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Effects of organic matter addition to landfill soil cover to enhance methane oxidation

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Abstract: Methane emission from landfills has become a major concern due to its derogatory effects to both environment and human health. This study was carried out to determine the optimum organic matter addition in the landfill cover soils to promote the methane oxidation process. The experiment was performed with methane gas introduced to the biofilter media in the soil columns and methane concentration at different profiles was analyzed daily. The results shows the unamended soil was able to degrade the methane concentration up to 87.33% but in the meantime, the total methane oxidation efficiency slightly increased to 94.09, 95.40 and 97.52% in the columns containing the mixture of soil to coconut husk at ratios of 30:70, 50:50 and 70:30 (v/v) respectively. These results suggest that addition of organic matter to landfill soil cover has the potential to induce the growth of methanotrophs populations and hence accelerate the methane oxidation process.

Key words: Biocover; CH4 mitigation; Landfill gas; Landfill soil cover; Methane oxidation

1. Introduction

In the sanitary landfills, landfill gas (LFG) emitted is usually collected as a source of energy to generate electricity or burnt by flaring. However, landfills or dumpsites that are not equipped with the facilities for the above purposes only depend on the soil cover to reduce the landfill gas emission. Landfill gas seeps through the fissures of the landfill soil cover and finally escapes to the atmosphere. The accumulation of these fugitive landfill gases in the environment has known to be the contributor to depletion of ozone layer and a threat to the public health. LFG emission from the landfill stems from the anaerobic decomposition of organic waste by methanogenic bacteria and the waste sector generates 18% of anthropogenic methane emission at global scale (Yucheng & Cel, 2015). The constituents of LFG, on average, are primarily 55-60% volume of carbon dioxide (CO_2) and 40-45% volume of methane (CH_4) (Raco et al., 2010). These gases pile up in the biosphere as Green House Gases (GHG) and it was reported that methane is more potent than carbon dioxide at trapping heat in the atmosphere, comparatively 25 times higher in 100-year extrapolation. Besides, methane ability to reside persistently in the atmosphere for 9 to 15 years makes it the second largest contributor of global warming behind carbon dioxide (Barker, 2007).

Owing to these facts, researchers have come up with numerous studies on the abatement of methane emission from the landfills and one of the approached methods was using engineered soil oxidise methane. The addition of green waste that rich in organic matter content has been proven to extend the soil cover performance at removing methane by supplying nutrients to the soil-borne bacteria called methanotrophs (Albanna & Fernandes, 2009; Siva Shangari & Agamuthu, 2012). Additionally, the incorporation of organic matter into the soil cover was recorded to increase porosity of the biofilter media to allow gas diffusivity (Cao & Staszewska, 2013; Hettiaratchi et al., 1998). Hence, the experiment was carried out to determine the optimum volume of organic addition to the landfill soil cover to maximize methane oxidation.

2. Method

2.1. Biofilter media preparation

The main biofilter media used in this experiment was obtained from Jeram Sanitary Landfill located in Jeram, Kuala Selangor, Malaysia. The 647947 m^2 landfill has the capacity of 2,500 tonne of wastes per day and is operated by Worldwide Landfills Sdn. Bhd. Soil sample was collected at 30 cm from the surface after removing the unwanted debris (Mei, Wang, Han, & Zhao, 2011; Moon, Lee, Lee, Ryu, & Cho, 2010). In the lab, the moisture content of the soil sample was adjusted to 12% (w/w) which was found to be the optimum moisture content for methane oxidation to take place in the previous study (Faeiza et al., 2014). Meanwhile, the coconut husk was air-dried and ground. Both soil and ground coconut husk were sifted through 2 mm aperture size of sieve to provide large surface area of the biofilter media.

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2.2. Column reactor experiment

Cylindrical soil columns were fabricated from polyvinyl chloride (PVC) pipes with the dimensions of height, inner diameter and thickness at 107, 15 and 0.5 cm respectively (Kightley et al., 1995). In order to determine the concentration of methane by profiles, sampling ports secured with cap-sleeved Suba-Seal rubber septa were embedded at 10 cm intervals along the soil columns. The gas chromatography analyses were conducted for 14 consecutive days on methane samples retrieved from these ports. Besides, 17 cm gas distribution layers made up of gravels were fitted below the packed media which were supported by perforated plastic net with 0.5 mm aperture size. The gravels selected were 3 to 5 cm in diameter.

