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Methane Emission Mitigation from Landfill by Microbial Oxidation in Landfill Cover

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Abstract. Landfill gas is one of the major anthropogenic sources of methane that has global warming potential over 20 times greater than carbon dioxide. Microbial methane oxidation has been universally observed in different types of soils including landfill cover soil. In this work, the methane oxidation activities in landfill cover and the underlying kinetics were described. The influence of methane and oxygen concentrations, cover materials, temperature, water content and pH on the methane oxidation was discussed. It can be suggested that the microorganism-based methane oxidation is one of the most efficient approaches to mitigate the residue methane emission in landfill to the atmosphere.

Keywords: greenhouse effects, methane emission, landfill cover, methane reduction.

1. Introduction

Methane (CH_4) is a greenhouse gas that can remain persistently in the atmosphere for approximately 9-15 years. According to Environmental Protection Agency of USA [1], methane is over 20 times more effective in trapping heat in the atmosphere than carbon dioxide (CO_2) over a 100-year period. Although the concentration of CH_4 in the atmosphere is rather low (about 1.8 ppm), its current contribution to global warming reach as much as 15 % [2]. More unfavourably, this contribution is believed to remain escalating as a result of a growing CH_4 emission to the atmosphere. Unarguably, anthropogenic activities are the main cause of the increasing CH_4 level. Human-influenced sources include, but not limited to, landfills, natural gas and petroleum systems, agricultural activities, coal mining, stationary and mobile combustion, wastewater treatment, and certain industrial process.

The biogas emitted from landfills is one of the major sources of methane, accounting for approximately 24-30 % of total anthropogenic methane emissions [3]. Therefore, reduction of its emission to the atmosphere is of great importance for mitigation of climate change. One win-win strategy is to use the landfill gas as substitute fuel for heat or electricity generation. This strategy has been widely recognized and receiving an increasing attention, since its successful practice can not only lead to a net mitigation of the greenhouse gas, but could ease our reliance on the limited fossil fuel.

In general, the calorific value of the landfill gas ranges between 18 and 25 MJ/m^3 [4], depending on how much CH_4 it contains. For comparison purposes, the heat value of the natural gas used in Poland is 31.0 MJ/m^3, while the heat value of pure CH_4 is 35.9 MJ/m^3. Therefore, it can be indicative that the biogas has the potential to allow its energetic quality to be enhanced, and to make it comparable with fossil fuel. A simple option towards this purpose is to upgrade the biogas by removal of CO_2 from the biogas discharged. However, this option has no economical feasibility when CO_2 accounts for major volume share in the gas mixture.

There are numerous scenarios under which utilization of the biogas for energy generation can not be available, technically or economically. For example, some old landfills were not equipped with gas

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collection system; the abandoned landfills still emit more or less methane-dilute biogas. Even in modern landfills, the biogas produced can not be collected sufficiently, primarily limited by the gas collection system, in particular the number of gas wells. This is demonstrative by the study evaluating the amounts of the methane collected and lost for 25 landfills in California [2]; the result found the methane loss amount was approximately two times greater than the collected methane amount on the basis of per tonne of MSW. In these cases that the utilization of the biogas is not accessible, how to control and mitigate the undesirable emission of the gas to the atmosphere needs to be addressed.

A variety of field and laboratory studies have confirmed that soils, precisely the methanotrphic microorganisms living in soils, have the ability to oxidize CH_4 to CO_2 [5-23]. The underlying bio-reaction pattern can be simplified as [24]

$$CH_4 + O_2 \xrightarrow{\text{Methanotrphic microorganisms}} CO_2 + 2H_2O\,.$$

This microorganism-based methane oxidation has been observed in different types of soils and under different climatic conditions [20-23]. Similar activity has also been observed in ocean and lake sediments [25-28]. Since the biological methane oxidation has been universally observed, particularly in the soil system, it is possible to employ and engineer this activity in landfill to reduce the undesirable methane emission. Currently, this approach is experienced in two processes: by use of special biofilters [5-11], or by forming a special soil bio-cover layer [11-18].

