Methane Oxidation in Landfill Cover Soils: A Review

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ABSTRACT

Migration of methane (CH_4) gas from landfills to the surrounding environment negatively affects both humankind and the environment. It is therefore essential to develop management techniques to reduce CH_4 emissions from landfills to minimize global warming and to reduce the human risks associated with CH_4 gas migration. Oxidation of CH_4 in landfill cover soil is the most important strategy for CH_4 emissions mitigation. CH_4 oxidation occurs naturally in landfill cover soils due to the abundance of methanotrophic bacteria. However, the activities of these bacteria are influenced by several controlling factors. This study attempts to review the important issues associated with the CH_4 oxidation process in landfill cover soils. The CH_4 oxidation process is highly sensitive to environmental factors and cover soil properties. The comparison of various biotic system techniques indicated that each technique has unique advantages and disadvantages, and the choice of the best technique for a specific application depends on economic constraints, treatment efficiency and landfill operations.

Key words: Methane emissions, Methane oxidation, Mitigation, Methanotrophic bacteria, Cover soils

1. INTRODUCTION

Methane (CH_4) gas is one of the most important greenhouse gases (GHGs). As a result of human activities, CH_4 emission concentrations in the atmosphere have increased from 715 ppb during the pre-industrial age to 1,732 ppb in the early 1990s and 1,774 ppb in 2005 (IPCC, 2007). Although the CH_4 concentration in the atmosphere is much lower than that of carbon dioxide (CO_2), its global warming potential is 25 times greater (IPCC, 2007). A study by Henckel *et al.* (2001) showed that the global CH_4 concentration is approximately 1.8 ppmv, which represents a doubling during the last 200 years.

Landfills rank as the third major anthropogenic source of CH_4 emissions after rice paddies and ruminant manure (Qingxian *et al.*, 2007; Ritzkowski *et al.*, 2007). A total of 40-60 metric tons of CH_4 are emitted from landfills worldwide, accounting for approximately 11-12% of the global anthropogenic CH_4 emissions (Ritzkowski *et al.*, 2007). CH_4 gas migration from landfills to the surrounding environment negatively affects both humankind and the environment. Gas explosion disasters due to landfill gas (LFG) migration resulting from variations in atmospheric pressure were reported in the village of Loscoe in England in 1986 and at Skellingsted Landfill in Denmark (Christophersen *et al.*, 2001).

Mitigation of landfill CH_4 emissions has been conducted using two approaches. The first approach uses gas collection systems for recovering or burning LFG, while the second approach seeks to reduce the emissions by various means, including waste recycling, composting and incineration. The first approach is more prevalent because it is cost-effective for large sanitary landfills. However, it is considered to be too costly and infeasible for older and smaller landfills whose CH_4 emission rates are much lower. Although major sanitary landfills utilizes gas collection systems, small quantities of LFG still escape into the atmosphere or migrate into the surrounding soil through the topmost layer of cover soil. Some researchers have found that conventional gas recovery systems only capture 50 to 90% of the CH_4 generated in landfills (Augenstein and Pacey, 1996). Therefore, the development and application of techniques for effectively reducing CH_4 emissions from landfills are required to minimize both the future global warming potential and the human risks associated with CH_4 gas emissions.

Microbial CH_4 oxidation in landfill cover soil may provide a means of controlling CH_4 emissions. Several studies have shown that the CH_4 oxidation process in landfill cover is an efficient method of CH_4 emission mitigation (Abushammala *et al.*, 2013a; Huber-Humer *et al.*, 2008; Stern *et al.*, 2007; Huber-Humer, 2004;

Hilger and Humer, 2003; Humer and Lechner, 1999). This process takes place in many natural systems and soils without human interference, due to the abundance of several groups of bacteria requiring oxygen (O_2) for the oxidation process. This process may be exploited to reduce CH_4 emissions at landfill sites where gas recovery systems are nonexistent or alongside existing gas collection systems to complement emissions control. A value of 0 to 10% of CH_4 oxidation has been recommended by the Intergovernmental Panel on Climate Change (IPCC) guidelines for national GHG inventories. However, laboratory and field studies indicates that the CH_4 oxidation capacity is between 0 and 100% (Jugnia *et al.*, 2008). Conversely, Bogner *et al.* (1995) stated that landfill cover soil under certain conditions can be a sink for atmospheric CH_4. Currently, there is insufficient information available regarding CH_4 oxidation capacity due to the lack of a standard method to determine the oxidation rate.

This study discusses the CH_4 oxidation process, which mitigates CH_4 emissions associated with LFG production. First, the mechanisms of CH_4 oxidation by methanotrophic bacteria in landfill cover soils are identified. Second, the key factors that control the CH_4 oxidation process in landfill cover soils are discussed. Finally, current techniques for mitigating CH_4 emissions using biotic systems are compared to investigate their key features and examine how they can be incorporated into the future design of landfill soil covers.

2. METHANE OXIDATION BACTERIA

The CH_4 oxidation process in landfill cover soils is facilitated by a group of methanotrophic bacteria that live in landfill cover soil (Huber-Humer, 2004). For simplicity, previous studies have reported that the CH_4 oxidation process in landfill cover soils is accomplished by methanotrophic bacteria (Abushammala *et al.*, 2012; Huber-Humer *et al.*, 2008; Albanna *et al.*, 2007; Stern *et al.*, 2007; Kettunen *et al.*, 2006). Methanotrophic bacteria (Fig. 1) are a group of obligate aerobes that have the ability to oxidize CH_4 under natural conditions to produce CO_2, water (H_2O), and microbial biomass (Eq. 1). Other organic compounds in LFG, such as aromatic and halogenated hydrocarbons, can be partially or fully degraded by methanotrophic bacteria that have the ability to co-metabolize substrates other than CH_4 (CLEAR, 2009; Scheutz and Bogner, 2003).

$$CH_4 + 2O_2 \rightarrow CO_2 + 2H_2O \ microbial \ biomass \qquad (1)$$
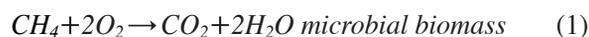
There are several complex enzymatic pathways for CH_4 oxidation. Methanotrophs are divided into three

Fig. 1. Methanotrophic bacteria (Huber-Humer, 2004).

types: type I methanotrophs follow a ribulose monophosphate (Ru MP) pathway, type II methanotrophs follow a serine pathway, and type X methanotrophs follow both pathways (Bogner, 1996). These classifications are based on their carbon assimilation pathways, intracytoplasmic membrane arrangements, cell morphology and the specific protein content of their DNA. In general, all three types of methanotrophs possess the CH_4 monooxygenase (MMO) enzyme, which assists them in oxidizing CH_4 for energy yield (Fig. 2) (Bogner, 1996).

