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Enhanced Methane Oxidation Potential of Landfill Cover Soil Modified with Aged Refuse

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Abstract: Aged refuse with a landfill age of 1.5 years was collected from a municipal solid waste landfill with high kitchen waste content and mixed with soil as biocover material for landfill. A series of laboratory batch tests was performed to determine the methane oxidation potential and optimal mixing ratio of landfill cover soil modified with aged refuse, and the effects of water content, temperature, CO_2/CH_4, and O_2/CH_4 ratios on its methane oxidation capacity were analyzed. The microbial community analysis of aged refuse showed that the proportions of type I and type II methane-oxidizing bacteria were 56.27% and 43.73%, respectively. Aged refuse could significantly enhance the methane oxidation potential of cover soil, and the optimal mixing ratio was approximately 1:1. The optimal temperature and water content were about 25 °C and 30%, respectively. Under the conditions of an initial methane concentration of 15% and an O_2/CH_4 ratio of 0.8–1.2, the measured methane oxidation rate was negatively correlated with the O_2/CH_4 ratio. The maximum methane oxidation capacity measured in the test reached 308.5 (μg CH_4/g)/h, indicating that the low-age refuse in the landfill with high kitchen waste content is a biocover material with great application potential.

Keywords: methane oxidation; landfill cover; refuse; soil

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1. Introduction

Landfill remains the main treatment method of municipal solid waste (MSW) in China and other developing countries [1]. As leachate and landfill gas (mainly methane and carbon dioxide) are produced in the anaerobic degradation process of MSW, landfills have become important sources of methane emissions from human activities [2,3]. In order to avoid uncontrolled methane emissions from landfill, landfill gas collection systems composed of wells and a horizontal collection system have been proposed as an effective way for controlling methane emissions and utilizing the collected methane resource [4–6]. However, in practical engineering application, it is found that it is usually difficult for this system to collect all landfill gas [7–9]. Spokas et al. [10], through field tests, concluded that the efficiency of the landfill gas collection system is related to the cover type, and the amount of methane collected is about 41–98% of the estimated methane production. Bian et al. [11] reported that the collection efficiency of landfill gas collection systems

in China might be lower due to the fast degradation of kitchen waste. Using methane-oxidizing bacteria in cover soil can consume uncontrolled methane emissions, making it a complementary means to control methane emissions [11–13].

Biocover is an important technology for reducing the uncontrolled methane emissions from landfills, especially for small and old landfills that are not suitable for establishing landfill gas collection systems [14–17]. Unlike the traditional landfill soil cover [18,19], the biocover generally consists of a methane oxidation layer and a gas diffusion layer [20,21]. The methane oxidation layer is usually composed of organic-rich materials (e.g., compost, sludge, and peat) and soil, which supports and promotes the growth of methane-oxidizing bacteria and has a certain methane oxidation capacity [22]. Landfill gas enters the methane oxidation layer uniformly through the gas diffusion layer, and methane is consumed by methane-oxidizing bacteria to produce carbon dioxide and water [23]. Cabral et al. [24] conducted field tests on methane oxidation of biocover in a landfill, using compost and coarse sand as the methane oxidation layer. The test results showed that the measured maximum methane oxidation rate of biocover could reach 804 $(g/m^2)/d$, indicating that biocover can achieve a high methane removal capacity through reasonable design. Previous studies have shown that biocover has the potential to become an economic and environmentally friendly method to reduce methane emissions from landfills [25–27].

The methane oxidation potential of biocover is closely related to the physicochemical properties of cover materials and environmental factors, including soil organic matter content, pH, water content, temperature, water-gas transport characteristics, methane concentration, and oxygen concentration [28–30]. The change of soil water content and temperature is considered to be an important reason for the seasonal change of methane oxidation capacity of biocover [31–33]. Ensuring that the cover material has sufficient water content is the basic condition for maintaining the activity of methane-oxidizing bacteria, but excess water will block the gas migration channel, limiting the migration of methane and oxygen and inhibiting methane oxidation [34–36]. The optimal temperature for methane oxidation in soil environments is generally considered to be 25–35 °C [23], but the existence of heat-resistant and thermophilic methane-oxidizing bacteria may also lead to optimal temperatures higher than 55 °C [37]. When the application area of the biocover is subject to large seasonal changes, it is necessary to determine the most suitable water content and temperature of the cover material in order to enhance the methane oxidation of the biocover [27].