The methane oxidation in landfill cover could be effective for the mitigation of the methane generated in landfilled waste. Nozhevnikova et al. [29] estimated that in favourable conditions about 70 % of methane generated in the waste layer could be oxidized to CO_2. Compared to such high methane oxidation efficiency, studies carried out by Pawłowska et al. [13-17] showed lower methane oxidation abilities measured in sand and gravel layers under simulated landfill condition. These simulated layers allowed 57 % of methane to be oxidized. The efficiency of methane oxidization is affected by the methane production rate in waste and oxidation rate in the cover layer. The rate of methane emission from deposited waste surface can reach the value up to 1613 $dm^3m^{-2}d^{-1}$ [30]. The maximum values of methane oxidation capacity measured in bed layer (60-80 cm depth) at laboratory and field scale ranged from 200 to 400 g $m^{-2}d^{-1}$ [31-33]. The methane oxidation in cover layer does not ensure complete methane reduction in the case of an intensive methanogenesis process. Therefore, an application of biochemical oxidation process for methane reduction can be applied to reduction residual methane emission from landfill. The aim of this work was to summarize the performances and characteristics of the methane oxidation in landfill cover. Specifically, the factors influencing the methanotrphic activities were discussed.

2. kinetics of the methane oxidation

Study on methanotrophic kinetics can not only provide the information on how fast the methanotrophic reaction occurs, but also allow the potential methanotrophic activity to be evaluated. In most cases, kinetic of the methane oxidatione can be described by Michaelis-Menten equation [9,16, 31-38], which can be given as

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

where $\boldsymbol{V}$ ($m^3.m^{-3}.\ s^{-1}$) is the actual methane oxidation rate, $\boldsymbol{V_{max}}$ ($m^3.m^{-3}.\ s^{-1}$) is the maximum methane oxidation rate, $\boldsymbol{K_M}$ (%) is Michaelis constant for CH_4, and $\boldsymbol{C}$ (%) is the CH_4 concentration. The kinetics parameter, $\boldsymbol{V_{max}}$, can be used to indicate the capacity of the methane oxidation. The half-saturation constants, $\boldsymbol{K_M}$, can be used to 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.

Table 1. Kinetic parameters of the methane oxidation

Material examined	Range of CH_4 concentration / vol. %	V_{max} / $cm^3\,kg^{-1}\,s^{-1}$	K_M/ %	Source
Landfill cover soil	$1.7 \cdot 10^{-4} – 1.0$	$0.88 – 1.09 \cdot 10^{-3\,a}$	0.18 – 0.7	[34]
Landfill cover topsoil	$1.6 \cdot 10^{-2} – 8.0$	$4.65 \cdot 10^{-3}$	2.54	[35]

Loam from Landfill cover	<10	$4.8 \cdot 10^{-3} – 6.2 \cdot 10^{-3}$	0.75	[32]
Coarse sand soi1 from landfiIl cover	0.05 – 5.0	$6.2 \cdot 10^{-3} \pm 0.36 \cdot 10^{-3}$	0.6 – 2.41	[36]
C1ay 1ayer in biofilter	0.2 – 10	$1.1 \cdot 10^{-2}$	1.2	[9]
Sand material (co1umn experiment)	1-16	$1.1 \cdot 10^{-4} – 8.3 \cdot 10^{-4}$	0.6 – 2.9	[16]
Sand loamy soil (co1umn experiment)	<3	$1.5 \cdot 10^{-3} – 1.7 \cdot 10^{-2}$	0.17 – 0.58	[31]
Forest cambisol	$0.2 \cdot 10^{-5} – 0.03$	$2.2 \cdot 10^{-5}$ [a]	$2.2 \cdot 10^{-3}$	[37]
Bog soil in Alaskan	$1.7 \cdot 10^{-4} – 0.1$	$1.48 \cdot 10^{-3}$	0.084	[38]
Forest soil in Alaskan		$4.9 \cdot 10^{-6} – 56.8 \cdot 10^{-6}$	$2.9 \cdot 10^{-3} – 9.9 \cdot 10^{-3}$	

[a] Per kg of wet weight

Table 1 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 were also listed in Table 1. The largest values of methanotrophic activity (V_{max}) oscillated at the scale of two orders of magnitude (Table 1), varying with the CH_4 concentration and the type of material. The K_M values calculated for the methane oxidation exposed to high CH_4 concentration in landfills and simulated biofilters ranged from 0.17 % up to 2.9 % (v/v), 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 $2.2 \cdot 10^{-3}$ to $9.9 \cdot 10^{-3}$ % (v/v) [38], while the $K_M$values measured in the sand material simulated by column experiment had a perk value of 2.9 % [16].

