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ROLE OF LANDFILL COVER IN REDUCING METHANE EMISSION

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Abstract: Uncontrolled emissions of landfill gas may contribute significantly to climate change, since its composition represents a high fraction of methane, a greenhouse gas with 100– year global warming potential 25 times that of carbon dioxide. Landfill cover could create favourable conditions for methanotrophy (microbial methane oxidation), an activity of using bacteria to oxidize methane to carbon dioxide. This paper presents a brief review of methanotrophic activities in landfill cover. Emphasis is given to the effects of cover materials, environmental conditions and landfill vegetation on the methane oxidation potential, and to their underlying effect mechanisms. Methanotrophs communities and methane oxidation kinetics are also discussed. Results from the overview suggest that well-engineered landfill cover can substantially increase its potential for reducing emissions of methane produced in landfill to the atmosphere.

INTRODUCTION

There is a growing consensus that the earth temperature is rising, and that the principle cause for this is the emission of greenhouse gases (e.g. CO_2, CH_4 and N_2O) [1–2]. Methane (CH_4), an important greenhouse gas that can remain persistently in the atmosphere for approximately 9–15 years, is 25 times more effective in trapping heat in the atmosphere than carbon dioxide (CO_2) over a 100– year period [1]. Although CH_4 concentration in the atmosphere is rather low, its current contribution to global warming reaches as much as 15% [3]. More unfavourably, this contribution is believed to remain escalating as a result of a growing CH_4 emission to the atmosphere.

Landfill is known as one of the major anthropogenic emissions sources of CH_4. According to the US EPA [4], the total anthropogenic emissions of CH_4 in 2000 were about 282.6 million tons, of which 13% was due to landfill emissions. Therefore, reduction of landfill gas emission to the atmosphere is of importance for mitigation of climate change. One win–win strategy toward this purpose is to collect landfill gas and use it as a substitute fuel for heat or electricity generation. However, there are several scenarios under which collection and utilization of the biogas is not available. For example, some

old landfills were not equipped with gas collection systems; some abandoned landfills still emit more or less methane. Even in modern landfills, the biogas produced cannot be collected sufficiently, primarily limited by the gas collection system, in particular the number of gas wells. This was demonstrated by the study evaluating the amounts of the methane collected and lost for 25 landfills in California [3]; the result showed that the amount of methane lost was approximately two times greater than the collected methane amount on the basis of per ton of municipal solids waste. Therefore, there is a need seeking for other pathway for suppression of landfill CH_4 emission.

Soils, more precisely the microorganisms living in soils, have been widely observed to have the unique ability of utilizing CH_4 as their carbon and energy source and oxidize it to CO_2 [5–6]. Landfill cover, where CH_4 is presented at high concentration and O_2 is partly available, has proven to possess impressive CH_4 oxidation potential [7–14]; recent study reported a mean value of 36 ±6% for CH_4 oxidation efficiency [15], although the default value for this parameter set by IPCC and the USEPA is relatively low (0%–10%) [16].

Landfill CH_4 oxidation potential varies largely, depending on a number of factors. Factors influencing CH_4 oxidation performance in landfill include cover materials (type and physical-chemical properties), landfill gas flux (particularly CH_4 concentration), O_2 availability, landfill vegetation and climatic variables [17–21] . The present work presents a brief overview on CH_4 oxidation potential in landfill cover, main limiting factors and their underlying effect mechanism. Specifically, the methanotrphic communities and CH_4 oxidation kinetics are discussed.

METHANOTROPH COMMUNITIES

CH_4-consuming bacteria (known as methanotroph) can grow under different environment conditions [5–6, 22], even extreme environments, e.g. in permafrost soils of Siberia (a mean annual temperature of –14.7°C) [23], at temperature as high as 72°C [24], and in extremely acidic (pH 2.0−2.5) [25] or alkalic environment (pH 9.5–10.5) [26]. Based on the differences in morphological and physiological characteristics, methanotrophs are divided into two groups: type I and type II, together comprising a total of 12 genera [27]. Among them, type I methanotrophs account for 8 genera: *Methylomonas*, *Methylobacter*, *Methylomicrobium*, *Methylosarcina*, *Methylosphaera*, *Methylococcus*, *Methylocaldum* and *Methylothermus*, while type II consists of the rest, that is, *Methylosinus*, *Methylocystis*, *Methylocapsa* and *Methylocella* [27–28]. Recently, several new members of methanotrophs, such as *Crenothrix polyspora* [29] and *Clonothrix fusca* [30], have been isolated and characterized, suggesting that the methanotroph communities are more diverse than were previously thought.

