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Exploring Effective Bio-Cover Materials for Mitigating Methane Emission at a Tropical Landfill

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Abstract: Methane emission and oxidation in different bio-cover materials, i.e., sandy loam, compost, and stabilized wastes, were investigated at a municipal solid waste landfill in Thailand. The bio-cover was purged with extracted landfill gas while methane reduction through biological oxidation was studied. The moisture content in bio-cover materials was maintained with natural rainwater during the wet period and leachate irrigation during the dry period. Methane emissions were found to vary between media and were influenced by rainfall. The methane loading rates of the bio-cover varied from 8.2–20.3 mol/m3/d, being higher during the dry period. Methane removal rates at the bottom part of the biofilter (0.4–0.6 m depth), the most active zone, were found to be from 6.4–10.9 and 7.8–11.4 mol/m^3/d during wet and dry periods. The highest methane removals were found in the lower part of sandy loam, followed sequentially by compost and stabilized wastes. Nevertheless, compost had the highest methane oxidation capacities and greater methanotroph population compared to sandy loam and stabilized wastes. Methanotroph type I was found to predominate during the dry period, whereas methanotroph type II was predominant during the wet period.

Keywords: bio-cover; compost; landfill gas; methane oxidation; methanotrophs; tropical climate

Citation: Chan, R.; Chiemchaisri, W.; Chiemchaisri, C. Exploring Effective Bio-Cover Materials for Mitigating Methane Emission at a Tropical Landfill. *Appl. Sci.* **2023**, *13*, 1990. https://doi.org/10.3390/app13031990

Academic Editors: Fulvia Chiampo and Micaela Demichela

Received: 30 December 2022
Revised: 28 January 2023
Accepted: 31 January 2023
Published: 3 February 2023

1. Introduction

The landfill is the most commonly used municipal solid waste (MSW) disposal method globally [1] due to its flexibility in MSW characteristics, operation simplicity, and low-cost association. Nevertheless, it can generate significant environmental impact in the forms of leachate and landfill gas which can adversely impact human health and the ecosystem if they are not properly managed. Methane (CH_4) and carbon dioxide (CO_2) are the major landfill gases produced in landfills. Among them, methane is an important greenhouse gas with an infrared activity up to 34 times that of CO_2 over a 100-year horizon [2]. Methane from waste accounted for about 18% of total anthropogenic methane emissions in which landfill methane emissions from developed countries have been largely stabilized; however, emissions from developing countries are increasing as more controlled (anaerobic) landfilling practices are implemented [3].

There are several techniques used for mitigating methane emission from landfills such as pre-treatment of organic wastes before landfilling, promotion of aerobic conditions in landfills, landfill gas collection, and utilization of biological methane oxidation in the final cover soil. Among them, biological treatment is the most cost-effective technique for the mitigation of methane emissions through the use of a natural microbial methane oxidation process. Practically, biological oxidation in the bio-filter, bio-window, or bio-cover is a conventional and effective technology for controlling methane emissions at older and smaller landfill sites with low gas potential [4]. During their application, low methane emissions were found in the bio-cover systems operated under passively loaded operation, but some hotspots were identified on actively loaded biofilters [5]. To promote methane

oxidation, several materials have been utilized such as soil, compost, and municipal solid waste [6,7]. Huber-Humer et al. [8] utilized organically labile materials such as compost and natural soil and commonly observed 100% methane oxidation but eventually also observed that the reaction would taper off to a lower level of performance during long-term operation.

Effective methane oxidation could be promoted when environmental factors are properly controlled for methanotrophic microorganisms [9]. Methanotrophic bacteria can be naturally grown in landfill cover materials provided that they are maintained under aerobic conditions. Therefore, it can be considered a complementary strategy for minimizing methane emissions from landfills. Methanotrophs are specific microorganisms that can utilize methane as a sole carbon and energy source under aerobic conditions [10]. The capacity of methane oxidation mainly depends on the characteristics of materials such as porosity, water/gas permeability, and nutrients, as well as environmental conditions such as temperature, soil moisture, presence of vegetation, etc. [9,11,12].

In tropical climate conditions, maintenance of methanotrophic activities in landfill cover is challenging due to distinct difference in climatic conditions between the monsoon (rainy) season with short-period intensive rain events and the other long dry seasons, i.e., summer and winter with extensive evaporation where moisture content in landfill cover drops significantly. Therefore, the selection of appropriate materials is essential for maintaining moisture content, especially in the dry period for which leachate irrigation can be considered [13]. Meanwhile, ambient temperatures play a less important role in regulating methane oxidation in a tropical landfill because there is not much difference in temperature (30–35 °C) between the seasons [14].

