

INSIGHTS INTO THE LANDFILL LEACHATE PROPERTIES AND BACTERIAL STRUCTURE SUCCESSION RESULTING FROM THE COLANDFILLING OF MUNICIPAL SOLID WASTE AND INCINERATION BOTTOM ASH

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ABSTRACT

Four simulated bioreactors were loaded with only MSW, 5% BA+MSW, 10% BA+MSW and 20% BA+MSW to investigate the leachate property and bacterial community change trends during the colandfilling process. The results showed that with increasing BA addition proportion (5%~20%), the leachate oxidation–reduction potential (ORP) was lower, the leachate pH quickly entered the neutral stage, and the chemical oxygen demand (COD), volatile fatty acids (VFA), $\text{NH}_4^+\text{-N}$, Ca^{2+} and SO_4^{2-} presented faster downward trends. BA can quickly increase bacterial diversity, and the higher the addition proportion of BA, the more significant the change in microbial community structure during the landfilling process. The leachate pH and COD greatly influenced the bacterial community structure. A low BA proportion can increase metabolism pathway abundance during the initial stage, but a high BA proportion had an inhibitory effect on the metabolism pathway.

KEYWORDS: Municipal solid waste (MSW), bottom ash (BA), colandfilling, leachate property, microbial community

1. INTRODUCTION

The increasing amount of municipal solid waste (MSW) has been one of the most serious environmental issues restricting economic and social development (Wijekoon et al., 2022). Landfilling is one of the main disposal methods for MSW due to its

low construction cost, simple operation and wide applicability; however, it also poses numerous problems, such as the occupation of a large amount of land, the requirement of long stabilization times and secondary pollution problems (Cheng et al., 2020). Therefore, the long-term stabilization process and operation maintenance of landfills are bottlenecks that restrict landfill technology development. MSW incineration generates a large amount of bottom ash (BA), accounting for approximately 20~25% of the MSW amount (Al-Ghouti et al., 2021). To facilitate the final disposal of slag, the “Standard for Pollution Control on the Landfill Site of Municipal Solid Waste (GB 16889-2008)” stipulated that BA can be directly disposed of in MSW landfill sites in China.

BA has a strong acid-neutralizing capacity, and the release of alkaline substances, such as CaO and K_2O , can effectively improve the low pH value of the leachate, eliminate organic acid accumulation, and potentially accelerate methane production (He et al., 2017). MSW degradation is the result of the synergistic effects of different groups of microorganisms. The microorganisms in landfills utilize organic matter to continuously grow and multiply and accelerate the biodegradation of organic components, which plays a key role in the landfill stabilization process (Wei et al., 2017). An investigation of the bacterial communities in landfills and leachates showed that Firmicutes was predominantly present in all landfills, while

Actinobacteria and Proteobacteria also played important roles in refuse degradation. Wang et al. (2021c) investigated the succession of the bacterial community in refuse of landfills of different ages and found that the bacterial community compositions in the refuse of landfills of similar ages were highly similar, and bacterial community succession occurred continuously with landfill aging. Nevertheless, the effects of BA under different codisposal ratios on the microbial community and the relation between microbial and leachate properties during the landfilling process remain unclear.

In this study, simulated bioreactor landfills in which BA and MSW were mixed at different ratios were constructed. The effects of different codisposal ratios of BA on the variations in leachate properties as well as the leachate stabilization degree were explored. Moreover, the succession of the microbial community and the relationships between the microbial community and the leachate properties were illuminated. In addition, the effects of BA on the microbial function characteristics and metabolism pathways were illustrated. This research can help further understand the impacts of BA on refuse degradation and landfill stabilization.

2. MATERIALS AND METHODS

2.1 Experimental device

Four simulated anaerobic bioreactor landfill columns denoted A, B, C, and D were constructed. A was a control group that was loaded with only 11.25 kg MSW, while B, C, and D were loaded with BA and MSW at mass ratios of 1:20 (0.56 kg BA and 11.25 kg MSW), 1:10 (1.13 kg BA and 11.25 kg MSW), and 1:5 (2.25 kg BA and 11.25 kg MSW)

2.2 Experimental procedure

Before the experiment, 3 L of tap water was added to each column to maintain a suitable moisture content for the refuse biodegradation. During stage I (1~155 days), the bioreactors were operated anaerobically without leachate recirculation, and the leachate samples were collected through the leachate collection pipe every month. During stage II

(156~365 days), a total volume of 3.0 L liquid (in order to avoid the long-term acidification of the system caused by leachate recirculation, a mixture of 2.5L leachate and 0.5L distilled water was used for recirculation) was recirculated into the column once a week by using a peristaltic pump, and the leachate samples were collected every week. After that, the leachate was recirculated, and then the columns were sealed until the next cycle.

2.3 Analytical methods

The physicochemical properties of the leachate during the landfilling process were determined. The pH was determined by using a pH meter. The EC was determined by using a conductivity meter. The ORP was determined by using an ORP meter. The alkalinity was determined by using an automatic potentiometric titrator. The chemical oxygen demand (COD) was determined by the potassium dichromate method. The Ca^{2+} and volatile fatty acids (VFA) were determined by ethylene diamine tetraacetic acid (EDTA) titration and distillation titration, respectively. $\text{NH}_4^+\text{-N}$ was determined by spectrophotometry.

Solid samples (10 g) were collected at different degradation stages, including the initial stage (on Day 7), middle stage (on Day 176), late stage (on Day 267) and final stage (on Day 351), to analyze the microbial community.

2.4 Data analysis

Redundancy analysis (RDA) was performed using the Vegan package in R, based on KEGG annotation and PICRUST software to analyze metabolic pathways. SPSS 22.0 was used to analyze the correlation between bacterial community structure and environmental factors.