The literature reports that aged refuse is a biocover material with application potential [38–40]. Aged refuse usually refers to MSW disposed of in landfills after it has undergone a long-term stabilization process, and this residue contains many nutrients and a rich microbial content [41]. Mei et al. [42] pointed out that the aged refuse buried for 8 years can be used for biological oxidation of methane produced by the landfill, and its biological oxidation capacity can be enhanced by leachate. Zhang et al. [43] conducted batch tests on the methane oxidation capacity of aged refuse with a landfill age of 12 years, and found that the maximum methane oxidation rate was 235.7 $(\mu g\ CH_4/g)/h$. Some previous studies mainly focused on the methane oxidation capacity of aged refuse with an age of more than 8 years [44–46], and few of them have used low-age MSW for methane oxidation related tests.

Due to the influence of local dietary habits, MSW in developing countries generally has kitchen waste content as high as 40–85%, which is characterized by fast hydrolysis, large amounts of leachate, and fast gas production [47–49]. Some studies have pointed out that MSW with high kitchen waste content hydrolyzes about 80% of degradable substances within 2 years after landfill, and then enters a slow degradation stage with a duration of about 13 years [50,51]. This paper explores the possibility of using low-age refuse with high kitchen waste content to enhance the methane oxidation performance of landfill cover soil. The main objectives of this study are: (I) to determine the physicochemical composition of low-age refuse, the community structure of methane-oxidizing bacteria, and the optimal mixing ratio of aged refuse and cover soil; (II) to study the effects of temperature and water

content on the methane oxidation potential of cover soil modified with aged refuse; and (III) to analyze the effects of CO_2/CH_4 and O_2/CH_4 ratios on methane oxidation.

2. Materials and Methods

2.1. Test Material Properties

The soil samples used in the test were collected from the Shabei MSW landfill, which is located in Jiaojiang District, Taizhou City, Zhejiang Province, China (121.45° E, 28.68° N). This landfill was built in 1998 and closed in 2018 for remediation. The soil samples were taken from the closure cover of the landfill, mainly within the cover depth range of 10–20 cm. After drying, these samples were crushed to remove grass roots and other sundries, and then screened through a 2 mm sieve. Afterwards, they were analyzed by X-ray diffraction with an X-ray diffractometer (D8 Advance, Bruker Corporation, Stuttgart, Germany). It was found that the main mineral components are quartz, anorthite, and calcite. According to the relevant provisions of the Standard for Geotechnical Testing Method (GB/T 50123-2019), the basic physicochemical properties of the soil were tested. Its liquid limit and plastic limit were 38.63% and 13.28%, respectively, placing it in the category of silty clay. Other basic characteristic parameters are shown in Table 1.

Table 1. Basic physicochemical properties of cover soil.

Natural Water Content (%)	Particle Composition (%)			Proportion	Organic Matter Content (%)	pH
	2–0.075 mm	0.075–0.005 mm	<0.005 mm			
23.2	70.3	25.0	4.7	2.74	0.9	7.9

The aged refuse used in the test was obtained from Hangzhou Tianziling MSW landfill (120.22° E, 30.39° N). This landfill was put into use in 2007. As the main sanitary landfill of MSW in Hangzhou, it currently disposes of about 4300 tons of landfill waste daily. The aged refuse used in the test was MSW with a landfill age of about 1.5 years in Tianziling landfill, and its main composition is shown in Table 2. The aged refuse was screened through a 3 mm sieve to remove plastics and other sundries. The organic matter content of the screened organic fine material was determined by the loss on ignition method [52], and the measured organic matter content was about 79.5%. In addition, a 20 g sample of aged refuse was placed in a 100 mL centrifuge bottle; then pure water was added for preservation, and the sample was frozen with dry ice. The sample was sent to Shanghai Wiki Biotechnology Co., Ltd., for microbial diversity assessment, and the Amplified 16S rRNA Gene Restriction Analysis (ARDRA) was used for the microbial identification and classification.

Table 2. Composition of the aged refuse.