Our previous investigations in laboratory-scale set-ups simulating tropical climate conditions have demonstrated high methane oxidation capacities, e.g., >80% of methane loaded in landfill cover materials [14,15]. Nevertheless, there was a wide fluctuation of reported methane oxidation capacities observed in the field varying from 27.5% to 52.5% depending on the waste disposal and climatic conditions [16–18]. The discrepancy in methane oxidation capacities between different scales could be attributed to differences in soil structure, water content, and temperature of landfill cover soils between laboratory- and field-scale operations [19].

Therefore, this study was conducted to evaluate actual methane emission and oxidation in bio-cover operated at a tropical municipal landfill in Thailand. Sandy loam (SL), compost (C), and stabilized waste (SW) were employed as alternative materials to clay, which is typically used as landfill cover. Enhanced methane oxidation in those media was anticipated as oxygen supply would be improved through their high porosity properties. Moreover, organic and nutrient supplements from alternative materials as well as leachate application would also help promote their methanotrophic activities. However, their performance would be affected by environmental condition changes during wet and dry seasons of tropical climate.

2. Methodology

2.1. Bio-Cover Unit Set-Up and Operation

Three bio-cover units (1.50 m diameter × 1.00 m height) with the schematic shown in Figure 1 were installed at a municipal landfill in Thailand. Each unit contained different packing materials, i.e., sandy loam, compost, or stabilized wastes, of 0.60 m depth providing a material volume of 1.0 m^3. The sandy loam was prepared by mixing with clay soil to obtain a final composition of 55.7% sand, 25.9% of silt, and 18.4% clay. Commercial compost prepared from animal manure and yard wastes was used whereas stabilized waste (>10 years old) was obtained from a closure waste disposal area of the studied landfill. Their chemical characteristics are shown in Table 1.

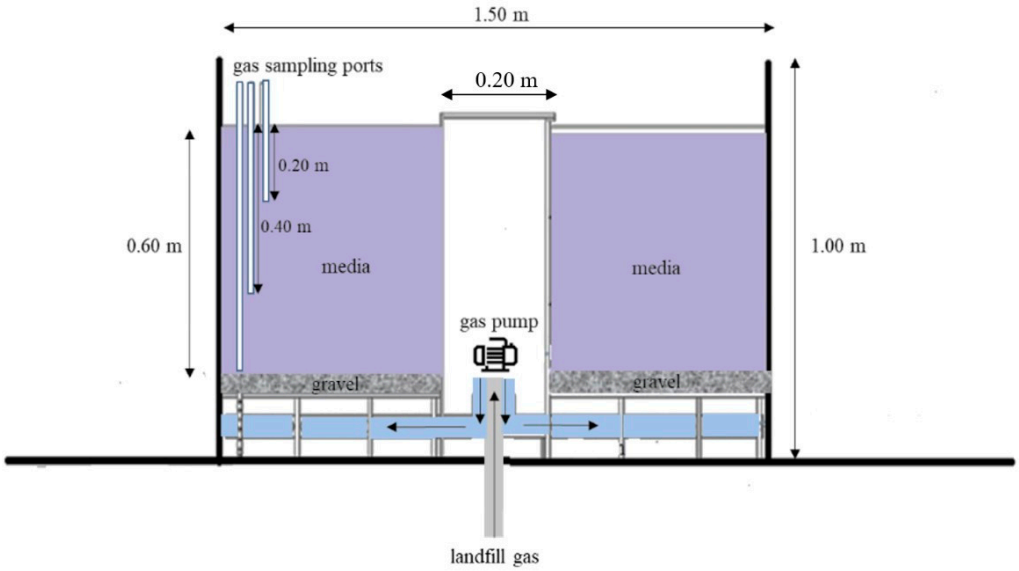

Figure 1. Schematic of bio-cover unit.

Table 1. Physical and chemical characteristics of bio-cover materials.

Parameter	Sandy Loam (SL)	Compost (C)	Stabilized Wastes (SW)
Bulk density (kg/m^3)	1178	595	189
Porosity (%)	55.5	71.4	75.5
pH	7.27	7.12	6.8
EC (µs/cm)	1766	1206	285
TOC (%)	0.6	30	71.1
NH_4^+-N (mg/kg)	1.38	4.74	0.6
NO_3^--N (mg/kg)	16.4	5.5	57.9
TN (g/kg)	1.08	0.18	0.15
TP (g/kg)	0.43	1.08	0.61