MMO can be found in two forms: particulate CH_4 monooxygenase (pMMO) and soluble CH_4 monooxygenase (sMMO). Most methanotrophic bacteria are known to express themselves as pMMO, while a few of them express themselves as sMMO, and some have the ability to express themselves in both forms (Lee, 2008). However, methanotrophic bacteria have broad differences with respect to their responses to different CH_4 concentrations (Reay and Nedwell, 2004), and they can be classified accordingly as high-affinity or low-affinity methanotrophic bacteria. High-affinity methanotrophic bacteria are characterized by low CH_4 oxidation capacity, which enables them to begin oxidation at low CH_4 concentrations (0.8-280 nmol L^{-1}) (Huber-Humer *et al.*, 2008). High-affinity methanotrophic bacteria exist in soils temporarily exposed to CH_4 concentration. Low-affinity methanotrophic bacteria exhibit a high oxidation capacity, with CH_4 levels in the range of 0.8-66 µmol L^{-1} (Huber-Humer *et al.*, 2008). Low-affinity methanotrophic bacteria are more prevalent in landfill cover soils than are the high-affinity variant (Kightley *et al.*, 1995).

Methanotrophic bacteria can use substrates other than CH_4 under certain conditions, resulting in a reduction in the CH_4 oxidation rate and oxidation of ammonia (NH_4^+) to nitrite and nitrous oxide, due to the non-
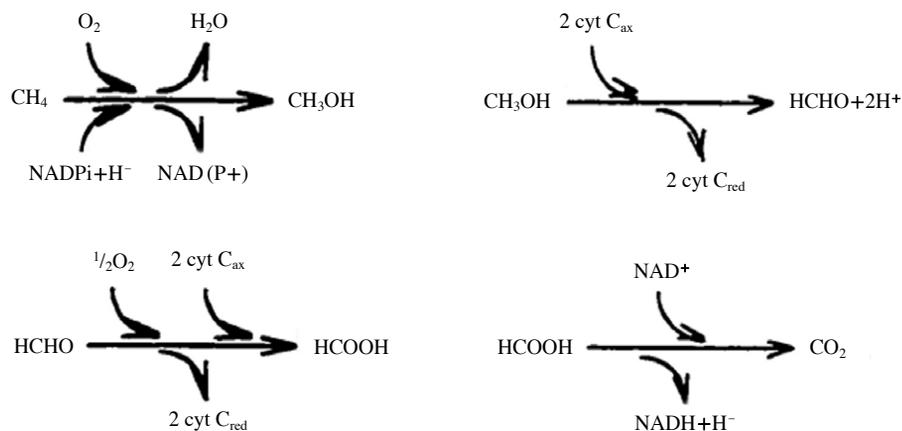
Fig. 2. CH_4 conversion into CO_2 by MMO enzyme.

specific nature of MMO (Knowles, 2005). Bogner (1996) documented inhibitions of methanotrophic activity due to nitrogen cycle processes that occur when hydroxylamine is produced by the oxidation of NH_4^+ by MMO, which inhibits MMO enzyme activity, when nitrite inhibits other enzyme activity necessary for CH_4 oxidation, and finally, when methanol is present in addition to NH_4^+.

3. FACTORS AFFECTING METHANE OXIDATION

The CH_4 oxidation capacity of landfill cover soils varies within and among landfills due to many factors that affect the oxidation process, such as seasonal variations (Abushammala *et al.*, 2013b; Einola *et al.*, 2007; Maurice and Lagerkvist, 2003; Börjesson *et al.*, 2001), physical and chemical heterogeneities of landfill cover soils (Tecle *et al.*, 2008; Albanna *et al.*, 2007; Visvanathan *et al.*, 1999), and the CH_4 concentrations in landfills (Boeckx *et al.*, 1996). According to Mosier *et al.* (2004), the major factors controlling CH_4 oxidation are potential biological demand and diffusion. The biological demand is regulated by both the physical and chemical environments, while the CH_4 diffusion rate is regulated by physical factors only. The reported values of landfill cover soils' CH_4 oxidizing efficiency vary widely in the literature. Albanna *et al.* (2007) reported that increasing the soil layer thickness from 15 to 20 cm increased the CH_4 oxidation values from 29% to 35% for a soil with 15% moisture content without nutrient addition, from 34% to 38% for a soil with a 30% moisture content without nutrient addition, and from 75% to 81% for a soil with a 30% moisture content with nutrient addition. However, in investigating the effect of bio-cover on CH_4 oxidation at the Leon

landfill in Florida, Stern *et al.* (2007) found that the efficiency of CH_4 oxidation can reach 64% with bio-cover utilization, while only 30% efficiency was reported for the control cell. Abichou *et al.* (2009) reported that at the same landfill, an average of 79% of CH_4 was oxidized in the bio-cover system and 29% was oxidized in the control cell. These wide variations can be attributed to the previously mentioned factors.

