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Evaluation of Methane Oxidation Efficiency of Landfill Soil Cover: Influence of Soil Texture

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ABSTRACT

Objective – Methane emission from landfills has come to a great attention over recent years for its adverse effect to environment and human health. Owing to the facts, researchers have come out with numerous studies to improve the capability of the soil cover to oxidise the fugitive landfill gas. This paper revolves around the determination of methane oxidation efficiency of different texture media via sand incorporation into clayey soil at laboratory scale.
Methodology/Technique – Addition of sand by 30, 50 and 70% of volume ratio resulted in clay transformation to clay loam, sandy clay loam and sandy loam respectively. Sandy clay loam exhibited the highest methane oxidation efficiency at 93% followed by sandy loam and clay loam at 88% and 87% respectively. Meanwhile, unamended clay reduced methane concentration by 81%. The biooxidation of methane by methanotrophs was influenced by the conducive environment for the propagation of the consortia. Coarser texture media facilitated gas migration throughout the soil matrix by providing connected macropores to allow oxygen penetration and dynamic mixing of gases during the residence time of reaction.
Findings – The study manifested the prospective added material to clayey soil to simulate soil bacterial growth and therefore enhance the capacity of methane sink of the media.
Novelty – Sand significantly can transform the soil to the coarser texture for its potential to reduce methane emission at the landfill. Subsequently, the natural and abundant factors of sand in the environment would allow the application of this method at landfill scale for its cost-effectiveness and environmental harmlessness.

Type of Paper: Empirical

Keywords: Biocover, CH4 Mitigation, Landfill Gas, Landfill Soil Cover, Methane Oxidation, Soil Texture.

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1. Introduction

Methane is a detrimental gas which concentration is noteworthy to increase since a few decades ago. Its derogatory impacts severely impose on human health and environment generally. Methane was cited to have more than twice of radiative efficiency with 3.7x10-4 W m-2 ppb-1 after carbon dioxide (1.4x10-5 W m-2 ppb-1). It also has the capacity at trapping heat 25 times greater than carbon dioxide in a 100-year projection [1]. In Malaysia, landfills account for 53% of total methane emission in the country behind palm oil sector, swine

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manure, and industrial sectors. In the extrapolation to the year 2010 until 2020, the figure is expected to magnify due to expansion of human population in the region that would lead to 1 kg day-1 of waste generation per capita [2]. Based on the facts, numerous studies have been carried out to develop soil cover with maximum capability to mitigate methane emission via biooxidation process by soil bacteria called methanotrophs. During the reaction, methane which stems from the degradation process of organic waste is consumed by the methanotrophic community as carbon source using an enzyme namely monooxygenase under aerobic condition. The stoichiometric of the oxidation process was recorded in the study by Hilger and Humer (2003) [3];

$$CH_4 + 1.50\ O_2 + 0.118\ NH_4^+ \rightarrow 0.118(C_4H_8O_2N) + 0.529\ CO_2 + 1.71\ H_2O + 0.118\ H^+ \qquad (1)$$

Whereby;

$C_4H_8O_2N$ = microbial biomass

The process correlates with providing a suitable environment for the methanotrophs within the soil matrix which is the key aspect for their growth and sustainability to ensure continuous methane oxidation to take place. Among other governing parameters are pH value, moisture content, temperature and methane concentration that are in need of attention while performing the experiment. Equally important, soil texture also contributes to the breathable property of the soil cover and for that reason, the study evaluated the effects of coarsening soil texture by sand addition to methane oxidation efficiency in a closed-system method.

2. Methodology

2.1. Media Preparation

Soil sample was collected from Jeram Sanitary Landfill (JSL) which is located at 03˚11’20”N and 101˚21’50”E with total capacity of 8 million tonnes of waste. It is managed by Worldwide Landfills Sdn. Bhd. to cater 7 municipalities in the state of Selangor. The sampling was done in a sunny day in January 2015 and the soil was dug to 30 cm depth after removing the debris at the surface [4], [5]. At the laboratory, the soil was air-dried to achieve 15% (w/w) of moisture content before the granular size was standardised by grinding and then sieving through 2.0mm mesh [6]. This was to ensure the media to have large surface area to retain the soil bacteria within the matrix. On top of that, fine beach sand was used in the addition to the clayey soil cover and the percentage composition is illustrated in the pie chart in Figure 1.

