

SULFATE REDUCTION BEHAVIOR IN PRESSURE-BEARING LEACHATE SATURATED ZONE

Dongsheng Shen¹, Haomin Zhou¹, Zhiyuan Jin¹, Wenyi Yang¹, Manting Ci¹, Yuyang Long^{1*}, Lifang Hu^{2*}

¹ Zhejiang Provincial Key Laboratory of Solid Waste Treatment and Recycling, School of Environmental Science and Engineering, Instrumental Analysis Center, Zhejiang Gongshang University, Hangzhou, 310012, China

² College of Quality and Safety Engineering, Institution of Industrial Carbon Metrology, China Jiliang University, Hangzhou, 310018, China

ABSTRACT

Attention should be paid to the sulfate reduction behavior in a pressure-bearing leachate saturated zone. In this study, within the relative pressure range of 0–0.6 MPa, the ambient temperature with the highest sulfate reduction rate of 50°C was selected to explore the difference in sulfate reduction behavior in a pressure-bearing leachate saturated zone. The results showed that the sulfate reduction rate might further increase with an increase in pressure; however, owing to the effect of pressure increase, the generated hydrogen sulfide (H₂S) could not be released on time, thereby decreasing its highest concentration by approximately 85%, and the duration extended to about two times that of the atmospheric pressure. Microbial community structure and functional gene abundance analyses showed that the community distribution of sulfate-reducing bacteria was significantly affected by pressure conditions, and there was a negative correlation between DsrB gene abundance and H₂S release rate. Other sulfate reduction processes that do not require DsrA and DsrB may be the key pathways affecting the sulfate reduction rate in the pressure-bearing leachate saturated zone. This study improves the understanding of sulfate reduction in landfills as well as provides a theoretical basis for the operation and management of landfills.

Keywords: Landfill, Pressure, Leachate saturated zone, Sulfate reduction, Odors, Microbial community

INTRODUCTION

Landfills have always been pertinent solutions for the final disposal of municipal solid wastes because of their advantages of simple construction and management, large processing capacity, and low treatment cost (Meyer-Dombard et al., 2020). With the continuous improvement of technical means and operation management levels, landfill sanitation level has significantly improved, but the odor pollution caused by landfill gas emissions continues to cause serious harm to the surrounding environment. Hydrogen sulfide (H₂S) has the characteristics of low odor threshold and high toxicity and is often considered as one of the main

pollutants causing landfill odor pollution (Long et al., 2016). Similarly, volatile organosulfur compounds (VOSCs), such as dimethyl sulfide (DMS) and methyl mercaptan (MM), are considered the main pollutants of landfill odor pollution (Zhang et al., 2017). The above sulfur-containing odorous gases are often found in the odorous pollution of landfills (Jin et al., 2020b). Notably, H₂S, DMS, and MM discharged from landfills come from the sulfate reduction behavior that occurs inside landfills.

Previous studies have shown that active sulfate reduction behavior exists in landfills, mainly due to the high sulfate content of landfills, as well as the anaerobic environment and widespread presence of sulfate-reducing bacteria (SRB) in landfills (Long et al., 2017). Our studies found that as the depth of landfill deepens, there were significant differences in internal water content (Ying et al., 2019), temperature (Jin et al., 2020a), and other environmental factors, affecting SRB to an extent, leading to an impact on sulfate reduction behavior.

In addition, there are shreds of evidence that because of the internal structure as well as material migration and transformation factors, the pressure field in the deep layer of a landfill is formed inside the landfill, making the pressure inside the landfill significantly higher than that on the surface (Giri and Reddy, 2014; Kadambala et al., 2011; Zhang et al., 2021). For the internal pressure field of landfills, there are three main sources of pressure, namely, landfill waste accumulation and extrusion (weight of solid matter) (Benson et al., 2012), leachate silting (pore water pressure) (Tupsakhare et al., 2020), and long-term detention of landfill gas (pore gas pressure) (Ma et al., 2019), which are influenced by multiple factors. First, owing to the accumulation and burial of landfill wastes in the landfill process, the pressure in the bottom layer of a landfill is significantly higher than that of the surface layer, whereas the surface layer's pressure is similar to the atmosphere. With an increase in depth, the pressure on a landfill waste and microorganism changes sequentially (Tupsakhare et al., 2020). Further, owing to the influence of factors, such as the high moisture content of deposited wastes and

landfill management, there is a widespread phenomenon of high leachate production in landfills, leading to the deposition of landfill leachate and formation of leachate saturation zone. Owing to the vertical migration of leachate, pore water pressure in a landfill also increases with an increase in depth (Ma et al., 2019). Finally, by measuring the gas production pressure of landfill gas well, the relative pressure of the gas production is generally less than 0.5 MPa, whereas, if the leachate is retained, the relative pressure of the gas production is between 0.7 and 1.6 MPa (Jafari et al., 2017), attributable to the accumulation of landfill gas due to the clogging of a gas-well pipeline, and the local pressure increases (Zhang et al., 2021). Therefore, the internal pressure field of landfills is common, and the pressure increases gradually in the longitudinal direction. We can assume that a sulfate reduction process dominated by SRB in an actual landfill is affected by a specific pressure field for a long time. Pressure as a physics and thermodynamics parameter, has been proven to have a certain impact on SRB and sulfate reduction behavior in various pressure-bearing environments, such as oceans (Bhattarai et al., 2018; Fichtel et al., 2015), lakes (Cassarini et al., 2019), and reservoirs (Williamson et al., 2018). Unfortunately, to the best of our knowledge, studies on sulfate reduction in pressure-bearing leachate saturated zones are inadequate.

