Laboratory Evaluation of Coupled Thermo-Hydro-ChemoBiological (THC-B) Processes in Compost based Biocover

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A. Khoshand and M. Fall
University of Ottawa, Ontario, Canada

Abstract

Anaerobic biodegradation of waste in landfills produces methane (CH\textsubscript{4}) which is a potent greenhouse gas. CH\textsubscript{4} can be oxidized by natural biological processing when it passes through reactive biological cover soils or biocovers. Previous studies have mostly focused on the isolated effects of influencing factors on biocover performance and there is a lack of understanding on the coupled thermal (T), hydraulic (H) and chemo-biological (C-B) (THC-B) processes that occur in biocovers. This paper presents the results of laboratory column experiments on a compost based biocover. The obtained results are presented and discussed with regard to the interactions between the THC-B processes that occur in the studied biocover.

Keywords: biocover, methane oxidation, temperature, moisture

1. Introduction

Landfill gas (LFG) is produced by the anaerobic biodegradation of the organic fraction of disposed waste in landfills [1]. LFG consists of about 55–60% methane (CH\textsubscript{4}), 40–45% carbon dioxide (CO\textsubscript{2}) [2, 3], and numerous volatile organic compounds (VOCs) [4, 5, 6]. CH\textsubscript{4} is a greenhouse gas with a global warming potential which is 25 times that of CO\textsubscript{2} [7]. Landfills are one of the significant sources of anthropogenic CH\textsubscript{4} emissions [8]. It is now estimated that 10–20% of the worldwide anthropogenic CH\textsubscript{4} emissions are attributed to landfills [9]. So, urgent control of CH\textsubscript{4} emissions in landfills is necessary for reducing CH\textsubscript{4} concentration in the atmosphere [10].

Some of the advanced technologies, such as the extraction of LFG and its utilization in the form of energy, can reduce the environmental impacts of landfill emissions [11, 12]. However, there is evidence of the escape of large amounts of CH\textsubscript{4} gas at sites which are equipped by the
mentioned technologies in [13]. Also, the costs involved in the technical degasification of old landfills are high [14] and these technologies are mainly feasible for large and new landfill sites [11]. Attention is now currently focused on the development of low cost alternative technologies where implementation of the conventional mentioned methods is not economically or technically feasible [10, 15, 16]. One of the most promising alternative approaches to conventional methods is passively venting the LFG through a reactive biological cover soil or biocover in order to oxidize CH\(_4\) by natural biological processing [12, 17, 18, 19]. The natural CH\(_4\) oxidation process that takes place in biocovers is mediated by methanotroph bacteria at the interface of the aerobic and anaerobic zones [20]. The methanotroph bacteria use molecular oxygen (O\(_2\)) to oxidize CH\(_4\) into CO\(_2\) [11] which has a lower global warming potential than CH\(_4\). The CH\(_4\) oxidation reaction through biocovers can be described by the following equation [21]:

\[
38.25 \text{CH}_4 + 3 \text{NH}_3 + 63.75 \text{O}_2 \rightarrow 3 \text{C}_4\text{H}_8\text{O}_2\text{N} + 26.25 \text{CO}_2 + 69 \text{H}_2\text{O} + \text{Heat} \quad (1)
\]

A suitable support medium for CH\(_4\) oxidation should have high porosity, high water holding capacity and appropriate stable nutrient levels [22, 23]. Previous laboratory research [14, 24] and field studies [15, 20, 25, 26] have reported that generally stabilised and sanitised compost materials meet these criteria and can be a suitable medium for CH\(_4\) oxidation. A relatively large number of previous research have studied the CH\(_4\) oxidation capacity of biocovers through laboratory continuous gas flow systems or column tests [14, 20, 24, 27, 28].