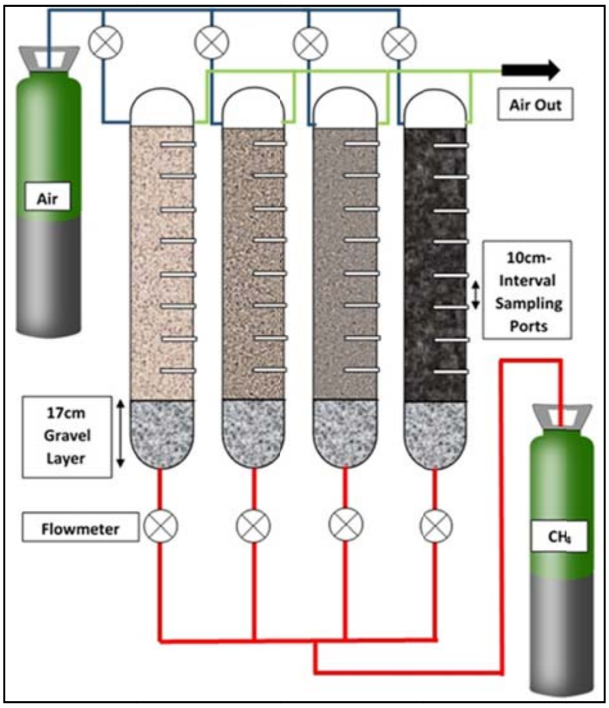

Fig. 1: Setup for soil columns with biofilter media of different ratios of soil to coconut husk (v/v).

The biofilter media was filled into the column up to 5 cm above the highest ports by random packing. The columns were agitated to allow the biofilter media to settle evenly throughout the columns. 99.99% purity of methane gas was fed from the bottom of the soil columns and was regulated at flow rate of 5 mL/min using Dwyer Flowmeters, model RMA-151-SSV. Further, the soil columns were cap-sealed with lids equipped with inlet and outlet for sweep air channeled at 100mL/min to imitate the real advection of air above the landfill surface using Dwyer Flowmeters, model RMA-11-SSV. The interstices on the columns were glued by using silicone sealant to prevent any gas penetrations. The soil columns were also bubble-checked for leakage prior to usage. The set-up as illustrated in Fig. 1 was placed outdoor to equate the temperature in actual landfill environment.Perkin Elmer Clarus 600 Gas chromatography equipped with Flame Ionisation Detector GC-FID was used to analyse the gas samples. The injector, oven and detector temperature were set at 65, 55, 200°C respectively whilst nitrogen gas was applied as carrier gas at flow rate 60 mL/min (Park, Brown, & Thomas, 2002). The 30 m length of Agilent J&W GC column with 0.530 mm diameter was also utilised for gas separation in the machine. Methane oxidation efficiency was determined by Equation 1.

$$\text{Methane Oxidation Efficiency (\%)} = \frac{[CH4]_{in} - [CH4]_{out}}{[CH4]_{in}} \text{x100} \quad (1)$$

Whereby;
$[CH_4]_{in}$ = initial concentration of methane at inlet
$[CH_4]$ = concentration of methane collected at respective port

2.3. Control study

The probability of methane loss due to sorption and non-microbial degradation was also taken into account. Thus, the decline in methane concentration along the soil columns by microbial oxidation was verified by control experiment. Biofilter media was first sterilized by autoclaving them for 4 days in succession at 118°C and 0.138MPa for 45 minutes (Park et al., 2002). Sodium aside was then added in proportion of 25mg/kg biofilter media to hinder the microbial growth (Scheutz & Kjeldsen, 1997). The sterilized biofilter media was then packed into the soil column and was flushed with nitrogen gas to expel the oxygen that would initiate the growth of the remaining bacteria. The experimental regime was continued by purging the biofilter media with 99.99% methane at 5 mL/min for 24 hours and any methane disappearance were examined by using gas chromatography thereafter.

3. Results and discussion

3.1. Properties of soil cover

The clayey soil cover was tested on its physical and chemical properties. The results were tabulated as shown in Table 1. Via soil texture analysis using hydrometer, the soil cover was classified as clay with the composition of clay; silt and sand were 73.18, 9.90, and 14.43% respectively. It was dark yellowish brown in color (10YR 4/4) with reference to Munsell System and on-site dry bulk density value, 2.32g/cm3 was determined by applying core method. On top of that, both on-site moisture content and organic matter content were 71.51 and 3.42% respectively. The soil was also found acidic with pH 4.92, however, it was not a restricting determinant for biooxidation of methane by methanotrophs for these bacteria can survive in a broad range of soil pH (Scheutz et al., 2009).