Methanotrophs oxidize methane to methanol by using enzyme methane monooxygenase (MMO). Two types of MMO have been isolated from methanotrophs, including a soluble cytoplasmic MMO (sMMO) and a membrane-bound particulate MMO (pMMO). The pMMO is presented in all methanotrophs (except *Methylocella*), while the sMMO only in a few methanotrophic genera [28]. The pMMO-containing cells have better growth capabilities and higher affinity for methane than the cells containing sMMO [27]. Copper ions were suggested to play a significant role in both pMMO regulation and the enzyme catalysis [31].

METHANE OXIDATION POTENTIAL

In general, methane oxidation potential is parameterized by methane oxidation rate and/or methane oxidation efficiency. The former is generally expressed on an area or mass basis (expressed as g $CH_4 \cdot m^{-2} \cdot d^{-1}$ or g $CH_4 \cdot g^{-1} \cdot d^{-1}$) while the latter expressed as percentage (% methane oxidized). Conventional method to determine these parameters is the static flux chamber technique based on mass balance. In some cases stable isotopes measurement was also employed as an alternative or confirmatory approach. More recently, push-pull tests, which were initiated to determine reaction rates of pollutant degradation in groundwater aquifers, were adapted as a possible method for in situ measurement of methane oxidation rates in landfill [32–33].

Table 1 summarizes the methane oxidation capacities measured in different landfill cover materials under different conditions. The potential of the methane oxidation in landfill cover is substantially impressive. As seen from Table 1, up to 100% of CH_4 emissions from landfill can be oxidized to CO_2 and H_2O, if landfill cover is well designed and constructed. The maximum methane oxidation capacity measured in bed layer (60–80 cm depth) at laboratory and field scales ranged from 200 to 400 g $m^{-2}d^{-1}$ [34–37]. Under certain conditions, methanotrophs in landfill cover not only oxidize the CH_4 produced from landfill, but also consume atmospheric CH_4 [38–39].

Table 1. Methane oxidation potential measured in landfill covers

Landfill cover material	CH_4 Loading	Methane oxidation potential		Source
	g $CH_4 \cdot m^{-2} \cdot d^{-1}$	Oxidation rate	Oxidation efficiency	
		g $CH_4 \cdot m^{-2} \cdot d^{-1}$	%	
Four terrestrial mineral soils Sediment rich in organic matter	25–100			[40]
Garden waste composts Sewage sludge compost	179–201	45–112	23–56	[41]
Cover with compost layer	2.69	2.69	100	[42]
Control (without compost layer)	29.4	19.5	63	
Landfill cover soil	233.6	118	51	[43]
Mixture of soil and earthworm cast	233.6	232	99–100	
Mixture of soil and PAC	233.6	232	99–100	
Mechanically–biologically treated municipal solid waste	30–78	22–82		[44]
Landfill cover soil	35.3–84.7		20–100	[45]

FACTORS AFFECTING METHANE OXIDATION

Methane concentration

The underlying biochemical reaction process of methane oxidation can be simplified as follows [46]:

$$CH_4 + O_2 \xrightarrow{\text{Methanotrphic microorganisms}} CO_2 + 2H_2O \quad (1)$$

Apparently, the methane oxidation activities are intrinsically dependent on CH_4 and O_2 concentrations. Various laboratory and filed experiments have shown that higher CH_4 concentration led to an increase in the CH_4 oxidation rate, up to a certain constant level [40].

