Journal of Hazardous Materials 191 (2011) 228–233

Contents lists available at ScienceDirect

Journal of Hazardous Materials

journal homepage: www.elsevier.com/locate/jhazmat

Can a breathing biocover system enhance methane emission reduction from landfill?

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ARTICLE INFO

Article history:
Received 26 January 2011
Received in revised form 24 March 2011
Accepted 16 April 2011
Available online 22 April 2011

Keywords:
Passive air venting
Biocover
Landfill
Methane oxidation
Aerothermodynamic

ABSTRACT

Based on the aerothermodynamic principles, a kind of breathing biocover system was designed to enhance O_2 supply efficiency and methane (CH_4) oxidation capacity. The research showed that O_2 concentration (v/v) considerably increased throughout whole profiles of the microcosm (1 m) equipped with passive air venting system (MPAVS). When the simulated landfill gas SLFG flow was 771 g m^{-3} d^{-1} and 1028 g m^{-3} d^{-1}, the O_2 concentration in MPAVS increased gradually and tended to be stable at the atmospheric level after 10 days. The CH_4 oxidation rate was 100% when the SLFG flow rate was no more than 1285 g m^{-3} d^{-1}, which also was confirmed by the mass balance calculations. The breathing biocover system with *in situ* self-oxygen supply can address the problem of O_2 insufficient in conventional landfill covers and/or biocovers. The proposed system presents high potential for improving CH_4 emission reduction in landfills.

1. Introduction

The Intergovernmental Panel on Climate Change 2007 defines methane (CH_4) as an important greenhouse gas with a global warming potential 25 times greater than carbon dioxide. Its current contribution potential to global warming is estimated at 15% and continues to escalate [1]. Landfill is considered the fourth largest anthropogenic source of CH_4 worldwide. It contributes 20–70 Tg of CH_4 to annual global CH_4 emissions [2]. In 2005, CH_4 accounted for 25% of anthropogenic CH_4 emissions in the US [3], while that in China was 11%, and is expected to reach 12.6% by 2010 [4]. CH_4 production and emission from landfills in Beijing accounted for 39.5% of the total CH_4 emissions in the city in 2005 – a value considerably higher than those generated from other pollutants because of the large number of landfills (>88%) used for municipal solid waste treatment [5].

Landfill CH_4 emissions require urgent mitigation to lower the total CH_4 concentration in the atmosphere [6]. A possible solution is the oxidation of CH_4 by bacteria residing in soil, which represents an important terrestrial CH_4 sink. A number of studies have focused on CH_4 biotransformation by indigenous microorganisms, and numerous methods such as biofiltration [6–8] and biocover technology [9–14] have been developed. Recent, specialized landfill biofilter designs for optimizing CH_4 oxidation have demonstrated a potential to mitigate landfill CH_4 emissions as high as 1900 g m^{-3} d^{-1} [15].

The application of landfill cover layers for CH_4 mitigation is influenced by two key parameters: temperature and available O_2 concentration [16]. To reduce problems related to O_2 diffusion in landfill covers and open biofilters, many researches prefer using multi-layer beds [15,17], composts [18], soil [19], or a mixture of compost and soil [20]. These porous materials can improve O_2 distribution but do not overcome the problem of insufficient O_2 concentration because downward diffusion capacity of atmospheric O_2 is limited: generally, an oxygenated zone of no more than 0.60 m of the top cover is observed [21–24]. Though forced-aerated cover can overcome the bottleneck, the continuous air supplied by compressor pump means more energy should be introduced. Therefore, a continuous O_2 supply through passive diffusion without energy consuming, would be an attractive approach.