Landfill gas extracted from the closed landfill area was supplied at a constant flow rate of 576 L/d, equivalent to an average methane flux of 16 mol/m^3 of media volume/d. The gas pump was operated with battery cells contained in a gas-tight box. The air flow rate was adjusted using a valve, regularly checked and calibrated with a drum-type gas meter (Ritter, TG05/3) to ensure a constant flow rate to the bio-cover. The gas residence time in the bio-cover varied from 0.9–1.3 days depending on its porosity. The gas was supplied through a gas distribution pipe located at the bottom of each bio-cover unit. During the operation, precipitation at the studied site was recorded using an automatic rain gauge station. The temperature and moisture content of bio-cover materials were regularly monitored using portable meters (ECHO, EC-5) and the moisture reduction through evaporation during non-raining periods was determined as about 3% per day. To maintain the optimum moisture content (wet weight basis) in those materials, i.e., 10–20% in sandy loam and stabilized waste and 50–55% in compost found in our previous studies [15,20,21], 20 L of landfill leachate taken from the leachate storage pond with the characteristics shown in Table 2 was sprayed onto bio-cover materials once every two days.

Table 2. Characteristics of leachate.

Parameters	Range	Average
pH	8.6–8.9	8.76
EC (μs/cm)	256–262	259
BOD (mg/L)	100–200	150
COD (mg/L)	1300–2000	1725
TKN (mg/L)	50.4–70	68
NH_4^+ (mg N/L)	2.24–5.6	4.5
NO_2^- (mg N/L)	1.615–1.664	1.639
NO_3^- (mg N/L)	44.64–52.63	46.47
PO_4^{3-} (mg P/L)	0.249–0.818	0.418

2.2. Samplings and Analyses

Gas samples were taken at different depths of the bio-cover material layer, e.g., 0 m (surface), 0.20 m, 0.40 m, and 0.60 m distance (inflow gas at gravel bed), respectively. The sampling was performed during the daytime on a weekly basis and the gas samples were analyzed for their composition (CH_4, CO_2, O_2) using a gas chromatograph (GC 6890, Shimadzu, Kyoto, Japan) equipped with a CRT1 column (Alltech, Nicholasville, KY, USA), and a thermal conductivity detector (TCD). The gas analyses were performed in triplicate using 300 μL injection volume, inlet temperature 105 °C, column temperature 35 °C, detector temperature 150 °C, and helium carrier gas of 65 mL/min.

The biofilter media were also analyzed monthly for their bulk density, porosity, moisture content, pH, electrical conductivity (EC), total organic carbon (TOC), NH_4^+, NO_3^-, total nitrogen (TN), and total phosphorus (TP). The initial physical and chemical characteristics of bio-cover materials used in this study are presented in Table 1. Their analyses were performed following the Standard Methods of Soil Analysis [22].

2.3. Evaluation of Bio-Cover Performance

The performance of bio-cover was evaluated in terms of methane loading rate (MLR) and methane removal rate (MRR) using the following equations.

$$MLR = \frac{Q \times C_i}{V_m} \quad (1)$$

$$MRR = \frac{Q \times (C_i - C_o)}{V_m} \quad (2)$$

where Q is the gas flow rate (m^3/d), C_i and C_o are the inflow and outflow methane concentrations (mol/m^3), and V_m is the material volume in the corresponding section of the bio-cover unit (m^3). Average and standard deviation of the *MLR* and *MRR* in each bio-cover section were determined on a weekly basis during the wet and dry operation periods.

The methane mass leaving the bio-cover units was also quantified using close-flux chamber methodology on a weekly basis. The procedures for the methane emission rate (MER) measurement are described in Chiemchaisri et al. [23]. An acrylic chamber (0.2 m diameter, 0.25 m height) was temporally installed at the surface of bio-cover materials during which the increasing rate of methane concentration in the chamber was determined. The MER was then calculated using the following equation.

$$MER = \frac{V}{A}\left(\frac{dC}{dt}\right) \quad (3)$$

where MER = methane emission rate ($mol/m^2/d$), V = chamber volume (m^3), A = footprint area of chamber (m^2), and dC/dt = linear slope of methane concentration over time ($mol/m^3/d$).

The outflow methane mass over time (*MER* multiplied by bio-cover unit area) was then calculated and presented as the percentage of methane mass loading into the bio-cover

unit ($Q \times C_i$). The difference between inflow (loading) and outflow methane mass was considered as the removal in the bio-cover unit.