Organic Fines	Plastics	Glass and Metal	Animal Bone	Large Stone	Plant Rhizome
53.38%	21.65%	2.02%	10.95%	10.65%	1.36%

2.2. Methane Oxidation Potential Test

The methane oxidation potential of the samples was determined through batch incubation tests [27]. Each sample was composed of cover soil and aged refuse based on the target proportion, and the dry weight of the sample was set to 4 g. After drying and crushing, the cover soil was screened through a 2 mm sieve to remove grass roots and other sundries. After the aged refuse was dried at 70 °C, it was screened through a 3 mm sieve to remove plastics and other sundries and retain organic fine materials. According to the target proportions, the dry weights of cover soil and aged refuse were determined, and the materials were mixed; then distilled water was added to prepare the sample with the target water content.

The prepared sample was placed in a glass serum bottle with a volume of 100 mL to measure its methane oxidation potential. Three replicates were set in parallel for each group of tests, and the average value of the final results was obtained. The serum bottle was sealed with a butyl rubber plug and a bakelite cap; the target volume of air was extracted from the serum bottle through an air tight syringe, and then the same volume of simulated landfill gas was injected into the serum bottle. The serum bottle was placed in a constant temperature water bath shaker, and the temperature was set to the target temperature for incubation under a constant temperature environment. The gas in the serum bottle was sampled every 24 h, and the volume fractions of oxygen, nitrogen, methane, and carbon dioxide in the gas sample were determined by a gas chromatograph (GC-7890A, Agilent Technology Co., Ltd., Santa Clara, CA, USA). The GC was configured with a molecular sieve column flame ionized detector (FID), using hydrogen as the carrier gas. After each sampling, the serum bottle was no longer sealed, and the gas in the bottle was washed repeatedly with air to make the gas in the bottle consistent with the laboratory air. Then, a butyl rubber plug and a bakelite cap were used to seal the serum bottle; the target volume of air was extracted from the bottle again, and the same volume of simulated landfill gas was injected. We continued keep the samples in a constant temperature water bath shaker for incubation, with each test lasting for 15 days. When the methane oxidation rate of the sample was high, in order to ensure that there was always methane and oxygen in the serum bottle, the sampling interval was shortened accordingly, and the serum bottle was ventilated at shorter intervals.

In the process of methane oxidation, methane and oxygen are consumed to produce carbon dioxide, which changes the total volume of gas in the incubation bottle. However, since nitrogen does not participate in any reaction, its quantity remains unchanged. The volume change of each gas can be determined according to the change of volume fraction of the gas and the amount of nitrogen in the bottle during the incubation process, and thereby, determine the methane oxidation rate of the sample.

2.3. Test Scheme

In order to determine the methane oxidation potential of cover soil modified with aged refuse and its influencing factors, four variables were measured in the batch incubation tests: the amount of aged refuse, water content, temperature, and landfill gas composition. The settings of the four test conditions are shown in Table 3. For working conditions I–III, the initial gas composition in the serum bottle was approximately 15% (mol%) CH_4, 17.9% (mol%) O_2, and 67.1% (mol%) N_2. The methane concentration in the serum bottle was close to the oxygen concentration, which is to ensure that methane was sufficient during the incubation process and would not become a limiting factor [27]. For working condition IV, the ratio of carbon dioxide to methane was set as 0:1, 0.5:1, 1:1, and 2:1, respectively. At this time, the initial volume fraction of methane in the serum bottle was always 15%, while the initial volume fraction of oxygen was 17.9%, 16.3%, 14.7%, and 11.6%, respectively. In order to avoid the oxygen supply becoming the limiting factor for the methane oxidation process of the sample, the sampling interval could be adjusted according to the gas composition analysis in the serum bottle. When the oxygen concentration in the serum bottle was lower than 3%, sampling was conducted and the serum bottle was ventilated [23].

Table 3. Incubation scheme of cover soil modified with aged refuse.

Working Condition	Soil Sample No.	Amount of Aged Refuse (%)	Water Content (%)	Temperature (°C)	CO_2 Volume (mL)	CH_4 Volume (mL)
I	R0	0	30	30	0	15
	R5	5				
	R10	10				
	R25	25				
	R50	50				
	R100	100				

Table 3. *Cont.*

Working Condition	Soil Sample No.	Amount of Aged Refuse (%)	Water Content (%)	Temperature (°C)	CO_2 Volume (mL)	CH_4 Volume (mL)
II	W5 W15 W25 W35 W45	50	5 15 25 35 45	30	0	15
III	T15 T25 T35	50	30	15 25 35	0	15
IV	C0 C0.5 C1 C2	50	30	30	0 7.5 15 30	15

