

# Methane Oxidation in Landfill Cover Soils: A Review

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## ABSTRACT

Migration of methane (CH<sub>4</sub>) 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<sub>4</sub> emissions from landfills to minimize global warming and to reduce the human risks associated with CH<sub>4</sub> gas migration. Oxidation of CH<sub>4</sub> in landfill cover soil is the most important strategy for CH<sub>4</sub> emissions mitigation. CH<sub>4</sub> 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<sub>4</sub> oxidation process in landfill cover soils. The CH<sub>4</sub> 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

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## 1. INTRODUCTION

Methane (CH<sub>4</sub>) gas is one of the most important greenhouse gases (GHGs). As a result of human activities, CH<sub>4</sub> 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<sub>4</sub> concentration in the atmosphere is much lower than that of carbon dioxide (CO<sub>2</sub>), its global warming potential is 25 times greater (IPCC, 2007). A study by Henckel *et al.* (2001) showed that the global CH<sub>4</sub> 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<sub>4</sub> emissions after rice paddies and ruminant manure (Qingxian *et al.*, 2007; Ritzkowski *et al.*, 2007). A total of 40-60 metric tons of CH<sub>4</sub> are emitted from landfills worldwide, accounting for approximately 11-12% of the global anthropogenic CH<sub>4</sub> emissions (Ritzkowski *et al.*, 2007). CH<sub>4</sub> 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<sub>4</sub> 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<sub>4</sub> emission rates are much lower. Although major sanitary landfills utilize gas collection systems, small quantities of LFG still escape into the atmosphere or migrate into the surrounding soil through the top-most layer of cover soil. Some researchers have found that conventional gas recovery systems only capture 50 to 90% of the CH<sub>4</sub> generated in landfills (Augenstein and Pacey, 1996). Therefore, the development and application of techniques for effectively reducing CH<sub>4</sub> emissions from landfills are required to minimize both the future global warming potential and the human risks associated with CH<sub>4</sub> gas emissions.

Microbial CH<sub>4</sub> oxidation in landfill cover soil may provide a means of controlling CH<sub>4</sub> emissions. Several studies have shown that the CH<sub>4</sub> oxidation process in landfill cover is an efficient method of CH<sub>4</sub> 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<sub>2</sub>) for the oxidation process. This process may be exploited to reduce CH<sub>4</sub> 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<sub>4</sub> oxidation has been recommended by the Intergovernmental Panel on Climate Change (IPCC) guidelines for national GHG inventories. However, laboratory and field studies indicate that the CH<sub>4</sub> 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<sub>4</sub>. Currently, there is insufficient information available regarding CH<sub>4</sub> oxidation capacity due to the lack of a standard method to determine the oxidation rate.

This study discusses the CH<sub>4</sub> oxidation process, which mitigates CH<sub>4</sub> emissions associated with LFG production. First, the mechanisms of CH<sub>4</sub> oxidation by methanotrophic bacteria in landfill cover soils are identified. Second, the key factors that control the CH<sub>4</sub> oxidation process in landfill cover soils are discussed. Finally, current techniques for mitigating CH<sub>4</sub> 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<sub>4</sub> 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<sub>4</sub> 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<sub>4</sub> under natural conditions to produce CO<sub>2</sub>, water (H<sub>2</sub>O), 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<sub>4</sub> (CLEAR, 2009; Scheutz and Bogner, 2003).