Heavy metal contents in the soil were analyzed using Inductively Coupled Plasma Optical Emission Spectrometry, ICP-OES (Perkin Elmer 7000). The results were demonstrated in Table 2 in which Aluminium (Al) and Iron (Fe) were significantly high with value greater than 100 ppm/g_{soil}. It was found by Tamai et al. (2007) that Aluminum (Al) showed inhibition on methane uptake (Tamai et al., 2007).

Table 1: Physicochemical properties of soil cover at Jeram Sanitary Landfill.

Properties of Soil	Characteristics / Value
Colour (ASTM D1535) –Munsell System	Dark Yellowish Brown 10YR 4/4
Texture (ASTM D422) Sand Silt Clay	Clay 14.43% 9.90% 73.18% Error = 2.49%
pH (ASTM D4972)	4.92
On-site bulk density (ASTM D2937)	2.32 g/cm³
On-site moisture content (ASTM D2216)	71%
Organic matter content (ASTM D2974) – Loss-on-Ignition	3.40%
Heavy Metal Content (USEPA Method 3050B)	Refer Table 2

Meanwhile, the elevated concentration of Iron (Fe) and Manganese (Mn) explained the reduced amount of Phosphorous (P) by immobilizing it in the insoluble complexes (Hupfer & Dollan, 2003). As represented in Table 2, Arsenic (Ar), Boron (B), Cadmium (Cd), Cobalt (Co), Lithium (Li), Mercury (Hg), and Nickel (Ni) detected were very low in concentration, less than 0.01 ppm/g_{soil}. This value suggests that these heavy metals are negligible to inhibit the oxidation of methane by methanotrophs.

Table 2: Heavy metals concentration of soil cover at Jeram Sanitary Landfill

Element	Concentration (ppm/g_{soil})	Element	Concentration (ppm/g_{soil})
Aluminium (Al)	150.95	Lithium (Li)	<0.01
Arsenic (As)	<0.01	Magnesium (Mg)	10.64
Boron (B)	<0.01	Manganese (Mn)	2.62
Cadmium (Cd)	<0.01	Mercury (Hg)	<0.01
Calcium (Ca)	14.60	Nickel (Ni)	<0.01
Chromium (Cr)	0.72	Phosphorous (P)	0.48
Cobalt (Co)	<0.01	Potassium (K)	11.09
Copper (Cu)	0.08	Sodium (Na)	12.66
Iron (Fe)	135.96	Sulphur (S)	19.58
Lead (Pb)	0.55	Zinc (Zn)	0.95

It was reported by Mohanty et al., (2000) that Nickel (Ni) and Cadmium (Cd) inhibited methane oxidation process at minor influence. It was also mentioned in their study that Chromium (Cr) imposed a great impact on the methanotrophic activity. Hence, the presence of 0.72 ppm/g_{soil} Chromium (Cr) can be omitted as well as 0.95 ppm/g_{soil} that has no inhibitory effect on the methane oxidation process (Mohanty et al., 2000). On top of that, 0.08 ppm/g_{soil} of Copper (Cu) quantified in the soil sample was considered insignificant to hinder the biooxidation mechanism. This fact was supported by (Scheutz & Kjeldsen, (1997) that low concentration of Copper (Cu) allowed better activation of soluble monooxygenaseenzyme which facilitates the oxidation of methane by methanatrophs (Scheutz & Kjeldsen, 1997).

Besides, the contents of both Phosphorous (P) and Potassium (K) were in acceptable concentration that both of these elements are putative for their impacts on soil fertility and hence the microbial growth in the soil (Jugnia et al., 2012). Calcium (Ca) at 14.60 ppm/g_{soil} was also significant for the adhesion effect of the biofilm within the biofilter media (Hilger, Cranford, & Barlaz, 2000). Sulphur (S) which was possibly derived from the traces in LFG emitted was quantified to be 19.58 ppm/g_{soil} (Huitric & Kong, 2006). Its volatile compounds were proposed to have inhibitory effects on methane oxidation (Gunnar Börjesson, 2007). Magnesium (Mg) and Sodium (Na) were also moderately low present (below 15 ppm/g_{soil}) as bioavailable form to provide nutrients to the soil and thus the methane-oxidising consortia (Nikiema, Girard, & Brzezinski, 2009).