In this study, the sulfate reduction behavior in a landfill leachate saturated zone was simulated under different pressures, especially at a high temperature (50°C), where sulfate reduction behavior was found to be extremely active in previous studies (Jin et al., 2020a). H₂S, MM, and DMS production were investigated, and microbial diversity and functional gene abundance were analyzed. We aim to elucidate the sulfate reduction behavior in landfills as well as provide a reliable theoretical basis for landfill gas management and control strategies.

1. MATERIALS AND METHODS

1.1. Materials

Mineralized wastes used in our experiment were collected from a real landfill site in Beijing, an anaerobic municipal solid waste landfill site. Two parallel sampling points were selected in a closed landfill cell, which was strictly operated based on the landfill process from historical records. Waste samples of different ages were collected at various depths from the two points at the same pace using the well-sampling method (Hu et al., 2019). Holes were drilled at the two sampling points. Subsamples of landfilled wastes from the same depth in each hole were mixed to obtain representative samples. The mineralized wastes were collected at 12 and 15 m and fully mixed.

The inert components, including stone, plastic, glass, and fabric, were removed, and the remaining mineralized wastes were cut into fragments of no more than 1 cm in diameter. All mineralized wastes after

crushing were collected and fully mixed, sealed in a ziplock bag, and stored in a refrigerator at 0°C. Because it is difficult to collect landfill leachate and mineralized wastes simultaneously in an actual landfill site, simulated leachate prepared according to actual leachate is used in the experiment. Table 1 lists the main characteristics of landfills and simulated leachate.

Table 1 The main characteristics of landfill waste and configured leachate.

Components	SO ₄ ²⁻	S ₂ O ₃ ²⁻	S ⁰	S ²⁻
Mineralized waste (mg/kg)	2584.3	/	/	131.4
leachate (mg/L)	1032.2	/	/	/
Components	DOC	NH ₄ ⁺	NO ₃ ⁻	NO ₂ ⁻
Mineralized waste (mg/kg)	1425.7	2021.5	44.9	/
leachate (mg/L)	1212.6	4.42	2.10	/

1.2. Experimental design

Pressure-bearing leachate saturated zone tests were simulated by stainless steel pressure tank reactors. Meanwhile, the normal atmospheric pressure tests were performed in glass tank reactors. All reactors' volumes were 1 L. The lid of the pressure reactor tanks was designed as four connecting ports, connecting pressure gauge, pressure-reducing valve, safety valve, and spherical exhaust valve (Fig. 1).

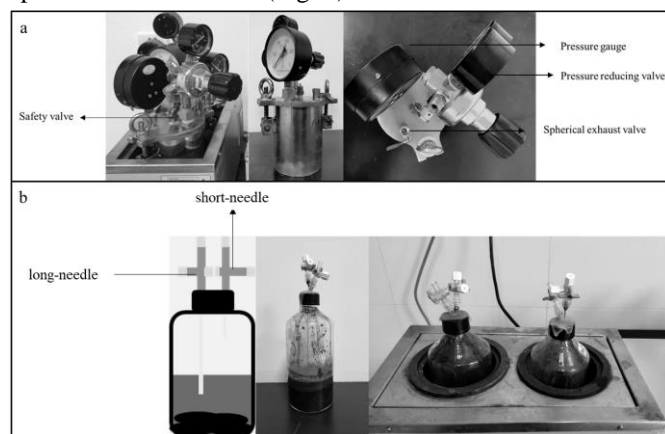


Fig. 1. (a) Pressure reactor diagram; (b) Atmospheric pressure reactor diagram.

The tank lid was sealed with two spiral valves. A pretreated mineralized waste sample (100 g) and 300-mL leachate were added to each reactor, and the initial headspace volume was recorded. The pressure condition of an atmospheric pressure reactor was simulated by

purging nitrogen to the headspace. The relative pressures of the reactors were set to 0.0, 0.2, 0.4, and 0.6 MPa, corresponding to landfill depths of about 0, 20, 40, and 60 m, respectively. The atmospheric pressure reactor used a butyl rubber plug and plastic cap to seal the bottle, and two three-way valves were arranged, connecting long and short-needle tubes, which were connected to each bottle through a bottle cap, and the long needle tube could reach the leachate surface. Meanwhile, the short-needle tube was used to connect the Schiller fermenter to the three-way valve to obtain gas production (Jin et al., 2020a). Two parallel groups were set for each pressure. Pressure reactor sampling, relying on the pressure-reducing valve-fixed release of 0.01 MPa gas, employed a gas collection bag and the same method to restore the pressure in the reactor. Gas-phase samples in the atmospheric pressure reactor are collected via a syringe through a short-needle three-way valve. To make SRB achieve a high sulfate reduction ability in this reaction system and conform to the actual internal landfill temperature, the experimental temperature was set to 50°C (Jin et al., 2020a) and heated in a constant temperature water bath. The test was completed when odor-containing sulfur could not be detected in the reactors.