The major controlling environmental factors governing the CH_4 oxidation process in landfill cover soils, such as soil texture, organic content, moisture content, temperature, pH, nutrients, and O_2 and CH_4 concentrations (Wilshusen *et al.*, 2004a; Börjesson *et al.*, 2001; Boeckx *et al.*, 1996) are briefly discussed in this section. Applying knowledge about these controlling factors can optimize the process of mitigating CH_4 emissions from landfills.

3.1 Soil Texture
Soil texture affects LFG transport and atmospheric O_2 penetration. It therefore controls both CH_4 emission and oxidation rates. The CH_4 oxidation capacity in soils of various textures was investigated by Kightley *et al.* (1995), and it was found that higher oxidation efficiency occurs in coarse sand (61%) than in fine sand or clay (40-41%). Boeckx *et al.* (1997) concluded that coarse soils have higher oxidizing capacities than fine soils. Gebert and Grongroft (2009) recommended the use of coarse-textured soils with more than 17% air-filled pores by volume, such as sands, loamy sands, sandy loams and some of the coarsely textured loams, for use as CH_4 oxidizing bio-cover.

3.2 Soil Organic Content
The CH_4 oxidation rate increases with increasing soil organic content (Humer and Lechner, 2001; Christophersen *et al.*, 2000; Visvanathan *et al.*, 1999). Through

soil incubation tests, Christophersen et al. (2000) found that soils containing more organic matter more effectively mitigate CH_4 emissions through oxidation. They also found a relationship between the optimal soil moisture content and the organic matter content. The water content provides optimal oxidation increases with increasing organic matter content. Visvanathan et al. (1999) found that higher soil organic contents resulted in higher CH_4 oxidation rates in column assays. High-organic-content materials, such as compost, are widely used in landfill cover systems to enrich their CH_4 oxidation capacity (Abichou et al., 2009; Huber-Humer et al., 2008; Gebert and Grongroft, 2006a; Wilshusen et al., 2004a; Streese and Stegmann, 2003). Materials with high organic contents, high levels of nutrients, and high porosity have been proven to have high CH_4 oxidation capacities, which in some cases, tends to oxidize atmospheric CH_4. However, De Visscher et al. (2001) found that adding compost materials enhanced CH_4 oxidation, after a brief period of inhibition.

3.3 Moisture Content

There are several sources of water in landfill soil cover, including surface water infiltration, precipitation, water from manmade sources (leachate recirculation) and the decomposition reaction within the soil cover. As reported previously, a high moisture content in landfill soil cover reduces the available pore space for gaseous transport and diffusion. A high moisture content also reduces O_2 penetration into the soil cover, which is the main reactor for the CH_4 oxidation process. A low soil moisture content reduces the biological activity in soil cover and results in a reduction in CH_4 oxidation capacity (Tecle et al., 2008). The combination of soil drying due to low moisture content and the heat generated by CH_4 oxidation are likely to reduce the pore water content of soil, which may facilitate LFG transport through the shallow soil cover and reduce the oxidation capacity, due to the inhibition of microbiological activities that require a certain amount of water (Maurice and Lagerkvist, 2003). The desirable moisture content for high CH_4 oxidation activity is in the range of 11-25% by volume (Tecle et al., 2008). Boeckx et al. (1996) studied the effect of the soil moisture content on the CH_4 oxidation capacity of a landfill soil cover 30 cm thick. In his laboratory test, the moisture content of the soil was tested at 5, 10, 15, 20, 25 and 30% by weight, and the optimum moisture content was found to be between 15.6 and 18.8% by weight. Visvanathan et al. (1999) reported ideal moisture contents of 15% and 15 to 20% for maximum CH_4 oxidation in column and batch experiments, respectively. They stated that a negligible amount of CH_4 oxidation might occur at a 6% moisture content and that zero oxidation would occur at a 1.5% moisture content. Lee et al. (2009) found that the highest CH_4 oxidation rates occurred at a moisture content of 5% in a sandy landfill soil cover, with CH_4 oxidation rates decreasing as the moisture content increased.

Four sandy soils from two landfills in Denmark were investigated in batch experiments by Christophersen et al. (2000) to determine the effects of soil moisture on CH_4 oxidation. The results showed that the optimum moisture content range from 11 to 32% in all samples. It was also found that both moisture content and CH_4 oxidation increased as the organic matter content increased. More recently, work has been conducted by Park et al. (2002) to test the effect of the moisture content of loamy sandy soil on CH_4 oxidation capacity. They found that 13% by weight was the optimum moisture content for CH_4 oxidation in this soil. Another study conducted by Park et al. (2005) concluded that moisture content is the most important factor controlling the CH_4 oxidation rate is a sandy soil landfill cover. Mor et al. (2006) found that the effect of the soil moisture content on CH_4 oxidation in various types of compost was time-dependent and that the optimum moisture content ranges between 45 and 110% (dry weight basis).

3.4 Temperature

CH_4 oxidation in landfill soil cover is a biological process, and soil temperature is an important factor affecting this process (Streese and Stegmann, 2003). The methanotrophic community structure changes due to temperature variations, rather the quantity of type II methanotrophs decreasing with increasing temperature and precipitation (Horz et al., 2005). Several studies have reported on the optimum temperature for CH_4 oxidation in soil cover. Castro et al. (1995) found that soil temperature is an important factor in CH_4 oxidation at temperatures between $-5°C$ and $10°C$ but has no effect on CH_4 oxidation at temperatures between $10°C$ and $20°C$. Visvanathan et al. (1999) documented inhibition of CH_4 oxidation at temperatures higher than the optimum temperature, which they found in laboratory experiments to be in the range of 30 to $36°C$. De Visscher et al. (2001) confirmed these results in reporting that $35°C$ was found to be the optimum temperature for CH_4 oxidation activity in a sandy loamy soil from a landfill in Belgium. They also concluded that soil temperatures in excess of $30°C$ for long periods can lead to a reduction in CH_4 oxidation activity. Scheutz and Kjeldsen (2004) reported that CH_4 oxidation increased exponentially (with $R^2 > 0.91$) with increases in soil temperature from 2 to $25°C$. The maximum CH_4 oxidation rate occurred at $30°C$, and the oxidation rate

started to decline at 40°C. The effect of temperature on CH_4 oxidation in various types of compost was studied by Mor *et al*. (2006), who found that the effect of temperature on CH_4 oxidation is time-dependent and that the optimum temperature range is between 15 to 30°C. Borken *et al*. (2006) found that in forest soils, summer drought may increase CH_4 oxidation.