2.4. Methane Oxidation Capacities and Methanotroph Population

The samples were collected from three depths of 0–0.20 m, 0.20–0.40 m, and 0.40–0.60 m in the biofilters operated under stable conditions (6th months of operation). Their methanotrophic activities and in situ microbial population were determined. To evaluate methane oxidation capacities, 10 g of wet media sample was placed in a 188 mL serum bottle capped with rubber septa and an aluminum ring. Then, 10 mL of 99.9% methane gas was injected into each bottle. All tests were incubated at room temperature (28–30 °C). The headspace gas composition in the bottle was determined at different times (0, 5, 24, 48, and 72 h) using the gas chromatograph. The methane oxidation rate (*MOR*) was then calculated using the following equation.

$$MOR = \frac{V_g}{W_m}\left(\frac{dC}{dt}\right) \quad (4)$$

where V_g = gas volume in headspace (cm^3), W_m = dry weight of media (g), and dC/dt = linear slope of methane concentration over time ($\mu mol/cm^3/h$).

Moreover, the carbon dioxide production rate (CPR) was also determined from its increasing rate during the batch experiment in the same manner as *MOR* to confirm the stoichiometric ratio of methane consumption and carbon dioxide production.

For the microbial study, the methanotrophic consortium including Methanotroph types I and II were examined using the fluorescence in situ hybridization (FISH) technique. The fluorescent probes Mγ84 + Mγ705 were used for counting the methanotroph type I population, whereas that of Mα450 was used for the methanotroph type II population [24]. The use of polymerase chain reaction (PCR)-denaturing gradient gel electrophoresis (DGGE) techniques followed by the determination of DNA copies in each DGGE band was performed to quantify the methanotroph populations. The detailed analytical procedures are described in Muenmee et al. [25].

3. Results and Discussion

3.1. Environmental Conditions at the Site

The environmental conditions of the site were monitored while the study was performed over six months covering three months of wet and dry periods. Several factors such as ambient temperatures, rainfall, and moisture content of biological media could influence their methane oxidation capacities. Based on monitored records, the ambient temperature varied from 30–35 °C whereas they fluctuated less from 28–30 °C in the bio-cover materials. This temperature range did not adversely affect the activities of methanotrophic microorganisms [26].

The recorded precipitation data suggested weekly rainfall intensities varied from 0 to 0.67 m during the wet period (September–November) with peak rainfall observed during week 8 (October) and no precipitation taking place during week 7. For the dry season (December–February), rainfall intensities were 0 in all weeks except week 8 (0.10 m).

The moisture content at the upper part of bio-cover material was found to be 52.1–56.2% in sandy loam, 58.6–60.3% in compost, and 32–39.1% in stabilized waste during the wet period. During their operation with leachate irrigation over the dry period, their moisture content was 20–25%, 53–57%, and 19–27%, respectively. The moisture content in all materials was higher in the wet period than in the dry period. Methane emission and oxidation in bio-cover materials were expected to be influenced by the rainfall events, especially those observed during wet periods as higher water retention in the media could reduce gas flow through media and oxygen availability required for methane oxidation [14].

3.2. Observed Methane Removals in Bio-Cover

During the six months of operation, methane loading rates (MLRs) to the bio-cover were found to vary from 8.2–20.3 $mol/m^3/d$ (Table 3). The MLRs to the biofilters dur-

ing the dry period (13.7–20.3 mol/m^3/d) were higher than those during the wet period (8.2–11.5 mol/m^3/d). It was due to higher methane content in the extracted landfill gas attained during the dry period, i.e., 20.6–29.6% than those during the wet period (12.9–19.2%). Chiemchaisri et al. [26] reported that higher methane emission during the dry period in tropical landfills could be associated with an increase in gas-filled porosity in cover soil as well as the presence of cracks as soil moisture fell below its shrinkage limit thus facilitating gas transport from the landfill. After bio-cover treatment, the residual methane concentrations observed in the outflow gas during the wet period were not detected (ND) to 0.15% whereas they were 0.6 to 0.7%. during the dry period. Therefore, observed methane removal efficiencies in those bio-covers were >99% during the wet period and 97–98% during the dry period with only small differences between them.

Table 3. Observed methane loading and removal rates in bio-cover materials.

Period	Media Depth (m from Top)	MLR (mol/m^3/d)			MRR (mol/m^3/d)		
		SL	C	SW	SL	C	SW
Wet	0–0.20	0.4 ± 0.1	0.4 ± 0.2	0.2 ± 0.1	0.3 ± 0.1	0.3 ± 0.2	0.2 ± 0.1
	0.20–0.40	0.6 ± 0.2	2.2 ± 0.7	1.7 ± 0.6	0.2 ± 0.1	1.8 ± 0.5	1.6 ± 0.6
	0.40–0.60	11.5 ± 1.7	10.5 ± 0.9	8.2 ± 1.4	10.9 ± 1.5	8.3 ± 0.8	6.4 ± 1.2
Dry	0–0.20	1.5 ± 0.1	1.4 ± 0.2	0.9 ± 0.1	1.0 ± 0.1	1.0 ± 0.2	0.6 ± 0.1
	0.20–0.40	8.9 ± 0.6	5.2 ± 0.7	5.8 ± 0.2	7.4 ± 0.5	3.9 ± 0.6	4.9 ± 0.2
	0.40–0.60	20.3 ± 0.9	15.3 ± 1.2	13.7 ± 0.4	11.4 ± 0.8	10.1 ± 0.9	7.8 ± 0.5

Remark: MLR, MRR: Methane Loading, Removal Rate; SL: Sandy Loam; C: Compost; SW: Stabilized Wastes.