Semi-aerobic landfill technology was developed more than 20 years ago in Japan [25]. It has been reported as an effective method for reducing *in situ* CH_4 emission. A semi-aerobic status is achieved through a continuous supply of fresh air sucked passively through the open end of a leachate collection pipe (diameter $\geq$ 0.6 m), with a temperature gradient created by spontaneous heating of microorganisms during biodegradation (Fig. 1). The constant supply of fresh air not only enhances biodegradation and reduces CH_4 production but also improves leachate quality [26]. The technology that has been proven and tested in numerous areas in Japan, also shows high feasibility in other countries, such as Malaysia, Iran, and China [27]. It has been well-established that a semi-aerobic landfill technology creates partial aeration at the bottom of the landfill and prevents

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doi:10.1016/j.jhazmat.2011.04.068

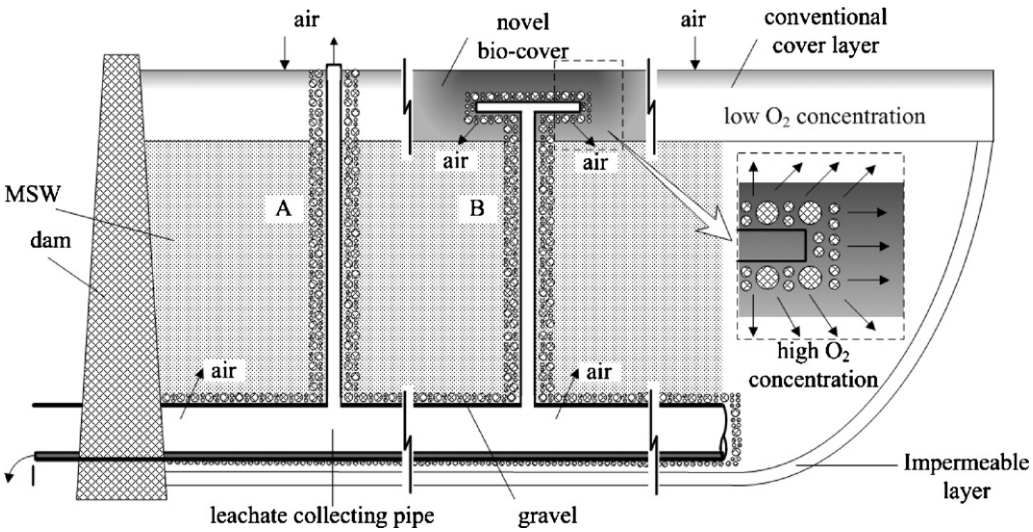

Fig. 1. A novel biocover in a semi-aerobic landfill.

methanogenic activity. However, it confines its further effect on upper layer because its capacity of air/O_2 upward transportation is very limited. Its effect to the activity of methanotrophs in cover layer is therefore negligible.

In this paper, we designed a kind of breathing bicover system including a passive air venting system (PAVS) using the aerothermodynamic principles that underlie semi-aerobic landfill to solve the problem of O_2 limitation. Its ability of enhancing O_2 supply efficiency and CH_4 transformation was investigated by lab-scale experiment and mass balance analyses.

2. Materials and methods

2.1. Experimental setup

Two sets of simulated biocover microcosms were used in the experiments. Each microcosm had a volume of 88.4 L (ø0.32 m × 1.10 m). The first breathing biocover system equipped with PAVS was labeled "microcosm with passive air venting system" (MPAVS). While the second one served as the control without PAVS was labeled microcosm of simulated conventional biocover system (MSCBS). Diagrams of the two microcosms are shown in Fig. 2.

PAVS comprises a bottom drainage pipe (i.d. = 0.057 m), a 1.50 m vertical venting pipe (i.d. = 0.05 m), and an air-distributing head wheel (i.d. = 0.20 m). The head wheel is made of a ring-shaped pipe and six spokes (i.d. = 0.02 m). Fig. 1 shows that the six spokes are all connected to the vertical venting pipe. Moreover, perforations were drilled evenly onto the spokes for air distribution. The head wheel and part of the vertical venting pipe (ca. 0.15 m) were placed inside the microcosm; the rest of the vertical venting pipe and the bottom drainage pipe were kept outside. All parts of the set-up are made of polyvinyl chloride.

At the bottom of the two microcosms, a ring-shaped air-distributing pipe with perforations was installed and connected to a pump that brought simulated landfill gas (SLFG) into the microcosms. A 0.10 m layer of gravel was placed on the top of the air-distributing pipe to ensure homogenous distribution of gas. This simulates the transport of landfill gas venting from the deposited refuse to the cover layer. Each microcosm had seven gas sampling ports with 0.15 m intervals at the sides and an outlet port on top. All these ports were sealed, except during sampling.

