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In-situ neutralize methane emission from landfills in loess regions using leachate

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In loess regions, landfilling is the predominant solid waste disposal and loess is usually used as landfill cover soil. However, the methane (CH_4) bio-oxidation activity of virgin loess is usually below 0.01 μmol/(h g-soil). In this study, we proposed a method to improve CH_4 removal capacity of loess by amelioration with mature landfill leachate, which is *in-situ*, easily available, and appropriate. The organic matter content of the ameliorated loess increased by 180%, reaching 19.69–24.88 g/kg-soil, with more than 90% being non-leachable. The abundance of type I methane-oxidizing bacteria and methane monooxygenase gene pmoA increased by 5.0 and 79 times, respectively. Consequently, the maximum CH_4 removal rate of ameliorated loess reached 0.74–1.41 μmol/(h g-soil) at 25°C, which was 4-fold higher than that of water-irrigated loess. Besides, the CH_4 removal rate peaked at 10 vt% CH_4 concentration and remained at around 1.4 μmol/(h g-soil) at 15°C–35°C. The column test confirmed that the highest CH_4 removal efficiency was at 30–40 cm below the surface, reaching 26.1%±0.4%, and the 50-cm-thick loess layer irrigated with leachate achieved more than 85% CH_4 removal efficiency. These results could help to realize carbon neutrality in landfill sites of global loess regions.

methane bio-oxidation, leachate irrigation, loess improvement, landfill cover soil, greenhouse gas emission, biocover, solid waste

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1 Introduction

Loess is widely distributed on earth, accounting for 10% of the global land area [1]. The Loess Plateau in Northwest China, covering an area of 630000 km^2, is the largest loess region in the world [2], and the long-term arid climate and serious water-soil loss in this region result in a low potential ecological carrying capacity and high sensitivity to climate change [3]. Therefore, the Loess Plateau is one of the most ecologically vulnerable areas in the world.

The Loess Plateau has a vast territory, but underdeveloped economy. As a result, landfilling has become the dominant method for the treatment and disposal of mixed municipal solid waste (MSW) in this region owing to its relatively low cost. For instance, in Gansu Province in Northwest China, 56.8% of urban MSW was treated by landfilling in 2018 [4]. However, landfill is well known to be a huge greenhouse gas source, accounting for 16% of global anthropogenic methane (CH_4) [5]. Although landfill cover soil (LCS) is conventionally regarded as a barrier for mitigating CH_4 emissions from landfills, the barrier using loess as LCS is fragile because bioactivity, including CH_4 oxidation ability, of loess is weak and usually leads to a higher amount of CH_4 release [6,7]. Therefore, to abate the disorganized release of CH_4 from landfill in loess regions, local ecological restoration is in huge demand.

CH_4 released from landfills can be significantly mitigated by well-structured LCS through bio-oxidation of soil me-

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thanotrophs [8–10]. In general, the CH_4 bio-oxidation capacity of LCS can be enhanced by the following factors: (1) soil microbial communities in CH_4-rich environment have higher CH_4 affinity [11,12]; (2) ammonia nitrogen (NH_4^+-N) can stimulate the abundance of methanotrophs by improving nitrogen availability for type I methane-oxidizing bacteria (MOB), and enhance the transcription of methane monooxygenase gene pmoA. Besides, NH_4^+-N can increase the abundance of ammonia-oxidizing bacteria (AOB) that can co-oxidize CH_4 [13,14]; (3) the aggregate structure of soil can improve landfill gas adsorption of LSC [15]; and (4) temperature, aeration, pH and water content of soil can affect the activity of MOB as well [16]. Accordingly, bioactive materials like sludge [17] and compost [10] are used to supplement microorganisms and stimulate microbial activity. Biochar addition has been reported to lead to a more balanced ratio of homogeneous microbial community structures across different soil depths and 90% CH_4 removal efficiency by LCS [18]. However, owing to the arid climate of the Loess Plateau, these bioactive materials can lose moisture, making it difficult to maintain high bioactivity. Additionally, substantial efforts are required to apply these solid materials to the LCS. Furthermore, the amount of bioactive materials needed for remediation is huge, which is economically unaffordable from the point of material cost as well as transportation cost. Therefore, a more sustainable method is needed to obtain appropriate landfill covering materials.

Leachate irrigation is another alternative to improve the CH_4 oxidation capacity of LCS because mature leachate contains a large amount of nitrogen and humus [19,20], which can increase the soil organic matter content, aggregate structure, and water holding capacity [21]. Consequently, leachate has been applied to irrigate LCS such as silt [22], sandy soil [9], loam [23], and vegetated LCS [24]. However, when compared with these LCSs, loess presents lower organic matter contents (<1%), low nitrogen contents (<1‰) [25], and more serious salinization (pH>8) [26]. Straw incorporation and plastic-film mulching have been used to enhance bioactivity and organic matter content of loess [27,28]. Nevertheless, the extent to which CH_4 bio-oxidation activity of barren soil such as loess could be improved by leachate remediation is still unclear.

The present study aimed to determine the remediation effect of leachate on the CH_4 bio-oxidation activity of loess, and applied mature leachate to irrigate loess at different irrigation levels. In addition to tracking the changes in the physicochemical soil quality along the irrigation incubation period, soil microorganisms and related genes were also quantified to address the underlying biological rationale leading to enhanced CH_4 bio-oxidation activity of the ameliorated loess. Finally, greenhouse gas reduction through leachate remediation in the loess region was estimated.

2 Materials and methods

2.1 Loess and leachate

Loess soil was collected from a storage site for LCS in Lanzhou, Gansu Province, China (103.7°E, 36.5°N). The soil was directly procured with a ferruginous spade after removing about 5 cm of sand and gravel from the surface. Subsequently, the soil was mixed evenly and stored in a polypropylene bag at room temperature (around 25°C). Prior to the experiments, the soil was further screened using a 2 mm sifter to remove large gravel and plant roots. The basic physicochemical properties of loess soil are listed in Table 1.

The leachate was sampled from the outlet of a landfill regulating tank and stored in plastic buckets at room temperature (around 25°C). Table 2 shows the physicochemical characteristics of the stored leachate. The ratio of biological oxygen demand over 5 days (BOD_5) to chemical oxygen demand (COD) was lower than 0.1 (0.029–0.036), suggesting that the leachate was sufficiently stabilized [29]. The value variation of 0–90 days indicated that the characteristics of the leachate did not significantly change during the entire irrigation period (P>0.05).

2.2 Experimental setup

A total of five experiments were scheduled as follows, and the research scheme is illustrated in Figure S1 in Supporting information.