3. Results and Discussion

3.1. Community Structure of Methane-Oxidizing Bacteria in Aged Refuse

The test results showed that the aged refuse used in the test contained 269 *Methylobacter* sequences, 166 *Methylocaldum* sequences, and 338 *Methylocystis* sequences. The proportion of each methane-oxidizing bacteria is shown in Figure 1. *Methylobacter* and *Methyllocaldum* belong to type I methane-oxidizing bacteria [53], accounting for 56.27% of the total methane-oxidizing bacteria, while *Methylocystis* belongs to type II methane-oxidizing bacteria, accounting for 43.73% of the total methane-oxidizing bacteria.

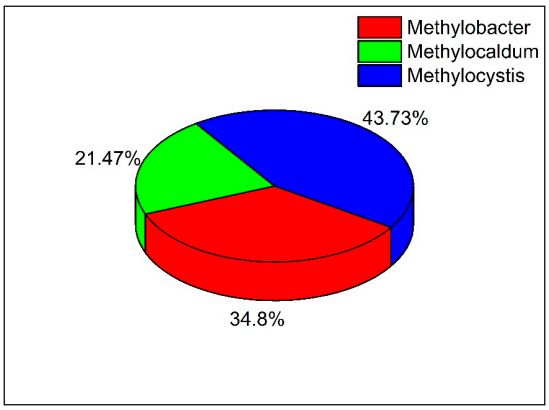

Figure 1. Types and proportions of methane-oxidizing bacteria in aged refuse.

3.2. Effect of the Amount of Aged Refuse on the Methane Oxidation Potential of Cover Soil

Under the conditions of a constant temperature of 30 °C and a water content of 30%, the variation in the methane oxidation rate with time of cover soil with different amounts of aged refuse is shown in Figure 2. The methane oxidation rate of the sample generally increases to the maximum value with incubation time, and then gradually decreases to a lower value. This is due to the accumulation of extracellular polymeric substances (EPSs) produced during methane oxidation, which hinders the diffusion of gas and inhibits methane oxidation [22]. When the amount of aged refuse was 0%, the methane oxidation rate of cover soil was close to 0 (μg CH_4/g)/h; that is, the pure cover soil exhibited hardly any methane oxidation capacity during the test. When the amount of aged refuse increased from 5% to 100%, the methane oxidation rate of the sample increased from 103.1 (μg CH_4/g)/h to 190.7 (μg CH_4/g)/h. With the increase in the amount of aged refuse, the methane oxidation rate of cover soil increases. This is because the organic matter content and nutrients in the cover soil increase with the increase of the amount of aged

refuse, which promotes the activity of methane-oxidizing bacteria and improves methane oxidation [12]. Moreover, the abundance of microorganisms brought by the aged refuse may be a key reason for the enhanced methane oxidation capacity.

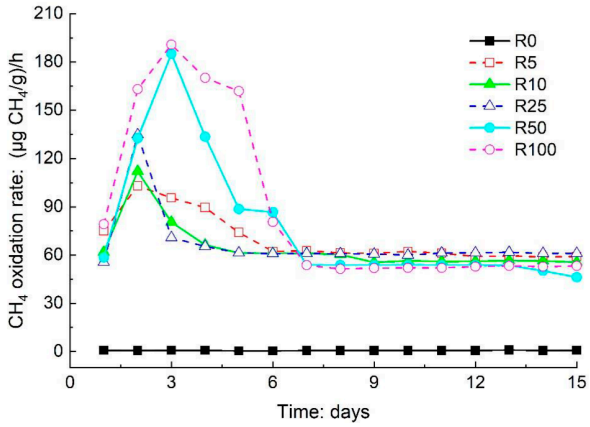

Figure 2. Methane oxidation rate of cover soil with different amounts of aged refuse versus time.

The relationship between the maximum methane oxidation rate of cover soil measured in the test and amount of aged refuse is shown in Figure 3. When a certain amount of aged refuse is added to the pure cover soil, the methane oxidation rate can be greatly improved. This is mainly because the lower organic matter content and nutrients in the pure cover soil inhibit methane oxidation, and the higher organic matter content and amount of nutrients of aged refuse remove this restriction. When the amount of aged refuse gradually increased from 5% to 50%, the methane oxidation capacity of cover soil was basically linear and positively correlated with the amount of aged refuse. When the amount of aged refuse was further increased from 50% to 100%, the maximum methane oxidation rate of the cover soil did not increase significantly. This shows that when the amount of aged refuse reaches 50%, increasing the amount of aged refuse cannot further promote methane oxidation in cover soil. The reason may be that the test conditions limit the water content and gas supply of the sample. With the increase in the amount of aged refuse, the test conditions of our incubation tests may limit methane oxidation [54,55]. Similar results were also observed by other researchers in methane oxidation incubation tests [56].