It should be mentioned that the Michaelis-Menten equation can not universally describe the kinetics of the methane oxidation. According to Bender and Conrad [37] and Streese and Stegmann [39], the kinetics of methane oxidation follows first order reaction, when the methane concentration is below substrate saturation level.

3. Factors influencing methane oxidation activity

3.1. Methane concentration

Various laboratory experiments have shown that the rate of methane oxidation grows with the increase of methane concentration, up to a certain constant level. Pawłowska and Stepniewski [14,16] studied the methanotrophic activity in the profile of a simulated landfill cover as a function CH_4 concentration. As shown in Fig.1, CH_4 concentration had a significant influence of on the methanotrophic activity. The increase of CH_4 concentrations from 2 to 16 % leaded to 1.1 - 2.5 fold increase of the methanotrophic activity measured at different depths in the column. A similar value (2.3-fold) in the methanotrophic activity increase was observed in forest cambisoil, where the measured CH_4 concentration varied from 25 to 200 ppm [37].

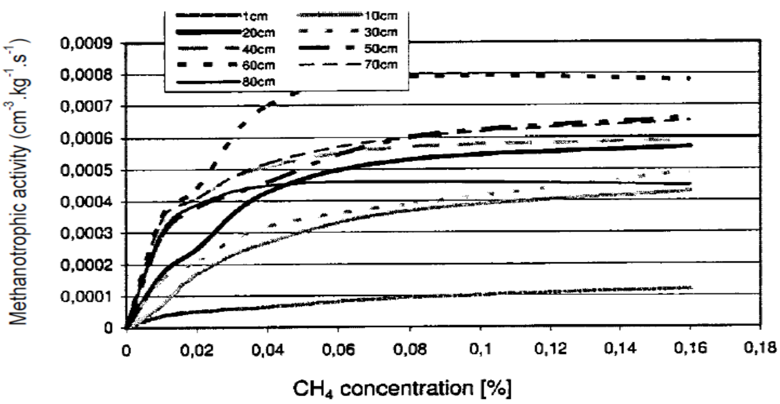

Fig. 1. The CH_4 concentration dependence of methanotrophic activity observed in sand materials taken from 9 different depths in the column [16]

3.2. Oxygen concentration

Oxygen accessibility is a significant factor influencing the methane oxidation process. All types of methane-oxidizing bacteria are aerobic, and prefer O_2 concentrations lower than the atmospheric level [40]. Schnell and King [41] noticed that the decrease of oxygen concentration from the atmospheric level to 0.2 % (v/v) caused the decrease of methanotrophic activity in forest soils. Similar result was found by Pawłowska and Stępniewski [15] who experimentally investigated the effect of oxygen concentration on methanotrophic activity in sand material (Figure 2). As shown in Figure 2, the CH_4 oxidation rate experienced a gradual increase when the O_2 concentrations increased from 2.5% to 15 %, followed by a slow approach to a constant oxidation rate. The study additionally observed that the O_2 dependency of the CH_4 oxidation rate can also be described by Michaelis-Menten reaction.

It is interesting to mention that, the fact that the methanotrophic bacteria are aerobic, does not indicate the methanotrophic abilities not to be allowed. There are large amounts of reports that methane oxidation has been observed under anaerobic conditions at the bottom part of landfills or simulated landfills. This could be evident by the findings of the previous study comparing the different methanotrophic abilities at different depths in simulated landfill (Fig. 1) [16]. The result found that the largest methanotrophic ability was at the 60 cm depth close the bottom of the column. According to Dalton and Hocknull [42], this may result from the activity of the bacteria reducing the sulphates or other compounds. The findings of the methanotrophic ability under anaerobic conditions suggest that a further investigation into the biological mechanism and conversion pathways remains necessary.