On the other hand, it has been reported that there is an interdependency between the effects of soil temperature and water content on CH_4 oxidation. Visvanathan *et al*. (1999) found that a sufficient moisture content combined with an appropriate temperature (approximately 20°C) could result in higher CH_4 oxidation. However, Castaldi and Fierro (2005) found that CH_4 oxidation rates were maximized when the water content was very low and the temperature was high. Einola *et al*. (2007) have reported an interdependency between soil temperature and water content, the most important factors controlling CH_4 oxidation capacity, and their effects on CH_4 oxidation.

3.5 pH

Variation in the pH value of a landfill soil cover affects CH_4 oxidation activities (Hutsch *et al*., 1994). According to Whittenbury *et al*. (1970), all types of methanotrophic bacteria can grow in pH values ranging from 5.8 to 7.4, with the optimum pH value being in the range of 6.6 to 6.8. However, Saari *et al*. (2004), found the optimum pH for CH_4 oxidation to vary from 4 to 7.5 in tests of CH_4 oxidation capacity in different type of soils with pH values ranging from 3 to 7.5. They also found that for some soils, the optimum pH for CH_4 oxidation is greater than the natural pH. The optimal pH value for CH_4 oxidation in soil samples collected from the Skellingsted Landfill in Denmark was found by Scheutz and Kjeldsen (2004) to be 6.9.

Methanotrophic bacteria are sensitive to the acidification of surrounding soils. Mer and Roger (2001) observed that the oxidation rate of non-fertilized permanent grassland at the Rothamsted experimental station in England decreased from -67 to -35 nL $CH_4.L^{-1}.h^{-1}$ (nL=nanoliter) when the pH of the cover soil at the site decreased from 6.3 to 5.6. Others have reported that the CH_4 oxidation decreases to zero at pH values between 5.6 and 5.1 (Huetsch *et al*., 1994). According to Hanson and Hanson (1996), methanotrophic bacteria cannot grow at pH values below 5. Numerous attempts to isolate or obtain enrichments for methanotrophic bacteria that would grow at pH values below 5.5 from acidic peat samples have failed.

3.6 Nutrients

Aside from the carbon substrate from CH_4 oxidation, bacteria in landfill soil cover require other nutrients for their cellular metabolism. The addition of nutrients to a soil cover system results in activation of methanotrophic bacteria, thus enhancing the CH_4 oxidation rate and oxidation efficiency (Lee *et al*., 2009; Albanna *et al*., 2007; Börjesson *et al*., 1998).

Albanna *et al*. (2007) found that soil moisture and the addition of nutrients have a combined effect on CH_4 oxidation in soil cover, and they reported that adding nutrients to incubated soil with a 32% average moisture content doubles the oxidation efficiency. However, adding nutrients to a soil with a low moisture content (15%) was found to have a negative effect on the oxidation efficiency. Lee *et al*. (2009) found that the CH_4 oxidation capacity of sandy soil cover increased by approximately 60% with the addition of 100 mg-N NH_4^+ per kg of soil.

Vegetation might affect on the growth and activity of methanotrophic bacteria in a variety of ways (Wang *et al*., 2008). Vegetation roots assist the process of transporting O_2 from the atmosphere into deeper soil layers (Fig. 3) (Tanthachoon *et al*., 2007). Furthermore, exudates that are supportive nutrients for methanotrophic bacteria are released to the root zone, which enhances CH_4 oxidation (Tanthachoon *et al*., 2007). Therefore, vegetation on the surfaces of landfill covers encourages methanotrophic activities throughout the soil depth profile. However, vegetation might compete with microorganisms for nutrients and water, which might result in an overall decrease in CH_4 oxidation (Hilger and Humer, 2003). Bohn and Jager (2009) found that the CH_4 oxidation rate can be enhanced by at least 50% by vegetation growth on landfill cover soils.

In engineered biological treatment systems, nitrogen and phosphorous is added in the form of NH_4^+ and orthophosphate. Adding NH_4^+ reduces the CH_4 oxidation capacity due to NH_4^+ inhibiting the activities of methanotrophic bacteria (Reay and Nedwell, 2004; Wang and Ineson, 2003; Hanson and Hanson, 1996). However, as discussed previously, the oxidation of NH_4^+ produces nitrite, which has an inhibitory effect on the MMO enzyme. Bosse *et al*. (1993) found that the CH_4 oxidation rate decreases at NH_4^+ concentrations ≥ 4 mM (mM=millimolar) and is completely inhibited at NH_4^+ concentrations > 20 mM. Keller *et al*. (2006) reported that nutrients (nitrogen and phosphorus) are important in the control of peat land microbial carbon cycling and that the roles of these nutrients differ with short- and long-term incubation.

