With the increase in initial headspace gas concentration to a higher value, the increase in oxidation capacity was minimal. The oxidation capacity at 1% initial headspace gas concentration in column I soil (150 days) was 4.3×10^{-6} g CH_4 (g soil)$^{-1}$ h^{-1}, whereas the oxidation capacity in the same soil at 5.6% (5.6 times) the initial headspace gas concentration was 10.8×10^{-6} g CH_4 (g soil)$^{-1}$ h^{-1} (only 2.5 times).

The oxidation rate was higher at higher initial methane concentrations. However, the rate of increase in the oxidation rate was not proportional to the increase in initial headspace gas concentration. This could indicate the possibility of constant oxidation rates at higher initial headspace gas concentrations irrespective of the initial headspace methane concentration.

Gas supply rates in the column system

As indicated by the oxidation rate results in Fig. 7, although some increase was observed, an increase in the gas supply rate did not substantially increase the methane oxidation rate. However, some increase in oxidation rate was observed when the gas supply rate was increased. This could be explained by the oxidation rate approaching zero order. Except at 3.7 ml min^{-1}, the flow-rate for column I (observed headspace methane concentration 0%), all the other supply rates followed a similar pattern of an increase in oxidation rates at higher supply rates (although not proportional to the supply rates). An increase in oxidation rates corresponding to an increase in supply rate from 5 to 9 ml min^{-1} was very small (50 to 63 g CH_4 m^{-2} d^{-1}). At higher moisture contents the oxidation rates at 6 ml min^{-1}, 100 g CH_4 m^{-2} d^{-1}, were found to be more than the oxidation rates for a flow-rate of 9 ml min^{-1} with a low moisture content. Bogner (1997) reported the oxidation rate in a US landfill as 48 g CH_4 m^{-2} d^{-1} and suggested that the different trophic groups of methanotrophs may function over different dynamic ranges of methane concentrations. Our oxidation rate (about 100 g CH_4 m^{-2} d^{-1}) is higher than the 45 g CH_4 m^{-2} d^{-1} determined by Whalen *et al.* (1990). The maximum reported oxidation rate is 166 g CH_4 m^{-2} d^{-1} by Kightley *et al.* (1995) with coarse sand columns and 108 g CH_4 m^{-2} d^{-1} with fine sand and clay purged with 5 ml min^{-1} of pure methane in a core of 15 cm diameter (equivalent to a 13.4 ml min^{-1} gas flow-rate for our experiment).

Both batch and column systems showed a similar pattern of oxidation rates, with an increase in oxidation rates at low supply rates (or headspace gas concentrations) and almost constant oxidation rates at higher supply rates (or headspace gas concentration). The oxidation kinetics also resemble the findings of Roslev *et al.* (1997) with low flow-rates resulting in higher order oxidation kinetics. The results indicate the potential for a constant oxidation capacity of soil. Therefore, increased methane flow-rates could cause higher methane emissions from landfills in their mature stage.

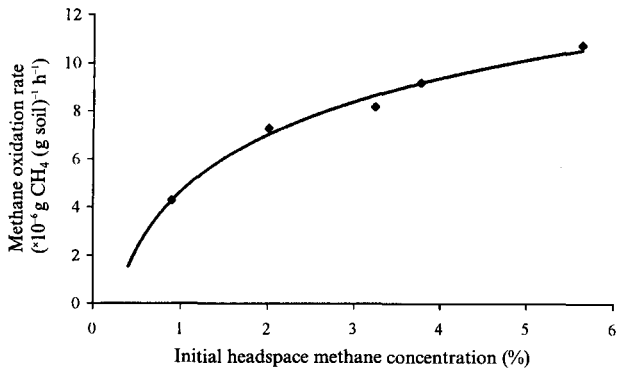

Fig. 7. Batch experiment on test column I soil (150 day old) with 15% moisture contents and incubated at 30°C and different initial headspace methane concentrations.

Depth profiles

The depth profiles of various parameters were monitored daily in the column experiments.