1.3. Analyses

During the tests, the headspace H_2S , MM, and DMS concentrations of all reactors were determined using a gas chromatograph equipped with a flame photometric detector (GC 7890A, Agilent Technologies, USA) (Zhang et al., 2017).

At the beginning and end of the tests, landfill waste and leachate samples were analyzed for sulfate (SO_4^{2-}), sulfite (SO_3^{2-}), thiosulfate ($\text{S}_2\text{O}_3^{2-}$), sulfur (S^0), sulfide (S^{2-}), nitrate (NO_3^-), nitrite (NO_2^-), and dissolved organic carbon (DOC) after the samples were passed through a 0.22- μm filter. The methods used to determine the concentrations were in accordance with previous studies (Fang et al., 2016; Ying et al., 2019). The abovementioned analyses were performed in triplicate. DNA was extracted from each landfill waste sample (0.1–0.6 g per sample) using a FastDNA Spin kit (MP Biomedicals, USA) according to the manufacturer's instructions. After extraction, the DNA concentration was determined using a NanoDrop2000 ultraviolet-visible (UV – Vis) spectrophotometer (Wilmington, USA). 16S rRNA gene was amplified via polymerase chain reaction (PCR) using 515FmodF (5' - GTGYCAGCMGCCGCGGTAA-3') and 806RmodR (5' -GGACTACNVGGGTWTCTAT-3') primers, and the PCR reaction was in accordance with a previous study (Liu et al., 2018).

In this study, Mothur (Version: 1.30.2) software was used to calculate the α -diversity of microorganisms in samples based on the OTU level. Principal coordinate

analysis (PCoA) was employed to analyze the similarity or difference in microbial community compositions of samples treated with different pressures. We adopted the Bray–Curtis distance algorithm. Microbial β -diversity was analyzed on the basis of the phylum level.

DNA isolation, detection, and PCR amplification were in accordance with Section 2.4. The PCR primers for DsrA and DsrB were DsrA_F (5' - ACSCACTGGAAGCACG-3') and DsrA_R (5' - CAACATCGTYCAYACCCAGGG-3'), respectively. Finally, the abundance of the amplified genes was quantified via real-time quantitative PCR (ABI7500, Applied Biosystems, USA), as previously reported (Hu et al., 2021).

2. Results and discussion

2.1 Release behavior of H_2S and VOSCs under pressure

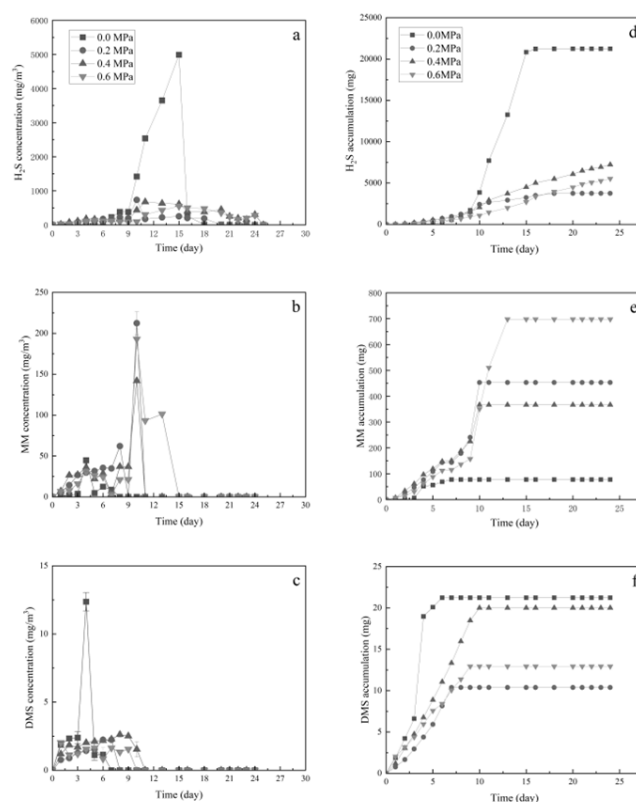


Fig. 2. Emission of (a) hydrogen sulfide (H_2S), (b) methyl mercaptan (MM), and (c) dimethyl sulfide (DMS) under various pressures and accumulation of (d) H_2S , (e) MM, and (f) DMS under various pressures.

The pressure environment in the pressure-bearing leachate saturated zone is harsher than that in the surface saturated zone. The release behaviors of H_2S ,

MM, and DMS under the influence of pressure are shown in Fig. 2. From Fig. 2 (a), H₂S release behavior was significantly inhibited under high pressure. When it was atmospheric pressure, the highest H₂S concentration was in the same order of magnitude as that of the surface saturated zone, which was 4,991.0 mg/m³ on the 15th day. The highest H₂S concentration decreased with an increase in pressure. When the ambient pressure was in the range of 0.2–0.6 MPa, the highest concentration of H₂S was 733.9, 674.1, and 546.2 mg/m³, on the 10th, 11th, and 15th days, respectively, proving that the H₂S release rate would increase in a pressure environment of 0.2–0.4 MPa, but would decrease under higher pressures. In addition, when the pressure was higher than the atmospheric pressure, H₂S concentration maintained the same order of magnitude range for about 10 days after reaching the peak and then decrease to below the detection limit. The decrease rate also showed a downward trend with an increase in pressure, and the decrease process only lasted for two days at 0 MPa, probably due to the increase in the headspace pressure, which limited the release of H₂S and kept it at a relatively low and constant concentration for a long time (Huttenhuis et al., 2008).