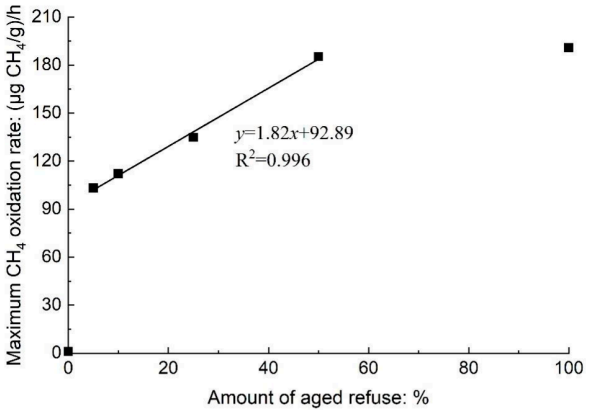

Figure 3. Relationship between maximum methane oxidation rate and amount of aged refuse.

3.3. Effect of Water Content on the Methane Oxidation Rate of Cover Soil Modified with Aged Refuse

Under a constant temperature of 30 °C and with a 50% proportion of aged refuse, the variation the methane oxidation rate with time for cover soil with different water contents is shown in Figure 4. It can be seen from the figure that the incubation time required for samples with different water contents to reach the maximum methane oxidation rate is

significantly different. The incubation time required for the samples with water contents of 5%, 15%, 25%, 35%, and 45% to reach the maximum methane oxidation rate was 8 d, 4 d, 3 d, 3 d, and 4 d, respectively. This shows that soil water is conducive to maintaining microbial activities, and the increase of soil water content shortens the time for the cover soil to reach the maximum methane oxidation rate. However, excess water will limit the diffusion and supply of methane and oxygen, resulting in the inhibition of methane oxidation of cover soil, so that the time for cover soil to reach the maximum methane oxidation rate will be prolonged with the further increase of water content [14].

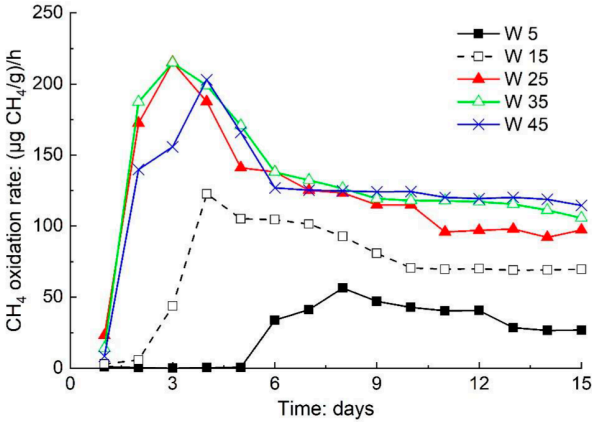

Figure 4. Methane oxidation rate of cover soil with different water contents versus time.

The relationship between the maximum methane oxidation rate of cover soil measured in the test and water content is shown in Figure 5. It can be seen that with the increase of water content from 5% to 45%, the maximum methane oxidation rate first increases to the highest value (215.3 (μg CH_4/g)/h)) and then decreases. Based on the results of this study, the optimal water content of cover soil containing 50% aged waste is approximately 30%. The optimal water content of cover materials is related to the material characteristics and ambient temperature, and the materials with higher water holding capacity and better gas conductivity may have higher optimal water content [27,54,57]. Conversely, unlike the soil samples containing less soil used in the incubation test, the cover is usually compacted in the actual project, and the change of water content has a more significant impact on the gas supply [36]. Therefore, the optimal water content of biocover is not constant, and its value shows spatiotemporal variability according to environmental conditions and cover material characteristics.