Depth profile of gases

Figure 8a shows the depth profile of various gases in the two test columns and the one control column (measured on the 50th day of the experiment). A decrease in oxidation concentration with depth in the two test columns compared with the control column indicates the presence of oxygen-depleting bacteria in the main column. Although a minor decrease in oxygen concentration and an increase in carbon dioxide concentration in the control column was observed, the amount of decrease was insignificant. This could be the

result of some other aerobic bacteria using oxygen. A decrease in methane concentration with depth in the test columns reflects the presence of methane-utilizing bacteria in the soil. A rapid decrease in the concentration of methane and oxygen at a depth of 35 cm (Fig. 8a) also shows that it is the depth of maximum oxidation. The concentrations of methane and oxygen overlapped at this depth. The whole depth of the soil is under oxidation conditions, with a small concentration of oxygen present at 80 cm depth.

The depth profile of all the gases determined by Kightley *et al.* (1995) was similar to those found in the research reported in this paper, except for carbon dioxide. The use of a carbon dioxide–methane mixture in this experiment is the reason for obtaining a different carbon dioxide profile from that reported by Kightley *et al.* (1995). Nozhevnikova *et al.* (1993), through *in situ* experiments in landfill cover soil, also found a decrease in the concentration of carbon dioxide upwards Barratt (1995) also describes a similar characteristic in an experiment with a column system.

At 35 cm depth the concentration of methane decreases rapidly. A decrease in the relative concentrations of oxygen and methane in opposite directions could also be due to high diffusion at this depth. However, the same soil, when purged with an equal supply of nitrogen in the control column, did not produce such a high rate of decrease in concentration at 35 cm depth. Some decrease in the concentration of oxygen in this column was observed, which could be due to diffusion as well as outgassing by upward flowing gases. A rapid decrease in concentration at this depth indicates high methane oxidation. Also, the methane and oxygen concentrations overlapped (equal concentrations) at this depth, indicating the requirement of an equal concentration of oxygen and methane for maximum oxidation. A comparison of the depth profiles of gases for both the test columns produced a similar pattern of gas concentration at various depths, reflecting a similar behavior in both soils (Fig. 8a). The depth of maximum oxidation reported by Nozhevnikova *et al.* (1993) and Borjesson & Svensson (1997) was between 40 and 60 cm. Kightley *et al.* (1995), however, found maximum oxidation at 25 cm depth in a laboratory experiment with sandy soil. Barratt (1995) reported a wide range of values between 15 and 60 cm for the depth of maximum methane oxidation.

A comparison of the oxygen concentrations in the test columns (2.4 and 2.3%) with the control column (14%) indicates the consumption of oxygen in the test columns. The oxygen consumption could also be confirmed by comparing the aeration coefficient (the ratio between nitrogen and oxygen) for columns I and II alone, which increased from 3.8 at the top to 16.7 at 80 cm depth. The presence of a small concentration of oxygen at 80 cm depth and the absence of methanotrophic bacteria at this depth indicate the requirements of minimum oxygen for effective oxidation as described by William & Zobell (1949).

Depth profile of the oxidation capacity of column soil

The oxidation capacity measured in the batch experiments for soil samples collected from different depths of column is shown in Fig. 8b. The maximum oxidation capacity of the soil was observed with samples collected from 10 to 35 cm depth. The oxidation capacity measured at the top and at 80 cm depth was very low due to the unfavorable conditions (moisture content and oxygen) at these depths. This result indicates the presence of maximum methanotrophic activities at 10 to 35 cm depth. This result also supports the depth profile of gases as shown in Fig. 8a, indicating 35 cm depth as the depth of maximum oxidation.

Depth profile of methane carbon dioxide ratio

The initial methane and carbon dioxide ratio of 1.5 in the test columns was found to decrease upwards along the column. The ratio was 0.1 and 0.2 in column I and II, respectively, indicating the decreasing concentration of methane and increasing concentration of carbon dioxide. Figure 8c shows the depth profile of methane to carbon dioxide ratio for a 5 ml min^{-1} gas supply and 15% average moisture content. This figure indicates the rapid decrease in the ratio at depths between 10 and 35 cm, indicating high methanotrophic activity at this depth.