There are several complex enzymatic pathways for CH<sub>4</sub> oxidation. Methanotrophs are divided into three

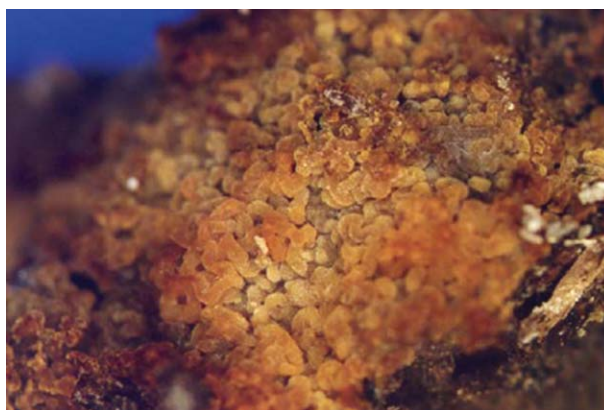
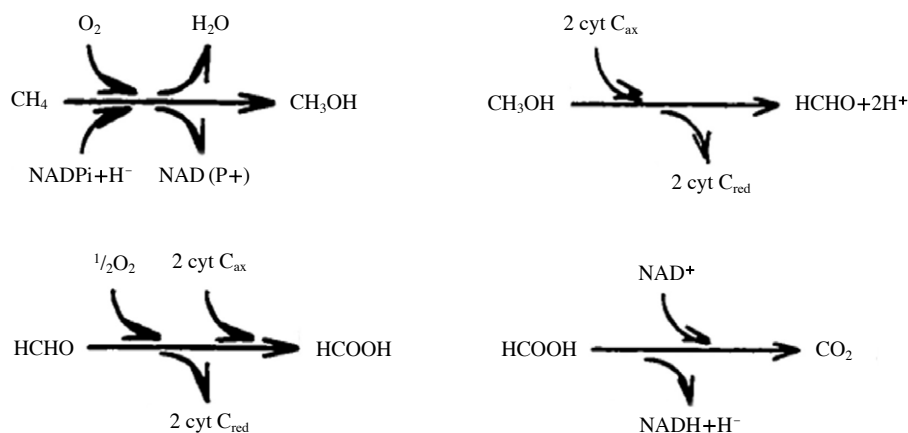


Fig. 1. Methanotrophic bacteria (Huber-Humer, 2004).

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<sub>4</sub> monooxygenase (MMO) enzyme, which assists them in oxidizing CH<sub>4</sub> for energy yield (Fig. 2) (Bogner, 1996).

MMO can be found in two forms: particulate CH<sub>4</sub> monooxygenase (pMMO) and soluble CH<sub>4</sub> 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<sub>4</sub> 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<sub>4</sub> oxidation capacity, which enables them to begin oxidation at low CH<sub>4</sub> concentrations (0.8-280 nmol L<sup>-1</sup>) (Huber-Humer *et al.*, 2008). High-affinity methanotrophic bacteria exist in soils temporarily exposed to CH<sub>4</sub> concentration. Low-affinity methanotrophic bacteria exhibit a high oxidation capacity, with CH<sub>4</sub> levels in the range of 0.8-66 μmol L<sup>-1</sup> (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<sub>4</sub> under certain conditions, resulting in a reduction in the CH<sub>4</sub> oxidation rate and oxidation of ammonia (NH<sub>4</sub><sup>+</sup>) to nitrite and nitrous oxide, due to the non-



**Fig. 2.** CH<sub>4</sub> conversion into CO<sub>2</sub> 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<sub>4</sub><sup>+</sup> by MMO, which inhibits MMO enzyme activity, when nitrite inhibits other enzyme activity necessary for CH<sub>4</sub> oxidation, and finally, when methanol is present in addition to NH<sub>4</sub><sup>+</sup>.

### 3. FACTORS AFFECTING METHANE OXIDATION

The CH<sub>4</sub> 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 (Tecele *et al.*, 2008; Albanna *et al.*, 2007; Visvanathan *et al.*, 1999), and the CH<sub>4</sub> concentrations in landfills (Boeckx *et al.*, 1996). According to Mosier *et al.* (2004), the major factors controlling CH<sub>4</sub> oxidation are potential biological demand and diffusion. The biological demand is regulated by both the physical and chemical environments, while the CH<sub>4</sub> diffusion rate is regulated by physical factors only. The reported values of landfill cover soils' CH<sub>4</sub> 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<sub>4</sub> 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<sub>4</sub> oxidation at the Leon

landfill in Florida, Stern *et al.* (2007) found that the efficiency of CH<sub>4</sub> 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<sub>4</sub> 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<sub>4</sub> oxidation process in landfill cover soils, such as soil texture, organic content, moisture content, temperature, pH, nutrients, and O<sub>2</sub> and CH<sub>4</sub> 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<sub>4</sub> emissions from landfills.