Figs. 1 (b) and (c) show the MM and DMS release behaviors, respectively. Unlike with H₂S, the release rates of MM and DMS did not decrease significantly when the pressure was 0 MPa. The highest MM concentration occurred on the 4th day, but the concentration was only 44.7 mg/m³. The highest DMS concentration of 12.4 mg/m³ also appeared on the 4th day. When the pressure was in the range of 0.2–0.6 MPa, the highest MM concentration appeared on the 10th day, corresponding to 62.1, 141.9, and 192.9 mg/m³. The DMS concentration peaked at 2.2 and 2.3, and 1.6 mg/m³ on days 6–7. The highest MM and DMS concentrations generally occurred earlier than the highest H₂S concentration. The releases of H₂S, MM, and DMS were not synchronous, confirming that the main sources of H₂S and VOSCs differed. During microbial activation, some microorganisms that could degrade amino acids were activated first, releasing MM, DMS, and a small amount of H₂S through the degradation process (Jin et al., 2020b). After SRB was fully activated, H₂S began to dominate. This view can be verified by the change in DOC content. Compared with the release behavior of sulfur-containing odorous gas in the leachate saturated zone at different temperatures in the surface layer (Jin et al., 2020a), the DOC content in the landfill waste samples decreased, and some amino acids were decomposed in the preservation process, decreasing the highest MM and DMS concentrations.

Because the H₂S concentration remained at the highest for a long time under high pressure, to eliminate the influence of pressure difference on the apparent concentration value, we analyzed the release capacity of sulfur-containing odorous gas at various pressures from

the cumulative release amount of gas.

The results of significance analysis showed that the cumulative release amounts of H₂S and MM differed significantly when the pressure was 0 MPa compared with other pressure conditions ($P < 0.05$). There was no significant difference between the cumulative release of DMS under 0 and 0.4 MPa pressure environments ($P > 0.05$), significantly different from 0.2 to 0.6 MPa ($P < 0.05$). When the pressure was in the range of 0.2–0.6 MPa, there was no significant difference in H₂S accumulation ($P > 0.05$). According to the above results, when the ambient pressure was higher than the atmospheric pressure, the release of sulfur-containing odorous gas would be immediately affected, the release capacity of H₂S and DMS would decrease, and the release capacity of MM would increase. From the accumulation result of H₂S, the accumulation amount was the lowest at 0.2 MPa. However, while the sulfate reduction behavior was affected, the subsequent transformation of H₂S, including dissolution, oxidation, and precipitation, might also be affected.

As shown in Fig. 2, we can obtain an early indication of the concentration change. When the pressure was 0.2 MPa, H₂S would immediately decline after the peak, but it did not directly return to zero, proving that the phenomenon of concentration decline was due to the increase in the H₂S conversion rate. Combined with the conclusion that the H₂S release rate was fast at 0.2 MPa, the H₂S conversion rate was also higher at higher pressure.

2.2 Sulfate reduction behavior discrepancy

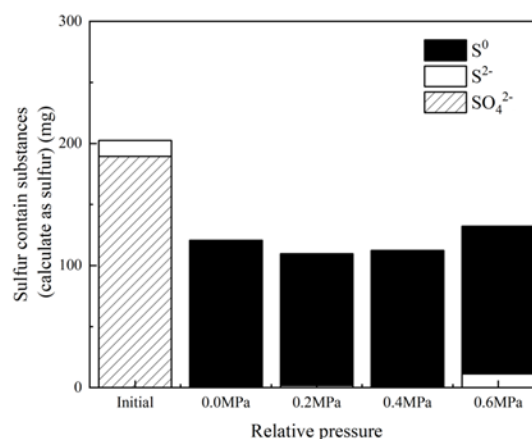


Fig. 3. Sulfate fate under various pressures (calculated as sulfur) after sulfate reduction.

These results proved that there was no difference in the cumulative release amount of H₂S at a pressure higher than the atmospheric pressure. Similar to the sulfate

reduction behavior at different temperatures in the surface saturated zone (Jin et al., 2020a), the reduction product was dominated by sulfur. The accumulation of sulfur in each reactor was 120.1, 107.9, 111.4, and 120.0 mg, with no significant difference ($P > 0.05$) (Fig. 3). The accumulation amount of sulfide was 0.5, 1.8, 0.9, and 11.2 mg, and there was more sulfide accumulation in a 0.6-MPa environment.

Further analyses of the contribution rate of sulfate to sulfide and sulfur are shown in Fig. 3. The analyses showed that it is not difficult to find that the contribution rate of sulfate exceeded 50% under all tested pressures and reached over 60% under the 0.6 MPa pressure. The results showed that pressure did not significantly affect the reducing ability of sulfate and could only fluctuate within a certain range.