The two microcosms were filled with a mixture of yard waste compost and landfill cover soil (3:7, w/w). The yard waste compost was prepared from leaves mixed with manure and soil and allowed to mature for more than six months. The absence of odor confirmed compost maturity. The landfill cover soil was collected from the Zhuozhou municipal solid waste Landfill (Hebei Province, China). Before loading, the compost and cover soil were mixed to come up with a homogeneous material. The density, as well as the moisture and organic contents of the final filling biocover were 1.6 g cm^{-3}, 26.0%, and 41.5% tested by standard methods [28], respectively. The height of the loaded biocover material was 1.0 m.

2.2. Ventilation efficiency test for PAVS

In order to avoid the interference of air retention and physic-chemical reactions, the MPAVS was first filled with inert material (glass beads) and inert gas (N_2) instead of biocover materials and landfill gas, respectively. Gas supply (Q_{vp}) was controlled by pumping certain amount of N_2 (V_{in}, ranging from 0 to 2.8×10^{-4} m^3 s^{-1}) from the bottom drainage pipe of PAVS. The N_2 flow rate at the outlet port of the microcosm (V_{out}, m^3 s^{-1}) was measured with a gas flow meter (W-NK, China). Because the N_2 is neither consumed nor lost in microcosm since the filler is inert glass bead, the measured gas quantity at the outlet port is regarded as the effective quantity (Q_{bioc}) of gas filled into the microcosm through PAVS. The ventilation efficiency (η) of PAVS was calculated by Q_{vp} and Q_{bioc} as showed in Eq. (1).

$$\eta = \frac{Q_{bioc}}{Q_{vp}} \times 100\% = \frac{V_{out}}{V_{in}} \times 100\% \quad (1)$$

2.3. Aerothermodynamic test for PAVS

Determine of the aerothermodynamic relation between the temperature and passive venting rate is key for a passive venting system as it is impossible to measure its gas flow once it was applied to an actual landfill. But the measurement of temperature of an actual landfill is feasible.

The same setup as the above test was used. However, the controlled parameter is the temperature instead of the supplied gas. The vertical venting pipe outside of the microcosm was heated by a heating tape to simulate spontaneous heating by microbes during biodegradation of organic matter in an actual landfill. The temperature inside the vertical venting pipe (T_{inside}) was adjusted from 20 to 60 °C at intervals of 2–3 °C using a temperature controller (XMT122, Chinlt, China). The ambient temperature ($T_{ambient}$) was maintained