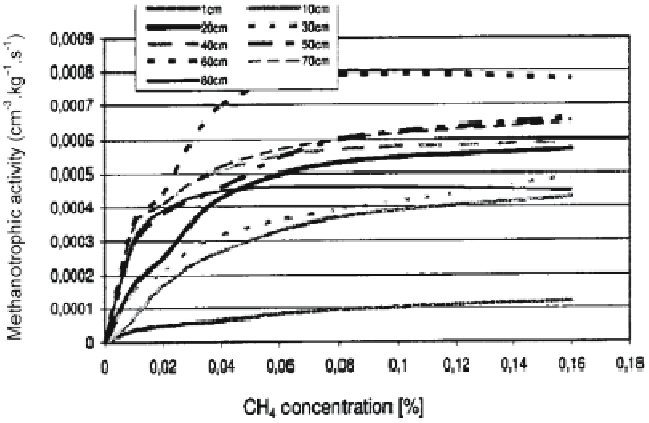

Fig. 1. The CH_4 concentration dependence of methanotrophic activity observed in sand materials taken from 9 different depths in the column [14]

Pawłowska and Stepniewski [8, 14] studied the methanotrophic activity in the vertical profile of a simulated landfill cover as a function of CH_4 concentration (Figure 1). As shown in Figure 1, the increase of CH_4 concentrations from 2 to 16% resulted in a 1.1–2.5 fold increase in the CH_4 oxidation rate measured at different depths in the column. A similar value (2.3 – fold) was observed in forest cambisoil, where the measured CH_4 concentration varied from 25 to 200 ppm [47]. The reason for the increase in CH_4 oxidation capacity can be partly explained by the fact that higher populations of CH_4 oxidation bacteria are achieved with the presence of higher CH_4 concentrations, leading to more CH_4 consumed.

Oxygen concentration

Another limiting factor influencing the methane oxidation process is O_2 supply. Pawłowska and Stępniewski [48] investigated the effect of oxygen concentration on methanotrophic activity in sand material. It was found that the CH_4 oxidation rate almost linearly increased when O_2 concentrations increased from 2.5 to 15%, followed by a slow increase approaching to a constant value (Figure 2). Similar result was observed by Schnell and King [49] who investigated the effect of O_2 concentration ranging from the atmospheric level to 0.2% (v/v) on the methanotrophic activity in forest soils. Results from these studies also indicate that the O_2 dependency of the CH_4 oxidation rate can be described by Michaelis-Menten reaction.

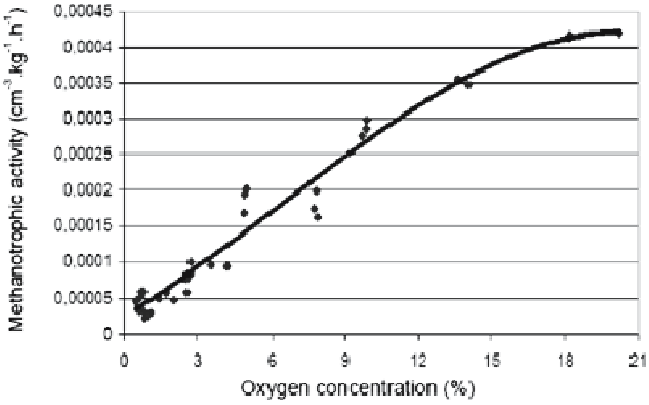

Fig. 2. The influence of O_2 concentration on the methanotropic activity in sand material, measured as a rate of CH_4 consumption [48]

It is important to note that the fact that the methanotrophic bacteria recognized are aerobic, does not indicate the methanotrophic abilities cannot occur in anaerobic conditions. There are numerous reports that have observed the methane oxidation activity at the bottom part of landfills cover or simulated landfills cover where O_2 concentration is very low. In fact, the phenomena of anaerobic oxidation of methane have been widely observed in CH_4 – rich bearing marine sediments [50–51]. The SO_4^{-2}–CH_4 interface and reaction is recognized as the fundamental mechanism of the CH_4 oxidation under anaerobic conditions. The SO_4^{-2}–CH_4 interface is a thin interval at the base of the SO_4^{-2} reduction zone that separates SO_4^{-2} – containing sediments above from CH_4 – rich sediments below [50–51]. During anaerobic oxidation of methane, CH_4 and SO_4^{-2} are consumed at the interface, leading to the production of HCO_3^{-1} and HS^-. The net reaction for the anaerobic oxidation of methane is formulated as the following equation [51]:

$$CH_4 + SO_4^{-2} \xrightarrow{\text{microorganisms}} HCO_3^{-1} + HS^{-1} + H_2O \quad (2)$$