Accordingly, wastes under low pressure, in the leachate saturated environment, could release more sulfur-containing odorous gas, increasing the highest concentration of odorous gas. However, for wastes under high pressure, the odorous gas concentration was diluted on the time scale, but it could maintain the odor pollution for a long time; at this pollution level, all sulfates would be reduced.

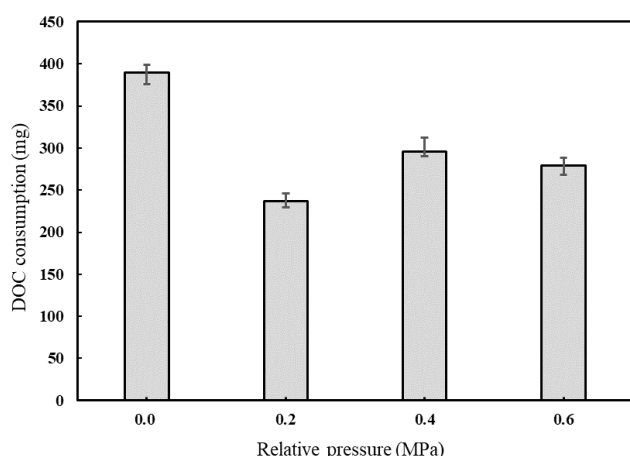


Fig. 4. Dissolved organic carbon (DOC) consumption under various pressures.

Fig. 4 shows DOC consumption under various pressures. Although microorganisms need a certain time to be activated, DOC consumption did not significantly decrease at 0 MPa and was still about 400 mg. However, under higher pressure, DOC consumption decreased to different degrees, and the DOC consumption of 0.2–0.6 MPa was 236.5, 296.1, and 279.1 mg. We found that only DOC consumption at 0 MPa had significant difference ($P < 0.05$). The results showed that high pressure influenced the activity of microorganisms, but the influence degree was not correlated with pressure. That is, the total activity of microorganisms was similar under high pressure. Combined with the difference of sulfate

reduction behavior, we could infer that the difference in microbial community structure under higher pressure was not less obvious than other pressure conditions.

2.3 Functional microbial community structure under different pressures

PCoA was employed to investigate the variation in microbial β -diversity in the pressure environments. After dimension reduction, the explanatory rate of variables was 81.4%, and that of main axes 1 and 2 were 20% and 61.4%, respectively (Fig. 5). The results showed that under different pressure conditions, the microbial community structure in the saturated leachate zone presented three conditions: 0.0 (unpressured), 0.2 (micropressure), 0.4–0.6 MPa (high pressure). Under the high-pressure condition, the distance between quadrats was almost the same as the distance within quadrats, indicating that the change in pressure had little influence on the microbial community structure under the high-pressure condition.

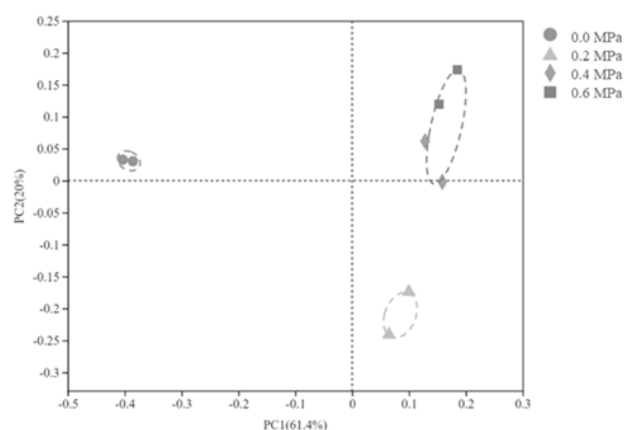


Fig. 5. PCoA analysis on OTU level of samples from various pressures.

Thus, under high pressure, microorganisms in the leachate saturated zone have a strong ability to adapt to pressure, so that pressure cannot affect their community structure. Therefore, in a follow-up study of this study, 0.4 and 0.6 MPa were combined as high-pressure groups, and the mechanism of microbial-mediated sulfate reduction under different pressures was discussed under the unpressured and micropressure conditions.

From the viewpoint of microbial community distribution characteristics, the microbial community structure of the landfill leachate saturated zone under different pressures was further analyzed (Fig. 6). Firmicutes were the dominant microorganisms in the landfill leachate saturated zone, and the abundance of Firmicutes decreased with an increase in environmental pressure. The relative abundance of Firmicutes was as high as 80% in an unconfined environment, but it decreased to 66% and 72% in the micropressure and high-pressure

a

Relative abundance on Phylum level

- Firmicutes
- Proteobacteria
- Chloroflexi
- Haloanaerobactota
- Synergistota
- Halo bacterota
- Bacteroidota
- Actinobacteriota
- others