#### 3.1 Soil Texture

Soil texture affects LFG transport and atmospheric O<sub>2</sub> penetration. It therefore controls both CH<sub>4</sub> emission and oxidation rates. The CH<sub>4</sub> 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<sub>4</sub> oxidizing bio-cover.

#### 3.2 Soil Organic Content

The CH<sub>4</sub> 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<sub>4</sub> 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<sub>4</sub> oxidation rates in column assays. High-organic-content materials, such as compost, are widely used in landfill cover systems to enrich their CH<sub>4</sub> 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<sub>4</sub> oxidation capacities, which in some cases, tends to oxidize atmospheric CH<sub>4</sub>. However, De Visscher *et al.* (2001) found that adding compost materials enhanced CH<sub>4</sub> 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<sub>2</sub> penetration into the soil cover, which is the main reactor for the CH<sub>4</sub> oxidation process. A low soil moisture content reduces the biological activity in soil cover and results in a reduction in CH<sub>4</sub> oxidation capacity (Teclé *et al.*, 2008). The combination of soil drying due to low moisture content and the heat generated by CH<sub>4</sub> 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<sub>4</sub> oxidation activity is in the range of 11-25% by volume (Teclé *et al.*, 2008). Boeckx *et al.* (1996) studied the effect of the soil moisture content on the CH<sub>4</sub> 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<sub>4</sub> oxidation in column and batch experiments, respectively. They stated that a negligible

amount of CH<sub>4</sub> 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<sub>4</sub> oxidation rates occurred at a moisture content of 5% in a sandy landfill soil cover, with CH<sub>4</sub> 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<sub>4</sub> 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<sub>4</sub> 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<sub>4</sub> oxidation capacity. They found that 13% by weight was the optimum moisture content for CH<sub>4</sub> oxidation in this soil. Another study conducted by Park *et al.* (2005) concluded that moisture content is the most important factor controlling the CH<sub>4</sub> oxidation rate in a sandy soil landfill cover. Mor *et al.* (2006) found that the effect of the soil moisture content on CH<sub>4</sub> 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<sub>4</sub> 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<sub>4</sub> oxidation in soil cover. Castro *et al.* (1995) found that soil temperature is an important factor in CH<sub>4</sub> oxidation at temperatures between -5°C and 10°C but has no effect on CH<sub>4</sub> oxidation at temperatures between 10°C and 20°C. Visvanathan *et al.* (1999) documented inhibition of CH<sub>4</sub> 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<sub>4</sub> 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<sub>4</sub> oxidation activity. Schütz and Kjeldsen (2004) reported that CH<sub>4</sub> oxidation increased exponentially (with R<sup>2</sup> > 0.91) with increases in soil temperature from 2 to 25°C. The maximum CH<sub>4</sub> oxidation rate occurred at 30°C, and the oxidation rate

started to decline at 40°C. The effect of temperature on CH<sub>4</sub> oxidation in various types of compost was studied by Mor *et al.* (2006), who found that the effect of temperature on CH<sub>4</sub> 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<sub>4</sub> 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<sub>4</sub> oxidation. Visvanathan *et al.* (1999) found that a sufficient moisture content combined with an appropriate temperature (approximately 20°C) could result in higher CH<sub>4</sub> oxidation. However, Castaldi and Fierro (2005) found that CH<sub>4</sub> 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<sub>4</sub> oxidation capacity, and their effects on CH<sub>4</sub> oxidation.