(1) Loess amelioration with leachate irrigation

To explore the amelioration effect of long-term mature leachate irrigation on the properties of loess, the soil was divided into three groups according to the leachate holding capacity of the loess employed (Table 1). The water content of the groups reached 16 wt% (named as **L16**, representing 60% of leachate holding capacity of the soil), 22 wt% (named as **L22**, representing the plastic limit water content of the soil) [30,31], and 28 wt% (named as **L28**, representing the maximum leachate holding capacity of the soil), respectively, by leachate irrigation. Distilled water was used as the negative control group to adjust the water content of the soil to 16 wt% (named as **W16**). These four groups were

Table 1 Basic physicochemical properties of loess sample

Parameter	Value
Bulk density (g/cm^3)	1.21±0.006
Water content (%)	6.29±0.08
Water holding capacity (g/kg)	303.5±2.1
Leachate holding capacity (g/kg)	378.2±2.4
pH	8.37±0.06
Organic matter content (g/kg)	6.98±0.29
TN (g/kg)	0.89±0.08
NH_4^+-N (g/kg)	0.62±0.01

Table 2 Physicochemical characteristics of the leachate during 90 days' storage

Parameter	Value			
	0 d	30 d	60 d	90 d
pH	8.11±0.01	8.12±0.01	8.17±0.02	8.21±0.02
Alkalinity (mg$CaCO_3$/L)	7554±27	7694±23	7706±19	7754±22
COD (mg/L)	4026±26	3765±23	3554±21	3570±16
BOD_5 (mg/L)	146±7	136±8	122±10	102±10
TOC (mg/L)	1280±9	1062±12	992±10	951±8
TC (mg/L)	3116±10	2969±11	2748±13	2733±8
TN (mg/L)	2067±24	1984±20	1964±38	1897±24
NH_4^+-N (mg/L)	2041±39	1940±35	1933±29	1824±30
BOD_5/COD	0.036	0.036	0.034	0.029

spread on four enamel pans to form a soil layer of about 100 cm×80 cm×3 cm and incubated in an aerobic incubator (SPX-250-Z-S, Shanghai Yuejin Medical Instruments Co., Ltd, China) at 25°C±2°C. The water content of the four groups was monitored regularly, and leachate or distilled water was added to maintain constant water content.

On days 0, 4, 11, 18, 25, 36, 44, 61, 79, and 95 after the start of irrigation, three parallel samples were collected from each enamel pan to test the pH, organic matter content, total nitrogen (TN) content, and NH_4^+-N content of the soil. Besides, to determine the stability of the combination between organic matter and loess, rainwater leaching test was also performed with these samples. To evaluate the changes in soil microorganisms during the amelioration process, DNA was extracted from the initial soil sample, leachate, and soil after leachate irrigation, and subjected to 16S rRNA high-throughput sequencing and quantitative real-time PCR (qPCR) assays.

(2) Acclimation of ameliorated loess in a CH_4-rich environment

With the aim to analyze the changes in CH_4 oxidation capacity of leachate-ameliorated loess, CH_4-rich acclimation of the leachate-ameliorated loess soil was performed. After 95 days of leachate irrigation, 2 kg of loess samples were collected from **L16** and **W16**, respectively, and placed in two plexiglass cylinders with an inner diameter of 20 cm and a height of 40 cm. The soils in the cylinders were placed under atmospheric O_2 and sparged with 5 vt% CH_4 at 25°C±2°C for acclimation of CH_4 oxidation for 3 weeks. The concentrations of CH_4 and CO_2 in the headspace of the cylinders were determined every day. After daily gas determination, the cylinder headspace was flushed completely with air and CH_4 was sparged again to maintain the inflow CH_4 concentration of 5 vt% with a O_2 concentration of 20 vt% [22]. The leachate-ameliorated loess after CH_4 acclimation was named as MLA and distilled water irrigated loess was named as MWA. Subsequently, the DNA from MLA and MWA was extracted and subjected to 16S rRNA high-throughput sequencing and qPCR assays.

(3) CH_4 removal capacity of loess at different CH_4 concentrations

Batch experiments were conducted to determine the CH_4 removal capacity of leachate-ameliorated and CH_4-acclimated loess at different CH_4 concentrations at 25°C. A total of 50 g of loess was collected from MLA and MWA, respectively, and sealed in a 250 mL serum bottle with butyl rubber stopper and aluminium to form 2–3-cm-thick soil layers. The concentration of CH_4 in serum bottles was adjusted to 2, 5, 10, 20, 30, 40, and 50 vt%, respectively. Accordingly, the concentration of O_2 was 21, 20, 19, 17, 15, 13, and 11 vt%. After 0, 1, 3, and 8 h, 1 mL of headspace gas was extracted from the serum bottle with a syringe and the concentrations of CH_4 and CO_2 were measured. For each group, three parallel samples were established and the CH_4 removal rates were calculated using eq. (1):

$$R = \frac{(C_1 - C_2) \times V}{V_m \times \Delta t \times m} \times 10^6, \quad (1)$$

where R is the CH_4 removal rate per unit mass of loess (μmol/(h g-soil)), C_1 and C_2 are the initial and final CH_4 concentrations (vt%) respectively in serum bottle, V is the volume of gas in serum bottle under standard condition (L), V_m is molar volume of gas (22.4 L/mol), Δt is time elapsed (h), and m is the mass of loess (g).

(4) CH_4 removal capacity of loess at different temperatures

Another batch experiment series was established to determine the CH_4 removal capacity of loess at different temperatures. The experimental procedure was the same as that employed for determining the CH_4 removal capacity of loess at different CH_4 concentrations, except for the variations in temperature (5°C, 15°C, 25°C, and 35°C, respectively). The input CH_4 concentration was initially adjusted to 10 vt% with a O_2 concentration of 19 vt% and the headspace gas was measured after 1 h. The CH_4 removal rates were also calculated using eq. (1).

(5) CH_4 removal capacity of loess layers in column experiments

To simulate the dynamic changes in CH_4 when passing through the loess layer, column experiments were performed. Raw loess was respectively placed in two plexiglass cylindrical columns (Figure S2). The inner diameter of the column was 20 cm and the thickness of the loess layer was 50 cm. Gas sampling orifices were established at every 10 cm along the column height. For that loess soil settled naturally in the columns, the density of the loess inside the columns was the same as that of virgin loess, which is about 1.2 g/cm^3. The loess in one column was irrigated with mature leachate (named as CL), while that in the other column was irrigated with distilled water (named as CW). The water content in both the columns was 16 wt%. A mixed gas of 10 vt% CH_4, 10 vt% CO_2, and 80 vt% N_2 was prepared and circulated through a peristaltic pump to the bottom of the two columns. After the gas state in the two columns was stable, gas samples were collected from the sampling orifices to test the concentrations of CH_4, CO_2, N_2, and O_2 along the columns. The CH_4 removal efficiency of the whole loess layer was calculated using eq. (2), and the CH_4 removal efficiencies of different depth of loess layers were calculated using eq. (3):

$$E_T = \frac{C_b - C_a}{C_b} \times 100\%, \quad (2)$$

$$E_X = \frac{C_{xb} - C_{xa}}{C_b} \times 100\%, \quad (3)$$

where E_T is the CH_4 removal efficiency (%) of whole loess layer, E_X is the CH_4 removal efficiencies (%) of different depth of loess layer, C_b and C_a are the CH_4 concentrations (vt%) below and above whole loess layer, respectively, C_{xb} and C_{xa} are the CH_4 concentrations (vt%) below and above different depth of loess layer, respectively.

2.3 Analytical methods

(1) Physicochemical properties of soil

The bulk density and water holding capacity of the soil were analyzed by the ring-knife method (NY/T 1121.4-2006; NY/T 1121.22-2010). The soil water content was determined by drying at 105°C (GB 7172-87). The pH of the soil was measured using digital pH meter (PHS-25, Shanghai Precision Scientific Instrument Co., Ltd, China) in 1:2.5 (*w*/*v*) soil:water mixture (NY/T 1121.2-2006). The soil organic matter was oxidized with excess potassium dichromate-sulfuric acid solution, and determined by titration with ferrous sulfate standard solution (NY/T 1121.6-2006). The soil TN content was measured using an automatic Kjeldahl analyzer (UDK 159, Velp Scientifica Srl, Italy) after digestion (NY/T 1121.24-2012). To determine the soil NH_4^+-N content, the loess:KCl solution (2 mol/L) was mixed at a ratio of 1:5 (*w*/*v*) for 1 h, filtered (HJ 634-2012), and analyzed by spectrophotometry (AQ2, SEAL Analytical, Germany). All the above-mentioned soil testing methods were performed according to the Chinese standards.