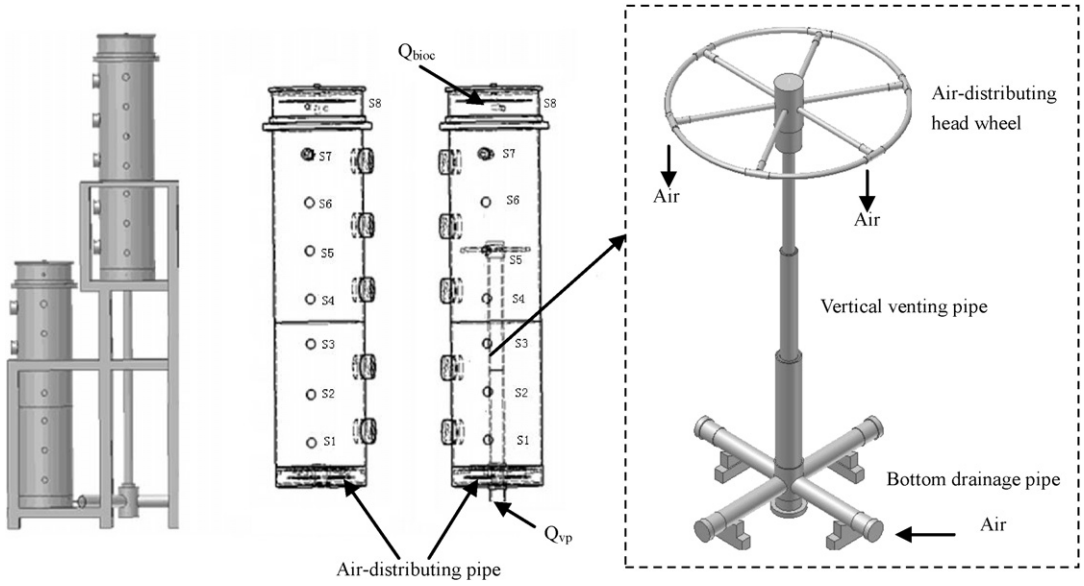

Fig. 2. Simulated biocover microcosms (Left, MSCBS; Right, MPAVS).

at 8 °C during the experiment. The temperature discrepancy (ΔT) of PAVS can therefore be calculated as

$$\Delta T = T_{\text{inside}} - 8 \tag{2}$$

Once the temperature raised, the fresh air was then passively sucked through the bottom drainage pipe and transported through venting pipe to the head wheel. The maximum air flow rate in the vertical venting pipe (V_{max}) was measured by a hot wire anemometer (Kanomax KA22, Japan) that placed at the center of the pipe. The relationship between V_{max} and ΔT was established from the experimental data, i.e. $f(\Delta T)$.

According to the basic model in hydromechanics, the average rate is equal to half of the V_{max} in a circular pipe filled at a steady flow rate. Then, Q_{vp} can be calculated (Eq. (3)) [29].

$$Q_{vp} = \frac{1}{2} V_{\max}(\Delta T) \cdot S_{vp} = \frac{1}{2} f(\Delta T) \cdot S_{vp} \tag{3}$$

where S_{vp} is the sectional area of the vertical venting pipe.

Finally, the Q_{bioc} was obtained by combing Eqs. (1) and (3):

$$Q_{bioc} = Q_{vp} \cdot \eta = \frac{1}{2} f(\Delta T) \cdot S_{vp} \cdot \eta \tag{4}$$

2.4. Effect of PAVS on O_2 supply and gas components in the microcosm

After the aerothermodynamic study, both microcosms were loaded with the biocver material as described above. Moreover, they also were loaded with SLFG (CH_4:CO_2 = 1:1, v/v) to simulate landfill gas interference from the deposited refuse below the cover layer. SLFG was pumped from gas cylinders through the air-distributing pipe at flow rates of 771, 1028, and 1285 $g\,m^{-3}\,d^{-1}$ respectively. In MPAVS, the vertical venting pipe of the PAVS was heated and the temperature was controlled at 50 ± 1 °C by a heating tap and a temperature controller to obtain passive air (O_2) supply driven by the aerothermal force. That means the gas sources in MPAVS included SLFG delivered through the pump, and fresh air passively sucked through the bottom drainage pipe. The air passed through the vertical venting pipe and finally diffused into the cover layer through the holes on the six spokes of the head wheel. As a control, the gas source of MSCBS was only from SLFG delivered through the pump. The test period of each SLFG flow rate was 15 days, and the entire experiment lasted 45 days.

Gases in different layers labeled S1 to S8 of both microcosms were sampled by syringe (1 mL) once a day. A gas chromatograph (Agilent 6890N) with a thermal conductivity detector was used to measure CH_4, O_2, and CO_2 in the gas samples. The detection program consisted of a carrier gas (H_2) flow of 30 mL min^{-1}, an oven temperature of 120 °C, and injector and detector temperatures of 160 °C.

2.5. Mass balance calculation of CH_4 oxidation efficiency

Because of the influence of supplied air in MPAVS, the SLFG concentration under a steady flux differ with that in MSCBS although the flow of SLFG was set at equivalent level in each test. Taking into account additional air from PAVS, the CH_4 oxidation efficiency had to be modified.

Assuming only physical diffusion of SLFG exists in MPAVS, the theoretical CH_4 concentration ($C_{outlet\cdot CH_4}$) (v/v) in the outlet can be calculated as

$$C_{outlet\cdot CH_4} = \frac{1/2 Q_{SLFG}}{(Q_{bioc} + Q_{SLFG})} \times 100\% \tag{5}$$

where Q_{SLFG} is the filling quantity of SLFG.