2.2. Soil Column Set Up

Soil microcosms in the experiment were represented in soil columns fabricated using polyvinyl chloride (PVC) pipes with dimensions 0.5 cm thickness, 15 cm inner diameter and 107 cm length [7]. The columns were embedded with sampling ports at 10 cm intervals secured with Suba-Seal rubber septa and free-draining layer of 17 cm gravel in the range of 3 to 5 cm at the bottom as shown in Figure 2. Media were filled into the columns by random packing up to 5 cm above the highest sampling ports. The packed columns were agitated and dropped 5 times from the height of 3 to 5cm on the thermoplactic elastomers (TPE) pad to settle the media down the bottom of the columns which were hold by perforated plastic net with 0.5mm mesh. Methane gas of 99.99% purity was then purged from the column bases through the inlets at 5 mL/min while the headspace of the columns were channelled with advection of sweep air at 100 mL/min to represent the air movement on the landfill surface [8]. Both gases were controlled by Dwyer Flowmeter, model RMA-151-SSV and RMA-11-SSA respectively. In addition, by bubble test, the soil columns were examined for leakage at all penetration points and interstices that previously overlaid with silicone sealant. The soil columns were also securely placed outside the laboratory to obtain the equivalence temperature at the landfill.

2.3. Control Experiment

In order to confirm the methane loss by the microbial oxidation, a set of control columns were prepared. The BFM used was sterile which was autoclaved for 4 days in succession at 118°C and 0.138 MPa for 45 minutes [9]. It was then mixed with sodium azide at proportion of 25 mg/kgmedia to deactivate the soil microflora [10]. The columns were then packed with the sterile media and purged with nitrogen gas for 2 hours at 50 mL/min to displace trapped oxygen within the soil matrix that would initiate the bacterial growth. The methane concentration was determined according to the campaign performed on the samples.

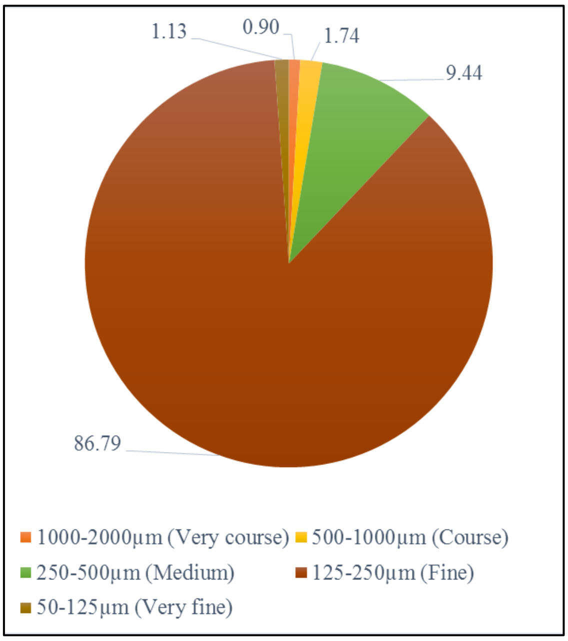

Figure 1: Size composition of sand used (%).

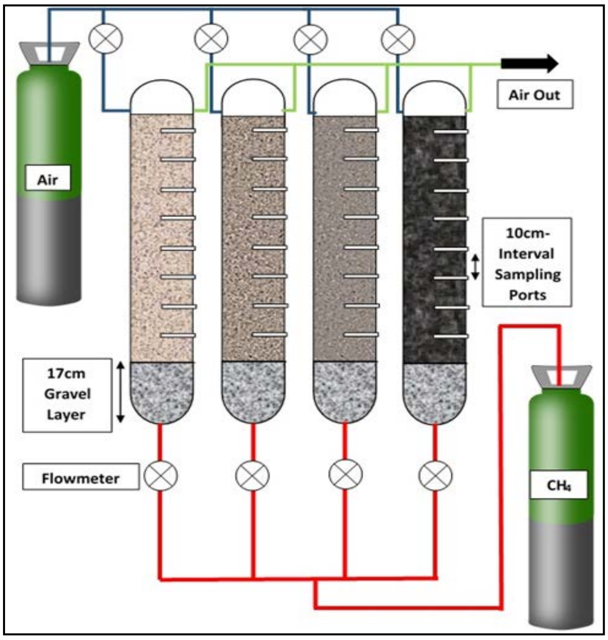

Figure 2: Schematic diagram of soil columns set up.