(2) Physicochemical properties of leachate

The pH of the leachate was measured using a digital pH meter (PHS-25, Shanghai Precision Scientific Instrument Co., Ltd., China) (GB 6920-86). The alkalinity of the leachate was ascertained by standard acid solution titration (GB/T 9736-2008) with an automatic titrator (800 Dosino, Metrohm, Switzerland). The COD was evaluated by fast digestion spectrophotometry (DR3900, HACH, USA) (HJ/T 399-2007), and BOD_5 was tested by dilution and inoculation (GB/T 7488-1987) in a respirometer (BSBdigi CO_2, SELUTEC, Germany). Total organic carbon (TOC) and total carbon (TC) contents were ascertained by the nondispersive infrared absorption method (GB/T 13193-1991) using a TOC analyzer (TOC-V_{CPH}, Shimadzu, Japan). The TN content was determined by thermocatalytic decomposition and chemiluminescence at 720°C using a TN measuring unit (TNM-l, Shimadzu, Japan). The NH_4^+-N content was analyzed by salicylic acid spectrophotometry (AQ2, SEAL Analytical, Germany) (GB/T 7481-1987). All the above-mentioned water quality testing methods were conducted according to the Chinese standards.

(3) Rainwater leaching test

To simulate the rainwater environment, sulfuric acid-nitric acid solution (pH of 3.20±0.05) was used with a liquid:solid ratio of 10:1 (L/kg) according to the Chinese Standard (HJ/T299-2007), and the mixture was shaken for 18±2 h at 25°C ±2°C on a flip oscillation device at a speed of 30±2 r/min. After leaching, the mixture was filtered with a mixed cellulose membrane. Subsequently, the organic matter content of the sulfuric acid-nitric acid leaching residue and lixivium was measured for calculating the proportion of organic matter that had leached out.

(4) Concentrations of gaseous CH_4, CO_2, N_2, and O_2

The concentrations of CH_4, CO_2, and N_2 were measured by gas chromatograph (GC Trace 1300, Thermo Fisher Scientific, USA) equipped with a thermal conductivity detector and flame ionization detector. A 5 m×2.1 mm carboxen-1000 column with argon as the carrier gas was employed, and the column operating temperature was held at 55°C for 3 min and increased at a rate of 10°C/min to 75°C and held for 3 min [32]. The O_2 concentration was determined by using a digital oxygen meter (CYS-1, Shanghai Jiading Xuelian Instrument panel Factory, China), and the concentrations of CH_4, CO_2, N_2, and O_2 were corrected for standard temperature and pressure conditions (0°C and 101 kPa, respectively).

(5) DNA extraction and 16S rRNA high-throughput sequencing

For microbial analysis, soil samples were collected after leachate irrigation and CH_4 acclimation, and DNA was extracted with PowerSoilTM DNA isolation kit (Mo-Bio Laboratories Inc., CA, USA) [33]. The 16S rRNA high-

throughput sequencing was performed on an Illumina platform (Illumina MiSeq PE300) by Major Bio-Medicine Technology Co., Ltd., China, to characterize the microbial communities. To obtain the relative abundances of bacteria and archaea, the universal primers 515F and 806R were selected to amplify the variable regions V4 of the microbial 16S rRNA gene by PCR [32]. The reads obtained from the MiSeq runs were demultiplexed based on exact matching. The demultiplexed raw amplicon reads were dereplicated and denoised into amplicon sequence variants (ASVs) using DADA2. QIIME2 software was employed to perform taxonomy identification [34], and the taxonomic composition of the samples was explored using pre-fitted sklearn-based taxonomy classifier against SILVA database 132 [35].

(6) qPCR assays

The qPCR assays were performed for quantifying the 16S rRNA gene, pmoA gene, and amoA gene. The primer pair 341F and 515R was used for bacterial gene amplification [36], and the reaction conditions were as follows: initial denaturation step at 95°C for 3 min, followed by 40 cycles of denaturation at 94°C for 20 s, annealing at 58°C for 30 s, and extension at 72°C for 30 s, and a final extension at 72°C for 7 min. For the amplification of archaeal genes, the primer pair Ar364f and Ar934r was used [37], and the reaction conditions were as follows: initial denaturation at 94°C for 1 min, followed by 40 cycles of denaturation at 94°C for 20 s, annealing at 59°C for 30 s, and extension at 72°C for 30 s, and a final extension at 83°C for 1 min. For the analysis of methanotrophic community, the primer pair A189F and mb661R was employed [38], and the reaction conditions were as follows: initial denaturation at 95°C for 5 min, followed by 40 cycles of denaturation at 95°C for 20 s, annealing at 60°C for 30 s, and extension at 72°C for 30 s, and a final extension at 72°C for 7 min. For the amplification of genes related to ammonia-oxidizing bacteria (AOB), the primer pair amoA-1F and amoA-2R was used [39] under the following reaction conditions: initial denaturation at 95°C for 5 min, followed by 40 cycles of denaturation at 95°C for 30 s, annealing at 58°C for 30 s, and extension at 72°C for 45 s, and a final extension at 72°C for 1 min. The qPCR assays were conducted in a cycler (Mastercycler@ realplex^2, Eppendorf, Germany), and the reaction mixture contained 2 μL of the extracted DNA sample, 9.5 μL of ddH_2O, 12.5 μL of SYBR Green Mix, 0.5 μL of Primer 1 (10 μmol/L), and 0.5 μL of Primer 2 (10 μmol/L).

3 Results

3.1 Variations in the physicochemical properties of loess during leachate irrigation

During the process of leachate irrigation, the organic matter content in the three leachate-irrigated soils dramatically increased after the first three leachate additions, as shown in Figure 1(a) (P<0.001). The organic matter content in **L16**,