3.2. Enhancing methane oxidation by addition of coconut husk

Coconut husk is derived from the outer part of coconut fruit with thickness 5-6 cm. It includes the waxy outer thin layer (exocarpium) and fibrous inner layer (encarpium) which make 30% fibre and 70% dust (Tooy et al., 2014). Via this study, the addition of ground coconut husk was proven to enhance the performance of the clayey landfill soil cover at oxidizing methane. More than 94% of methane concentration was reduced with the addition of coconut husk and the percentage of methane oxidation efficiency slightly increased with the amount of coconut husk added. The coconut husk applied in the experiment was significantly high in both moisture content (16.89% of dry weight) and organic matter content (91.33% by loss-on-ignition). On top of that, as measured using CHNS Elemental analyzer, the values of total carbon and total nitrogen were 42.93 and 0.40% respectively to result in 107.33 of C: N ratio. The data are presented in Table 3.

Table 3: Properties of coconut husk

Properties of Coconut Husk	Percent (w/w)
Moisture content (ASTM D2216)	16.89
Organic matter content (ASTM D2974) – Loss-on-Ignition	91.33
Total Carbon (C)	42.93
Total Nitrogen (N)	0.40

This lab-scale landfill soil cover study encompasses the effects of supplementation of clayey landfill soil cover with coconut husk to the methane oxidation efficiency. The experiment on the clayey soil at adjusted moisture content to 12% with no addition of coconut husk showed the methane oxidation efficiency only reached 9.10% at the height of 10 cm, nevertheless, the value increased gradually at every 10 cm intervals. At 80 cm height, the total methane reduction obtained was 87.33%. The 12% moisture content (w/w) was chosen based on the studies by Faeiza et al. (2014). This finding was supported by literature that the optimum moisture content of the biofilter media to cater the methane oxidation was in the range 10 to 20% (Cao & Staszewska, 2011; Huber-Humer et al., 2008; Sadasivam & Reddy, 2014). Besides, it was found by Philopoulos et al. (2009) that the increase in moisture content caused the higher level of compaction of the soil that hinder gas distribution. Thus, optimum water content is vital for the soil-borne bacteria called methanotrophs to flourish by allowing the diffusion of oxygen through the macropores and micropores within the biofilter media. In the presence of oxygen, this microbial community consumes methane as their carbon and energy source and convert it to carbon dioxide (Chi, Lu, Li, & Wang, 2012; Pack et al., 2011; Reddy et al., 2014). The equation of the metabolic pathway is represented in the following equation by Hanson & Hanson (1996) :

$$CH_4 + 2O_2 \rightarrow CO_2 + 2H_2O\ (cell\ biomass) \quad (2)$$

The increasing number of the studies on the effects of organic matter enrichment to the landfill soil cover has been recorded. Organic matter in the forms of compost or mulch were added to the soil cover in pre-determined fraction or composition to enhance the biooxidation of methane (Cabral et al., 2010; Mostafid et al., 2012; Nguyen et al., 2013; Streese & Stegmann, 2003; Wilshusen et al., 2004).

The addition of coconut husk into clayey soil exhibited noticeable effects on the assessment. Fig. 2 lays out the amelioration on the methane oxidation efficiency with the association of larger portion of coconut husk to the soil. At 30:70 ratio of soil to coconut husk, the mixture manifested the greater potential to reduce methane concentration exceedingly more than the other ratios and the unamended soil. The improvement on methane oxidation was also observed at as lowest as 10cm thickness of the biofilter media whereby 35.52% efficiency was obtained. However, there were differences on the methane oxidation efficiency of the other two mixing ratios of soil to coconut husk; 50:50 and 70:30 which were merely 23.27 and 17.83% respectively.