### 3.5 pH

Variation in the pH value of a landfill soil cover affects CH<sub>4</sub> 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<sub>4</sub> oxidation to vary from 4 to 7.5 in tests of CH<sub>4</sub> 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<sub>4</sub> oxidation is greater than the natural pH. The optimal pH value for CH<sub>4</sub> 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<sub>4</sub>.L<sup>-1</sup>.h<sup>-1</sup> (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<sub>4</sub> 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<sub>4</sub> 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<sub>4</sub> 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<sub>4</sub> 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<sub>4</sub> oxidation capacity of sandy soil cover increased by approximately 60% with the addition of 100 mg-N NH<sub>4</sub><sup>+</sup> 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<sub>2</sub> 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<sub>4</sub> 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<sub>4</sub> oxidation (Hilger and Humer, 2003). Bohn and Jager (2009) found that the CH<sub>4</sub> 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<sub>4</sub><sup>+</sup> and orthophosphate. Adding NH<sub>4</sub><sup>+</sup> reduces the CH<sub>4</sub> oxidation capacity due to NH<sub>4</sub><sup>+</sup> 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<sub>4</sub><sup>+</sup> produces nitrite, which has an inhibitory effect on the MMO enzyme. Bosse *et al.* (1993) found that the CH<sub>4</sub> oxidation rate decreases at NH<sub>4</sub><sup>+</sup> concentrations ≥ 4 mM (mM=millimolar) and is completely inhibited at NH<sub>4</sub><sup>+</sup> 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<sub>4</sub> oxidation process in landfill cover soils (Berger *et al.*, 2005). The O<sub>2</sub> concentration



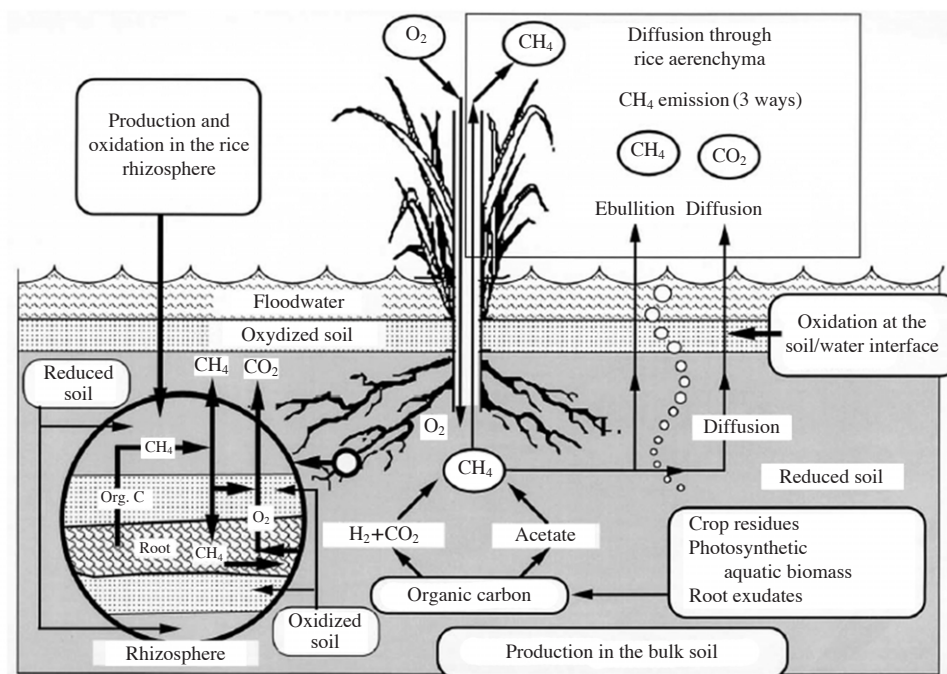


Fig. 3. Mechanism of production, oxidation and emission of CH<sub>4</sub> from rice fields (Mer and Roger, 2001).