Although numerous studies have examined the anaerobic oxidation of methane in marine environments, few have studied this activity in landfill cover. Recent studies have observed the presence of anaerobic oxidation of methane in drained peat and automorphic – sodpodzol soils [52] and landfill – leachate plume [53], suggesting that this process could occur in landfill cover.

Cover materials

Land cover materials used are of particular importance for landfill CH_4 oxidation systems. Previous studies have revealed that the type and physical-chemical properties of cover materials (e.g. particle size, porosity, moisture, and organic matter content) have a multi--dimensional effect on gas transfer and distribution, methane and oxygen availability, methanotrophs community structure and population, and nutrients supply [18, 54–55]. He et al. [54] investigated the CH_4 oxidation capacities and microbial community structures for two types of cover materials: a stabilized waste and an ordinary landfill cover material

(clay soil). It was found that type II methanotrophs were more abundant in the waste relative to the clay soil, while type I methanotrophes were predominant in the clay soil. Results from the study also suggest that the waste favours the development and growth of methanotrophs in comparison with the clay soil.

Pawłowska et al. [20] carried out an examination of methanotrophic performance on four types of mineral materials with different grain size. Results from their study showed that the grain size of materials had an influence on CH_4, O_2 and CO_2 profiles, water and organic carbon content, and redox profiles. The maximum value of methane oxidation capacity (227.4 ± 10.6 dm^3 m^{-2} d^{-1}) was achieved for the coarse sand material with grain size ranging from 0.5 to 1.0 mm. Further increase or decrease of the grain size resulted in reduced methane oxidation capacity. Gebert et al. [45] pointed out that the methane oxidation performance in landfill cover is governed by the share of pores available for gas transport, if other environmental variables (e.g. pH and nutrients) are not limiting. The authors conducted diffusion tests to investigate the effect of air-filled porosity of cover soil and degree of compaction on diffusivity and methane oxidation efficiency. It was suggested that soils used as methane-oxidizing cover material need to maintain an air-filled porosity of at least 14 vol. %.

Water content of cover soils influences the methanotrophic process, via modification of the conditions for methanotrophs growth and the effect on gas diffusion. Excessive water content can decrease the CH_4 oxidizing capacity of landfill cover soils; gas diffusion is limited when the soil pores are water saturated. On the other hand, insufficient moisture content can also lead to the decrease in the oxidation capacity, presumably due to the response to water stress, which will result in lower microbial activities. Whalen et al. [21] investigated the influence of water contents in the range of 30–50% (v/v) on methanotrophic activity. The optimum moisture content for forest soils was observed in the range of 21– 27% of total water retention, whereas the optimum for flooded soil was about 50%. Einola et al. [56] examined the responses of methane oxidation to temperature and water content in cover soil of a boreal landfill. They found that the CH_4 oxidation response to water content varied largely with temperature: at 1–6°C, CH_4 consumption increased with water content (33–67% water-holding capacity), while at 12–19°C the response trend was curvilinear with peak value at 50% water-holding capacity.

The optimal pH value for methanotrophs growth is in the range of between 6 and 8 [57–58]. An increase of pH value caused by the liming of acid soils (pH increase from 3.6 to 4.7) had no visible effect on methanotrophic activity at the atmospheric CH_4 level [59]. It can be concluded that the pH of the landfill cover soil is not a limiting factor for methane oxidation process as it generally varies slightly at around 7 [55].