3.7 Oxygen Concentration

Oxygen is one of the main reactors and limiting factors controlling the CH_4 oxidation process in landfill cover soils (Berger *et al*., 2005). The O_2 concentration
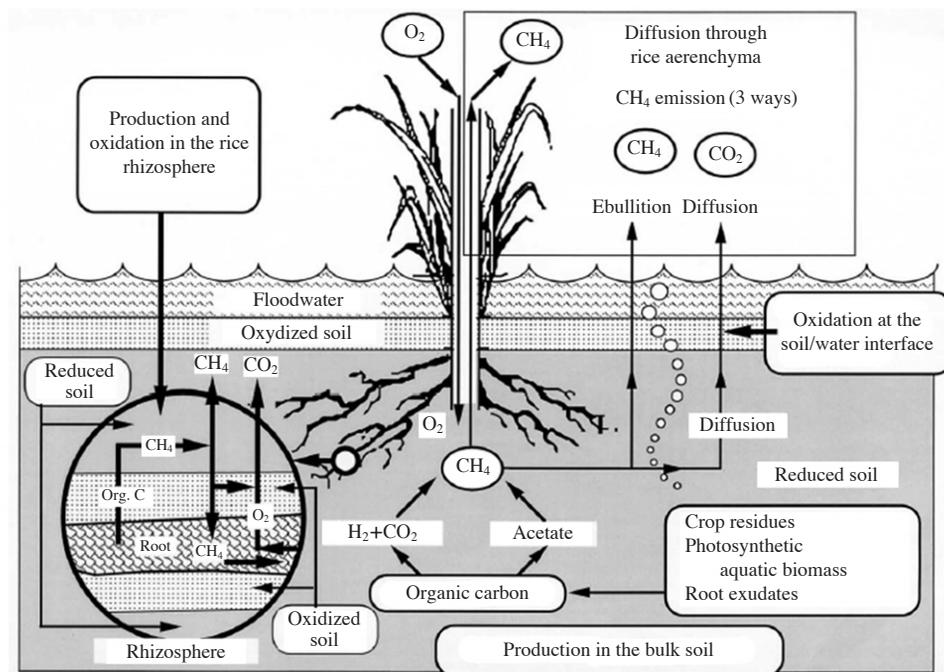
Fig. 3. Mechanism of production, oxidation and emission of CH_4 from rice fields (Mer and Roger, 2001).

Table 1. CH_4 consumption at various O_2 and N_2 concentrations over a 6-day assay period at 32°C.

Partial pressure		CH_4 consumed per day	
Oxygen (%)	Nitrogen (%)	Sample A (mL)	Sample B (mL)
0.0	70	0.0	0.0
10	60	1.05	0.94
20	50	0.88	0.88
30	40	1.05	1.05
40	30	–	0.94
60	10	0.35	0.52
70	0.0	0.23	0.29

Source: William and Zobell (1949)

varies with the depth of soil cover and is influenced by many variables, including gas characteristics, meteorological conditions, the microbial CH_4 oxidation rate, the soil texture and the cover thickness. Soil porosity controls the depth of O_2 penetration into soil (Humer and Lechner, 1999). The overlapping of the gradients of the CH_4 and O_2 concentrations in a soil profile occurs at the point of maximum CH_4 oxidation, and the depth at which this overlapping occurs is the optimum depth for maximum CH_4 oxidation. Several researchers have found different maximum CH_4 oxidation zones. Visvanathan *et al.* (1999) found that maximum oxidation occurs at depths of 15 to 40 cm, while Börjesson and Svensson (1997) found that 50 to 60 cm

is the optimum depth for maximum CH_4 oxidation. A study conducted by Jones and Nedwell (2006) stated that the maximum CH_4 oxidation occurred at depths from 10 to 30 cm, while Jugnia *et al.* (2008) stated that 0-10 cm is the optimal depth for CH_4 oxidation activity. William and Zobell (1949) reported that O_2 concentrations between 10 to 40% produce the highest range of CH_4 oxidation rates (Table 1), with an increase or decrease in O_2 concentration outside this range decreasing the CH_4 oxidation rate.

3. 8 Methane Concentration

The influence of the CH_4 concentration on the CH_4 oxidation capacity can be described using the Michaelis-Menten equation (Eq. 2):

$$V = V_{max} \frac{1}{1 + (K_M/C)} \tag{2}$$

where V is the actual CH_4 oxidation rate (m³ m⁻³ s⁻¹), V_{max} is the maximum CH_4 oxidation rate (m³ m⁻³ s⁻¹), K_M is the Michaelis constant for CH_4 (%) and C is the CH_4 concentration (%). Many researchers have reported the effect of the CH_4 concentration on the CH_4 oxidation capacity (Pawlowska and Stepniewski, 2006; Visvanathan *et al.*, 1999; Bogner *et al.*, 1997). Pawlowska and Stepniewski (2006) documented a significant influence of CH_4 concentration on the CH_4 oxidation capacity through a bio-filter model assay. They found

that an eightfold increase in CH_4 concentration caused the CH_4 oxidation capacity to increase by a factor of 1.1 to 2.5. Visvanathan *et al.* (1999) studied, in both column and batch assays, the effects of different environmental factors, such as soil temperature, moisture content and CH_4 concentration on the CH_4 oxidation capacity of landfill cover soils. They found that the CH_4 supply rate in column assays and the CH_4 concentration in the headspace of batch assays conflicts were different for low and high CH_4 oxidation capacities, due to the effects of both soil moisture content and temperature on the CH_4 oxidation capacity.

4. BIOTIC SYSTEMS FOR CH_4 OXIDATION

LFG treatment using a variety of types of biotic systems, including bio-washers (Figueroa, 1996), bio-membranes (Figueroa, 1996), bio-filters (Huber-Humer *et al.*, 2008; Gebert and Grongroft, 2006a; Wilshusen *et al.*, 2004b; Streese and Stegmann, 2003; Figueroa, 1996), bio-windows (Huber-Humer *et al.*, 2008), bio-covers (Shangari and Agamuthu, 2012; Huber-Humer *et al.*, 2008) and bio-tarps (Huber-Humer *et al.*, 2008), has been discussed in the literature. The first two types of systems (bio-washers and bio-membranes) for landfill emissions treatment are not discussed in this section because of their limited use. Biotic systems such as bio-filters, bio-windows, bio-covers and bio-tarps are discussed in more detail as they are the most widely used types of systems.

Biotic systems are economical options for controlling low levels of CH_4 emissions from landfills. Biotic systems can be used in many applications in landfills, in addition to gas collection systems for trapping CH_4 emissions at old landfills, at small landfill sites at which

gas collection systems are not economical options and during landfill site postclosure and aftercare processes.