Table 1. CH<sub>4</sub> consumption at various O<sub>2</sub> and N<sub>2</sub> concentrations over a 6-day assay period at 32°C.

Partial pressure		CH <sub>4</sub> 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<sub>4</sub> oxidation rate, the soil texture and the cover thickness. Soil porosity controls the depth of O<sub>2</sub> penetration into soil (Humer and Lechner, 1999). The overlapping of the gradients of the CH<sub>4</sub> and O<sub>2</sub> concentrations in a soil profile occurs at the point of maximum CH<sub>4</sub> oxidation, and the depth at which this overlapping occurs is the optimum depth for maximum CH<sub>4</sub> oxidation. Several researchers have found different maximum CH<sub>4</sub> 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<sub>4</sub> oxidation. A study conducted by Jones and Nedwell (2006) stated that the maximum CH<sub>4</sub> 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<sub>4</sub> oxidation activity. William and Zobell (1949) reported that O<sub>2</sub> concentrations between 10 to 40% produce the highest range of CH<sub>4</sub> oxidation rates (Table 1), with an increase or decrease in O<sub>2</sub> concentration outside this range decreasing the CH<sub>4</sub> oxidation rate.

### 3.8 Methane Concentration

The influence of the CH<sub>4</sub> concentration on the CH<sub>4</sub> oxidation capacity can be described using the Michaelis-Menten equation (Eq. 2):

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

where  $V$  is the actual CH<sub>4</sub> oxidation rate (m<sup>3</sup> m<sup>-3</sup> s<sup>-1</sup>),  $V_{max}$  is the maximum CH<sub>4</sub> oxidation rate (m<sup>3</sup> m<sup>-3</sup> s<sup>-1</sup>),  $K_M$  is the Michaelis constant for CH<sub>4</sub> (%) and  $C$  is the CH<sub>4</sub> concentration (%). Many researchers have reported the effect of the CH<sub>4</sub> concentration on the CH<sub>4</sub> oxidation capacity (Pawlowska and Stepniewski, 2006; Visvanathan *et al.*, 1999; Bogner *et al.*, 1997). Pawlowska and Stepniewski (2006) documented a significant influence of CH<sub>4</sub> concentration on the CH<sub>4</sub> oxidation capacity through a bio-filter model assay. They found

that an eightfold increase in  $\text{CH}_4$  concentration caused the  $\text{CH}_4$  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  $\text{CH}_4$  concentration on the  $\text{CH}_4$  oxidation capacity of landfill cover soils. They found that the  $\text{CH}_4$  supply rate in column assays and the  $\text{CH}_4$  concentration in the headspace of batch assays conflicts were different for low and high  $\text{CH}_4$  oxidation capacities, due to the effects of both soil moisture content and temperature on the  $\text{CH}_4$  oxidation capacity.

#### 4. BIOTIC SYSTEMS FOR $\text{CH}_4$ 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  $\text{CH}_4$  emissions from landfills. Biotic systems can be used in many applications in landfills, in addition to gas collection systems for trapping  $\text{CH}_4$  emissions at old landfills, at small landfill sites at which