Climatic conditions

Temperature has a significant effect on the methanotrophic activity, especially when the process is not limited by gas diffusion. An exponential increase in CH_4 oxidation rate was observed in response to temperatures ranging from 4–30°C [18]. Further increase in temperature leads to rapidly declined CH_4 oxidation rate. The temperature effect can be described by a parameter referring to the Van't Hoff Q_{10} temperature coefficient. When the Q_{10} value is below 2, the processes of methane oxidation is limited by diffusion. Conversely, the process is determined by biochemical factors.

Table 2. Q_{10} coefficients for the methane oxidation process in sand material [48]

Range of temperature change (°C)	Methanotrophic activity change ($cm^3 kg^{-1} s^{-1}$)	Temperature coefficient of Van't Hoff Q_{10}
7–14	4.8	7.3
14–21	2	2.7
14–7	1.84	2.3
21–14	1.72	2.1

The results presented in Table 2 show different behaviour of the methanotrophic activity during the increase and decrease of temperature [48]. These Q_{10} values were higher than the temperature coefficient measured in landfill cover soils by other authors. Whalen and Reeburgh [60] found Q_{10} equal to 1.9 (at the temperature range from 5°C to 26°C), indicating that the process of methane oxidation was limited by diffusion. The examined material in their study was heterogeneous soils with different grain sizes. While the sand materials examined by Pawłowska and Stępniewski et al. [48] had homogenous granulometric composition (without silt and clay fractions), the diffusion limitation was not observed.

Vegetation

Plants are known to play a considerable role in CH4 oxidation process occurring in landfill. Several studies have unanimously showed that the type of plant has a significant influence on methanotrophs populations (both type I and type II) and methane oxidation potential [61]. In a study [61] comparing the effect of four different plants (Miscanthus, poplar, grass, alfalfa–grass mixture) and an unplanted control, the alfalfa–grass mixture cover was shown to have the best performance, with a high relative abundance of Methylocystis. Wang et al. [19] investigated the effect of landfill vegetation of a plant (Chenopodium album L, tolerant to high concentrations of landfill gas) on the methane oxidation potential and bacterial community in the presence and absence of landfill gas. The co-presence of the plant and landfill gas was found to significantly enhance the population of methanotrophic bacteria and their methane oxidation potential. The study also revealed that there were interactive effects of landfill gas and vegetation on methanotrophic bacterial activity and community composition.

Several mechanisms have been proposed to explain the positive effect of plant vegetation on landfill CH_4 oxidation activities [19, 61–62]. First, plant vegetation leads to the form of rhizosphere, a soil zone that surrounds and is influenced by the roots of plants, creating a favourable habitat for methane oxidizing bacteria. Second, the spread of plant roots loosens the soil structure, and thus benefits landfill gas diffusion and facilitates the transport of oxygen from the atmosphere. Another possibility is that plant root exudates serve as selective substrates for methanotrophic bacteria and promote their growth.

KINETICS OF METHANE OXIDATION

Studies of methane oxidation kinetics can not only provide the information on how fast the methanotrophic reaction occurs, but also allow for the potential of methane oxidation

to be evaluated. In most cases, kinetics of the methane oxidation can be described by Michaelis-Menten equation, which is given as:

$$V = \frac{V_{\max}}{1 + K_M / C} \tag{3}$$

where V ($m^3 \cdot m^{-3} \cdot s^{-1}$) is the actual methane oxidation rate, V_{max} ($m^3 \cdot m^{-3} \cdot s^{-1}$) is the maximum methane oxidation rate, K_M (%) is the Michaelis constant for CH_4, and C (%) is the CH_4 concentration.

The kinetics parameter of V_{max} can be used to indicate the capacity of the methane oxidation. The half-saturation constants, K_M, can characterize the affinity (reciprocal of K_M) of methanotrophs to CH_4; a high K_M value indicates a poor affinity (reciprocal of K_M) of methanotrophs to CH_4. Based on the difference in K_M [63], the CH_4 oxidation and CH_4-consuming bacteria are grouped into two distinct forms. The first form occurs at low CH_4 concentrations (atmosphere-level), commonly known as high affinity oxidation (low capacity CH_4 oxidizer) [64]. The second form is typical of those encountered in landfill cover soils where methanotrophs oxidize high CH_4 concentrations, known as low affinity (high capacity CH_4 oxidizer) [64].

