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ORIGINAL ARTICLE

Enhancement of landfill methane oxidation using different types of organic wastes

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Abstract Methane (CH_4) is a greenhouse gas with a global warming potential that is 25 times higher than carbon dioxide (CO_2). Microbial oxidation of landfill CH_4 is about 5 % and it plays a significant role in reducing emissions to the atmosphere. Previous studies have shown that microbial oxidation of CH_4 in landfill cover soil can be enhanced using organic matter-rich substrates. This study aims to investigate the CH_4 oxidation activity with the addition of different organic wastes in different combinations. Batch experiments with different organic wastes for CH_4 oxidation activity showed that compost and sawdust required the shortest time to oxidize CH_4 compared to empty fruit bunch and black soil. However, other organic wastes (spent yeast, sewage sludge and spent tea leaves) tested in this study produced CH_4 instead. Compost and sawdust took 4 days for complete CH_4 oxidation. Batch experiments with different combinations of organic waste showed potential CH_4 reduction when combined with compost. The combination of 20 % spent yeast + 80 % compost was able to completely oxidize CH_4 at day 16. CH_4 was fully oxidized on day 1 by 20 % sewage sludge + 80 % compost, showing good CH_4 oxidation potential; 100 % sawdust fully oxidized CH_4 within 4 days, while a combination of 20 % sawdust + 80 % compost and 40 % sawdust + 60 % compost took 2 days for complete CH_4 oxidation. From this study, it can be concluded that the addition of organic wastes at optimum ratio and combination with compost as landfill cover material will have a significant effect on CH_4 emission reduction.

Keywords CH_4 oxidation · Landfill cover · Greenhouse gas reduction · Microbial oxidation

Introduction

The generation of municipal solid waste (MSW) is increasing due to population growth, urbanization, economic level and community living standard (Zamali et al. 2009). In a developed country such as the USA, the daily per capita MSW generation has been increasing from 1.2 kg per capita per day in the year 1960 to 2.0 kg per capita per day in the year 2010. In other words, the total MSW generated in the USA had increased from 88.1 million tons to 249.9 million tons throughout the duration of 50 years (USEPA 2012). Meanwhile, in a developing country such as Malaysia, MSW was generated at 1.2 kg per capita per day in the year 2007 and increased to 1.5 kg per capita per day in year 2010 (Fauziah and Agamuthu 2012). This increasing trend in the daily generation of MSW shows an alarming need for proper waste management to minimize the impacts to the environment and all inhabitants on the earth. Final disposal is an important part of waste management hierarchy, because there are no technologies available to avoid the entire unwanted residue from the waste sector and no endowment is available for zero waste (Fauziah and Agamuthu 2012). Landfill is the ultimate disposal method chosen by most developed and developing countries to manage the MSW generated. Landfill gas (LFG) is generated when the organic materials from MSW in the landfill undergo the decomposition process. There are four main phases involved in the

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production of LFG after waste is deposited into the landfill. Phase I is aerobic decomposition; Phase II is acid genesis; Phase III is acetogenesis; and finally Phase IV is methanogenesis. The LFG consists of CH_4 (50–60 % by volume), CO_2 (35 %), volatile organic compounds (VOC) which is less than 1 % and small amounts of nitrogen, oxygen, hydrogen and trace amounts of inorganic compounds (Kettunen 2003). The constant production of LFG increases the pressure within the landfill and causes the gas to be released into the atmosphere. Improper gas collection systems in landfills also result in passive release of LFG to the atmosphere causing huge impacts to the environment. Landfills are also identified as a significant source of atmospheric greenhouse gas (GHG) which contributes to global warming (Bogner et al. 2007). Both CH_4 and CO_2 are the landfill gases recognized as primary contributors to global warming. CH_4 is a particularly potent GHG and is currently considered to have a global warming potential (GWP) that is 25 times that of CO_2 in a time horizon of 100 years (IPCC 2007). CH_4 is found to have a strong absorption band in the infrared region of the solar spectrum (Pawloska 2008). Therefore, CH_4 poses a more serious threat to the environment compared to other gases as it contributes largely to global warming. The annual global CH_4 emission from landfills is estimated to be in the range of 500–800 MT CO_2-Eq and represents the highest source of GHG within the waste sector (Bogner et al. 2007). The reduction of CH_4 from landfill is necessary to reduce and mitigate GHG emissions. CH_4, a potent GHG, can be converted to CO_2 which is a less harmful GHG through the oxidation process. CH_4 oxidation is the process of converting CH_4 into CO_2, water and biomass through microbial activity and plays an important role in the mitigation of atmospheric CH_4 emission from landfills (Kightley et al. 1995). The CH_4 oxidation process depends on several factors such as moisture content, temperature, CH_4 concentration, O_2 availability and nutrient concentrations (Kettunen 2003). Emission of CH_4 can be oxidized by the addition of small amounts of organic wastes in biologically active biocovers (Hilger and Humer 2003; Barlaz et al. 2004). The addition of organic wastes to the compost not only reduces the amount of compost needed, but also improves the microbial community (Isabel et al. 2010) within the biocover. The development of biocover materials will also reduce the carbon foot print of landfills and promote the recycling and re-use of waste materials (Reddy and Sadasivam 2013). Passively aerated low-maintenance biocovers are potentially good for CH_4 mitigation. The aim of this study is to test different cover materials for potential CH_4 oxidation activity with the addition of different types of organic wastes in different combinations for CH_4 oxidation enhancement.