When biodegradation is considered (Eq. (6)) aside from physical diffusion, the reduction in gas quantity in MPAVS is equal to double that of the CH_4 quantity transformed. Therefore, the theoretical CH_4 quantity ($Q_{outlet\cdot CH_4}$) in the MPAVS outlet can be calculated by Eq. (7) [30]

$$CH_4(g) + 2O_2(g) \xrightarrow{MB} CO_2(g) + 2H_2O(l) \tag{6}$$

$$Q_{outlet\cdot CH_4} = \frac{1}{2} Q_{SLFG} - Q_{reaction\cdot CH_4} \tag{7}$$

where MB is methanotrophic bacteria and $Q_{reaction\cdot CH_4}$ is the quantity of CH_4 oxidized by methanotrophs.

Consequently, the theoretical $C_{outlet\cdot CH_4}$ can be calculated using

$$C_{outlet\cdot CH_4} = \frac{Q_{outlet\cdot CH_4}}{Q_{bioc} + Q_{SLFG} - 2Q_{reaction\cdot CH_4}} \times 100\% \tag{8}$$

A zero $C_{outlet\cdot CH_4}$ implies that CH_4 is completely oxidized by methanotrophs in the biocover.

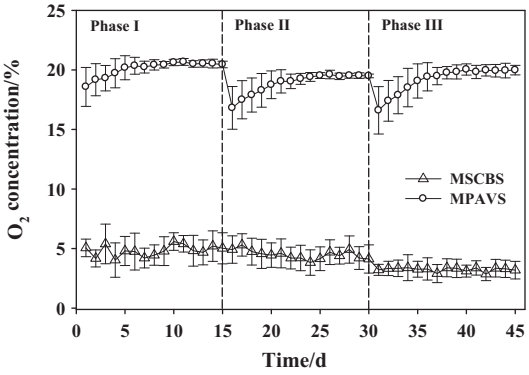

Fig. 3. O_2 concentration in microcosms at different SLFG flow rates (SLFG flow were 771, 1028, and 1285 g m^{-3} d^{-1} at phases I, II, and III, respectively).

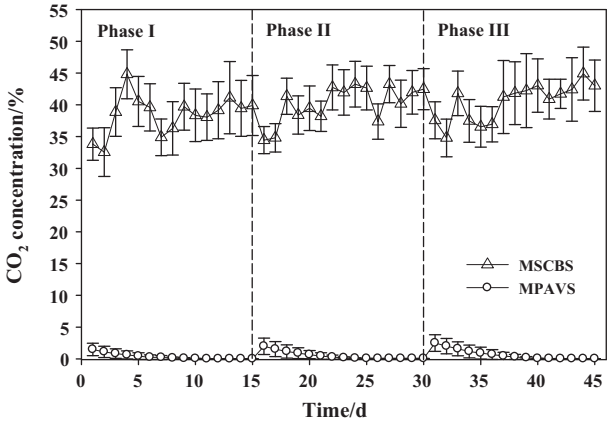

Fig. 4. CO_2 concentration in microcosms at different SLFG flow rates (SLFG flow were 771, 1028, and 1285 g m^{-3} d^{-1} at phases I, II, and III, respectively).

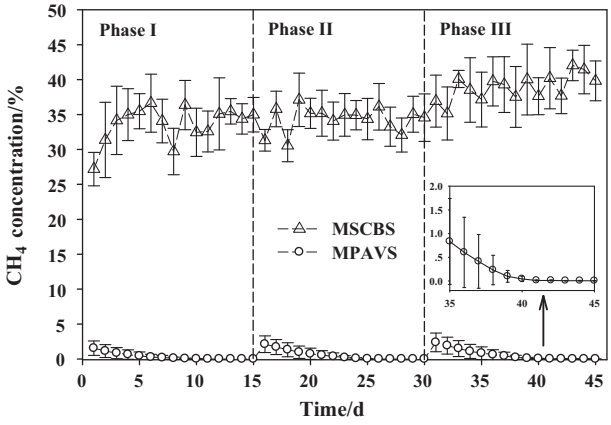

Fig. 5. CH_4 concentration in microcosms at different SLFG flow rates (SLFG flow were 771, 1028, and 1285 g m^{-3} d^{-1} at phases I, II, and III, respectively).