Depth profiles of total organic carbon and microbial population

Figure 8d represents the depth profile of the total organic carbon and the microbial population for column I. A comparatively higher value of the total organic carbon was noticed at a depth between 30 and 60 cm, although the difference was not substantial. The bacteriological analysis to determine the presence of indicators of methanotrophs in the soil gave the following results. The soil incubated under a methanol atmosphere indicated the possibility of higher concentrations of methanotrophic bacteria at a depth of 20 to 40 cm. Higher values of the bacteriological population in a column at a depth of 35 to 60 cm could be related to higher values of organic carbon (although very small differences) due to the maximum rate of microbial activities at this depth. Kightley *et al.* (1995) also reported higher concen-

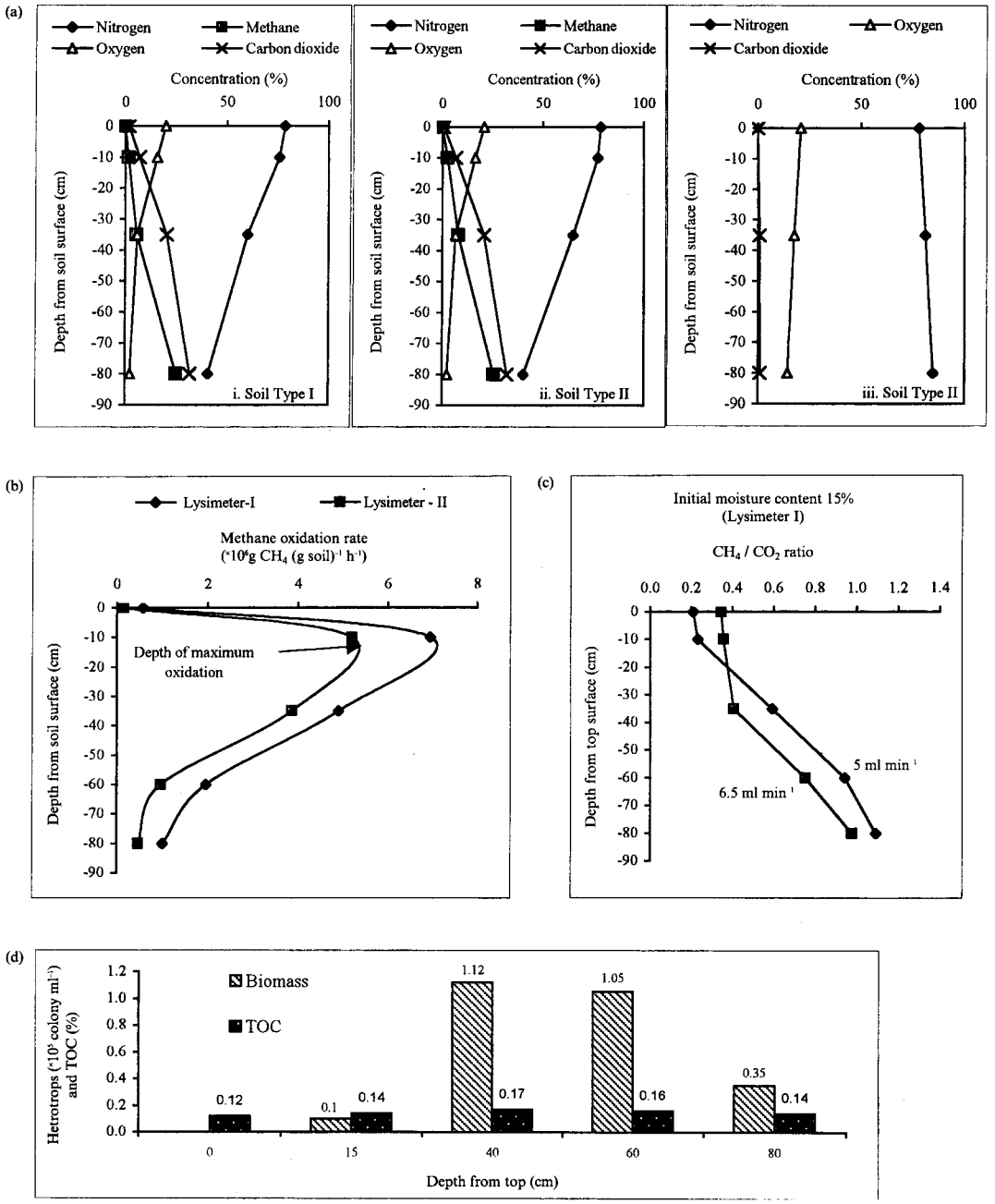

Fig. 8. Depth profiles of various parameters. (a) Depth profile of gases: (i), (ii) lysimeters I and II purged with gas at 5 ml min^{-1} and (iii) lysimeter III purged with nitrogen at 5 ml min^{-1} (after 50 days of experiment). (b) Depth profile of oxidation potential of soil removed from various depth of lysimeters (after 55 days of experiment). (c) Depth profiles of methane to carbon dioxide ratio for test column I with a moisture content of 15%. (d) Depth profile of microbial number and total organic carbon (60 days).

trations of carbon at a depth of high oxidation. Little difference was observed in the measurement of the redox potential of soil with depth, although a gradual decrease in the oxidation reduction potential (ORP) was noticed with depth, indicating the presence of oxidizing conditions at the top. The ORP was +452 at the top and +311 at 80 cm depth, measured on the 65th day. William & Zobell (1949) reported higher oxidation at higher values of ORP in batch experiments.

Interrelationship between various parameters

Experimental studies on methane oxidation in the columns and the batch experiments show similar results under various environmental conditions. A low moisture content inhibited methane oxidation in both batch and column experiments. The optimum moisture content for both experiments was around 15%, and higher moisture contents up to 25% produced significant oxidation measured in the batch experiments. Higher moisture contents could not be