Materials and methods

Compost

Compost was obtained by composting a mixture of 75 % grass clippings and 25 % cow manure. The grass clippings and cow manure were uniformly mixed to ensure an even distribution of microbes for optimum composting. The heap method was employed and the composting was carried out under a cover. The heap was 1 m high and 2 m wide at the base. Water was added to the compost mixture regularly to ensure an optimum moisture level of 50–60 %. Manual turning of compost mixture once daily for the first 8 days and once every 2 days thereafter was necessary to maintain the aerobic condition. The daily temperature of the composting mixture was measured using thermometer.

Organic waste

In this study, different types of organic wastes were collected. Sewage sludge (SS) was obtained from Indah Water Konsortium (IWK), Pantai Dalam, Kuala Lumpur, Malaysia. Fine sawdust (SD) was obtained from Boon Sawmill, Klang Valley, Malaysia. Spent yeast (SY) was collected from Carlsberg Brewery, located at Shah Alam, Malaysia. Black soil was obtained from a nursery in Sungai Buloh, Kuala Lumpur, Malaysia. Shredded empty fruit bunch (EFB) was obtained from Seri Ulu Langat Palm Oil Mill Sdn Bhd, while the spent tea leaves (STL) were obtained from the Kompleks Perdana Siswa (KPS), University of Malaya, Malaysia cafeteria.

Physiochemical properties

To determine the gravimetric moisture content, the biocover materials were oven dried for 24 h at 100 °C and expressed as the mass ratio of water. Organic contents were determined by loss on ignition. Electronic pH meter was used to measure the pH of the biocover materials. Further chemical analysis on the biocover materials was carried out in duplicates according to the ASTM standard procedure.

Batch experiment

To investigate the performance of the biocover in oxidizing CH_4, the Wheaton bottle experiments were carried out. Wheaton bottles with a total volume of 125 mL were used to conduct the batch experiments in laboratory conditions. Each bottle was filled with 20 g of biocover materials (different ratios or different combinations) and sealed with rubber septum and aluminum seal to ensure gas tight condition. Next, 15 mL of air from the headspace was withdrawn using a syringe and replaced with 10 mL of O_2

(99.8 % purity) and 5 mL of CH_4. These amounts provided a mixing ratio of approximately 4 % of CH_4 (v/v) and 8 % of O_2 (v/v) of the total headspace. The addition of O_2 into the Wheaton bottles was to ensure that aerobic conditions prevailed during the experiments. For every variable and parameter studied, the experimental runs were conducted in duplicate. Gas chromatography, SHIMADZU GC-8A, was used to measure the concentrations of CH_4, O_2 and CO_2 in the headspace daily.

Biocover performance index

The performance of the biocover was expressed in terms of biocover performance index (BPI). A high value of BPI indicates a high CH_4 oxidation rate in biocover performance as the BPI is inversely proportional to the period of CH_4 oxidation. Equation 1 was used to derive the BPI:

$$\mathrm{BPI} = \frac{(\mathrm{CH_4})_0 - (\mathrm{CH_4})_n}{W \times N} \qquad (1)$$

where

$(\mathrm{CH_4})_0$ = initial concentration of CH_4 (μg),
W = amount of biocover (g),

$(\mathrm{CH_4})_n$ = concentration of CH_4 at time n (μg),
N = time taken (hours).