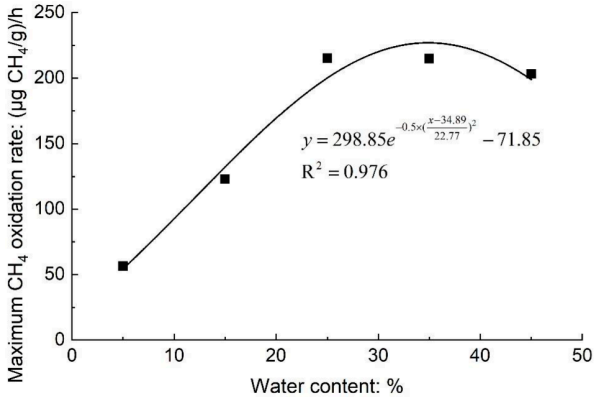

Figure 5. Relationship between maximum methane oxidation rate and water content.

3.4. Effect of Temperature on the Methane Oxidation Rate of Cover Soil Modified with Aged Refuse

Under the conditions of 30% water content and a 50% proportion of aged refuse, the variation of the methane oxidation rate with time for cover soil under different temperatures

is shown in Figure 6. The maximum methane oxidation rates of samples with the incubation temperature of 15 °C, 25 °C, and 35 °C were 210.0 (μg CH_4/g)/h, 308.5 (μg CH_4/g)/h, and 232.1 (μg CH_4/g)/h, respectively, indicating that the optimal temperature is around 25 °C. Scheutz et al. [23] summarized a large number of methane oxidation incubation experiments and pointed out that the optimal temperature for methane oxidation in soil environment is usually 25–35 °C, and the results of this test fall exactly within this range. However, the incubation time for the sample to reach the maximum methane oxidation rate at 15 °C is 4 days, which is the lowest in the three working conditions. This may be related to the types of methane-oxidizing bacteria in the sample. The detection results of the aged refuse showed that the methane-oxidizing bacteria contained is composed of about 56.27% of type I methane-oxidizing bacteria and 43.73% of type II methane-oxidizing bacteria. Type I methane-oxidizing bacteria can adapt to lower temperatures than type II methane-oxidizing bacteria [58], which enables them to quickly acquire the dominant position at lower temperatures and cause the sample to quickly reach the maximum methane oxidation rate. It should be noted that temperature can affect the selection of the methane-oxidizing bacterial population by environment [59]. The optimal temperature of the cover layer is affected by the service environment. The effect of the climatic environment should be considered to determine the optimal temperature of biocover in field applications [14].

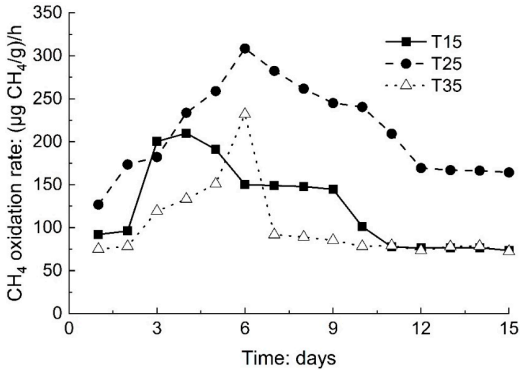

Figure 6. Methane oxidation rate of cover soil under different temperatures versus time.

3.5. Effect of Gas Composition on the Methane Reaction Rate of Cover Soil Modified with Aged Refuse

Under the conditions of 50% aged refuse, 30%, water content and a temperature of 30 °C, the variation of the methane oxidation rate with time for cover soil with different gas compositions is shown in Figure 7. With the increase of the initial concentration of CO_2 and the decrease of the initial concentration of O_2 in the serum bottle, the incubation time required for the sample to reach the maximum methane oxidation rate increases. When the ratio between the three gases was approximately CO_2: CH_4: O_2 = 1:1:1, the measured methane oxidation rate was the highest (260.6 (μg CH_4/g)/h).

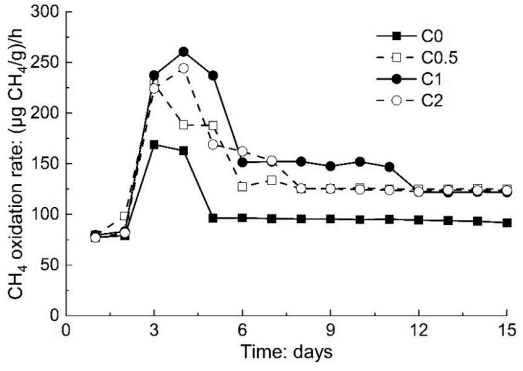

Figure 7. Methane oxidation rate of cover soil with different gas compositions versus time.