Table 3 summarizes the results of several kinetics studies on methane oxidation in landfill cover soils and in materials tested. For comparison, the kinetic characteristics of the methane oxidation under natural conditions with the atmospheric level of methane concentration are also listed.

Table 3. Kinetic parameters of the methane oxidation

Materialexamined	CH_4 concentration (vol.%)	V_{max} ($cm^3 \cdot kg^{-1} \cdot s^{-1}$)	K_M (%)	Source
Sand material (co1umn experiment)	1–16	$1.1 \cdot 10^{-4}$– $8.3 \cdot 10^{-4}$	0.6–2.9	[8]
Landfillcoversoil	$1.7 \cdot 10^{-4}$–1.0	$0.88–1.09 \cdot 10^{-3}$ [a]	0.18–0.7	[21]
Landfillcovertopsoil	$1.6 \cdot 10^{-2}$–8.0	$4.65 \cdot 10^{-3}$	2.54	[65]
Loam from Landfillcover	<10	$4.8 \cdot 10^{-3}$–$6.2 \cdot 10^{-3}$	0.75	[35]
Coarse sand soi1 from landfiII cover	0.05–5.0	$6.2 \cdot 10^{-3} \pm 0.36 \cdot 10^{-3}$	0.6–2.41	[64]
C1ay 1ayer in biofilter	0.2–10	$1.1 \cdot 10^{-2}$	1.2	[66]
Sand loamy soil (co1umn experiment)	<3	$1.5 \cdot 10^{-3}$–$1.7 \cdot 10^{-2}$	0.17–0.58	[36]
Forestcambisol	$0.2 \cdot 10^{-5}$–0.03	$2.2 \cdot 10^{-5}$ [a]	$2.2 \cdot 10^{-3}$	[47]
Bogsoil in Alaskan	$1.7 \cdot 10^{-4}$–0.1	$1.48 \cdot 10^{-3}$	0.084	[60]
Forestsoil in Alaskan		$4.9 \cdot 10^{-6}$–$56.8 \cdot 10^{-6}$	$2.9 \cdot 10^{-3}$–$9.9 \cdot 10^{-3}$	

The largest values of methanotrophic activity (V_{max}) oscillate at the scale of two orders of magnitude (Table 3), depending on CH_4 concentration and type of cover material. The K_M values calculated for the methane oxidation exposed to high CH_4 concentration

in landfills and simulated biofilters ranges from 0.17% up to 2.9% (v/v), and are two to three orders of magnitude greater as compared to the methane oxidation process exposed to the atmospheric level of methane concentration. For example, the K_M values measured in the box and forest soils ranged from 2.2·10–3 to 9.9·10–3 % (v/v), while the K_M values measured in the sand material simulated by column experiment had a perk value of 2.9% (Table 3).

It should be mentioned that the Michaelis-Menten equation cannot universally describe the kinetics of the methane oxidation. According to Bender and Conrad [47], and Streese and Stegmann [67], the kinetics of methane oxidation follows first order reaction, when the methane concentration is below substrate saturation level.

CONCLUSION

Microbial methane oxidation is a promising way to control methane emission from landfill, but still having significant undeveloped potential. The landfill methane oxidation capacity has been found to be affected by a range of intrinsic and extrinsic factors, such as landfill gas flux and oxygen availability, type and properties of cover soil, ambient conditions and landfill vegetation. An understanding of how these factors influence the performance of methane oxidation in landfill cover allows for this undeveloped potential to be better exploited. Different from climatic conditions and landfill gas flux and oxygen availability, which are not easy to be artificially managed, landfill cover material and associated physical-chemical features can be effectively controlled. An in-depth understanding of the effect of properties of cover materials and their underlying effect mechanisms appears to be needed to optimize the potential of landfill methane oxidation. In addition, further research needs to be conducted on the kinetics of the landfill methane oxidation to develop design requirements for an in situ application of this approach.

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