Results and discussion

Table 1 shows the characteristics of organic materials used in this study. Sewage sludge has the highest moisture content (82.85 %) among the different organic materials used, while EFB has the lowest moisture content (37.6 %). This is important for biocover performance, as water retention ability is necessary to sustain the microbial population for CH_4 oxidation. CH_4 oxidation activity will become limited due to physiological stress to methanotrophs if moisture content is low (Pawloska 2008).

Based on a previous study by Jayanthi and Agamuthu (2011), the optimum CH_4 oxidation activity was observed when the moisture content of the biocover was 60 %. However, excess water content will affect the penetration of O_2 into the soils, which is the main reactor for CH_4 oxidation. This also affects the porosity of the soil and influences gas transport through the soil. According to Moldes et al. (2007), the pH of the compost should be neutral to slightly acidic to optimize CH_4 oxidation activity. All the organic materials used in this study fall in this pH range and show suitability for CH_4 oxidation. Spent yeast has the highest content of organic matter (78 %) and, according to Chanton and Liptay (2000), CH_4 oxidation activity increases in organic matter-rich soils. This is because organic matter serves as a carrier for microorganisms and improves the soil properties and substrate (Christensen et al. 1996). Lizik et al. (2013) have also concluded that the addition of nutrients to soil was useful in the reduction of CH_4 emissions by 50–60 %.

The C:N ratio of the organic materials used in this study ranges from 16.3 to 33.4. Different types of biocover materials used by Mor et al. (2006) have C:N ratio in the range of 11.2–26.9. Boeckx et al. (1996) stated that, at low value of C:N ratio, the CH_4 oxidation activity will be affected. Sewage sludge has a high copper content (41.7 ppm) compared to other organic materials tested, while compost has a 0.627 ppm of copper content. According to Kjeldsen (1996), CH_4 oxidation will be higher when the copper content is low, because methanotrophs only express CH_4 oxidizing capability at low copper concentration. (see Tables 2, 3).

Batch experiment carried out with different types of organic waste

Figure 1 shows the CH_4 oxidation activity of different types of organic wastes. Compost and sawdust took the shortest period to oxidize CH_4. Complete CH_4 oxidation activity took place on day 4. According to Humer and Lechner (1999); Wilshusen et al. (2004), optimal CH_4 oxidation activity was obtained from diverse, mature and well-structured compost materials compared to other cover

Table 1 Characteristics of different types of organic materials

Test parameter	Test method	Compost	Black soil	Spent yeast	EFB	Sawdust	Sewage sludge	Spent tea leaves
Moisture content	ASTM 2004	62.17 %	43 %	74.7 %	37.6 %	38.74 %	82.85 %	72.1 %
pH	ASTM 2004	6.33	6.02	5.29	5.24	–	–	5.42
Organic matter	ASTM 830-97	52 %	40 %	78 %	50.5 %	20.80 %	60.70 %	76.0 %
Total carbon	ASTM 777-87(96)	20.30 %	16.20 %	46.8 %	38.9 %	16.30 %	20.30 %	46.87 %
Total nitrogen	ASTM E778-87	1.20 %	1.10 %	1.59 %	1.86 %	1.0 %	1.20 %	1.4 %
C:N ratio	USEPA 3050B	17	14.70	29.4	20.9	16.3	16.9	33.4
Copper	USEPA 3050B	0.627 ppm	2.71 ppm	12.40 ppm	0.01	ND	41.70 ppm	–

ND Not detected

Table 2 Biocover performance index for different types of organic wastes

Types of organic waste	Biocover performance index ($\times 10^3 \mu g\ g^{-1}\ h^{-1}$)
Compost	2.08
Black soil	1.19
Empty fruit bunch	1.754
Sawdust	2.08

Table 3 Biocover performance index for the best combination of organic waste and compost