3. Results and discussion

3.1. Effect of O_2 supply by PAVS

With regard to the S1 and S2 are the neighboring areas of the SLFG inlet, the results of the two microcosms were presented by the average values of S3 to S8 (n = 6), and the detailed results of S1 and S2 are shown in Table 1. When SLFG flow was 771 g m^{-3} d^{-1} (phase I), the O_2 concentration in MPAVS gradually increased in the initial stage and stabilized after 10 days till the end of phase I. Namely, it increased from 18.6% to 20.5% during the 15 days (Fig. 3), almost near the atmospheric level. By contrast, the O_2 concentrations in MSCBS, ranging from 4.0% to 5.8%, were drastically lower than that in MPAVS at this phase. When SLFG flow increased to 1028 g m^{-3} d^{-1} (phase II), the variation trend of O_2 concentration in two microcosms remained similar to that at phase I. However, the O_2 concentration is slightly lower because of increased SLFG flow. In MPAVS, the O_2 concentration was 16.8% at the beginning of phase II, and gradually increased and finally stabilized at 19.5% till the end of phase II. The O_2 concentrations in MSCBS were again considerably lower than those in MPAVS in this phase. The highest level was no more than 5.3%. When SLFG flow increased to 1285 g m^{-3} d^{-1} (phase III), similar phenomena were observed like the two scenarios above. The only difference was that the average O_2 concentration slightly decreased because of high SLFG flow. For instance, O_2 concentration ranged from 16.6% to 19.9% in MPAVS, whereas it ranged from 2.8% to 3.4% in MSCBS.

The results demonstrated that the introduction of PAVS appreciably altered the aerobiotic situation in the landfill biocover. The use of compost resulted in high methanotrophic activity in the deeper active zones because of high porosity, which supports higher O_2 diffusion [31]. However, O_2 was considerably higher than the level of atmospheric diffusion (0.6–0.8 m) reported in other studies [20–23]. This indicates that PAVS became a second O_2 source for the biocover in addition to the natural atmospheric diffusion at the top layer. It is well documented that improved O_2 condition stimulates the growth of aerobic microbes such as methanotrophs [13]. Therefore, the presence of PAVS should encourage methanotroph activity throughout the whole profile (1 m in this test).

3.2. Effect of PAVS on gaseous components in the biocover microcosm

The effect of the PAVS on CH_4 and CO_2 in the microcosm was also studied. When SLFG flow was 771 g m^{-3} d^{-1}, the CO_2 concentrations rapidly decreased in the initial stage but remained stable at 0–1% after 10 days in MPAVS (Fig. 4). Correspondingly, the CO_2 concentrations in MSCBS were always higher than 35%, which are markedly higher than those in MPAVS. When SLFG flow increased to 1028 g m^{-3} d^{-1}, variations in the trend remained similar to those at the 771 g m^{-3} d^{-1}. The CO_2 concentrations decreased but stabilized at 0.2–1.1% in MPAVS. Meanwhile, the CO_2 concentrations in MSCBS ranged from 37% to 47%. When the SLFG flow increased to 1285 g m^{-3} d^{-1}, a trend in both microcosms were similar to those in the two previous scenarios. In MSCBS, the CO_2 concentration ranged from 36.4% to 47.7%, and ranged from 0.02% to 1.2% in MPAVS.

Correspondingly, the CH_4 concentration was also much lower in MPAVS than in MSCBS for equivalent layers. In the given SLFG flow rates of 771 and 1028 g m^{-3} d^{-1}, the CH_4 concentrations in MPAVS were below the detection level in later stages (Fig. 5). However, the trend differed when SLFG flow was further increased to 1285 g m^{-3} d^{-1}. CH_4 in SLFG of MPAVS was not completely transformed because the CH_4 concentrations at the outlet maintained at 0.2%–0.8%. By contrast, the CH_4 concentrations in MSCBS always maintained at 25%–45% in all three scenarios.