In the bio-covers, the majority of methane removals took place at the first 0.2 m from the inlet, i.e., 0.4–0.6 m depth for all media. The average MRRs of SL, C, and SW media in this active zone during the wet period were 10.9, 8.3, and 6.42 mol/m^3/d which accounted for 94.8%, 79.0%, and 78.0% removal efficiency, respectively. The average MRRs during the dry period in those materials at the same location were 11.4, 10.1, and 7.8 mol/m^3/d, or 56.2, 66.0, and 56.9% removal efficiency. Based on these results, the MRRs observed in bio-cover materials during the dry period were found to be slightly higher than those during the wet period. Nevertheless, their observed removal efficiencies were considerably lower due to the high MLRs attained during the dry period. The residual methane leftover from the lower part of the bio-cover was subsequently removed in the upper parts of the bio-cover material layer. Especially during the dry period, the remaining methane was largely removed at 0.2–0.4 m depth at the MRRs of 3.9–7.4 mol/m^3/d and observed removal efficiencies of 75.0–84.5%. Due to effective removals of methane in those active zones, the upper part of the material (0–0.2 m) was responsible for removing only a small amount of residual methane mass with the observed MRRs of 0.2–1.0 mol/m^3/d.

Figure 2 shows the gas profiles observed in different bio-cover materials during wet and dry periods. As methane was supplied to the bio-cover materials, it was reduced along the upward flow through media while oxygen, supplied through surface diffusion at the top of the bio-cover, was utilized for methane oxidation and thus produce carbon dioxide as the end product. From the results, it was found that the SL and C media at 0.40–0.60 m depth were most effective for methane removals. Similar gas profile patterns were also observed in those materials operated during the wet period. For SW material, methane removal was lower than those in the other materials, but the removal could still take place in the upper part thus yielding satisfactory total removal. In all cases, oxygen was found highly available throughout the depth of bio-cover for methane oxidation.