Types of organic waste	Biocover performance index ($\times 10^3 \mu g\ g^{-1}\ h^{-1}$)
20 % Sewage sludge + 80 % compost	8.33
20 % Spent yeast + 80 % compost	0.52
All ratios of EFB	1.754
20 % Sawdust + 80 % compost	4.16
40 % Sawdust + 60 % compost	4.16

materials. Also, according to Owolabi et al. (2003), an experiment carried out with soil in southwest of Nigeria showed that sawdust enhanced the soil porosity and nutrient status of the soil. This may also be one of the reasons for the higher CH_4 oxidation activity when sawdust was used as the biocover material. Different types of organic wastes will influence the nutrient property of the cover material differently. However, the types of organic wastes used in this study were by no means exhaustive as the organic wastes were selected for their availability and abundance in Malaysia. Meanwhile, EFB took 6 days for complete oxidation and black soil took 7 days. The slower CH_4 oxidation activity of EFB may be due to the low water content compared to the other organic materials as, according to Pawloska (2008), lack of water content in biocover material causes physiological stress to the methanotrophs. Low moisture content and organic matter in black soil also may be the cause of low CH_4 oxidation activity. Even though EFB and black soil showed a lower CH_4 oxidation activity than compost and sawdust, complete CH_4 oxidation still managed to take place. The high porosity of the shredded EFB increases the O_2 concentration in the material which promotes CH_4 oxidation. However, other organic materials (spent yeast, sewage sludge and spent tea leaves) tested in this study produced CH_4 instead of oxidizing CH_4. The high water content of more than 70 % in spent yeast, sewage sludge and spent tea leaves caused these materials to generate CH_4, because it encouraged the internal condition to become anaerobic. According to Barlaz et al. (2004), compost or other organic material can also produce CH_4 if the moisture content is

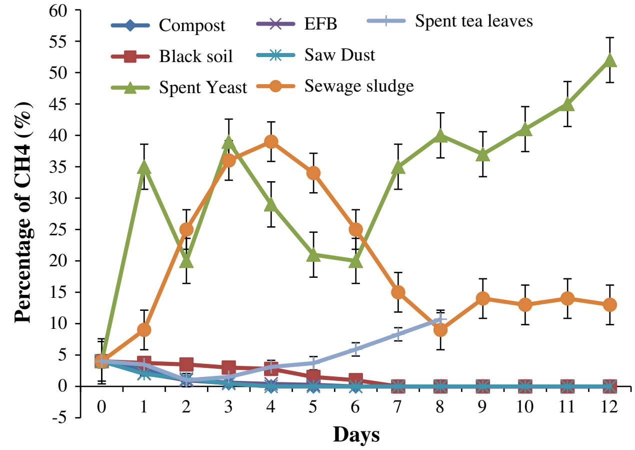

Fig. 1 Reduction of CH_4 by a single use of organic waste

too high. The process of gaseous movement will also slow down if the moisture content of the biocover material is too high, as molecular diffusion in water is 10^4 times slower than in air (Cabral et al. 2010). The differences within the CH_4 reduction performance observed in different organic wastes are also due to the uncontrolled nature of the materials, as the rate of CH_4 oxidation is influenced by many factors. Among them are pH, moisture, temperature and nutrient levels (Chanton et al. 2011).

Biocover performance index

The biocover performance index (BPI) was used to calculate the CH_4 oxidation rate for the batch experiments with different types of organic waste that achieved 100 % CH_4 oxidation activity. Compost and sawdust exhibited a high BPI of $2.08 \times 10^3\ \mu g\ g^{-1}\ h^{-1}$ compared to EFB and black soil which scored $1.754 \times 10^3\ \mu g\ g^{-1}\ h^{-1}$ and $1.19 \times 10^3\ \mu g\ g^{-1}\ h^{-1}$, respectively. Previous research also obtained BPI in the range of $1.19 \times 10^3 \mu g\ g^{-1}\ h^{-1}$–$8.33 \times 10^3 \mu g\ g^{-1}\ h^{-1}$ when compost was inoculated with CH_4-oxidizing bacteria (Jayanthi and Agamuthu 2011) (see Table 3).