The relationship between maximum methane oxidation rate of the sample with CO_2/CH_4 and O_2/CH_4 ratios is shown in Figure 8. Spokas and Bogner [30] studied the effect of CO_2 concentration on the methane oxidation rate of cover soil through batch incubation tests. The results showed that the increased CO_2 concentration does not change the methane oxidation rate of cover soil. As can be seen from Figure 8, the relationship between the maximum methane oxidation rate of the sample and the O_2/CH_4 ratio is significantly closer than that of the CO_2/CH_4 ratio. With the decrease of the O_2/CH_4 ratio from 1.2 to 0.8, the maximum methane oxidation rate of the sample increases; that is, the methane oxidation capacity of the sample has a negative correlation with the O_2/CH_4 ratio. The main reason for this result may be related to the EPS produced in the process of methane oxidation. More EPSs are produced at high oxygen concentration, which inhibits methane oxidation [60]. However, in the landfill cover, the diffusion depth of oxygen usually becomes an important factor limiting the methane oxidation performance of the cover. Moreover, the scale effects need to be considered when studying and designing the methane oxidation performance of the cover [61].

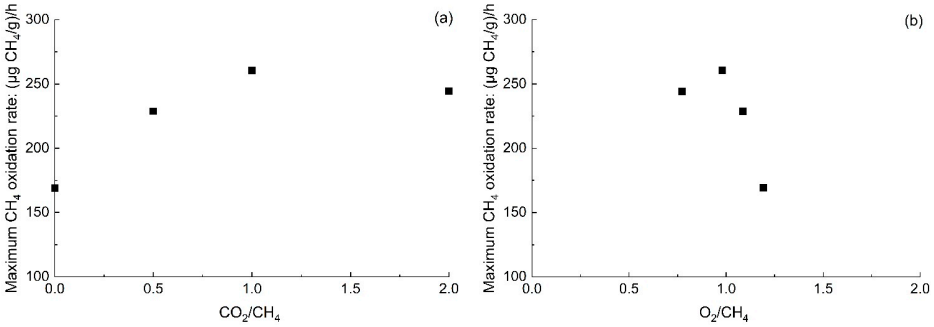

Figure 8. Relationship between maximum methane oxidation rate and (**a**) CO_2/CH_4 and (**b**) O_2/CH_4 in the serum bottle.

4. Conclusions

Based on the biochemical degradation characteristics of MSW containing high kitchen waste content in developing countries, a new method of using low-age refuse and cover soil as biocover material was proposed in this paper. Through a series of laboratory batch incubation experiments, the effects and characteristics of aged refuse that influence the methane oxidation potential of cover soil were explored. The main results and conclusions can be summarized as follows:

- The microbial community analysis of aged refuse shows that it mainly includes *Methylobacter*, *Methylocaldum*, and *Methylocaldum*. The type I and type II methane-oxidizing bacteria account for 56.27% and 43.73% of the total methane-oxidizing bacteria, respectively.
- Adding aged refuse to cover soil can significantly enhance its methane oxidation potential. When the amount of aged refuse gradually increases from 5% to 50%, the methane oxidation capacity of cover soil is basically linear and positively correlated with the amount of aged refuse. However, the methane oxidation capacity of cover soil does not increase with the further increase in the amount of aged refuse, indicating that the optimal mixing ratio between aged refuse and cover soil is approximately 1:1.
- When the amount of aged refuse reaches 50%, the optimal temperature and water content are approximately 25 °C and 30%, respectively, and the maximum methane oxidation rate measured in batch incubation tests is 308.5 (μg CH_4/g)/h.
- When the initial concentration of methane in the serum bottle is 15%, the methane oxidation capacity of cover soil modified with aged refuse is significantly affected by the O_2/CH_4 ratio, but is little affected by the CO_2/CH_4 ratio. When the O_2/CH_4 ratio is in the range of 0.8–1.2, the methane oxidation capacity of cover soil modified with aged refuse is negatively correlated with the O_2/CH_4 ratio. In follow-up research, it is

necessary to study the effect of oxygen concentration on the production of EPSs in the methane oxidation process of cover soil modified with aged refuse.

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