maintained in columns due to drainage problems. Even so, a high oxidation rate was observed at 18% moisture content.

Temperature was another important parameter controlling the methane oxidation rate. Temperatures below 20°C did not produce much methanotrophic activity. Higher temperatures (up to 37°C) were more effective. This condition could assist in effective oxidation in a tropical climate. However, very high temperatures (about 40°C) in tropical climates could also desiccate the surface moisture, producing less methanotrophic activity in surface soils.

Higher supply rates enhanced the methane oxidation rates, but the effect was not proportional to the supply rates. This could be because of the limiting capacity of the methanotrophs under this environmental condition. A low moisture content and high supply rate did not increase the methanotrophic activity, but high moisture contents (15%) increased the oxidation rates even at low supply rates. The oxidation rate at 9 ml min^{-1} gas supply with 11% moisture content (70 g m^{-2} d^{-1}) was lower than the oxidation rate at a gas supply rate of 5 ml min^{-1} and 14% moisture content (90 g m^{-2} d^{-1}). Higher supply rates and higher temperatures with low moisture contents always increased the headspace methane concentration in lysimeters, indicating limiting oxidation rates. Similarly, very high moisture contents (above 25%) also inhibited oxidation by restricting the diffusion of the atmospheric air required for oxidation.

The results of the experimental program on methane oxidation and parameters affecting oxidation are plotted and interrelated to determine the depth of maximum oxidation (Fig. 8). The depth of maximum oxidation under normal conditions is between 15 and 40 cm as a result of low moisture contents at the surface.

Oxidation kinetics at different temperatures

As described previously, temperature was one of the main environmental parameters controlling the methane oxidation rate. Calculation of the temperature dependence of the methane oxidation capacity was made in the batch experiment with the main column I topsoil sample collected after 150 days from the start of the experiment. The oxidation capacities of the topsoil with 16% moisture contents incubated at different temperatures (20, 30 and 37°C) under different initial headspace gas concentrations are shown in Fig. 9.