Batch experiment with different types of organic waste at different ratios

Based on the index above, it was observed that compost showed higher CH_4 oxidation activity compared to other types of organic waste. Figure 2 shows the CH_4 oxidation activity of sewage sludge and compost at different ratios. The combination of 20 % sewage sludge + 80 % compost showed the highest CH_4 oxidation capacity, which took only 1 day for complete CH_4 oxidation. This was followed by 40 % sewage sludge + 60 % compost which took 6 days for complete CH_4 oxidation. However, for 60 %

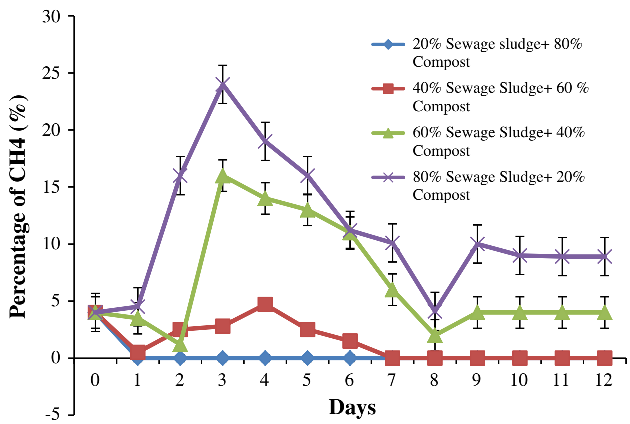

Fig. 2 Reduction of CH_4 at different ratios of sewage sludge and compost

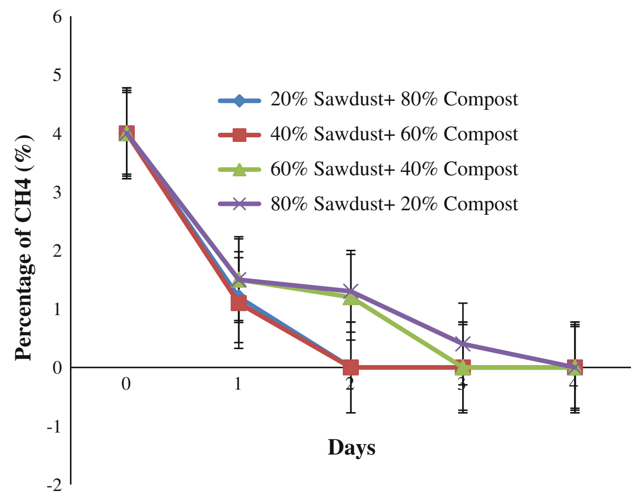

Fig. 3 Reduction of CH_4 at different ratios of sawdust and compost

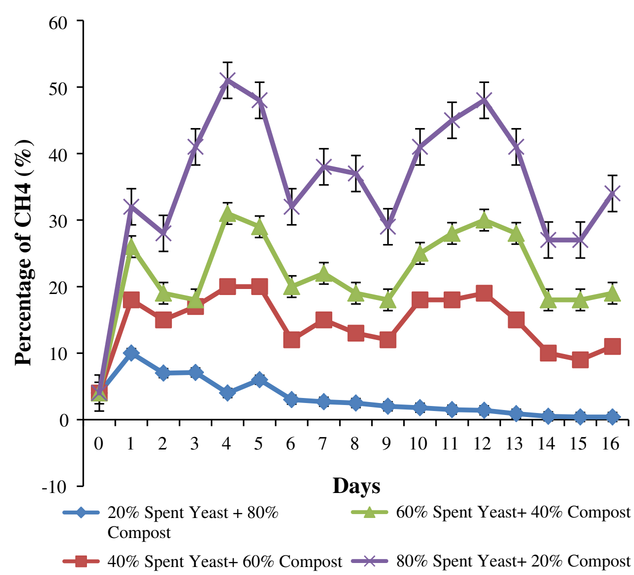

Fig. 4 Reduction of CH_4 at different ratios of spent yeast and compost

sewage sludge + 40 % compost and 80 % sewage sludge + 20 % compost, complete CH_4 oxidation did not manage to take place even after 12 days of experiment. Not only did they not manage to achieve complete CH_4 oxidation, but CH_4 was produced instead. The production of CH_4 may be due to the decrease in O_2 level and also the high content of water in sewage sludge which was more than 70 %. Very low O_2 content led to the anaerobic condition which resulted in fermentation and production of CH_4. Optimum water content is also essential for the activity of methanotrophs to carry out CH_4 oxidation. The microorganisms tend to be inactive when minimum and maximum humidities are not achieved (Bender 1992) and could be one of the reasons for lower oxidation activity when a higher composition of sewage sludge was used in this study.