Atmospheric pressure

Meso pressure

High pressure

b

Relative abundance on Genus level

- Bacillus*
- Hydrogenispira*
- norank_f_norank_s_M803*
- Pseudomonas*
- Ruminococcaceae*
- Halorubra*
- ECC-012*
- norank_f_norank_s_SBR101*
- Clostridium_sensu_strictu*
- Calditerrivactor*
- Haloplasma*
- unclassified_f_Firmicutes*
- HS-AF9186*
- Methanocaldococcus*
- Tepidimicrobium*
- norank_f_norank_s_Terrapinnabacteriota*
- unclassified_f_Bacillales*
- Aeromicrobium*
- Prostaphylinus*
- Delfiafilius*
- Melanconella*
- unclassified_b_norank_d_Bacteriota*
- norank_f_norank_s_MS5-021*
- norank_f_norank_s_norank_y_Limon*
- Syntrophoproduct*
- norank_f_Pseudomonadota-Spirillum*
- norank_f_norank_s_norank_y_norank_o*
- Syntrophomonas*
- Pseudogelatinifilum*
- Symbiodinium*
- others*

Atmospheric pressure

Meso pressure

High pressure

At the genus level, in the unpressured environment, *Bacillus* replaced *Lentimicrobiaceae* as the dominant bacterium in the saturated zone of the surface layer found in previous studies (Jin et al., 2020a) and became the dominant bacterium in the leachate saturated zone. Its relative abundance was close to 40%, indicating that *Bacillus* had good adaptability to temperature. However, *Bacillus* had a low ability to adapt to pressure, its abundance was significantly reduced in the micropressure environment, and its relative abundance was even reduced to less than 5% in the high-pressure environment. In addition, the relative abundance of *Hydrogenispora* and *Pusillimonas* increased in the micropressure environment, and *Hydrogenispora* could still maintain a relative abundance of 11% in the high-pressure environment, showing high adaptability to pressure. In addition to *Hydrogenispora*, the relative abundance of many microbial genera, including *Ruminiclostridium*, increased under the high-pressure condition. Notably, the relative abundance of *MBA03* genus remained between 5% and 10% at different pressures.

According to the Dsr function, SRB screened in GenBank, DDBJ, EMBL, and other databases and

Stacked bar chart showing the relative abundance of bacterial genera at different relative pressures (0.0, 0.2, 0.4, 0.6 MPa). The x-axis represents relative abundance on the genus level (0 to 1). The y-axis represents relative pressure (MPa). The legend identifies the following genera: *Desulfibacter*, *Desulfitobacterium*, *Dethiobacter*, *Desulfotomaculum*, *Candidatus_Desulforudis*, *Desulfosporosinus*, *Desulfovibrio*, and others.

Previous studies had proven that DsrA and DsrB abundances were positively correlated, and both exist continuously (Geets et al., 2006; Zeleke et al., 2013). The DsrB gene is a commonly used indicator gene. Therefore, this study only analyzed the differences in the DsrB functional gene content under different pressures (Fig. 9). At 0.0 MPa, the DsrB gene abundance reached 1.15×10^7 copies·g⁻¹, 57 times higher than that at 50°C in the shallow saturated zone and even higher than the maximum value at 30°C (4.0×10^6 copies·g⁻¹) (Jin et al., 2020a). However, the highest concentration of H₂S did not appear earlier but was delayed to the 15th day. At 0.2–0.6 MPa, the DsrB gene abundance was less than 4.0×10^6 copies·g⁻¹, indicating that SRB containing DsrB decreased in abundance at high pressures.

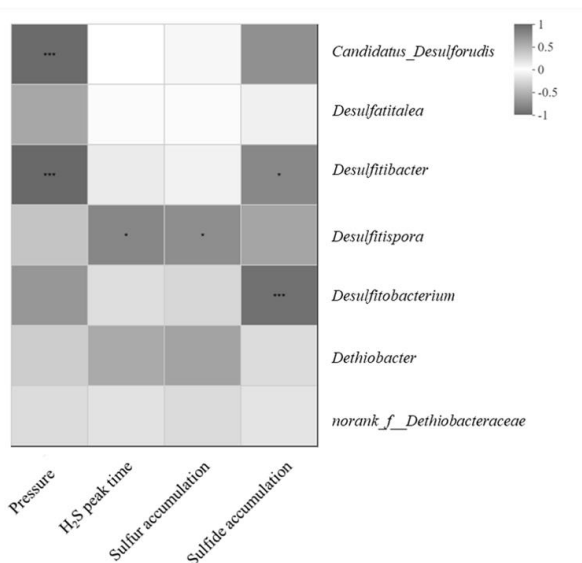


Fig. 8. Correlation analysis of environmental factors and SRB. * and *** denote significant correlation at $P < 0.05$ and $P < 0.001$, respectively (Student's t test); Red and blue respectively indicate the positive and negative correlation between environmental factors and SRB. The darker the color, the higher the correlation.

The abundance of the DsrB gene was negatively correlated with the H₂S release rate, confirming the conclusion in the shallow saturated zone. The direct relationship between the abundance of DsrB related to sulfite and sulfate reduction rates was not obvious. However, DsrA and DsrB were not completely unrelated to the sulfate reduction rate. Studies had shown that within a certain range, the sulfate reduction rate was positively correlated with DsrA abundance; however, when DsrA abundance continues to increase, the sulfate reduction rate did not increase further (Chin et al., 2008). Thus, some other pathway in the saturated zone increased the sulfate reduction rate.