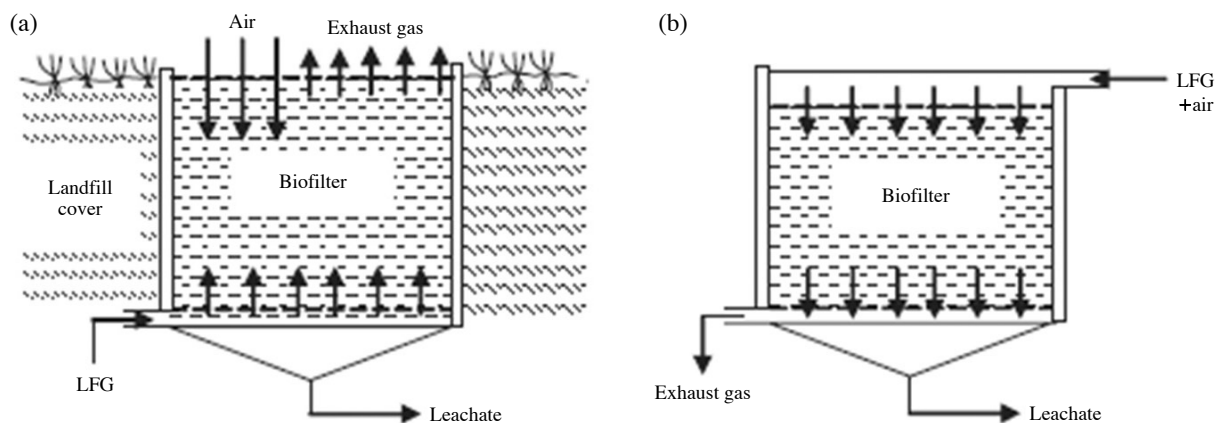
gas collection systems are not economical options and during landfill site postclosure and aftercare processes.

Biotic systems used for  $\text{CH}_4$  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  $\text{CH}_4$  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  $\text{H}_2\text{S}$  and  $\text{CH}_4$  was in 1979. Aerobic degradation of  $\text{CH}_4$  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



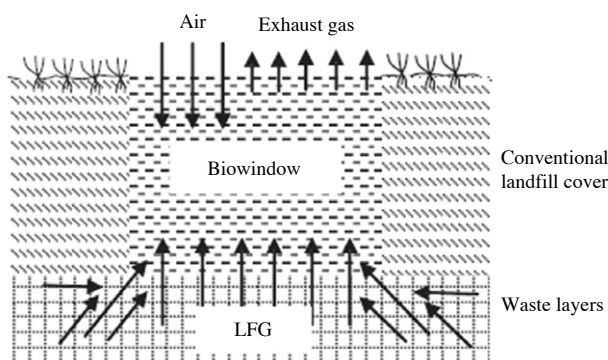
**Fig. 4.** Various integrated design of bio-filters and landfill soil cover: (a) upflow mode and (b) downflow mode.

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<sub>4</sub> 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<sub>4</sub> m<sup>-3</sup> h<sup>-1</sup> removal at a surface load of 5 m h<sup>-1</sup> and complete CH<sub>4</sub> removal at a surface load of 0.5 m h<sup>-1</sup>. 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<sub>4</sub> oxidation rates in the range of 20-60 g m<sup>-3</sup> h<sup>-1</sup> 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<sub>4</sub> oxidation potential. They observed that a maximum of 400 g CH<sub>4</sub> m<sup>-2</sup> day<sup>-1</sup> CH<sub>4</sub> oxidized over a period of 100 days, followed by a decrease in rate to approximately 100 g CH<sub>4</sub> m<sup>-2</sup> day<sup>-1</sup> 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<sub>4</sub> oxidation rate of approximately 20 g m<sup>-3</sup> h<sup>-1</sup> for a CH<sub>4</sub> 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<sub>4</sub> removal rate of up to 63 g m<sup>-3</sup> h<sup>-1</sup> in the first three months of the experiment for a CH<sub>4</sub> concentration of 2.5% by volume. Later, in the fifth month of the experiment, the decrease in the CH<sub>4</sub> oxidation rate was monitored. Both Wilshusen *et al.* (2004a) and Streese and Stegmann (2003) attributed the reduction in the CH<sub>4</sub> 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.