From this figure it can be observed that the oxidation capacity is augmented by an increase in temperature and initial headspace gas concentration. The oxidation rate was higher at 30°C than at 20°C. The kinetic parameters for

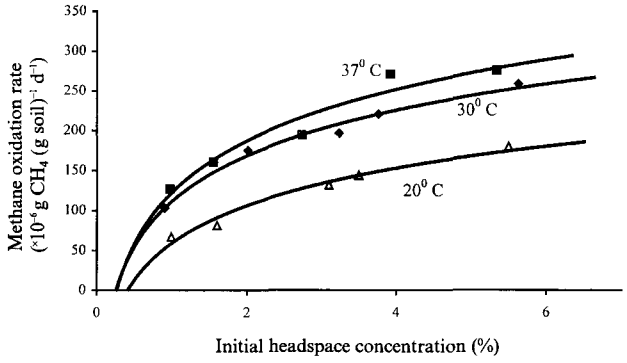

Fig. 9. Methane oxidation rates in soil samples collected from test column I and incubated under different headspace gas concentrations and different incubation temperatures.

methane oxidation are: K_m, 3.46 mg; K, 171.54 × 10^{-3} d^{-1}; and V_{max}, 430 × 10^{-6} g CH_4 (g dry soil)$^{-1}$ d^{-1}, about 1125 nmol (g dry soil)$^{-1}$ h^{-1}. Previously published values of V_{max} range from 5.6 to 3377 nmol (g dry soil)$^{-1}$ h^{-1} (Bogner *et al.* 1997).

Conclusions

Active methanotrophs are capable of utilizing methane as carbon and energy sources, producing carbon dioxide and water. These bacteria are found in soil environments containing high concentrations of methane and an abundance of oxygen. Different environmental parameters were found to be responsible for controlling methane oxidation rates. Low moisture contents are not favorable for methanotrophic bacteria. Temperature, on the other hand, is one of the most important parameters in controlling methane oxidation rates. Tropical climates with high soil temperatures are the most suitable for methane oxidation, but at the same time low moisture contents due to high temperatures at the soil surface may reduce methane oxidation rates. These two factors in tropical climates have the effect of encouraging a methanotrophic population to become established below the upper surface of a landfill.

From these findings it is believed that methane oxidation in tropical landfill covers is likely to be maximized immediately after a rainy season, with a sufficient moisture content (but not saturated) and optimum temperatures. One method of maintaining the moisture content is by recirculating leachate via the cover (spraying), which can serve two purposes. One purpose is to degrade the organic matter present to produce methane and the other is to provide suitable environmental conditions for oxidation in the top cover soil. An increase in the moisture content at the

top of the landfill to bring methanotrophs to the surface may be another alternative to attain maximum oxidation rates.

The current landfill cover system design is mainly based on the desire to minimize leachate production by restricting the infiltration of rainfall. Consequently, cover systems are designed to convert precipitation into surface runoff or evapo-transpiration. Therefore, a hydraulic barrier is an integral part of a cover. However, a gas collection and venting system has been introduced into many designs to address the off-site migration of methane due to pressure build-up in the landfill.

Incorporating a methane oxidative layer can cause a few problems in terms of the objectives of current designs of landfill covers. Optimum oxidation occurs when the soil is aerobic and there is enough moisture available. It has also been found that maximum oxidation occurs at a depth between 15 and 40 cm under normal conditions in tropical soils. The optimum soil moisture is around 15% (depending on the soil). To be aerobic the soil should be more permeable to allow oxygen to diffuse deep into the soil. Coarse-grained soil is best suited for this purpose. On the other hand, it allows water to infiltrate more easily and produce leachates. A drainage layer will not help much because gas might also migrate laterally. Evapo-transpiration should also be controlled otherwise the soil will dry below the optimum moisture content required for methanotrophy. Therefore there are some conflicts between minimizing the local (leachate) and global (methane) impacts. Hence the philosophy of the designs should change to strike a balance between these two interests.

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