The experiment on different ratios of sawdust (SD) and compost (Fig. 3) showed higher CH_4 oxidation activity with 20 % SD + 80 % compost and 40 % SD + 60 % compost. Both took 2 days for complete CH_4 oxidation, whereas 60 % SD + 40 % compost and 80 % SD + 20 % compost took 4 days for complete CH_4 oxidation. All the mixtures of sawdust and compost were able to oxidize CH_4 completely. The low moisture content of sawdust (38.74 %) could be one of the reasons for the low oxidation rate in 60 % SD + 40 % compost and 80 % SD + 20 % compost. According to Barlaz et al. (2004), low moisture content in a biocover system will inhibit microbial activity. On the other hand, the addition of spent yeast showed complete CH_4 oxidation when 20 % spent yeast + 80 % compost was used, while other ratios did not achieve complete CH_4 oxidation even until day 16 (Fig. 4). The addition of spent yeast recorded fluctuating amount of CH_4 throughout the incubation period of 16 days. Complete CH_4 oxidation activity did not take place when more than 20 % of spent yeast was used in this study. In fact, the concentration of CH_4 fluctuated alternatively every 2–3 days. Spent yeast contains high amount of anaerobic bacteria and this could be one of the reasons for the lower CH_4 oxidation activity. Moreover, the spent yeast used for the experiment was recovered by a brewing process after the fermentation process took place (Fillaudeau et al. 2006) and this could be the reason for the production of CH_4 in this study.

Figure 5 shows the optimum ratios of EFB and compost combinations for complete CH_4 oxidation. All combinations took 6 days to oxidize CH_4. However, the percentage of CH_4 reduction from day 1 to day 6 varied between different ratios of EFB and compost. The combination of

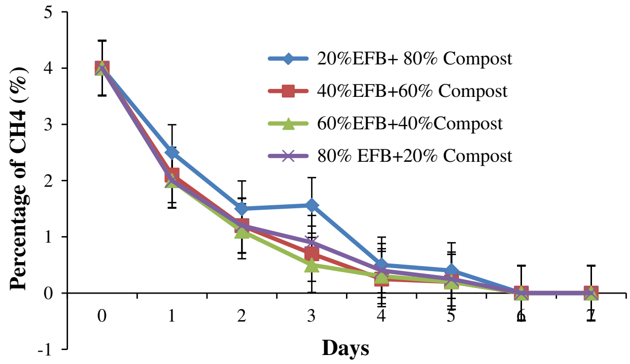

Fig. 5 Reduction of CH_4 at different ratios of EFB and compost

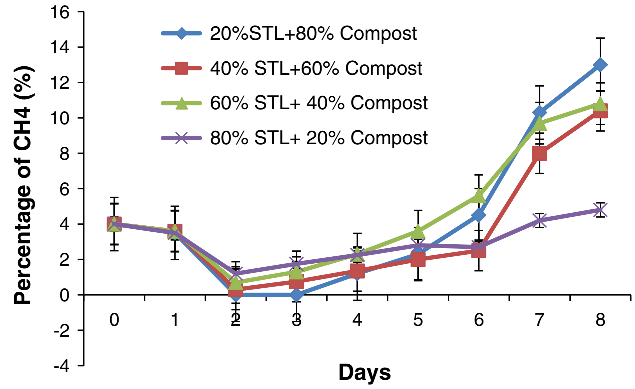

Fig. 6 Reduction of CH_4 at different ratios of STL and compost

60 % EFB + 40 % compost and 80 % EFB + 20 % compost was able to oxidize approximately 50 % of the CH_4 introduced within 1 day, but after day 1 the observed CH_4 oxidation rate became slower. This showed that both combinations required a short acclimatization period (1 day) and this might be due to the high content of organic matter in both the EFB and compost, hence enhancing the growth of methanotrophic microorganisms (Christensen et al. 1996) in the early stage of the CH_4 oxidation process. Organic components in the compost or biocover materials play an important role in the microbial activity and determination of the CH_4 oxidation process (Fauziah 2009; Insam and Wett 2008; He et al. 2008; Ritzkowski et al. 2006; Prant et al. 2006). Besides that, the porosity of EFB also increases the penetration of O_2 within the biocover material which leads to high activity of CH_4 oxidation (Kightley et al. 1995).