Generally, the performance of biocovers is strongly regulated by thermal (T), hydraulic (H), and chemobiological (C-B) factors and their coupled interactions (THC-B). Knowledge about coupled THC-B processes is essential for the design, construction and maintenance of biocovers. The current paper presents the results of an extensive laboratory study in order to discuss the evolution of the main influencing factors (T, H, C-B) and their coupled interactions (THC-B) in compost based biocover laboratory column tests. This approach provides evaluation of the temperature, moisture content, pH, organic content, gas profiles and CH\(_4\) oxidation rate properties of compost biocovers in two dimensions, time and depth.

2. Materials and Methods

2.1. Materials

In the current research, mature and stabilised compost is used as the biocover medium based on numerous literature recommendations [4, 15, 20, 24, 25, 26]. The used compost was collected from an open windrow operation at the Lafleche Landfill site in Moose Creek, Canada, which is owned and operated by Lafleche Environmental Inc. The compost material was sieved through a 9.5 mm mesh and any remaining plant material or large stones were removed by hand. Then, the compost material was moistened to reach a gravimetric moisture content of 35%. This moisture content was recommended by previous studies as the optimum moisture content for methanotrophic activity [14, 20, 29].

2.2. Methods

2.2.1. Column experiment set up and instrumentations

The column experiments were performed in identical Plexiglas tubes (inner diameter of 25.4 cm, 60 cm in height, and 6 mm in thickness). A perforated aluminium plate installed above a drain at the bottom of the column was used to support the compost material. A layer of fine and clean gravel was placed as the gas distribution layer to provide a uniform distribution of CH\(_4\) over the entire area of the biocover. The column was insulated with expansive insulation foam in order to
minimise the lateral heat exchange between the compost material within the column and the surrounding atmosphere.

The column was equipped with four gas sampling ports along the column at intervals of 10 cm from the first port. These ports included perforated acrylic pipes (with an inner diameter of 4 mm) with plastic fitting and septa which facilitate the taking of gas samples by a gastight syringe needle. The inlet CH\textsubscript{4} flow (99% purity) of 13.21 mL m\textsuperscript{-2} min\textsuperscript{-1} (equal to CH\textsubscript{4} flux of 250 g CH\textsubscript{4} m\textsuperscript{-2} day\textsuperscript{-1}) was uniformly injected in the middle of the gas distribution of each column. The selected inlet is similar to the mid to high range of reported landfill CH\textsubscript{4} fluxes. The inlet CH\textsubscript{4} flow rate was monitored by a Mass Stream D5111 mass flow controller (M+W Instruments) [30] which was connected to an NI 9211 data acquisition system (National Instruments Corporation) [31].

The volumetric water content (VWC) was monitored by using four calibrated EC-5 dielectric soil moisture sensors (Decagon Devices) [32] at heights of -5, -15, -25, and -35 cm from the surface of the biocover. EC-5 sensors were placed at the center of the column cross-section and connected to the EM 50 data loggers (Decagon Devices) [33]. Also, the evolution of the temperature at the depth of the column with time was measured by using TH-T temperature sensors (Roctest Ltd.) [34] which were installed at the same level where the EC-5 sensors were located. A digital temperature / humidity meter was also installed at the top of the column to monitor the relative humidity and room temperature during the experiment.

### 2.2.2 Material characterization tests

Extensive laboratory testing was carried out to evaluate the T, H, and C-B properties of the used compost material before beginning the column tests (Phase I). The column was dismantled after 153 days (Phase II) of the operation and the material was removed in the undisturbed layers in order to prepare samples for post analysis. The characterization of all the samples was performed in accordance with the procedures described by the American Society of Testing and Materials (ASTM) standards [35]. Measurements of Atterberg limits, organic content and pH were performed according to ASTM D4318, ASTM D 2974, and D 4972 respectively. A particle size analysis was carried out by following ASTM procedure D422 for the entire column at the mentioned phases of time.

### 2.2.3 Gas analysis

The gas samples (1 mL) were analysed by manual injection via a Series 400 isothermal gas chromatograph (GC) instrument (Gow-Mac Instrument Co.) [36] which was equipped with a thermal conductivity detector (TCD) and two stainless steel columns. O\textsubscript{2} and nitrogen (N\textsubscript{2}) were analysed on a Molsieve 13X, 80/100 mesh column, while a HayeSep Q, 80/100 mesh column was used to quantify CH\textsubscript{4} and CO\textsubscript{2}. The carrier gas was high purity helium (99.999% He, vol. basis), and the column, injector, and detector temperatures were respectively 120\degree C, 130\degree C, and 130\degree C. Gas standard samples which were made directly from a cylinder of a known volumetric concentration were used for calibration.