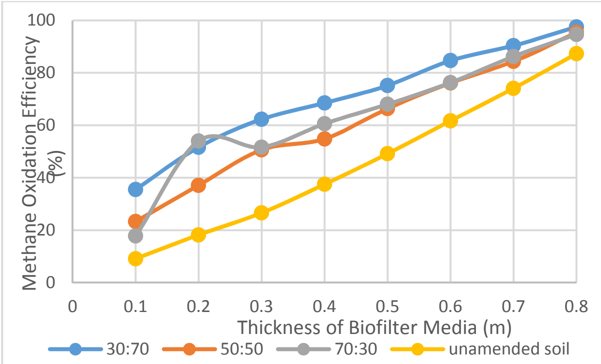

Fig. 2: Methane oxidation efficiency by biofilter media with different ratios of soil to coconut husk (v/v).

The distinctive values are explained with the fact that the biofilter media with 30:70 of soil to coconut husk ratio had the least compaction level at the lowest depth compared to the rest of soil columns to allow methane migration at optimum rate. Besides, the elevated amount of organic matter in the biofilter media induced the biooxidation of methane by promoting the growth of microbial community. The 70% volume ratio of coconut husk was the most favorable amount to supply nutrients to the methane-consuming bacterial flora to rapidly reproduce. It was also reviewed by Mer & Roger (2010) that high C:N ratio in organic matter corresponded to high labile carbon that was easily consumed by soil bacteria (Mer & Roger, 2010).

The percentage also steadily rose at every 10 cm thickness along all the soil columns and 97.52, 95.40 and 94.63% total oxidation were quantified at the highest port of 30:70, 50:50 and 70:30 ratios of soil to coconut husk respectively. The incremental pattern was also observed in the studies by Visvanathan et al. (1999) that used sandy loam and sandy clay loam as biofilter media in which the highest methane oxidation occurred at 55 to 80cm of the column height (Visvanathan et al., 1999). Another lab-scale experiment using taller and wider soil columns was carried out by Philopoulos et al. (2009) that sand-perlite-compost mixture and compost accomplished to reduce methane at 100% efficiency. The similar outcome of both mentioned studies and this experiment are reasoned by Gebert et al., (2011) that aeration throughout the soil profile permitted the activation of methanotrophic microorganisms to consume methane.

It was also found that there were no substantial differences between the methane oxidation efficiency by the biofilter media with mixing ratios of 50:50 and 70:30. The earlier was slightly lower than the later in methane oxidation from the height of 30 to 70cm albeit it managed to reduce methane more at 10cm layer. This phenomenon of impaired methane oxidation was suggested to cause by the formation and accumulation of extracellular exopolymeric substances (EPS) that hindered oxygen penetration (Hilger et al., 1999; Wilshusen et al., 2004). Although it was not measured, higher concentration of oxygen entering the biofilter media was taken as the probable factor to induce the secretion of excessive exopolymeric substances (Steinkamp et al., 2001).

Nonetheless, at 80cm of thickness, the biofilter media with 50:50 ratios exhibited slightly greater methane oxidation than 70:30 ratios owing to the facts it was more porous as a consequence of larger volume of coconut husk added. Therefore, the transfusion of oxygen into the body of biofilter media occurred at higher rate (Humer & Lechner, 2000).

The study also recorded that the methane oxidation was affected by the bulk density of the biofilter media. Soil bulk density which is correlated with soil compaction is subjected by governing parameters such as moisture content and thickness of biofilter media (Stern et al., 2007). Gebert et al. (2009) employed proctor percentage to measure soil compaction and documented that methane removal dropped with the increment of compaction level and consequently the diffusion coefficient (Gebert et al., 2009). In this study, dry bulk density was determined by core-method meanwhile the particle density was measured using pycnometer. The two densities then derived the percentage of porosity of the biofilter media and hence influence the rate of air-permeability and methane diffusion rate (Wickramarachchi et al., 2011). The data are tabulated in Table 4.

3.3. Methane oxidation efficiency in relation with bulk density

Table 4: Bulk density, particle density and porosity of biofilter media in each column

Column	Ratio of Soil to Coconut Husk	Bulk Density, ρb (g/cm³) –(ASTM D2937)	Particle Density, ρp (g/cm³) –(ASTM D854)	Porosity (%) $\left(1-\frac{\rho b}{\rho p}\right)\times 100$
1	30:70	0.3225	1.9862	83.76
2	50:50	0.4977	2.3088	78.44
3	70:30	0.6814	2.4246	71.90
4	Unamended soil	1.3717	2.8122	51.22

Double axes graph in Fig. 3 shows the relationship of bulk density of the biofilter media and the total methane oxidation in each soil column. Without amendment, the soil was quantified to have 1.3717 g/cm³ of bulk density and the total oxidation efficiency was 87.30%. However, the addition of coconut husk to the soil decreased the bulk density by the reason of high density ratio of soil to the coconut husk which was 2.04. The mixture of soil and coconut husk at ratios of 30:70, 50:50 and 70:30 by volume percentage resulted in 0.3225, 0.4977, 0.6814 g/cm³ of the bulk density of the biofilter media respectively. This increasing bulk density also influenced the porosity percentage that addition of more coconut husk resulted in increasing volume of pore and coarser texture of the biofilter media (Stern et al., 2007).