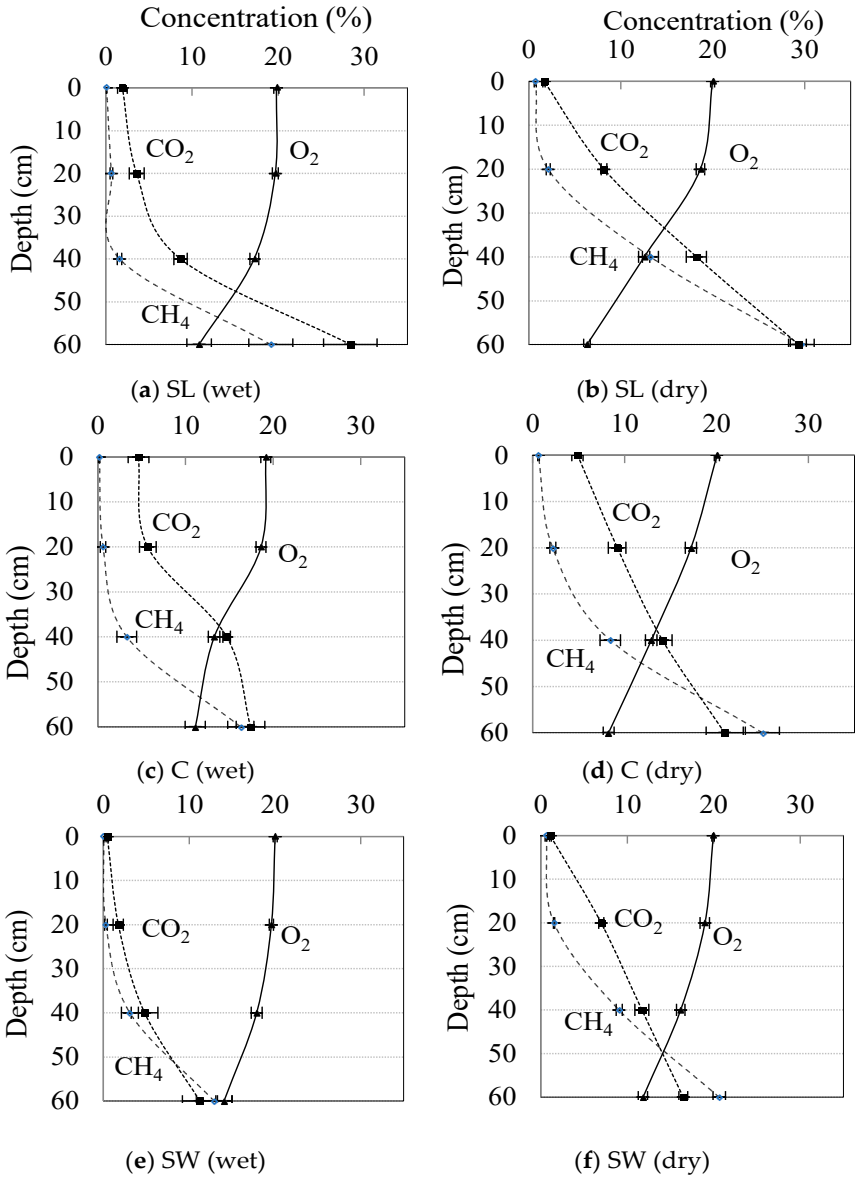

Figure 2. Gas concentration profiles observed in sandy loam (SL), compost (C), and stabilized waste (SW) media during wet and dry periods.

The overall MRRs in SL, C, and SW materials (0.6 m layer depth) observed during the wet period in this study were 11.4, 10.4, and 8.2 mol/m^3/d which are equivalent to 109.4, 99.8, and 78.7 g/m^2/d. The removals were increased to 19.8, 15.0, and 13.3 mol/m^3/d or 190.1, 144.0, and 127.7 g/m^2/d during dry period in those materials, respectively. Comparatively, a wide range of methane removals in bio-cover were reported in the literature. Abichou et al. [27] reported a much lower removal rate of 2.7 g/m^2/d in compost bio-cover whereas Berenjkar et al. [28] reported as high as 237 g/m^2/d in a bio-window filled with biosolids compost supplemented with yard waste and leaf compost. Similarly, Fjelsted et al. [29] reported a maximum methane oxidation rate of 460 g/m^2/d in an active loaded open compost filter treating diluted landfill gas. Meanwhile, Duan et al. [5] reported a comparable methane oxidation rate of 60.6–70.6 g/m^2/d when Danish biofilters were loaded at 60.7–70.7 g/m^2/d with an observed oxidation efficiency of 98.9–99.9%.

The average MER determined by the close flux chamber measurement at the surface of bio-cover material layers (weekly MER variation shown in Figure S3) were 0.007, 0.016, and 0 mol/m^2/d for SL, C, and SW materials during the wet period and 0.092, 0.080, and 0.069 mol/m^2/d for those respective materials during the dry period. Those emissions

accounted for 0–0.8% and 1.4–1.6% of methane loading to the bio-cover material layers during wet and dry periods, respectively. The results suggested that there was only a small amount of residual methane leaving as fugitive emission at the surface of the bio-cover material layer. Thus, more than 98% of methane mass entering the bio-cover unit was considered removed mainly through bio-oxidation. It should also be noted that the bio-cover system in this study was operated at a low loading rate to simulate the natural condition of tropical landfill emission. Therefore, the landfill gas was highly diluted as it entered the bio-cover material bed, noticeable from the significant reduction in methane and carbon dioxide concentrations especially at 0.4–0.6 m depth of the materials (Figure 2).

Figure 3 shows the relationship between MLRs and MRRs observed in bio-cover materials. It was clearly observed that higher MRR was achieved under higher MLR conditions. Nevertheless, MRRs were found to be more varied under high MLR conditions. Among them, the MRRs observed in C material were found to fluctuate less than the others at the highest MLR range. Meanwhile, SL material performance varied only under high MLR conditions (>15 mol/m^3/d) whereas MRRs in SW material fluctuated even at a MLR of 8–15 mol/m^3/d. These differences could be due to the heterogeneous nature of bio-cover materials which can possibly be higher for stabilized waste than sandy loam and compost materials. Fewer variations were mostly observed in all materials operated under lower MLRs (<8 mol/m^3/d).