### 3. Results and discussion

#### 3.1. Material characterization

The results of the material characterization tests for pH, bulk density, porosity (e), Atterberg limits, and organic content at different phases of time and depths are shown respectively in Figures 1a-e. As illustrated in Figure 1, the used compost in this study is a porous medium (initial porosity of 3.84), has high organic content (initial organic content of 29.4 %), and has a
neutral pH (initial pH of 6.7). These properties are similar to those which are recommended as the properties of suitable biocover materials in the literature [29]. It can be seen from Figure 1a (pH profiles) that the pH values remained relatively constant with time, but there is a slight decrease with depth throughout the study period. This trend is similar to those that have been reported by [37, 38]. The bulk density profiles (Figure 1b) indicate that the bulk density values are increased and the compost medium becomes compacted with depth. The bulk density of the compost medium also increases with time due to the rearrangement of the orientation of the compost particles and organic content degradation [39]. The compaction of the medium resulted in a general reduction of the porosity (Figure 1c) with depth and time, which can affect gas transport and CH$_4$ oxidation rate over time through the biocover medium [39]. It can be seen from Figure 1d that the liquid limit (LL) of the compost medium is 78% in Phase I and varies between 74.9-77% for Phase II.

There was no significant change in the LL values with depth. The high ranges of the LLs for the compost materials show good agreement with those quoted in the literature [40, 41]. It should be emphasized that the compost material is non plastic and measurement of the plastic limit is not possible. Figure 1e presents the depth profile of the organic content at different phases of time. The maximum organic content of the biocover medium after the column operation was carried out for 153 days is 40.8%, which is observed at a depth of 25 cm on the column. Methanotroph bacteria under certain environmental conditions produce exopolymeric
substances (EPS) which are long chain organic compounds [20, 39]. Therefore, higher organic content in the medium after 153 days (40.8%) in comparison to the initial value (29.4%) implies the presence of methanotrophic activities. Similar higher organic content at depths with maximum CH₄ oxidation were also reported by [24] in compost columns. The organic content profile can be a surrogate for determining the depth for maximum CH₄ oxidation [39].

Moreover, sieve analyses were performed to evaluate the grain size distributions of the samples, as presented in Figure 2. The compost samples at Phases I and II are classified respectively as organic well-graded sand (SW) and organic poor-graded sand (SP) based on the Unified Soil Classification System (USCS). It can be seen that samples become finer with time due to the mineralization of the compost layers over time [20]. The particle size reduction of the compost material as a biocover medium during column testing over time has also been reported by [20].

[Figure 2. Particle size distribution of studied compost material at different phases of time.]

3.3.  Moisture content

Moisture is essential for methanotrophic activity as a transport medium for nutrient supply and also metabolite removal [27]. Microorganisms are impacted by physiological stress under low moisture content which leads to the reduction of CH₄ uptake [20, 27]. Moreover, excessive moisture slows down gaseous transport processes and impacts CH₄ oxidation capacity. Excessive moisture content causes aqueous diffusion instead of gaseous diffusion of CH₄ and O₂ through the biocover. As aqueous diffusion is much slower (10000 folds less rapid) than gaseous diffusion, the CH₄ oxidation rate is drastically reduced in the biocover under high moisture content conditions [20]. Figure 3 presents variations of the VWC with time at different depths of the biocover. The initial VWC throughout the depth of the biocover is 19% which consistently decreases in the surface layer (-5 cm) with time and reaches a minimum value of 6.2% on day 153.