These results prove that the system can substantially overcome limited O_2 transport in biocover layers and considerably increase O_2 concentration throughout whole profiles. Thus, optimum conditions were achieved for efficient bio-transformation of CH_4.

3.3. CH_4 oxidation mass balance in MPAVS

As the results of above tests showed that the CO_2 and CH_4 concentrations were much lower in MPAVS than MSCBS for equivalent

Table 1
Results of ventilation efficiency test of PAVS.

No	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15
Q_{vp} (10^{-3} m^3 s^{-1})	0.0	1.2	2.6	5.2	6.8	9.6	11.2	13.6	15.6	17.4	19.4	22.8	23.6	26.2	27.8
Q_{bioc} (10^{-3} m^3 s^{-1})	0.00	0.18	0.42	0.77	1.06	1.47	1.65	2.05	2.38	2.70	3.02	3.48	3.61	3.98	4.26
η (%)	–	15.14	16.15	14.81	15.59	15.31	14.73	15.07	15.25	15.51	15.56	15.27	15.31	15.20	15.31

Table 2
Comparison between theoretical and measured value of CH_4 concentration in the MPAVS outlet.

Q_{SLFGt} (g m^{-3} d^{-1})	T_{inside} (°C)	ΔT (°C)	V_{max} (m s^{-1})	$C_{outlet\cdot CH_4}$ (%)		CH_4 oxidation efficiency (%)
				Theoretical value	Measured value	
771	50	35	0.46	0.85	0	100
1028	50	25	0.18	2.80	0	100
1285	50	30	0.32	1.98	0.83	60.6

layers, these attributed to two factors: (1) the high O_2 concentration throughout the whole profile of MPAVS stimulated the methanotrophs activity, with CH_4 becoming a limiting factor because of its efficient oxidation; and (2) the pumped CH_4 was diluted under the interference of supplied air (mainly as N_2 and O_2) in MPAVS. To address that question, the mass balance of CH_4 in MPAVS was therefore calculated and analyzed.

The results of the PAVS ventilation efficiency test are listed in Table 2. The ventilation efficiency (η) calculated from Q_{vp} and Q_{bioc} was 15.3%, indicating that 15.3% of air can be passively diffused into the biocover layer through PAVS.

Aerothermodynamic testing of PAVS resulted in a linear correlation between ΔT and V_{max} (Fig. 6).

$$V_{\max} = 0.0281 \cdot \Delta T - 0.5258 \quad R^2 = 0.9993 \tag{9}$$

Based on this equation, V_{max} at any ΔT can be calculated and used for further calculation to eventually obtain theoretical CH_4 concentrations in MPAVS under different SLFG flow rates. For example, when SLFG flow rates were set at 771, 1028, and 1285 g m^{-3} d^{-1}, the theoretical CH_4 concentrations in the MPAVS outlet were 0.85%, 2.80%, and 1.98%, respectively, considering only physical diffusion (Table 2). However, the actual CH_4 concentrations in the outlet of MPAVS were 0%, 0%, and 0.83%, respectively. Microbial analysis (data not shown) verified the assumption, which the difference between the measured and theoretical values was due to the biotransformation of CH_4 by methanotrophs. In particular, when SLFG flow was no more than 1028 g m^{-3} d^{-1}, the oxidation efficiency was 100%, indicating that when O_2 is no more the limiting factor in a system, microbial transformation is rapid and complete.

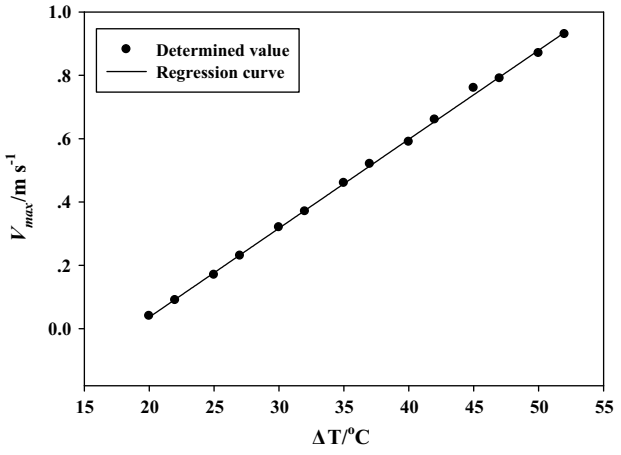

Fig. 6. Relationship between V and ΔT.