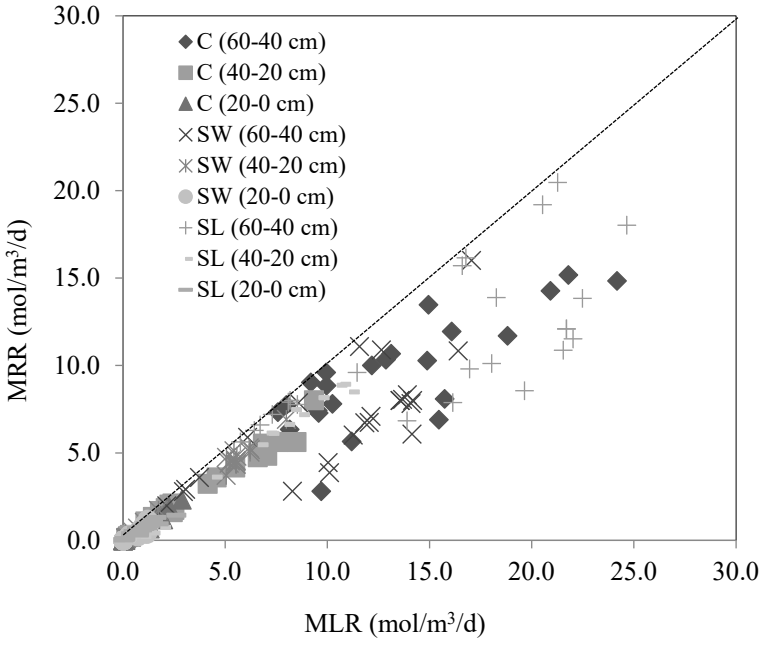

Figure 3. Relationship between methane loading and removal rates in bio-cover materials.

3.3. Methane Oxidation Capacities of Bio-Cover Materials

The presence of biological methane oxidation in bio-cover materials can also be confirmed through the determination of their methanotrophic activity in batch experiments. The moisture contents in SL, C, and SW materials examined in batch experiments were 23%, 58.5%, and 32%, respectively, which were in situ moisture contents of the bio-cover materials during the sampling time.

Table 4 presents the methanotrophic activities observed in the bio-cover materials. It appears that C material had the highest *MOR* among the types of materials examined. The observed *MOR* of C material was found highest (2.03–2.49 µmol/g.dry solids/h) and they were relatively consistent along the depth. For SL material, the most active zone for *MOR* was found at the top part (0–0.20 m) at 1.3 µmol CH_4/g dry soil/h, while SW material had the lowest *MOR* at 0.12–0.2 µmol CH_4/g dry solids/h, being highest at the bottom of the material layer. The MOR, to some extent, is related to the observed MRRs

in the bio-cover. Higher MORs were detected in SL and C materials than those found in SW material. Nevertheless, their *MOR* was found to be highest at the top of the material bed where oxygen is highly available. On the other hand, the MRRs in those materials were found to be highest at the bottom of the material bed where higher methane contents in gas were available. As the *MOR* were examined under high methane and oxygen condition to determine the maximum methane removal potential of the bio-cover material, the discrepancies between MORs obtained from batch experiments and MRRs observed in the field operation of bio-cover would be due to their differences in methane and oxygen conditions. The *MOR* data could also help to confirm the presence of microbial activities for methane removals in bio-cover materials. Based on the MOR/CPR ratio observed during batch experiments, higher values were found in C material (0.81–0.87) which suggested that methane oxidation is mainly responsible for methane removal as they are close to the stoichiometric ratio (1.0) required for the reaction. Meanwhile, the lower ratios observed in the other materials suggest the production of carbon dioxide from other reactions, e.g., oxidation of organic substances. According to previous research, methane in landfill gas could be completely consumed within 20 h in compost and 100 h in sandy loam cover material where methane oxidation was active [14]. For SW material, lower bulk density, as well as high porosity of media, could result in lower MORs but higher oxygen availability would be expected in the deeper zone of the material bed [21].

Table 4. Methanotrophic activity in the bio-cover materials.

Media	Depth (m)	MOR (μmol/g Dry Solids/h)	CPR (μmol/g Dry Solids/h)	MOR:CPR
SL	0–0.20	1.31	0.74	1.77
	0.20–0.40	0.50	1.03	0.48
	0.40–0.60	0.60	1.32	0.45
C	0–0.20	2.49	2.1	1.19
	0.20–0.40	2.08	2.4	0.87
	0.40–0.60	2.03	2.5	0.81
SW	0–0.20	0.13	0.44	0.3
	0.20–0.40	0.12	0.17	0.69
	0.40–0.60	0.20	0.27	0.74

Remark: MOR: Methane oxidation rate; CPR: Carbon dioxide production rate.