The top 5 cm of the biocover is noticeably drier due to surface desiccation [42, 43]. The low VWC of the surface layers indicates lack of methanotrophic activity within those depths which can affect the performance of the biocover [24]. Unlike the surface layer, the VWC at the bottom layer is continuously increased with time and higher than the initial VWC after 153 days (36.4% in comparison to 19%). This high VWC can be attributed to the downward migration of moisture [41, 43] and production of water during the CH₄ oxidation process [44].
3.4. Temperature

Temperature has a significant effect on methanotrophic activity [27]. CH$_4$ oxidation rate increases with temperature up to the maximum oxidation rate and then decreases with increasing temperature [45]. The temperature of a biocover is affected by the temperature of the LFG, atmospheric temperature, rainfall, and microbial activity [46]. Different research has reported different optimum temperatures for methanotrophic activity. [42, 47] have reported 30°C as the optimum temperature for CH$_4$ oxidation in loam soil. Laboratory studies conducted by [48] on landfill cover soil showed that the optimum temperature for CH$_4$ oxidation is in the range of 25-35°C. Figure 4 illustrates the development of temperature with time at selected depths. It can be seen that the average temperature of the studied material is 24.7°C at the beginning of the experiment. There is an increasing trend in the temperature for the 10 days at all depths. The increase of the temperature during the first 10 days can be attributed to the aerobic degradation of available carbon sources in the compost [49]. The CH$_4$ oxidation process is exothermic, and releases 780 kJ per mol of oxidized CH$_4$ [50]. This leads to an increase of temperature in the active oxidation layer.
temperature [12, 20]. A similar difference between ambient and column temperatures has been also reported in the literature [20, 44, 51].

3.5. ChemoBiological (C-B) properties

3.5.1. Methane oxidation rate

In this paper, the CH$_4$ oxidation rate at selected depths is defined based on CH$_4$ consumption, see Equation 2.

$$\text{CH}_4 \text{ oxidation rate} = \frac{(C_{\text{CH}_4} t=0) - (C_{\text{CH}_4} t=i)}{(C_{\text{CH}_4} t=0)} \times 250 \quad (2)$$

In the above equation, the CH$_4$ oxidation rate is in g/m$^2$day, $(C_{\text{CH}_4} t=0)$ is the CH$_4$ concentration (%v/v) which was fed into the column and assumed to be 100%, $(C_{\text{CH}_4} t=i)$ is the CH$_4$ concentration (%v/v) on the $i$th day. The adaptation phase is the time that it takes to reach a steady state of CH$_4$ oxidation in the biocover. In the literature, different adaptation periods (between 5 and 10 days) have been reported for laboratory experiments [20, 24, 52]. The adaptation phase for this study is relatively quick (10 days), which implies the presence of a high microbial mass that acclimatizes quickly [24]. The CH$_4$ oxidation rate of the studied column over 153 days is shown in Figure 5. The steady state, or the phase with a high rate of oxidation [24], continued for approximately 119 days after adaptation phase. In the phase with a high rate of oxidation, the CH$_4$ oxidation efficiency reached a maximum value of 209.7 g/m$^2$day which indicates the suitability of the used compost as a biocover medium.

![Figure 5. CH$_4$ oxidation at different depths.](image)

Moreover, the obtained results of the CH$_4$ oxidation rates are compared to the estimated CH$_4$ emissions for a 10 year old bioreactor landfill with assumed depth of 20 meters. It can be observed that the studied biocover is capable of entirely oxidising the released CH$_4$ emission from a 10 year old bioreactor landfill (107 g/m$^2$day [20]). After 119 days of high oxidation rates, a decline in the CH$_4$ oxidation was observed at a depth of 35 cm and the performance of the biocover slowly declined until the end of the experiment (153 days). This phase can be explained by the formation of EPS and is called the EPS-affected phase [24]. EPS formation leads to the clogging of media pores and hinders CH$_4$ and O$_2$ flow within biocovers [20, 49, 53]. The competition for available O$_2$ and nutrients is another factor which contributes to the
reduction in the CH₄ oxidation rate [49]. A similar trend for the CH₄ oxidation rate has also been observed in previous studies [20, 24, 49].