Methane movement from the bottom of the soil columns and the oxygen penetration from the headspace was the key feature of methane conversion to carbon dioxide by methanotrophic community. Hence, the breathing biofilter media allowed gas transportation throughout the micro and macropores of soil columns. This factor is important to provide sufficient retention time of methane molecule to reside in the biofilter media and therefore to allow microbial reaction to take place (Adams et al., 2011). As the experiment was carried out, the mixture of soil to coconut husk at 30:70 ratios served the most ideal compaction to maximize methane oxidation. Nevertheless, the incorporation of excess ground coconut husk would result in declining of methane reduction due to loosening structure of biofilter media and accelerating of methane flux. The evidence of the fact was registered in the study by Rachor et al. (2011). It was cited that in a system where the methane diffusion rate is greater than the oxygen permeation from the surface of biofilter media, the methane consumption is incomplete. They also justified the lowest methane concentrations at the height of 80 cm of all soil columns were due to lower bulk density at the top as compared to the bottom part due to increasing weight of the biofilter media added along soil profile.

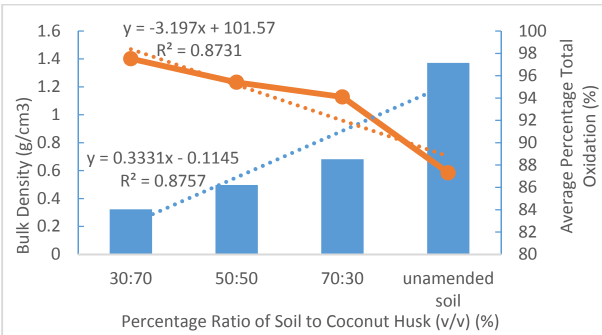

Fig. 2: Average total methane oxidation efficiency in relation with bulk density of biofilter media at different ratios (v/v) of soil to coconut husk.

Meanwhile, the unamended soil which had the highest bulk density charted the least methane uptake in the oxidation assessment. The occurrence of the inefficiency was relatively similar to the findings indicated by Prajapati & Jacinthe (2014) that methane oxidation rate was exacerbated in the intact cores (high bulk density) caused by the impeded oxygen percolation. For this reason, optimum gas blending only occurred at a few centimeters from the surface of the biofilter media (Pawłowska et al., 2011).

4. Conclusion

This experimental investigation plausibly shows that clayey soil can be applied as soil cover to mitigate the high concentration of methane emitted from the landfills. At the laboratory scale, the methane concentration was reduced up to 88.33% only by the unamended clayey soil providing the moisture content was maintained at 12% (v/v). However, the potential of the soil cover to oxidise methane was proven to increase with the supplement of organic matter whereby the biofilter media with 30:70 ratio was the best composition to oxidise methane up 97.52% efficiency. Coconut husk which is rich with organic matter and water content can facilitate the methane oxidation process by providing nutrients to the methanotrophs and hence encourage the growth of the microbial community (Albanna & Fernandes, 2009). Besides, coconut husk also increased the porosity of the clayey soil to make it a well-aerated soil cover by lessening the degree of bulk density. The mixing ratio 30:70 of soil to coconut husk reduced the bulk density from 1.3717 g/cm^3 of the unamended soil to 0.3225 g/cm^3. The pore size of the biofilter medium is vital for ensuring the transportation of methane from the base of the landfill is at suitable rate. This compaction factor is important in order to achieve optimum retention time for the reaction by microflora to occur whereby the increase in compaction leads to the receding in pore volume (Anlauf, 2012). In conclusion, coconut can be a promising element to enrich the landfill cover soil with organic matter and hence accelerate the methane biofiltration efficiency alongside the factors of availability, cost-effectiveness and harmful effects.

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