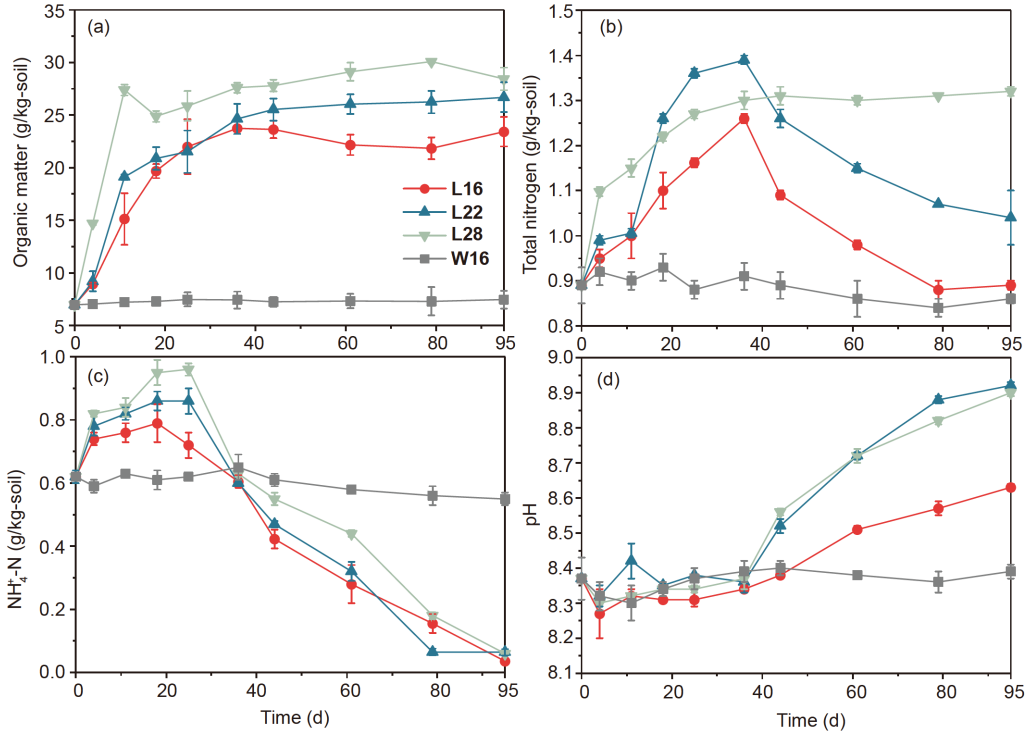

Figure 1 (Color online) Physicochemical properties variation of loess during leachate irrigation. (a) Organic matter; (b) TN; (c) NH_4^+-N; (d) pH. **L16**: the water content was adjusted to 16 wt% with leachate; **L22**: the water content was adjusted to 22 wt% with leachate; **L28**: the water content was adjusted to 28 wt% with leachate; **W16**: the water content was adjusted to 16 wt% with water.

L22, and **L28** improved to 19.69, 20.89, and 24.88 g/kg-soil from the initial 6.98 g/kg-soil, respectively, showing an increase of more than 180%, whereas there was no significant change in **W16** (P>0.05). Subsequently, the organic matter content in the three experimental groups remained constant at 20–30 g/kg-soil (P>0.05).

In contrast, the TN content in **L16**, **L22**, and **L28** (Figure 1 (b)) first increased to 1.26, 1.39, and 1.30 g/kg-soil from 0.89 g/kg-soil in the first five leachate additions, respectively, and then started to decline in **L16** and **L22**. Furthermore, the TN content in **L16** returned to its original level and that in **L22** returned to 1.04 g/kg-soil, whereas the TN content in **L28** remained high at 1.20–1.30 g/kg-soil. The decrease in the TN content could be explained by the NH_4^+-N loss owing to the enhanced pH resulting from leachate addition (Figure 1(c)). However, although **L28** and **L22** presented similar high pH of up to 8.9, its TN content did not decrease, and this phenomenon will be subsequently discussed.

The increase in organic matter content was relatively stable, and the leaching test demonstrated that the residues after leaching still retained most of the organic matter content of the soil samples (Figure 2). The proportion of organic matter content in the residues of **L16** (Figure 2(a)), **L22** (Figure 2(b)), and **L28** (Figure 2(c)) accounted for more than 60%, 65%, and 70% of the total organic matter content, respectively, while that in the corresponding lixivia only accounted for 8.1%, 9.4%, and 11%, respectively. The sum of the organic matter content in the residues and lixivia of **L16**, **L22**, and **L28** accounted for 68%, 74%, and 81% of the total organic matter, respectively, suggesting that the newly formed organic matter supplemented by leachate was closely bound to the soil particles and could not be easily leached out in the rainwater environment. Furthermore, the pH of the lixivia still remained at 8.6–8.9, indicating that the buffering property of the leachate-ameliorated soil was strong.

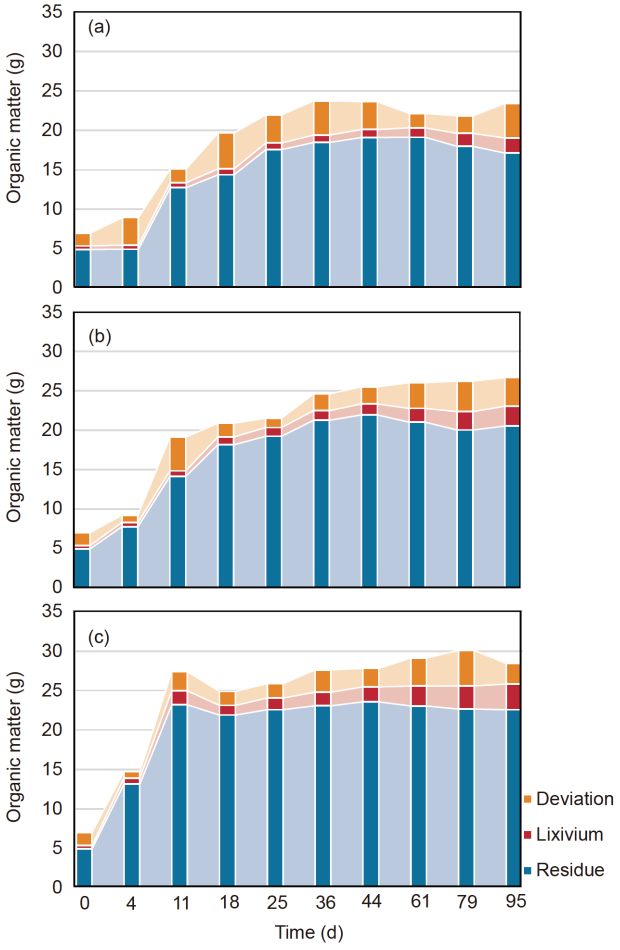

Figure 2 (Color online) Leaching test on the leachate ameliorated loess. Distribution of organic matter in (a) **L16**, (b) **L22**, and (c) **L28**.

3.2 CH_4 removal rate of ameliorated loess

Figure 3(a) shows that the CH_4 removal capacity of the leachate-ameliorated MLA reached up to 0.66 μmol/(h g-soil) after 20 days of CH_4 acclimation from the initial 0.067 μmol/(h g-soil) (P<0.001), whereas that of water-irri-

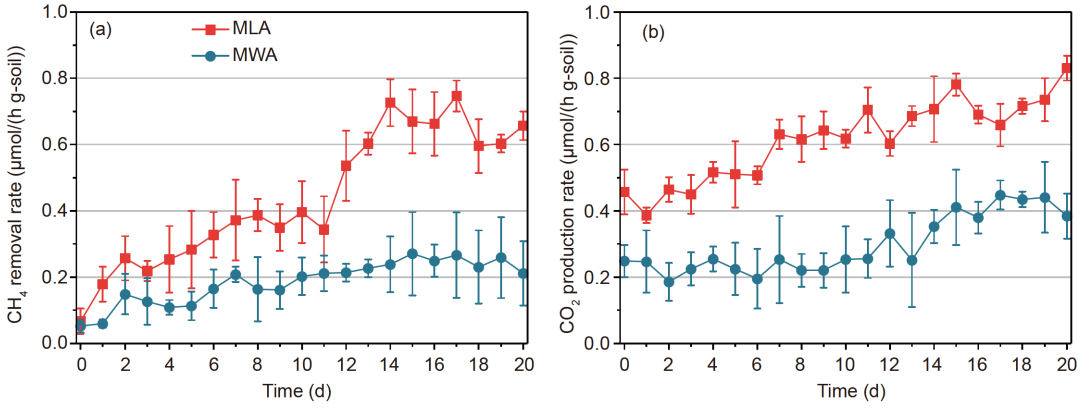

Figure 3 (Color online) Variation of CH_4 removal rate and CO_2 production rate of loess during the acclimation in CH_4-rich environment. (a) CH_4 removal rate variation in acclimation. (b) CO_2 production rate variation in acclimation. MLA: the water content was adjusted to 16 wt% with leachate and acclimated in 10 vt% CH_4-rich environment; MWA: the water content was adjusted to 16 wt% with distilled water and acclimated in 10 vt% CH_4-rich environment.

gated MWA was less obvious and reached only 0.21 μmol/(h g-soil) (P<0.01). Besides, the difference between respiratory activities of MLA and MWA remained relatively stable during the whole acclimation (Figure 3(b)), but the difference between CH_4 removal rates of MLA and MWA increased gradually, which means MOB in MLA gradually took a dominant position.