3.5.2. Gas profile

The gas profile indicates the concentration of O₂, N₂, CH₄ and CO₂ at selected depths (-5, -15, -25, -35 cm) within the column. The gas profile within the studied column is the result of the biochemical reaction, diffusion and advection of the gas flow [54]. The gas profiles at six selected different periods (after 25, 50, and 75, 101, 125 and 150 days of column operation) are shown in Figure 6. Generally, concentrations of O₂ and N₂ decline from the top to the bottom of the column while the opposite trend is true for CH₄ and CO₂ concentrations. It can be seen from Figure 6a that the CH₄ concentration becomes less than 20% at the surface layer (-5 cm) at the end of 25 days in the column operation.

![Gas composition graphs](image)

Figure 6. Gas composition for (a): 25th, (b): 50th, (c): 75th, (d): 101th, (e): 125th, (f): 150th day of column operation.

After 25 days of carrying out the column operation, O₂ could no longer be found in layers deeper than 25 cm and a high rate of CH₄ oxidation was observed at depths of 25 to 35 cm below the
surface. Low O$_2$ concentration in layers deeper than 25 cm may be attributed to the fact that O$_2$ was immediately consumed as it became available. As time progressed, the CH$_4$ oxidation layer shifted upwards towards the O$_2$ source located at a depth of 15 to 25 cm below the surface (after 69 days). The shifting of the CH$_4$ oxidation layer has also been reported by [20, 39] who studied the CH$_4$ oxidation of compost based biocovers in column experiments. The reason for the shifting of the CH$_4$ oxidation layer was due to the accumulation of EPS in the deeper layer (25 to 35 cm) of the column which prevented diffusion of O$_2$ further down [20]. It should be noted that shifting of the active CH$_4$ oxidation layer did not affect the overall CH$_4$ oxidation rate and there was still a high rate of oxidation after 69 days. Previous research reported that the active CH$_4$ oxidation layer is generally located at a depth of 20 to 30 cm of the soil profile [1, 47, 55, 56] which is similar to the obtained results in this current research.

In contrast to O$_2$, N$_2$ is not used for the CH$_4$ oxidation process. The presence of N$_2$ at a deeper depth (-35 cm) for an extended period of time can be used as an indicator of air intrusion [51] and also implies that the influent CH$_4$ flux is being diluted [12]. The gas profile changed at the end of the experiment (after 121 days). The CH$_4$ profile showed a gradual increase at the surface layer (Figures 6e and f) and the concentration of O$_2$ that was migrating downward was less than 2%. The reduction in the CH$_4$ oxidation rate can be attributed to unfavourable conditions for methanotrophic activity and EPS formation which developed a layer with low gas permeability at the depth of the biocover. This layer restricted the downward mitigation of O$_2$ and upward mitigation of CH$_4$ [57].

4. Conclusion

In this research, coupled THC-B processes of a compost based biocover are investigated through laboratory column experiments. Also, an extensive laboratory investigation has been performed in order to monitor the evolution of T, H, and C-B factors with two dimensions, time and depth. According to the derived results, the following conclusions can be made.

The used compost material is capable of removing a CH$_4$ influent flux of 209.7 g/m$^2$day. This indicates that the compost material used is able to sustain and enhance the growth of methanotrophic bacteria and can also be potentially used as a biocover medium.

The VWC of the biocover is a key influencing factor on CH$_4$ oxidation. Low efficiency of the surface layer (top 5 cm) in CH$_4$ oxidation especially after 119 days of the column experiment can be attributed to the low VWC of the surface layer (average of 8% in the last 23 days) due to surface desiccation.

Temperature has a significant impact on the performance of the biocover. The average temperature in the column is generally higher than the ambient temperature due to methanotrophic activity. Also, higher average temperatures in the middle layers (30.7°C and 30.27°C respectively at depths of -15 and -25 cm) in comparison to the bottom (-35 cm) and surface (-5 cm) layers (26.34 and 28°C respectively) is related to higher methanotrophic activity and consequently higher CH$_4$ oxidation rate in the middle layers.

5. References