Results from experiments with different ratio of spent tea leaves (STL) and compost are shown in Fig. 6. The mixture of STL and compost at all ratios showed fluctuating percentage of CH_4 in the headspace gas throughout the incubation period and complete CH_4 oxidation did not take place. From the graph, it was observed that the combination of 20 % STL + 80 % compost was able to fully oxidize the CH_4 present in the bottle within 1 day. However, after day 3, an increase in the CH_4 concentration was observed. Mixtures of higher than 40 % STL + 60 % compost did not manage to fully oxidize CH_4 throughout the experimental period. The presence of high amount of anaerobic bacteria in the STL and the insufficient supply of O_2 for aerobic condition at the initial phase of the batch experiments could be the reason for the fluctuation of CH_4 concentration in the experiment. The primary reason for the sharp increase in the CH_4 concentration is that the STL were not composted. Therefore, after day 1, the microorganisms in the bottle started decomposing the spent tea leaves, leading to anaerobic condition that caused the mixture to ferment and start producing CH_4. According to Kettunen (2003), an organic matter must be biochemically stable to avoid respiration by other microorganisms that can disturb the O_2 consumption of CH_4 oxidizers. Secondly, it could have also been influenced by the acidic condition of the soil. Since STL used was acidic, it triggered fermentation when O_2 was depleted. Furthermore, STL has been known to be used in the production of biogas. Moreover, the moisture content of STL is at 72.1 %, which is much higher than the suggested optimal moisture by Dan Wang et al. (2004) and Boeckx et al. (1996), ranging from 40 to 50 %. Higher moisture content will retard the gaseous transportation in the soil. In a highly saturated mixture, the air-filled voids in the soil are no longer interconnected and gases have to diffuse in liquid state, which cause reduced availability of CH_4 and O_2, thus limiting CH_4 oxidation.

Batch experiments with different combinations of organic waste showed potential CH_4 reduction when organic waste was combined with compost. Pure spent yeast was not able to oxidize CH_4 until day 16 and CH_4 concentration fluctuated every 2–3 days. However, 20 % of spent yeast + 80 % compost was able to completely oxidize CH_4 on day 16. Similarly, 100 % sewage sludge was also not able to oxidize CH_4 and the CH_4 concentration fluctuated. However, 20 % sewage sludge + 80 % compost was able to oxidize CH_4 within 1 day. This combination exhibited potent CH_4 oxidation activity in a very short period of time. While 100 % sawdust was able to fully oxidize CH_4 within 4 days, the combination of 20 % sawdust + 80 % compost and 40 % sawdust + 60 % compost took only 2 days for complete CH_4 oxidation. However, no difference was observed with different ratios of EFB and 100 % EFB, as all combinations took 6 days for complete CH4 oxidation. Spent tea leaves (STL) were not able to oxidize CH_4 at both different ratios and 100 % of STL, which due to their acidic properties started producing CH_4 when the atmospheric condition became anaerobic.

Biocover performance index

Biocover performance index was calculated to evaluate the efficiency of biocover materials at different ratios of organic waste and compost. Higher BPI value indicates better CH4-oxidizing performance of biocover materials and vice versa. From this experiment, we can observe that 20 % sewage sludge + 80 % compost (8.33×10^3 μg g^{-1} h^{-1}) records the highest BPI compared to other combinations. Meanwhile, 20 % sawdust + 80 % compost and 40 % sawdust + 60 % compost recorded BPI that were 50 % lower compared to 20 % sewage sludge + 80 % compost. The lowest BPI value was obtained for 20 % spent yeast + 80 % compost (0.52×10^3 μg g^{-1} h^{-1}) (See Table 3).

Conclusion

Batch experiment with different types of organic wastes shows that compost and sawdust took the shortest period of time to oxidize CH_4 compared to other organic wastes. Spent yeast, sewage sludge and spent tea leaves tested in this study produced CH_4 instead of oxidizing CH_4. Batch experiment with different combinations of organic wastes showed potential CH_4 reduction when the organic wastes were combined with compost. From this study it can be concluded that the addition of organic wastes at the best ratio with compost as landfill cover material will provide significant effect on CH_4 emission reduction.

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