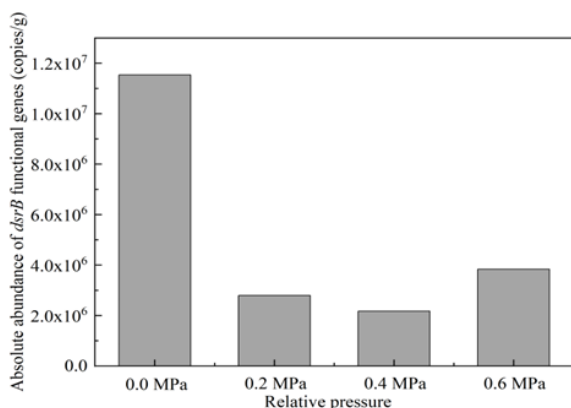


Fig. 9. Quantity DsrB in the samples from reactors under various pressures.

Notably, although the sulfate reduction behavior of the pressure-bearing leachate saturated zone might be performed by various SRB, some key SRB still play a paramount role in rate regulation, and pressure disturbance can change the dominant SRB, significantly affecting its reduction rate. *Dethiobacter* is a key SRB that affects the sulfate reduction rate. *Dethiobacter* can maintain a certain relative abundance in the unpressed and micropressure environments; however, in the high-pressure environments, the abundance of *Dethiobacter* decreased significantly, whereas *Desulfotibacter* had a constant abundance, which may be the main influencing factor.

3. CONCLUSIONS

Sulfate reduction behavior differs with respect to the pressure-bearing condition in a landfill leachate saturated zone. When the relative pressure is 0.0 MPa, the concentration of H₂S decreases rapidly within two days from the peak. In the range of 0.2–0.6 MPa, the highest concentration of H₂S can be further advanced, but its highest value decreases significantly, which may cause persistent odor pollution. The sequencing results of microbial community and functional genes showed that the SRB community structure differed significantly from that of other pressure environments under a relative pressure of 0.0 MPa, whereas the SRB community structure was similar under different pressures of 0.2–0.6 MPa. The content of SRB and the DsrB functional genes of *Dethiobacter* were positively and negatively correlated with the sulfate reduction rate, respectively. Other sulfate reduction processes without DsrA and DsrB enzymes may be the key microorganisms affecting the sulfate reduction rate in the leachate saturated zone. Finally, sulfate reduction behavior in pressure-bearing leachate saturated zone, which is the focus of this study, further indicates the long-term risk of odor contamination in the landfill leachate saturated zone. Therefore, strengthening the management of landfill gas collection and landfill leachate drainage and considering the elevation of landfill may provide some help to the source control of landfill odor pollution.

REFERENCES

- Benson, C.H., Edil, T.B., Wang, X., 2012. Evaluation of a final cover slide at a landfill with recirculating leachate. *Geotext. Geomembr.* 35, 100–106.
- Bhattacharai, S., Zhang, Y., Lens, P.N.L., 2018. Effect of pressure and temperature on anaerobic methanotrophic activities of a highly enriched ANME-2a community. *Environ. Sci. Pollut. Res. Int.* 25, 30031–30043.
- Cassarini, C., Zhang, Y., Lens, P.N.L., 2019. Pressure selects dominant anaerobic methanotrophic phylotype and sulfate reducing bacteria in coastal marine Lake Grevelingen sediment. *Front. Environ. Sci.* 6, 162.
- Chin, K.J., Sharma, M.L., Russell, L.A., O'Neill, K.R.,