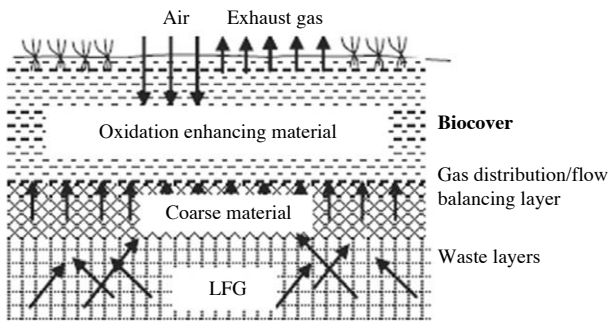
#### 4.2 Bio-Window

A bio-window is a system for mitigating landfill CH<sub>4</sub> emissions to the atmosphere. Composted materials with adopted environmental conditions are usually used as bio-window media to attain maximum CH<sub>4</sub> oxidation efficiency through enhanced microbial activity by CH<sub>4</sub> oxidation bacteria. The bio-window (Fig. 5) is integrated with the landfill soil cover in small regions of a landfill where high CH<sub>4</sub> emissions are observed. Measurements of the spatial variability of CH<sub>4</sub> emissions from landfill cover soils using the flux chamber technique and geo-statistical analysis are used to identify CH<sub>4</sub> emission hot spots within a landfill. Incorporation of a bio-window system into a landfill soil cover in these zones greatly mitigates the CH<sub>4</sub> 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

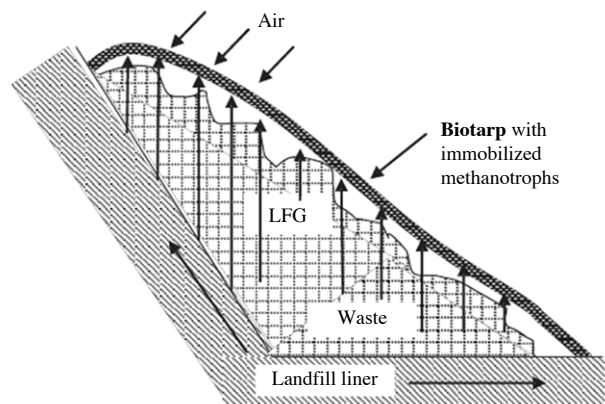




**Fig. 6.** Bio-cover system with gas distribution layer.

biotic  $\text{CH}_4$  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  $\text{CH}_4$  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  $\text{CH}_4$  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  $\text{CH}_4$  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%  $\text{CH}_4$  oxidation capacity can be achieved using compost bio-cover as a landfill cover. Humer and Lechner (2001) reported that the  $\text{CH}_4$  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  $\text{CH}_4$  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



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

its landfill  $\text{CH}_4$  emission reduction and  $\text{CH}_4$  oxidation capacity. They found that the bio-cover cells reduced  $\text{CH}_4$  emissions by a factor of 10 and doubled the percentage of  $\text{CH}_4$  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  $\text{CH}_4$  collection occurs. In this case, the use of a bio-tarp (Fig. 7) is a good strategy for mitigating  $\text{CH}_4$  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 geomembranes.
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 <sub>4</sub> concentration). *large surface area and thus high percent of oxidation. *low loading rate of CH <sub>4</sub> , 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 <sub>4</sub> 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); Streeze and Stegman (2003); Hilger *et al.* (2009)

that the use of multiple layers of water-absorbent geotextiles as bio-tarps removed 16% of CH<sub>4</sub>, while adding landfill cover soil, compost or shale amendments to the bio-tarp increased the CH<sub>4</sub> 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<sub>4</sub> oxidation process,

which mitigates CH<sub>4</sub> emissions associated with LFG production. Many factors affect the CH<sub>4</sub> 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<sub>4</sub> oxidation process and to mitigate landfill CH<sub>4</sub> emissions.

Biotic systems are economically feasible options for controlling low levels of CH<sub>4</sub> emissions from landfills. Based on the summary table (Table 2) in which the various types of biotic systems are compared, bio-filters appear to be appropriate at landfills where LFG collection is in operation because of their high CH<sub>4</sub> 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<sub>4</sub> emissions during landfill operations at times when no CH<sub>4</sub> 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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