2.4. Methane Concentration Assessment

The quantification of methane concentration was carried out using Perkin Elmer Clarus 600 gas chromatography equipped with flame-ionisation detector (FID). The temperature of injector, oven and detector were set at 65, 55 and 200°C respectively whilst the carrier gas of nitrogen was regulated at 60 mL/min [9]. The regime of sampling and analysis of methane was carried through for 14 days successively. Methane oxidation efficiency (MOE) of each designated thickness of the media was calculated as follows;

$$MOE(\%) = \frac{[CH_4]_{in} - [CH_4]_{out}}{[CH_4]_{in}} \times 100 \qquad (2)$$

in which;

$[CH4]in$ = initial concentration of methane (ppm)

$[CH4]out$ = concentration of methane measured at sampling port (ppm)

3. Results and Discussions

3.1. Characteristics of Soil Cover

The physicochemical characteristics of the soil were determined and the results are presented in Table 1. In accordance with Munsell System, the soil colour was Dark Yellowish Brown with description 10YR 4/4. The soil was acidic with pH value 4.92 and the clay texture was deduced from the percentages of sand, silt and clay which was respectively 19, 31 and 50%. On top of that, the on-site dry bulk density of the clayey soil was 2.32 g/cm3 while the organic matter content was 3.40%.

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)	Clay
Sand	19%
Silt	31%
Clay	50%
pH (ASTM D4972)	4.92
On-site bulk density (ASTM D2937)	2.32 g/cm^3
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

Soil heavy metals content was determined using Inductive Inductively Coupled Plasma Optical Emission Spectrometry, ICP-OES (Perkin Elmer 7000) and the results are tabulated as in Table 2. High concentration of Aluminium (Al) an Iron (Fe) were greater than 100ppm/gsoil. Tamai et al., (2007) found that Aluminium imposed a significant inhibition effect on methane uptake, meanwhile Hupfer and Dolan (2003) explained the reduction of Phosphorous (P) was caused by elevated amount of Iron (Fe) and Manganese (Mn) by immobilising the species in insoluble complex [11], [12]. Besides, low concentration of Copper (Cu) that was quantified at 0.08ppm/gsoil was preferable in methane oxidation process that it permitted the activation of monooxygenase enzyme to be used by methanotrophs [13]. On top of that, moderately low concentration of Calcium (Ca), 14.60ppm/gsoil, was essential in the adherence of biofilm on the soil particles surface [14]. Magnesium (Mg) (10.64ppm/gsoil) and Sodium (Na) (12.66ppm/gsoil) were suggested to enrich the soil with nutrients in bioavailable structure to be consumed by the methanotrophic consortia [15].

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

3.2. Application of Sand in Soil Texture Transformation

Sand is one of the alternatives in soil amendment either in the intermediate or the final soil cover at the landfill. It is due to its high permeable property that encourages gas vertical movement through the connected pores [16]. In this study, four types of media were prepared based on the addition of sand to the clayey soil that eventually altered the texture of the media. The incorporation of sand was performed according to the clay to sand ratio by volume percentage (%v/v) and it was observed the texture of each media changed from clay to clay loam, sandy clay loam and sandy loam as shown in Figure 3.

The percentages of sand, silt and clay of each media were also measured and compared to the soil texture triangle by USDA Textural Classification System as demonstrated in Figure 4 [17]. It was noticed that the increasing volume of added sand drew the points of soil texture towards the left vertex of the triangle; from clay to sandy loam.

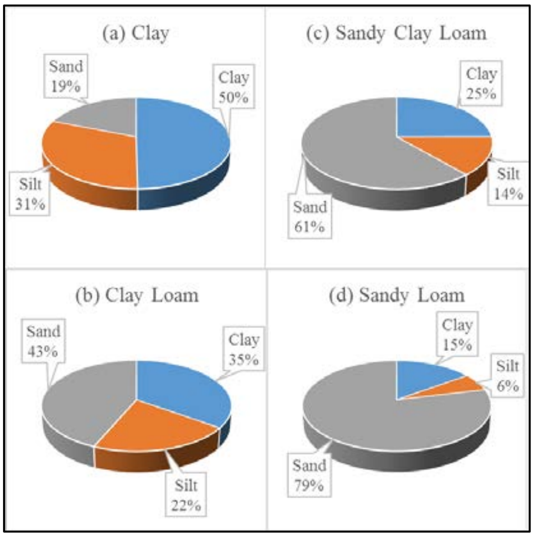

Figure 3: Composition and texture of each media with different ratios of Clay:Sand (%v/v); (a)unamended clay, (b)70:30, (c)50:50 and (d)30:70.