- Lovley, D.R., 2008. Quantifying expression of a dissimilatory (bi) sulfite reductase gene in petroleum-contaminated marine harbor sediments. *Microb. Ecol.* 55, 489-499.
- Fang, Y., Zhong, Z., Shen, D., Du, Y., Xu, J., Long, Y., 2016. Endogenous mitigation of H₂S inside of the landfills. *Environ. Sci. Pollut. Res. Int.* 23, 2505-2512.
- Fichtel, K., Logemann, J., Fichtel, J., Rullkötter, J., Cypionka, H., Engelen, B., 2015. Temperature and pressure adaptation of a sulfate reducer from the deep subsurface. *Front. Microbiol.* 6, 1078.
- Geets, J., Borremans, B., Diels, L., Springael, D., Vangronsveld, J., van der Lelie, D., Vanbroekhoven, K., 2006. DsrB gene-based DGGE for community and diversity surveys of sulfate-reducing bacteria. *J. Microbiol. Methods* 66, 194-205.
- Giri, R K, Reddy, K R, 2014. Slope stability of bioreactor landfills during leachate injection: Effects of heterogeneous and anisotropic municipal solid waste conditions. *Waste Manag. Res.* 32, 186-197.
- Hu, L.F., Nie, Z.Y., Wang, W.J., Zhang, D.C., Long, Y.Y., Fang, C.R., 2021. Arsenic transformation behavior mediated by arsenic functional genes in landfills. *J. Hazard. Mater.* 403, 123687.
- Huttenhuis, P.J.G., Agrawal, N.J., Versteeg, G.F., 2008. The solubility of hydrogen sulfide in aqueous N-methyldiethanolamine solutions. *Int. J. Oil. Gas. Coal. T.* 1, 399-424.
- Jafari, N.H., Stark, T.D., Thalhamer, T., 2017. Spatial and temporal characteristics of elevated temperatures in municipal solid waste landfills. *Waste Manage.* 59, 286-301.
- Jin, Z., Ci, M., Yang, W., Shen, D., Hu, L., Fang, C., Long, Y., 2020a. Sulfate reduction behavior in the leachate saturated zone of landfill sites. *Sci. Total Environ.* 730, 138946.
- Jin, Z.Y., Zhang, S.Y., Hu, L.F., Fang, C.R., Shen, D.S., Long, Y.Y., 2020b. Effect of substrate sulfur state on MM and DMS emissions in landfill. *Waste Manage.* 116, 112-119.
- Kadambala, R, Townsend, T G, Jain, P, Singh, K, 2011. Temporal and Spatial Pore Water Pressure Distribution Surrounding a Vertical Landfill Leachate Recirculation Well. *Int. J. Environ. Res. Public Health* 8(5), 1692-1706.
- Liu, W.J., Long, Y.Y., Fang, Y., Ying, L.Y., Shen, D.S., 2018. A novel aerobic sulfate reduction process in landfill mineralized refuse. *Sci. Total Environ.* 637, 174-181.
- Long, Y., Fang, Y., Shen, D., Feng, H., Chen, T., 2016. Hydrogen sulfide (H₂S) emission control by aerobic sulfate reduction in landfill. *Sci. Rep.* 6, 38103.
- Long, Y., Zhang, S., Fang, Y., Du, Y., Liu, W., Fang, C., Shen, D., 2017. Dimethyl sulfide emission behavior from landfill site with air and water control. *Biodegradation* 28, 327-335.
- Ma, P., Ke, H., Lan, J., Chen, Y., He, H., 2019. Field measurement of pore pressures and liquid-gas distribution using drilling and ERT in a high food waste content MSW landfill in Guangzhou, China. *Eng. Geol.* 250, 21-33.
- Melton, E.D., Sorokin, D.Y., Overmars, L., Lapidus, A.L., Pillay, M., Ivanova, N., del Rio, T.G., Kyrpides, N.C., Woyke, T., Muyzer, G., 2017. Draft genome sequence of *Dethiobacter alkaliphilus* strain AHT1(T), a gram-positive sulfidogenic polyextremophile. *Stand. Genom. Sci.* 12, 57.
- Meyer-Dombard, D.R., Bogner, J.E., Malas, J., 2020. A review of landfill microbiology and ecology: A call for modernization with 'next generation' technology. *Front. Microbiol.* 11, 1127.
- Nielsen, M.B., Kjeldsen, K.U., Ingvorsen, K., 2006. *Desulfitibacter alkalitolerans* gen. nov., sp. nov., an anaerobic, alkalitolerant, sulfite-reducing bacterium isolated from a district heating plant. *Int. J. Syst. Evol. Microbiol.* 56, 2831-2836.
- Sorokin, D.Y., Tourova, T.P., Muyzer, G., 2013. Isolation and characterization of two novel alkalitolerant sulfidogens from a Thiopaq bioreactor, *Desulfonatronum alkalitolerans* sp nov., and *Sulfurospirillum alkalitolerans* sp nov. *Extremophiles* 17, 535-543.
- Tupsakhare, S., Moutushi, T., Castaldi, M.J., Barlaz, M.A., Luettich, S., Benson, C.H., 2020. The impact of pressure, moisture and temperature on pyrolysis of municipal solid waste under simulated landfill conditions and relevance to the field data from elevated temperature landfill. *Sci. Total Environ.* 723, 138031.
- Williamson, A.J., Carlson, H.K., Kuehl, J.V., Huang, L.L., Iavarone, A.T., Deutschbauer, A., Coates, J.D., 2018. Dissimilatory sulfate reduction under high pressure by *Desulfovibrio alaskensis* G20. *Front. Microbiol.* 9, 1465.
- Ying, L.Y., Long, Y.Y., Yao, L.H., Liu, W.J., Hu, L.F., Fang, C.R., Shen, D.S., 2019. Sulfate reduction at micro-aerobic solid-liquid interface in landfill. *Sci. Total Environ.* 667, 545-551.
- Zelege, J., Sheng, Q., Wang, J.G., Huang, M.Y., Xia, F., Wu, J.H., Quan, Z.X., 2013. Effects of spartina alterniflora invasion on the communities of methanogens and sulfate-reducing bacteria in estuarine marsh sediments. *Front. Microbiol.* 4, 243.
- Zhang, S., Long, Y., Fang, Y., Du, Y., Liu, W., Shen, D., 2017. Effects of aeration and leachate recirculation on methyl mercaptan emissions from landfill. *Waste Manage.* 68, 337-343.
- Zhang, X., Wang, J., Bao, S., Zhang, X., 2021. Impact of internal conditions on the gas flow path in semi-aerobic landfill reactors. *Sci. Total Environ.* 770, 144673.
- Zhang, T, Shi, J Y, Wu, X, Lin, H, Li, X L, 2021. Simulation of gas transport in a landfill with layered new and old municipal solid waste. *Sci. Rep.* 11(1), 9436.