At different CH_4 concentrations at 25°C, the CH_4 removal rate of MLA was in the range of 0.74–1.41 μmol/(h g-soil) and peaked at 10 vt% CH_4, while that of MWA was in the range of 0.28–0.38 μmol/(h g-soil) and peaked at 20 vt% CH_4 (Figure 4(a)). Similarly, the CO_2 production rate of MLA was 0.17–1.09 μmol/(h g-soil) and peaked at 10 vt% CH_4, while that of MWA was 0.06–0.29 μmol/(h g-soil) and peaked at 5 vt% CH_4 (Figure 4(b)).

The effect of temperature was not as significant as that of CH_4 concentration, and at 10 vt% CH_4 concentration, the CH_4 removal rate of MLA was 1.17–1.40 μmol/(h g-soil) at various temperatures and reached the peak at 35°C. Similarly, the CH_4 removal rate of MWA was 0.20–0.37 μmol/(h g-soil) and peaked at 35°C (Figure 4(c)). Meanwhile, the CO_2 production rate of MLA was in the range of 0.95–1.34 μmol/(h g-soil) and peaked at 35°C, while that of MWA was 0.11–0.33 μmol/(h g-soil) and peaked at 35°C (Figure 4(d)). This result is in accordance with those reported in previous studies on the effect of temperature on CH_4 oxidation [40].

3.3 CH_4 removal efficiency along the depth of soil in column experiments

Figure 5 depicts the variation in the average concentration of four gases (CH_4, CO_2, O_2, and N_2) and CH_4 removal efficiency across 10 cm above the loess surface to 50 cm below the loess surface. In the CL column filled with leachate-ameliorated soil, the concentration of CH_4 was 10.2±0.2 vt% at 50 cm below the surface, and gradually declined to 8.2 ±0.2 vt%, 5.6±0.3 vt%, 4.2±0.2 vt%, and 3.2±0.07 vt% at 40, 30, 20, and 10 cm below the surface, respectively, finally reaching 1.3±0.05 vt% at the surface. This result suggested that more than 85% of CH_4 could be removed by 50-cm-thick ameliorated layer. In contrast, in the CW column filled with water-irrigated soil, the CH_4 concentration at the surface only reached 5.4±0.4 vt%, i.e., only 48% of CH_4 was removed. Thus, the removal efficiency of 50-cm-thick leachate-ameliorated loess layer was about 1.8 times higher

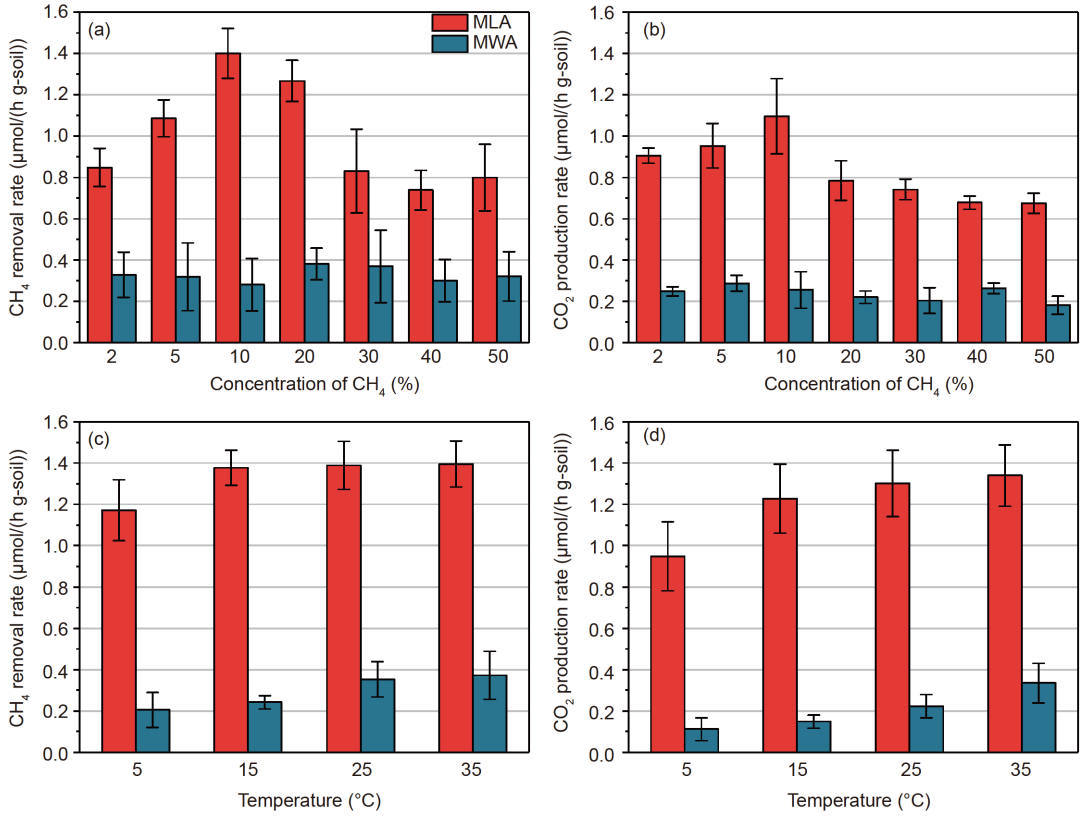

Figure 4 (Color online) Effect of CH_4 concentration and temperature on the CH_4 removal capacity and respiration activity of the ameliorated and methane acclimated loess. (a) CH_4 removal rate at different CH_4 concentrations at 25°C. (b) CO_2 production rate at different CH_4 concentration at 25°C. (c) CH_4 removal rate at different temperatures at 10 vt% CH_4 concentration. (d) CO_2 production rate at different temperatures at 10 vt% CH_4 concentration. MLA: the water content was adjusted to 16 wt% with leachate and acclimated in 10 vt% CH_4-rich environment; MWA: the water content was adjusted to 16 wt% with distilled water and acclimated in 10 vt% CH_4-rich environment.