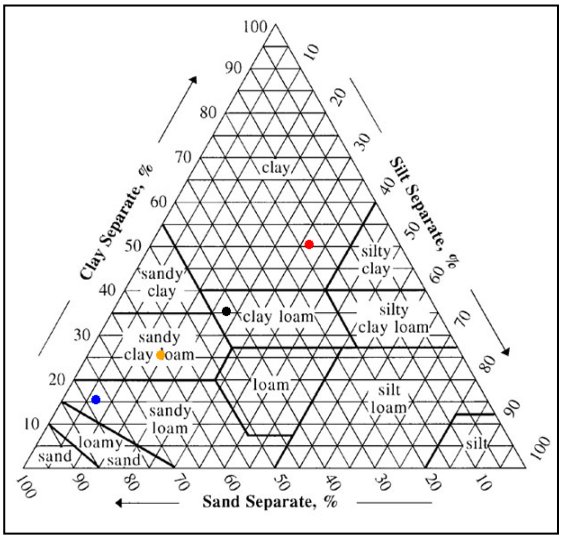

Figure 4: Reproduced soil texture triangle by USDA Textural Classification System. Coloured dots represent points of composition of each media.

3.3. Incorporation of Sand Into Soil Cover : Effects on Methane Oxidation Efficiency

Coarser texture of media was proven to provide congenial conditions for methanotrophic growth to elevate methane oxidation efficiency [18], [19]. This is shown in Figure 5 whereby the methane oxidation efficiency along the soil profile of clay loam, sandy clay loam and sandy loam were higher as compared to the unamended clay.

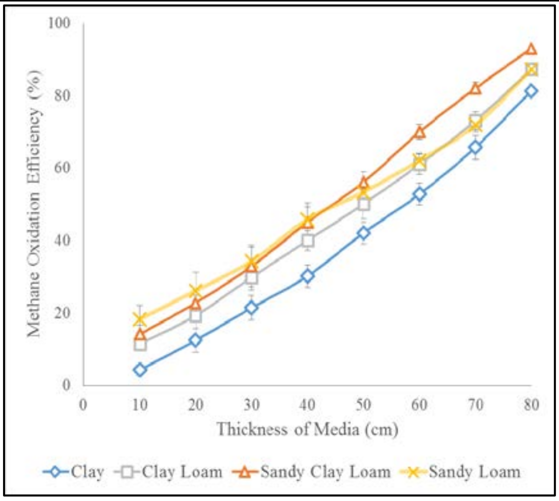

Figure 5: Methane oxidation efficiency by profiles of media with different textures.

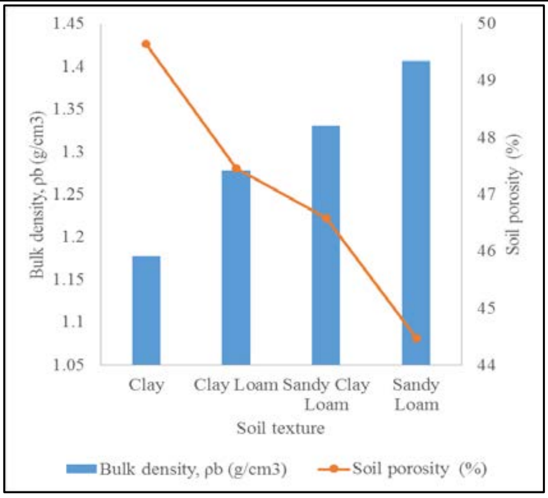

Figure 6: Bulk density and porosity of media with different textures.