3.4. Methanotroph Population and Microbial Consortium

The methanotroph population in the bio-cover material was quantified as shown in Table 5. Generally, methane, oxygen, and nutrient contents in the cover material are the regulator of the methanotrophic population. Previous research [30] reported that the growth of methanotroph type I was enhanced under high oxygen conditions in the landfill cover. In this study, both methanotroph type I and type II were observed abundant under both wet and dry conditions. During the wet period, a greater population of methanotroph type II than type I was found at all depths in the C and SL materials whereas they were quite similar in the SW material. During the dry period, the methanotroph type I population increased in the C and SL materials while type II decreased. The populations of both methanotroph types increased in SW material when the bio-cover was operated continuously from the wet period to the dry period. These results suggest that the methanotroph population in the bio-cover varied seasonally and the population also varied between the materials. The predominant groups of methanotrophs were also influenced by the environmental condition. In C material, methanotrophs of type II grew well during the wet period, especially in the upper part while they were found to be more prevalent at the middle and bottom parts of the material bed during the dry season. For SL material, the predominant methanotroph population shifted from type II during the wet period to type I during the dry period. Its methanotroph population was quite consistent along the bed depth except

for type II during the dry period. For SW material, the methanotroph population was found to be less than in other materials while it was greater at the deeper zone for all conditions.

Table 5. Methanotroph population in the bio-cover materials.

Period	Media Depth (m)	Number of Methanotrophic Cells (Cell × 10^6/g Dry Solids)					
		Type I			Type II		
		SL	C	SW	SL	C	SW
Wet	0–0.20	10.2	30.3	8.4	27.2	35.5	9.3
	0.20–0.40	10.1	9.33	10.4	28.5	32.8	9.6
	0.40–0.60	11.1	11.1	10.7	30.1	26.7	10.9
Dry	0–0.20	36.1	24.2	13.3	16.9	19.9	9.7
	0.20–0.40	35.5	28.7	13.8	17.2	24.9	14.7
	0.40–0.60	34.6	24.9	19.6	25.1	23.6	16.9

The PCR-DGGE profiles of microbial communities (shown in Figure S4) indicate the presence of five species of methanotroph type I and one methanotroph type II in the biofilter media. They are *Methylococcus*, *Methylomicrobium*, *Methylobacter*, *Methylomonas*, and *Methylocaldum* for type I and *Methylocystic* for type II. Other non-methanotroph species were also detected in SW media which may relate to plastic wastes containing the media, e.g., *Alcanivorax dieselolei* (an alkane degrading bacterium) and *Nitratifracter salsuginis* (a denitrifying bacterium). In previous research, it was also reported that the growth of *Methylobacter* and *Methylococcus* had a positive correlation with the oxygen available while that of *Methylocystic* was positively correlated with methane [30]. A high abundance of methanotroph type I over type II suggests a high oxygen environment available for methane oxidation in the media examined in this study which is well supported by the gas profile observed along the biofilter media bed (Figure 2).

4. Conclusions

Compost was found to be the most effective bio-cover material for mitigating methane emission from the tropical landfill examined in this study. Comparable field performance was also observed for sandy loam material. Stabilized waste could also be applied to mitigate methane at lower loading rates. Methane removals of 78.0–94.8% took place at the bottom part (0.20 m from inlet) of the material layer whereas the total removals of 97 to >99% were achieved in the whole material layer of 0.60 m depth. Higher methanotrophic activities and methanotrophic microorganisms were observed for compost and sandy loam materials compared to stabilized wastes. The majority of methane was possibly removed through biological oxidation in the bio-cover materials as fugitive emissions at the surface of the bio-cover material layer were low (<2% of methane loading).

Supplementary Materials: The following supporting information can be downloaded at: https://www.mdpi.com/article/10.3390/app13031990/s1, Figure S1. Variation of rainfall and ambient temperature; Figure S2. Variation of methane loadings (MLRs) and removals (MRRs) during bio-cover operation; Figure S3. Variation of methane emissions (MERs) during bio-cover operation; Figure S4. PCR-DGGE profiles of methanotrophic population observed in different materials (left) C (middle) SW (right) SL media; Table S1. Probes used in microbial population identification; Table S2. Moisture content (%) in bio-cover material; Table S3. Bio-cover material characteristics.

Author Contributions: Conceptualization, C.C. and W.C.; methodology, C.C. and W.C.; validation, R.C., W.C. and C.C.; investigation, R.C.; data curation, R.C.; writing—original draft preparation, R.C.; writing—review and editing; C.C.; funding acquisition, C.C. All authors have read and agreed to the published version of the manuscript.

Funding: This research was financially supported by the National Research Council of Thailand through the Kasetsart University Research and Development Institute (Research grant No. 79.60). Partial support through collaborative research from ASEAN University Network/Southeast Asia Engineering Education Development Network (AUN/SEED-Net) is also acknowledged.

Institutional Review Board Statement: Not applicable.

Informed Consent Statement: Not applicable.

Data Availability Statement: The data presented in this study are available in the Supplementary Materials. The other additional information is available upon request from the corresponding author.

Conflicts of Interest: The authors declare no conflict of interest.

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