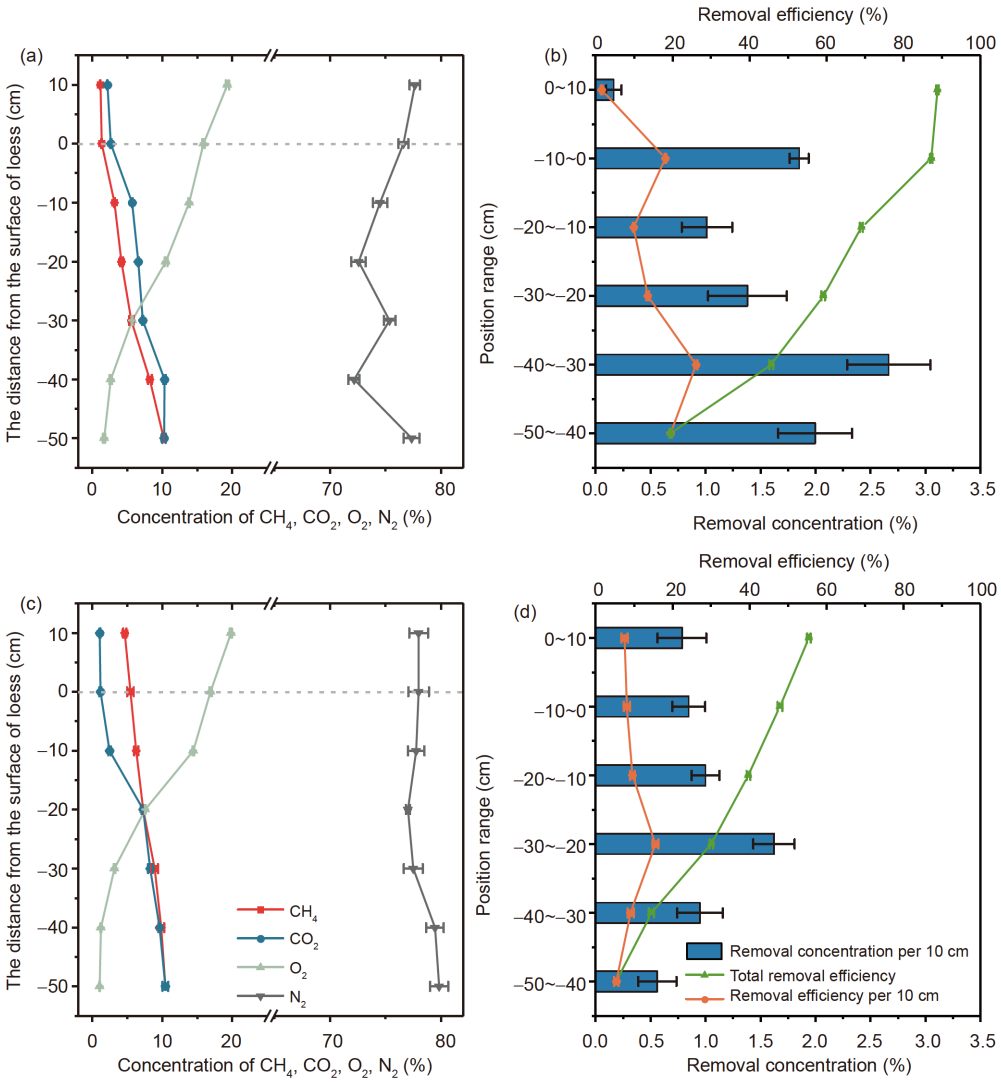

Figure 5 (Color online) The profiles of gas concentration in loess columns and CH_4 removal efficiency at different height. (a) The profile of gas concentration in the CL column; (b) CH_4 removal efficiency at different height in the CL column; (c) the profile of gas concentration in the CW column; (d) CH_4 removal efficiency at different height in the CW column.

than that of water-irrigated control (P<0.01).

In both the columns, the CH_4 removal efficiency of the bottom layer was higher than that of the top layer. The CH_4 removal efficiency of each 10 cm layer in the CL column was 17.4%±6.2% on an average, and the highest CH_4 removal efficiency was noted at 30–40 cm below the surface (26.1% ±0.4%). In the CW column, the CH_4 removal efficiency of each 10 cm layer was 9.6%±3.7% on an average, and reached the highest value of 15.6%±0.8% at 20–30 cm below the surface.

The O_2 concentrations in both the columns gradually dropped from 20% at the surface to less than 2% at the bottom, suggesting effective microbial activity. Furthermore, the concentrations of CO_2 in the CL column were always more than 1.5 vt% higher than those of CH_4 (P<0.01), whereas the concentrations of both CO_2 and CH_4 in the CW column were almost the same at 20 cm below the surface (P>0.05). This finding suggested that the leachate-ameliorated loess layer had stronger CH_4 oxidation capacity and respiratory rate than water-irrigated loess layer.

3.4 Quantity and composition of soil microbial communities

The relative abundance of prokaryotic communities at the phylum level is presented in Figure 6. A total of 54 ASVs were obtained in all the samples. In the original loess, the three bacterial phyla with the highest relative abundance were Actinobacteria (67.1%), α-Proteobacteria (10.0%), and Chloroflexi (8.7%). In the leachate, the three bacterial phyla with the highest relative abundance were Bacteroidetes (40.3%), Firmicutes (21.2%), and α-Proteobacteria (10.8%). In all the leachate-irrigated loess samples, the three bacterial phyla with the highest relative abundance were Actino-

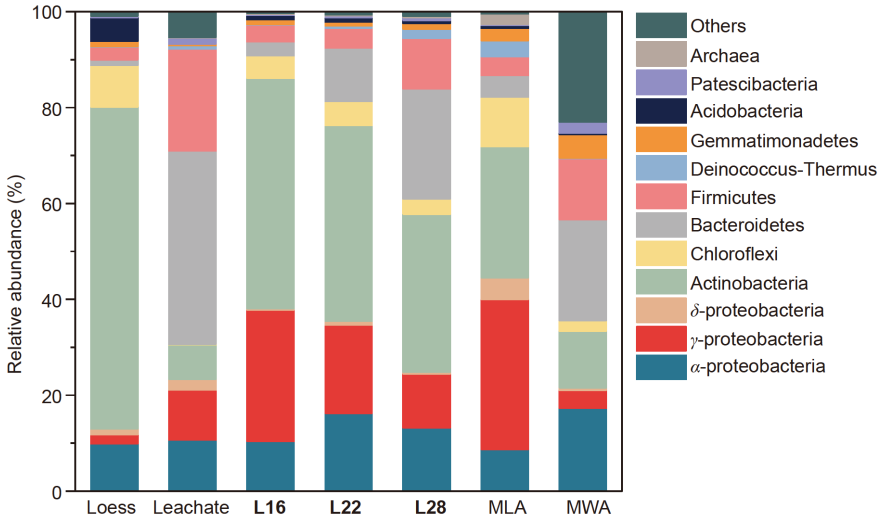

Figure 6 (Color online) Taxonomic composition of prokaryotic communities in different samples at phylum level. Loess: original loess sample; Leachate: mature leachate sample; **L16**: the water content was adjusted to 16 wt% with leachate; **L22**: the water content was adjusted to 22 wt% with leachate; **L28**: the water content was adjusted to 28 wt% with leachate; MLA: the water content was adjusted to 16 wt% with leachate and acclimated in 10 vt% CH_4-rich environment; MWA: the water content was adjusted to 16 wt% with distilled water and acclimated in 10 vt% CH_4-rich environment.

bacteria (32.9%–48.0%), Proteobacteria (24.9%–38.1%), and Bacteroidetes (2.9%–22.9%), implying microbial intervention from leachate irrigation. Furthermore, after acclimation in a CH_4-rich environment, the three bacterial phyla with the highest relative abundance were Proteobacteria (44.5%), Actinobacteria (27.3%), and Chloroflexi (10.4%). The relative abundance of archaea in all the loess samples, except MLA, was below 0.2%, while that in MLA was 2.2%, with the phylum Thaumarchaeota accounting for more than 95%.