It was apparent that soil texture gave profound impact on the methane oxidation process that it determined the distribution of intergranular micropores and macropores, and hence the tortuosity of the soil. Unamended clay with 50% of particles less than 0.2 µm was suggested to be more tortuous that distorted the flow of gases throughout the soil columns. This fact is an essential point to support the findings in this study in which unamended clay oxidised methane at the lowest efficiency in comparison with other types of soil [20]. On the contrary, methane was oxidised at higher efficiency in clay loam, sandy clay loam and sandy loam at all thickness levels. The difference in methane oxidation efficiency implied that the three types of soil were rich with interconnected pores to allow gas permeation within the soil crumbs [16], [21], [22].

Methane oxidation efficiency was found to increase along with the thickness of the soil matrix. The pattern was reasoned with the increasing volume of media in proportion of the soil thickness to provide more methanotrophs to oxidise the migrating methane from the bottom of the columns. Equally important, the lower methane oxidation efficiency at the lower horizon of the soil columns was reflected by higher degree of compaction due to media addition at the top. The weight imposed on the media reduced the pore volume through the depth of soil columns and hindered the penetration of oxygen. The concept was concurrent with the study by Gebert et al., (2009) that the findings demonstrated diffusivity of oxygen was impeded as a results of the increasing proctor density [23]. Despite this, all soil types showed better methane uptake at several centimeters of soil depth suggesting sufficient oxygen penetration from the surface to be well-blended with methane surged from the bottom of soil columns [24].

Noticeable accelerated methane oxidation efficiency of sandy clay loam at 60, 70 and 80cm thicknesses was also drawn on the graph showing greater rate of biooxidation process by methanotrophs. The percentage of efficiency recorded by sandy clay loam at the 3 points was 69, 82 and 93%, meanwhile sandy loam only achieved 62, 71 and 87% respectively. The oxidation potential by sandy clay loam seemed to outstrip the performance of sandy loam although the later demonstrated better methane removal at 10 to 40cm thicknesses. The distinguished gradients of efficiency line of the 2 types of soil intercepted at thickness 50cm suggested the need of optimum permeability for the oxygen to penetrate the soil body in the column. As explained by Rachor et al., (2011), the higher rate of methane migration from the base of landfill cover would decelerated the ingress of oxygen via atmospheric pressure [25]. Thus, over-porous aggregation at the top layer of sandy loam was not favourable for methane uptake due to incomplete oxidation.

In Figure 6, clay demonstrated the highest porosity level (49%) followed by clay loam, sandy clay loam and sandy loam (47, 46 and 44% respectively). Nevertheless, the pattern was inversely proportional to the degree of bulk density of each media. Clay, clay loam, sandy clay loam and sandy loam respectively had 1.18, 1.28, 1.33 and 1.41g/cm3 of bulk density and the correlation signified that the increasing weight of media was due to the additional sand. The concept was explained by Wickramarachchi et al., (2011) whereby the larger diameter of soil particles contributed to the higher cumulative mass fraction of the soil. The study also found that the addition of coarser particles in the clayey soil elevated the permeability of the soil matrix [26]. For that reason, the findings were taken as indication that soil porosity did not provide direct information on pore size, distribution and connectivity. Additionally, this was also an evidence for the clay loam, sandy clay loam and sandy loam to have better permeability rather than the unamended clay for the oxygen ingress as a results of the higher volume of added sand [27].

4. Conclusion

The application of engineered soil cover on at the landfill is vital to the abatement of methane emission to the atmosphere. Methane oxidation process mediated by the methanotrophic bacteria was found to enhance in the texture-modified of the clayey soil via sand addition at different volume ratio. The outcome of the experiment showed sandy clay loam oxidised the methane at 93% which was the highest as compared to unamended clay, clay loam and sandy loam. The 3 types of soil achieved 81, 87 and 88% of oxidation efficiency respectively. Coarsening the soil texture by incorporation of sand provided continuous and connected pores within the soil crumbs to make it a breathing soil cover that notably affects the rate of upward methane migration and the permeation of oxygen from the soil surface. At optimum condition, the well-blended gases had sufficient retention time to be intact with the methanotrophic consortia to convert methane to carbon dioxide and water. In brief, sand significantly can transform the soil to the coarser texture for its potential to reduce methane emission at the landfill. Subsequently, the natural and abundant factors of sand in the environment would allow the application of this method at landfill scale for its cost-effectiveness and environmental harmlessness.

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