Biotic systems used for CH_4 emissions mitigation are described in the following sections in terms of their key features and their incorporation into the design of future landfill cover soils.

4.1 Bio-Filter

Bio-filters were first used for contaminated gas treatment in the USA in 1966 to deodorize sewage sludge digestion gas. Recently, the application of bio-filters has expanded to CH_4 oxidation of LFG in addition to odor elimination. The first application of bio-filters for LFG treatment on a laboratory scale to investigate deodorization and the degradation of both H_2S and CH_4 was in 1979. Aerobic degradation of CH_4 in LFG using bio-filters was first investigated in 1986 (Figueroa, 1996).

Several laboratory and field experiments have been conducted to investigate bio-filter designs, media and gas flow. The filters are operated either in open or fully contained beds. A bio-filter consists primarily of a filter material that influences the performance of purification by its physical, chemical and biological properties (Figueroa, 1996). This filter material is considered to be the most important part of a bio-filter system because it supports bacteria cultures and is capable of sorption of contaminated gas. Bio-filter materials are primarily of biological origin, such as peat, compost from bio-waste, heather, shredded bark and sawdust (Huber-Humer *et al.*, 2008; Figueroa, 1996). Bio-filters have high water storage capacity and sufficient nutrients to facilitate biological processes. Admixtures such as expanded clay, polystyrene, lava and active carbon can be added to improve the structure of the filter material and increase its purification efficiency. LFG passively vented through the pressure gradient
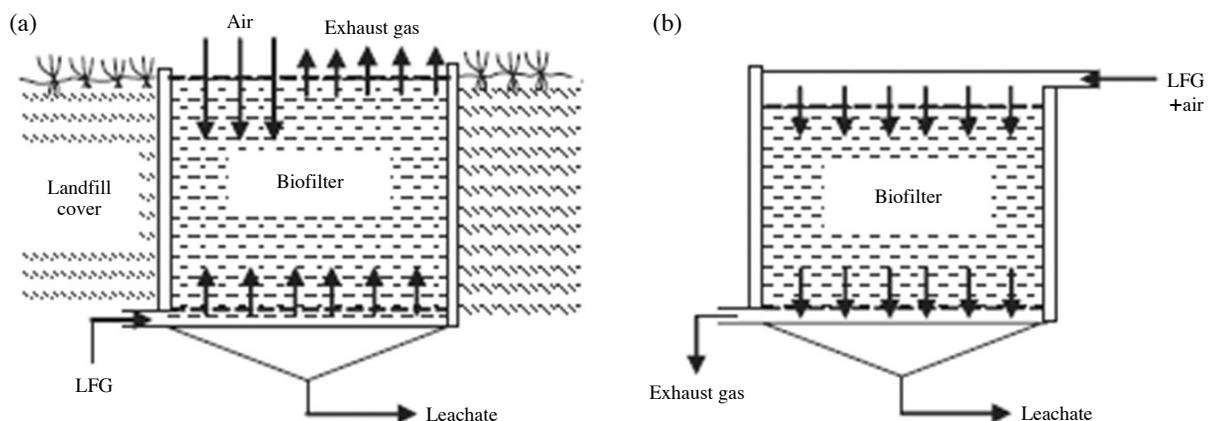

Fig. 4. Various integrated design of bio-filters and landfill soil cover: (a) upflow mode and (b) downflow mode.

between the landfill and the atmosphere (Gebert and Grongroft, 2006a) can be directed through the filter in either of two modes (Huber-Humer *et al.*, 2008): upflow or downflow (Fig. 4).

The CH_4 degradation process in a bio-filter is highly dependent on the retention time of LFG inside the filter (i.e., gas flow). Figueroa (1996) found 50 g CH_4 m^{-3} h^{-1} removal at a surface load of 5 m h^{-1} and complete CH_4 removal at a surface load of 0.5 m h^{-1}. However, several environmental conditions affect the filter efficiency (Figueroa, 1996), such as water content, temperature, pore volume or residence time and filter resistance. Good control of these environmental factors results in high filter efficiency and a positive effect on the functions of microorganisms.

CH_4 oxidation rates in the range of 20-60 g m^{-3} h^{-1} have been observed in a variety of laboratory column studies of bio-filters (Wilshusen *et al.*, 2004a; Streese and Stegmann, 2003; Park *et al.*, 2002), including studies up to one year in length. Wilshusen *et al.* (2004a) studied several types of compost filter material using column experiments conducted over periods up to 220 days on a laboratory scale to compare their CH_4 oxidation potential. They observed that a maximum of 400 g CH_4 m^{-2} day^{-1} CH_4 oxidized over a period of 100 days, followed by a decrease in rate to approximately 100 g CH_4 m^{-2} day^{-1} over the next 120 days. Various bio-filter materials for LFG treatment were tested by Streese and Stegmann (2003). They found that a mixture of compost, peat, and wood fibers exhibited a stable CH_4 oxidation rate of approximately 20 g m^{-3} h^{-1} for a CH_4 concentration of 3% by volume over a period of one year. On the other hand, fine-grained compost used as a bio-filter material was reported by the same authors to result in a CH_4 removal rate of up to 63 g m^{-3} h^{-1} in the first three months of the experiment for a CH_4 concentration of 2.5% by volume. Later, in the fifth month of the experiment, the decrease in the CH_4 oxidation rate was monitored. Both Wilshusen *et al.* (2004a) and Streese and Stegmann (2003) attributed the reduction in the CH_4 oxidation rate after reaching its maximum level to extracellular polymeric substances (EPS) formed by methanotrophic microorganisms.