With regard to the methanotrophic communities in Proteobacteria, four kinds of type II MOB and three kinds of type I MOB were observed (Figure 7). The total relative abundance of type II MOB in MLA was 2.1 times higher than that in MWA. The relative abundances of Pontibaca methylaminivorans, Methylocystis, and Methyloligellaceae in MLA were 5.8%, 2.0%, and 30% higher than those in MWA. Furthermore, Methylobacterium was not detected in MWA, while the relative abundance of this genus was 2.3% in MLA. The total relative abundance of type I MOB in MLA was 5.0 times higher than that in MWA. Leachate irrigation and CH_4-rich environment acclimation increased the relative abundance of the family Methylophagaceae in γ-Proteobacteria in MLA to 23.8%, which was 39 times higher than that in MWA. However, the relative abundances of Methylococcaceae and Methylophilaceae decreased to 0.4% and 0.2% after leachate irrigation and CH_4-rich environment acclimation, which were only one-fourth and one-tenth of those noted in MWA, respectively.

The total copy numbers of bacteria, archaea, pmoA gene, and amoA gene are shown in Figure 8. After leachate irrigation, the total copy number of bacteria in loess increased by more than 400% (Figure 8(a)). Besides, during CH_4-rich environment acclimation, the total copy number of bacteria kept increasing and reached the maximum on day 14, which was 119 times higher than that observed in the original loess. Leachate irrigation had no significant effect on the number of archaea, which remained at around 1.25×10^4 copies (P>0.05)

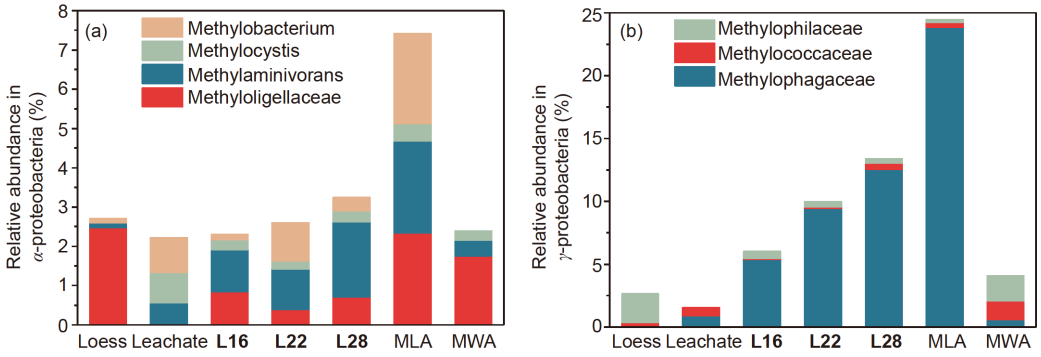

Figure 7 (Color online) Relative abundance of methanotrophic communities in proteobacteria. (a) Type II methane oxidizing bacteria in α-proteobacteria. (b) Type I methane oxidizing bacteria in γ-proteobacteria.

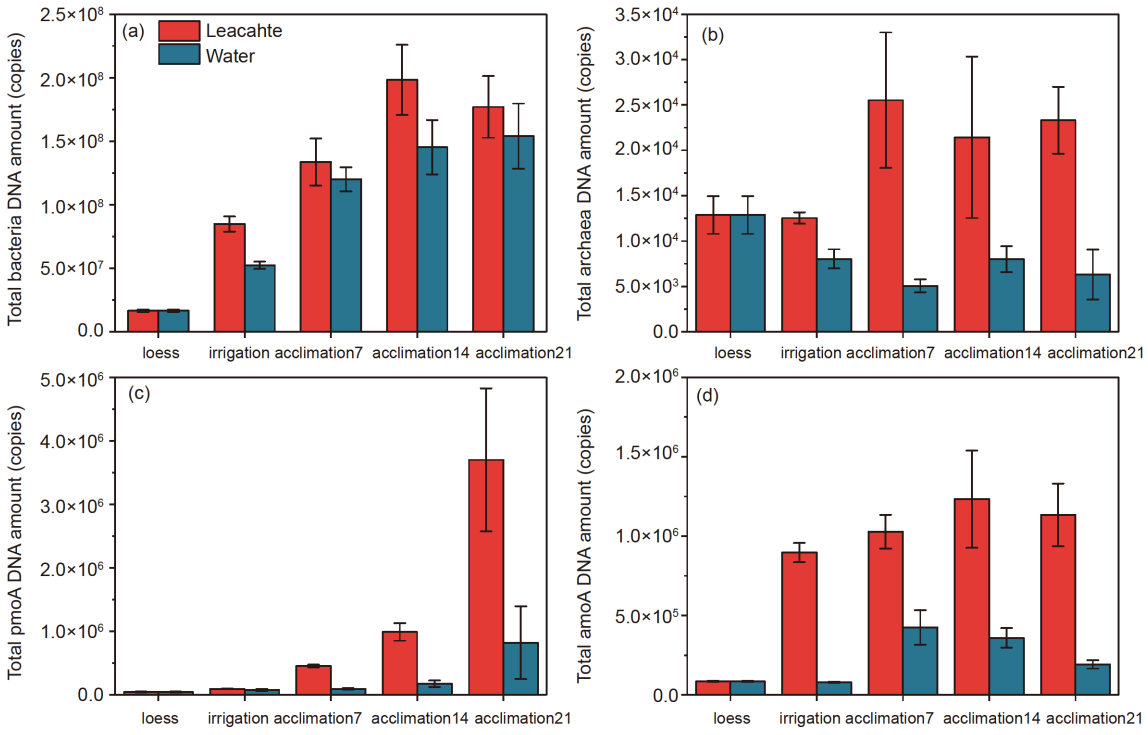

Figure 8 (Color online) Total copy number of bacteria, archaea, pmoA gene and amoA gene in different loess samples. Total copy number in 0.2 g of loess: (a) bacteria; (b) archaea; (c) pmoA gene; (d) amoA gene. Loess: original loess sample; irrigation: loess sample after 95 days' leachate irrigation (**L16**); acclimation7, acclimation14 and acclimation21: leachate-irrigated loess sample after 7, 14 and 21 days' CH_4-rich acclimation.

(Figure 8(b)), while CH_4-rich environment acclimation increased the total copy number of archaea to a maximum of 2.55×10^4 copies on day 7, which was more than 100% higher than that noted in the original loess. Furthermore, CH_4-rich environment acclimation increased the total copy number of pmoA gene to 3.70×10^6 copies from 4.62×10^4 copies, exhibiting more than 79-fold increase (Figure 8(c)). In contrast, leachate irrigation did not significantly increase the quantity of pmoA gene in loess ($P>0.05$). The total copy number of amoA gene was increased by more than 9 and 13 times after leachate irrigation and CH_4-rich environment acclimation, respectively, when compared with that noted in the original loess (8.50×10^4 copies in 0.2 g of original loess, Figure 8(d)).

4 Discussion

4.1 Increase in organic matter content could enhance CH_4 oxidation

The results of the present study revealed that the CH_4 removal ability of loess was significantly improved to 0.66 μmol/(h g-soil) from the initial 0.067 μmol/(h g-soil) ($P<0.001$) (Figure 3(a)) owing to the 5-fold increase in MOB (Figure 7) and 79-fold increase in pmoA gene (Figure 8). Besides, the increase in the soil organic matter could also be partly attributed to the enhanced CH_4 removal ability of loess. The organic matter content of leachate-ameliorated loess was increased by about 2.0 times (>20 g/kg-soil) (Figure 1(a)), reaching the level of some biochar-amended LCS (21.4 g/kg-soil) [41] and compost-amended agricultural soil (21.9 g/kg-soil) [42]. Besides, the organic matter introduced by leachate bound tightly with the soil particles (Figure 2), which was hardly leachable. Soil organic matter is composed of inherently stable and chemically unique compounds such as humus through different inputs [43], and the mature leachate used in the present study could directly increase the humus content of loess and bypass the decomposition process of biomass fragments. Moreover, organic matter can establish a favourable environment for microbial activities [44], and the ratio of BOD_5 to COD in mature leachate was lower than 0.1, which means there was little degradable organic matter in mature leachate so that heterotrophic microorganisms wouldn't accumulate significantly and therefore wouldn't cause extra O_2 consumption.