At an SLFG flow of 1285 g m^{-3} d^{-1}, $Q_{reaction\cdot CH_4}$ can be calculated using Eq. (10).

$$Q_{reaction\cdot CH_4} = \frac{1}{2} Q_{SLFG} - \frac{Q_{bioc} \cdot C_{outlet\cdot CH_4}}{1 - 2 \cdot C_{outlet\cdot CH_4}} \tag{10}$$

The result showed that the actual oxidation rate of CH_4 was 389 g m^{-3} d^{-1}, based on the measured $C_{outlet\cdot CH_4}$ (0.83%). As the pumped CH_4 flow in SLFG was 642 g m^{-3} d^{-1} (half of SLFG flow), the CH_4 oxidation efficiency was 60.6%.

CH_4 oxidation efficiency decreased when SLFG flow increased to 1285 g m^{-3} d^{-1}. A possible reason for this is that the residual time in the biocover of CH_4 was relatively short when the flow increased. Hence, the contact time for CH_4 and methanotrophs was insufficient for complete oxidation. The highest CH_4 oxidization efficiency (100%) in this study was obtained when the SLFG flow rate was 1028 g m^{-3} d^{-1}. The highest CH_4 oxidation rate was 610 g m^{-3} d^{-1}, lower than the reported value of 1900 g m^{-3} d^{-1} [15]. Compared with other studies, especially those conducted in pilot-scale facilities, the laboratory scale experiment did not exhibit appreciably high oxidation efficiencies because: (1) residual time in the biocover of CH_4 was too short due to high mass flow caused by both a high SLFG filling rate and air scour by PAVS. The contact time for CH_4 and methanotrophs was consequently insufficient for complete oxidation; thus, the optimum mass flow needs to be characterized to obtain sufficient reaction time for methanotrophs in the system. (2) In the laboratory set-up, O_2 very likely became excessive in contrast to the O_2 limitation. Therefore, the optimum ratio of SLFG flow quantity to that of O_2 needs further investigation. (3) Methanotrophs that acclimate O_2-rich environment need to be enriched and stimulated in a prolonged incubation time.

Nevertheless, enhanced CH_4 oxidation capacity is expected with improved O_2 supply and transfer efficiency brought about by a system such as PAVS. A demonstration engineering of this technology with land filling capacity of 10,000 m^3 has been carried out by the authors.

This is the first report that uses additional O_2 supply with no power consuming as basis in delving into a solution to O_2 bottlenecks in conventional landfill biocovers. This unique *in situ* O_2 supply method differs considerably from *ex situ* facilities (i.e. biofiltration). It is kind of all-purpose CH_4 emission reduction method for the landfills regardless of whether it is equipped with a gas collection system or not. And it especially could be an economically feasible approach in many developing countries.

4. Conclusions

The O_2 concentration (v/v) considerably increased throughout whole profiles of the microcosm (1 m) equipped with PAVS (MPAVS). When the simulated landfill gas SLFG flow was

771 g m^{-3} d^{-1} and 1028 g m^{-3} d^{-1}, the O_2 concentration in MPAVS gradually increased in the initial stage and stabilized at almost near the atmospheric level after 10 days. The CH_4 oxidation rate was 100% when the SLFG flow rate was no more than 1285 g m^{-3} d^{-1}, which also was confirmed by the mass balance calculations followed. This study provides a breathing biocover system to *in situ* self-oxygen supply addressing the problem of O_2 bottlenecks in conventional landfill covers/biocovers. The proposed system presents high potential for improving CH_4 emission reduction in landfills.

Acknowledgments

This research project was funded by the National High Technology Research and Development Program of China (No. 2007AA06Z350) and the State Key Project for National Science and Technology (No. 2006BAC06B05), and China Postdoctoral Science Foundation funded project (Nos. 20090460301 and 201003106).

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