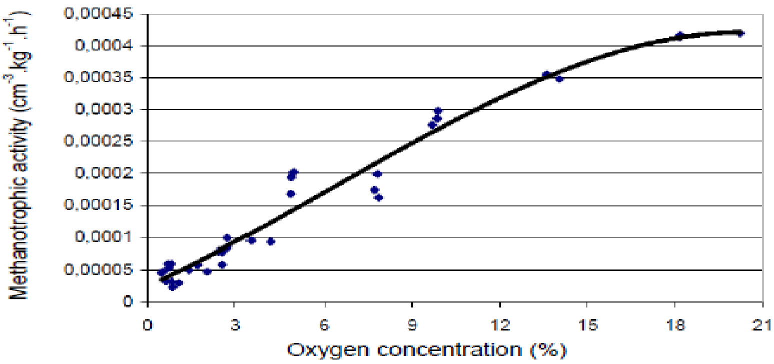

Fig. 2. The influence of O_2 concentration on the methanotropic activity in sand material, measured as a rate of CH4 consumption [15]

3.3. Properties of cover material

Grain size of the material used influences the gas transfer (substrates inflow and products outflow). Granulometric composition determines the porosity that influences the gas diffusion coefficients, as well as the specific surface of the material on which methanotrophs population grows. Pawłowska et al. [13] carried out an examination on four types of mineral materials with different grain size. Results showed that, the grain size of material 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, equal to 227.4 ± 10.6 $dm^3\ m^{-2}\ d^{-1}$, was achieved in the coarse sand material with 0.5-1.0 mm fraction. Further increase or decrease of the grain size resulted in reduction of methane oxidation capacity. Possible reason is associated with limitation of the specific surface area of the particles available for bacteria colonisation (in the case of the grain size increase), or with lowering of gas diffusion coefficient (in the case of the grain size decrease).

3.4. Water content.

Soil moisture content influences the methanotrophic process, directly via modification of the conditions for methanotrophs growth, and indirectly via the impact on gas diffusion. excessive moisture content levels can decrease the CH_4 oxidizing capacity of the landfill cover soil, due to the limiting of the gas diffusion in the soil system; Gas diffusion is limited when the soil pores are water saturated. On the other hand, insufficient moisture content can also decrease the oxidation capacity of the landfill cover soil, presumably due to the response to water stress, which will result in lower microbial activities. Whalen at al [34] investigated the influence of moisture content in range of 30-50 % (v/v) on methanotrophic activity. The optimum moisture content for forest soils was observed in range of 21-27 % of total water retention, whereas the optimum for flooded soil was about 50 %. The values of optimum moisture content for soil taken from a landfill cover varied from 11 to 20 %, equal to half of the total water retention of examined soil [38, 43-44].

3.5. Temperature

The temperature has an effect on the methanotrophic activity, especially when the process is not limited by gas diffusion. A parameter referring to the Van't Hoff Q_{10} temperature coefficient is generally used to characterize the temperature effect. 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. Temperature coefficients Q_{IO} for the methane oxidation process in sand material obtained in laboratory experiment [15].

Range of the 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 Q_{10} coefficient calculated for a temperature increase from 20 to 30°C was equal to 4.6, but Q_{10} calculated for the temperature decrease from 20 to l0 °C was only 2.3 (calculated by Van't Hoff equation). The results presented in Table 2 show different behavior of the methanotrophic activity during the increase and decrease of temperature [15]. These Q_{10} values were higher than the temperature coefficient measured in landfill cover soils by other authors. Whalen et al. [38] 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 et al. [15] had homogenous granulometric composition (without silt and clay fractions), the diffusion limitation was not observed.

3.6. The value of pH

The methane oxidation process was observed in a broad pH range, from pH< 4 in a sand soil to pH >9 in a bog soil. The optimal pH for methanotrophs growth is between 6 and 8 [20, 45-46]. The pH value of soil had an indirect impact on the methane oxidation rate by a reaction accompanying, for example, a release of toxic Al3+. 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 [47].

4. Conclusion

Application of the microbial oxidation is a promising way to control the methane emission from landfill. The microbial oxidation in landfill cover can be enhanced via appropriate measures and operations for the landfill cover soils. The kinetic of the methane oxidation in landfill cover can be described by Michaelis-Menten equation. The process have been found to be affected by granulometric composition of soil cover, pH, temperature, moisture content, methane concentration and oxygen accessibility. Different from other factors influencing the methane oxidation, moisture content of the landfill cover soil is easily accessible in practice to engineered control. While the moisture content has substantial impacts on the microbial oxidation, more attention should be given to this factor.

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