4.2 Increase in NH_4^+-N content could enhance CH_4 oxidation

A certain concentration of NH_4^+-N has been shown to improve CH_4 oxidation in landfill cover layer because AOB and MOB can co-oxidize CH_4 [45,46]. Previous studies on field and soil microcosm have also reported significant increases in AOB abundance at high NH_4^+-N concentration [47,48],

because in aerobic environment, NH_4^+-N could act as electron donors for AOB. In the present study, leachate amelioration introduced NH_4^+-N into the soil at a level of 0.79, 0.86, and 0.96 mg-N/kg-soil in **L16**, **L22**, and **L28**, resulting in the final N content of 1.26, 1.39, and 1.32 mg-N/kg-soil, respectively (Figure 1(a)). Figure 8(d) shows that the abundance of amoA gene increased by 9.5 times after leachate amelioration. During the entire process of leachate irrigation, the difference between the TN and NH_4^+-N contents gradually increased. The decrease in NH_4^+-N concentration could be owing to two main reasons, namely, ammoxidation by AOB [49] and volatilization of free ammonia triggered by increase in pH (Figure 1(d)). After leachate irrigation, the pH of both **L22** and **L28** increased to 8.5–8.9 and the TN content in **L22** correspondingly reduced to less than 1.0 g/kg-soil, whereas that in **L28** still remained at a high level of 1.3 g/kg-soil, indicating that the relative abundance of AOB, which could convert NH_4^+-N into nitrate nitrogen, was higher in **L28**.

4.3 Leachate amelioration could enhance CH_4 oxidation in CH_4-rich environment

CH_4-rich environment is a well-known factor that can enhance the CH_4 removal ability of soil. Methanotrophic communities are usually abundant and active in some CH_4-rich areas such as geothermal vents [50,51], showing high capacity and affinity kinetics [11]. In the present study, leachate-irrigated loess in CH_4-rich environment presented 2.7-fold increase in CH_4 removal capacity (Figure 3(a)), along with a 1.76- and 3.47-fold increase in CO_2 production and pmoA gene copy number, respectively (Figures 3(a) and 8(c)), when compared with water-irrigated loess. The reason for this increase might be the difference in O_2 availability in the leachate- and water-irrigated loess. Availability of O_2 is a major factor in determining the composition of microbial communities involved in methane oxidation [52]. Nevertheless, further research is needed to confirm whether leachate irrigation could change the granule structure of loess, causing enhancement of O_2 supply level for soil microorganisms. Besides, rainwater leaching test performed in the present study revealed that the pH of the lixivia of leachate-irrigated loess remained at 8.6–8.9, which was higher than that of the lixivia of water-irrigated loess. This result indicated that the buffering property of the leachate-irrigated loess was stronger than that of water-irrigated loess, which was beneficial to microbial growth.

4.4 Contribution of leachate amelioration to mitigate greenhouse gas emission from solid waste in global loess region

The present study proved that leachate amelioration is a feasible technology for soil remediation as LCS. As shown in Figure 5, when 50-cm-thick loess layer was irrigated with leachate, the CH_4 removal efficiency reached 88.8%, which was 1.6 times higher that of the original loess and comparable to that of biochar-amended soil inoculated with enriched MOB (CH_4 removal efficiency of up to 85.2%) [18], waste biocover soil (CH_4 removal efficiency of 94%–96%) [53], yard waste compost mixed soil (CH_4 removal efficiency of up to 55%) [54], or wormcast mixed soil (CH_4 removal efficiency of 53.8%–79.4%) [55]. Furthermore, batch experiments revealed that the CH_4 removal rate of leachate-irrigated loess was up to 1.4 μmol/(h g-soil), which is much higher than that achieved with biochar application (0.05–0.51 μmol/(h g-soil)) [40,56].

Based on the results of the present study, the ameliorated loess is suitable for temporary covering of landfills with thickness of 20–50 cm. If the bulk density of loess was 1.2 g/cm^3, unit CH_4 removal rate was 1.4 μmol/(h g-soil), and loess cover layer thickness was 20 cm, up to 129 g of CH_4 can be bio-oxidized per square meter in a single day; and if the loess cover layer thickness was 50 cm, CH_4 bio-oxidation can reach 322 g/(m^2 d). A full-scale research of CH_4 emission in a loess-gravel capillary barrier cover under semi-arid climates found the maximum CH_4 oxidation rate could reach 93.3 g/(m^2 d) [57]. It can be seen that the CH_4 oxidation rate of loess cover layer can be increased by mature leachate irrigation.

It has been reported that the global concentration of CH_4 in atmosphere has increased to a record level of 1845±2 ppb in 2016 [58], and a single CH_4 molecule has a century-old warming potential that is 25 times higher than that of CO_2. Landfills are the largest source of anthropogenic CH_4, and the CH_4 emissions from landfill working face in different regions range from 67 to 207 g/(m^2 d). In the cases of landfill sites with only cover and no active gas collection, the CH_4 emissions range from 11 to 127 g/(m^2 d) [59]. In particular, landfills in semi-arid climates have been reported to exhibit CH_4 emissions of 85 g/(m^2 d) [59]. Based on these results, the CH_4 emissions from landfills in loess region of semi-arid climate could possibly be mitigated to a carbon-neutral level. As loess region accounts for one-tenth of the world's land area with hundreds of millions of inhabitants, a decrease in the CH_4 emissions to a carbon-neutral level could have a significant positive effect on the control of greenhouse gases and mitigation of global warming.

5 Conclusions

This study presented a method to improve the CH_4 removal capacity of loess soil used as bioactive landfill cover. Overall, mature leachate irrigation increased the organic matter content of loess by 180% (>20 g/kg-soil), and the

organic matter was closely bound to loess soil particles with a leaching rate of less than 10%. The maximum CH_4 removal rate of the ameliorated loess reached 1.41 μmol/(h g-soil), which was 4 times higher than that of water-irrigated loess. Thus, 50-cm-thick mature leachate irrigated loess layer achieved more than 80% CH_4 removal efficiency. Moreover, the results of the present study indicated that mature leachate enhanced the CH_4 bio-oxidation ability of loess by increasing the abundance of type I MOB and methane monooxygenase gene pmoA by 5.0- and 79-fold, respectively. Thus, by using mature leachate irrigated loess as LCS, carbon neutrality can be accomplished for mitigating CH_4 emission from landfills in loess regions, which can have a positive effect on global greenhouse gas control. In addition, potential issues with this technology are worth further exploration. For example, what will be the possible impact of long-term leachate irrigation on loess and whether methane-oxidizing bacteria can maintain stable growth and activity in the dramatically changing soil environment.

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Supporting information

The supporting information is available online at tech.scichina.com and link.springerlink.com. The supporting materials are published as submitted, without typesetting or editing. The responsibility for scientific accuracy and content remains entirely with the authors.

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