EPS formation is a serious problem with bio-filters (Huber-Humer *et al.*, 2008; Gebert and Grongroft, 2006b; Wilshusen *et al.*, 2004a; Streese and Stegmann, 2003). These substances can block the pore space of the filter material and delay the substrate supplementation to the microorganisms inside the filter material, resulting in the deceleration of methanotrophic activity. EPS formation occurs primarily as a consequence of prolonged use of an active gas feed system (Wilshusen *et al.*, 2004a; Streese and Stegmann, 2003). Passive
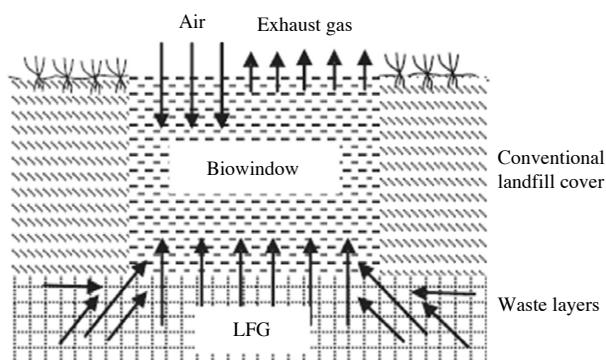

Fig. 5. Bio-window system incorporated into landfill soil cover (Huber-Humer *et al.*, 2008).

bio-filters tends to receive gas in an intermittent manner. However, by controlling the inlet flux rate to a landfill bio-filter, it may be possible to mitigate or prevent EPS formation (Huber-Humer *et al.*, 2008). Nonetheless, usage of additional gas distribution layers in bio-filter material optimizes mass transfer of gas components, thus reducing EPS formation (Streese and Stegmann, 2003). Hilger *et al.* (2009) reported that a nutrient imbalance could promote EPS formation in a bio-filter system.

4.2 Bio-Window

A bio-window is a system for mitigating landfill CH_4 emissions to the atmosphere. Composted materials with adopted environmental conditions are usually used as bio-window media to attain maximum CH_4 oxidation efficiency through enhanced microbial activity by CH_4 oxidation bacteria. The bio-window (Fig. 5) is integrated with the landfill soil cover in small regions of a landfill where high CH_4 emissions are observed. Measurements of the spatial variability of CH_4 emissions from landfill cover soils using the flux chamber technique and geo-statistical analysis are used to identify CH_4 emission hot spots within a landfill. Incorporation of a bio-window system into a landfill soil cover in these zones greatly mitigates the CH_4 emissions of the entire landfill. This technique is useful when the use of full-expanse compost materials is not economically feasible and when no gas collection system is available to feed a bio-filter system (Huber-Humer *et al.*, 2008). A bio-window receives passively vented LFG from the underlying waste, thereby offering flexible routes for gas movement.

4.3 Bio-Cover

In 2009, Huber-Humer *et al.* defined a landfill bio-cover as a top cover that optimizes the environmental conditions for methanotrophic bacteria and enhances
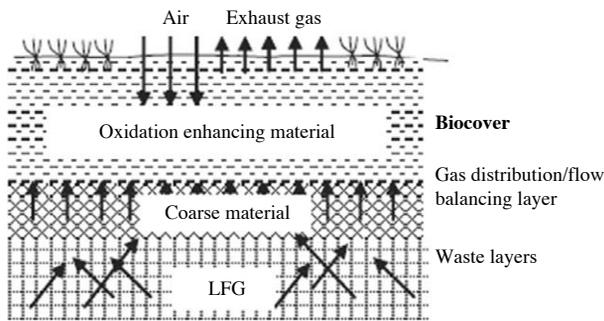
Fig. 6. Bio-cover system with gas distribution layer.

Fig. 7. Conceptual scheme of bio-tarp in landfill (Huber-Humer *et al.*, 2008).

biotic CH_4 consumption. A typical bio-cover system consists of a highly porous gas distribution layer above the waste, often gravel or crushed glass, followed by a compost-amended layer. The thickness of the gas distribution layer usually ranges from 10 to 30 cm (Jugnia *et al.*, 2008; Stern *et al.*, 2007), while the compost layer in the upper part is thicker, up to 100 cm or more, to attain high oxidation capacity. The gas distribution layer above the waste results in uniform LFG fluxes to the bio-cover layer, which permits biological activity to occur in a typical manner (Fig. 6).

Many researchers have attempted to reduce landfill CH_4 emissions to the atmosphere using bio-cover systems (Shangari and Agamuthu, 2012; Bogner *et al.*, 2010; Abichou *et al.*, 2009; Huber-Humer, 2009; Jugnia *et al.*, 2008; Stern *et al.*, 2007; Bogner *et al.*, 2005; Huber-Humer, 2004; Hilger and Humer, 2003; Humer and Lechner, 2001; Humer and Lechner, 1999). Their results show high CH_4 oxidation capacity in diverse, mature and well-structured compost materials, in both laboratory investigations (Abichou *et al.*, 2009; Stern *et al.*, 2007) and field trials (Bogner *et al.*, 2005; Huber-Humer, 2004; Humer and Lechner, 2001; Humer and Lechner, 1999). Shangari and Agamuthu (2012) found that CH_4 oxidation can reach 100% when a bio-cover of brewery spent grain and compost materials is used at a ratio of 7 : 3. Abichou *et al.* (2009) found that 100% CH_4 oxidation capacity can be achieved using compost bio-cover as a landfill cover. Humer and Lechner (2001) reported that the CH_4 oxidation capacity of compost landfill cover can reach 100% under optimum conditions of proper design and compost quality. Berger *et al.* (2005) found that in cover soil consisting of two layers, a mixture of compost plus sand (0.3 m) over a layer of loamy sand (0.9 m), the CH_4 oxidation capacity ranged from 98% to 57%. A system consisting of 50 cm of pre-composted yard bio-cover placed over 10-15 cm of crushed glass, utilized as a gas distribution layer, over a 40-100 cm interim cover, was used by Stern *et al.* (2007) to investigate
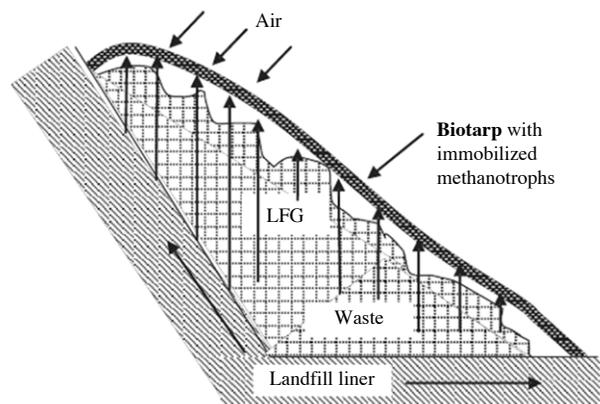
its landfill CH_4 emission reduction and CH_4 oxidation capacity. They found that the bio-cover cells reduced CH_4 emissions by a factor of 10 and doubled the percentage of CH_4 oxidation relative to control cells.

4.4 Bio-Tarp

There are two types of cover that are used in landfills before final capping. The first type is referred to as a daily cover and the second type is referred to as an intermediate cover. On an operational landfill site, a daily cover is used to cover the in-place waste at the end of each working day. An intermediate soil cover is used after a cell is completed and is awaiting final capping.

The daily cover functions to prevent interaction between the waste and air, thereby reducing odors. Furthermore, the daily cover is important to prevent wind-blown litter, minimize the risk of fire within the site, and discourage scavengers and flies. Most landfills use a 15-cm soil layer as a daily cover (Hilger *et al.*, 2009; Huber-Humer *et al.*, 2008). Alternative daily cover (ADC) materials, such as green and brown waste, sewage sludge, water slurries or commercial products such as foams and canvas, can also be used. The use of ADC materials are appropriate at some sites where local soils are unavailable and additional air space is required. Tarps are one type of ADC that maximizes airspace and thereby minimizes the required volume required of any other daily cover. Tarps are placed at the end of the working day and removed the next day to allow for further waste deposition. The filling of an active landfill cell may take a long period of time, during which no CH_4 collection occurs. In this case, the use of a bio-tarp (Fig. 7) is a good strategy for mitigating CH_4 emissions via methanotrophic bacteria impregnated in its material. Adams *et al.* (2011) found

Table 2. Comparison of different biotic system techniques.

| | Bio-filters | | Bio-windows | Bio-covers | Bio-tarps |
	Actively vented	Passively vented			
Field of application	*with a gas collection system, appropriate at old landfills where gas concentration has declined. *located within, on or adjacent to landfilled waste.	*without a gas collection system, appropriate at smaller and old landfills. *located within or under a landfill capping layer, within or adjacent to landfilled waste.	*usually used in hotspot areas in landfills. *can be used as interim or final cover.	*usually used over large areas such as an entire landfill. *used as an interim or final cover. *can be used with or without gas extraction. *can be used during landfill operation, aftercare or remediation.	*used as a daily cover. *used during the active phase of the landfill lifespan.
Materials used (examples)	Inorganic or organic engineered waste materials (e.g., compost, green or brown waste, manufactured clay, pellets, peat, wood chips, peat and sand mixtures, sewage sludge, water slurries).				*made of various types of polypropylene or polyethylene geo-membranes.
Advantages	*greater treatment of LFG emissions and therefore lower GHG emissions. *operation parameters are more controllable than bio-filter, bio-cover and bio-window.	*much less expensive than actively vented system. *no electricity is required, minimal maintenance, and lower operating costs than actively vented systems. *operation parameters are more controllable than bio-filter, bio-cover, and bio-window.	*simple and easy to install. *used in hotspot areas. *lower in cost. *no gas collection system needed.	*suitable for long-term operation (after landfill closure with low CH_4 concentration). *large surface area and thus high percent of oxidation. *low loading rate of CH_4, resulting in less EPS formation as bio-filter. *supports vegetation.	*mitigates emissions during landfill operation. *provides daily cover during routine landfill operation. *conserves landfill storage capacity.
Disadvantages	*have higher capital and operating costs than passively vented systems. *requires higher levels of operation and maintenance inputs than passively vented systems. *EPS formation occurs rapidly.	*The system may not ensure the prevention of surrounding gas migration. *EPS formation is slower than in an actively vented system.	*risk of CH_4 overload and EPS formation.	*limited control of operational conditions. *limited by materials demand.	*more expensive than conventional ADC. *no field data available.

Sources: Adapted from Huber-Humer *et al.* (2008); Streese and Stegman (2003); Hilger *et al.* (2009)

that the use of multiple layers of water-absorbent geo-textiles as bio-tarps removed 16% of CH_4, while adding landfill cover soil, compost or shale amendments to the bio-tarp increased the CH_4 removal by up to 32%.

Unlike bio-filters, bio-windows and bio-covers, bio-tarps can be removed and re-activated and can serve as a portable emissions reduction strategy. A comparison of the aforementioned biotic systems is provided in Table 2.

5. CONCLUSIONS

This study discusses the CH_4 oxidation process,

which mitigates CH_4 emissions associated with LFG production. Many factors affect the CH_4 oxidation capacity of landfill soil cover. The most important factors are environmental factors and the properties of the cover soil. Special consideration must be given to those factors to enhance the CH_4 oxidation process and to mitigate landfill CH_4 emissions.

Biotic systems are economically feasible options for controlling low levels of CH_4 emissions from landfills. Based on the summary table (Table 2) in which the various types of biotic systems are compared, biofilters appear to be appropriate at landfills where LFG collection is in operation because of their high CH_4 uptake capacity. Bio-covers offer the advantage of covering an entire landfill while simultaneously providing good water-holding capacity and porosity for vegetation and evapotranspiration. Bio-windows can be used at landfill hotspots. Bio-tarps can be appropriate alternative daily covers for use in mitigating CH_4 emissions during landfill operations at times when no CH_4 collection occurs. Each type of biotic system has advantages and disadvantages, and the choice of which method to apply depends on economic constraints, treatment efficiency and landfill operations.

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