3. RESULTS AND DISCUSSION

3.1 Changing leachate property trends during the colandfilling process

3.1.1 ORP, EC, pH and alkalinity

The leachate ORP, EC, pH and alkalinity change trend over time in each column is shown in Fig. 1. During stage I, the ORP change trend in column B was similar to that in column A, with values

fluctuating in the range of 0~-100 mV, while the ORP values in columns C and D were much lower than those in columns A and B. During stage II, the ORP values in columns C and D increased during Days 155~176 and then presented decreasing trends. Overall, the order of the ORP values was $D < C < B < A$. The results showed that the columns loaded with BA and MSW had lower reduction potentials, which was conducive to the refuse entering the anaerobic biodegradation process. During stage I, the order of the EC values in each column was $D > C > B > A$, which was greatly related to the initial BA addition amount. During stage II, the EC values in all columns presented gradually decreasing trends, and the order of the corresponding decreasing rates was $A < B < C < D$. During stage I, the leachate pH in each column

varied from 5.3 to 5.7, 5.3 to 6.1, 5.9 to 6.2, and 6.7 to 7.0, and the order of the leachate pH values in each column was $A < B < C < D$. During stage II, the leachate pH in column A was always acidic and significantly lower than that in columns B, C, and D. The results indicated that BA addition can effectively improve the system pH to prevent landfill system instability due to acid accumulation (He et al., 2017). During stage I, the leachate alkalinity in all columns showed increasing trends, and the values were in the order of $A < B < D < C$. During stage II, The increased alkalinity of the recirculated leachate played a buffering role and alleviated the initial acidification phenomenon, which further accelerated organic matter degradation (He et al., 2017; Wang et al., 2021b).

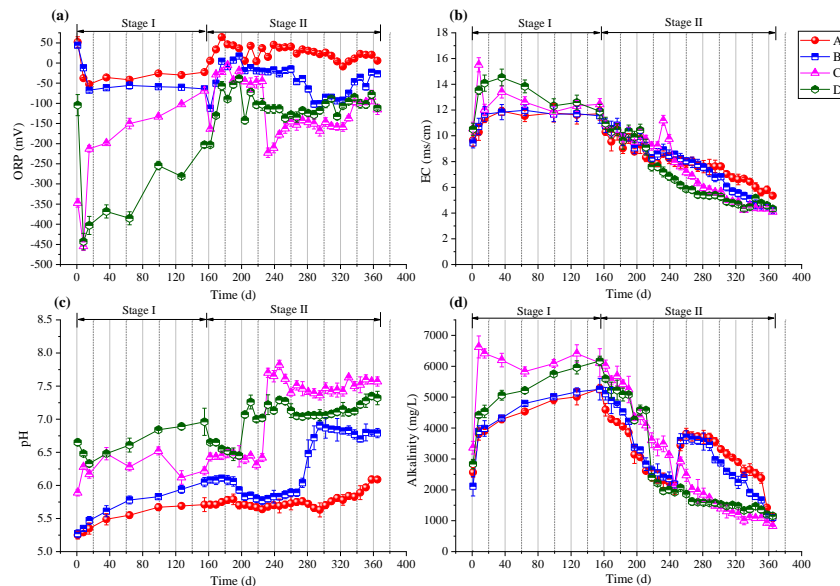


Fig. 1 The changing trends of leachate ORP (a), EC (b), pH (c), and alkalinity (d) in each column during the colandfilling of MSW and BA (A, B, C and D represented the columns that loaded with only MSW, 5% BA+MSW, 10% BA+MSW and 20% BA+MSW, respectively).

3.1.2 COD, VFA, Ca^{2+} , $\text{NH}_4^+\text{-N}$, SO_4^{2-} and S^{2-}

The leachate COD, VFA, Ca^{2+} , $\text{NH}_4^+\text{-N}$, SO_4^{2-} , and S^{2-} change trend in each column over time is shown in **Fig. 2**. During stage I, the leachate VFA increasing trends in all columns occurred because the complex organic matter in the refuse was hydrolyzed and fermented into soluble small-molecule organic matter under the action of hydrolysis and

fermentation microorganisms (Luo et al., 2022). During stage II, the leachate VFA concentration in each column decreased because a large amount of acid was consumed by methanogens. In the late landfill period, the order of the leachate VFA concentration was $D < C < B < A$, which also indicated that the addition of slag can reduce the leachate VFA concentration and accelerate the refuse

stabilization process (Lo and Liao, 2007). During stage I, the leachate Ca^{2+} concentration in all columns showed increasing trends, and the peak concentrations in columns C and D of 5438 mg/L and 5410 mg/L were slightly higher than those in columns A and B. During stage II, the leachate Ca^{2+} concentration showed decreasing trends in all columns. The leachate Ca^{2+} concentration gradually decreased to 1400 mg/L and 900 mg/L in columns A and B, respectively, by the end of the experiment.

The decreasing leachate $\text{NH}_4^+\text{-N}$ concentration was mainly ascribed to the dilution effect of recirculated leachate with 0.5 L distilled water every

cycle. The decreasing trends in the other three columns were more obvious than that in column A, which was related to the absorption effect of BA (Yao et al., 2015). During stage I, the leachate SO_4^{2-} concentration in all columns increased rapidly to peak values during the first several days, after which it gradually decreased. During stage II, the leachate SO_4^{2-} concentration drastically fluctuated and presented sharp increases, with values of 483, 644, 759 and 379 mg/L in each column during Days 162~176. The leachate S^{2-} concentration was relatively low during the whole process.

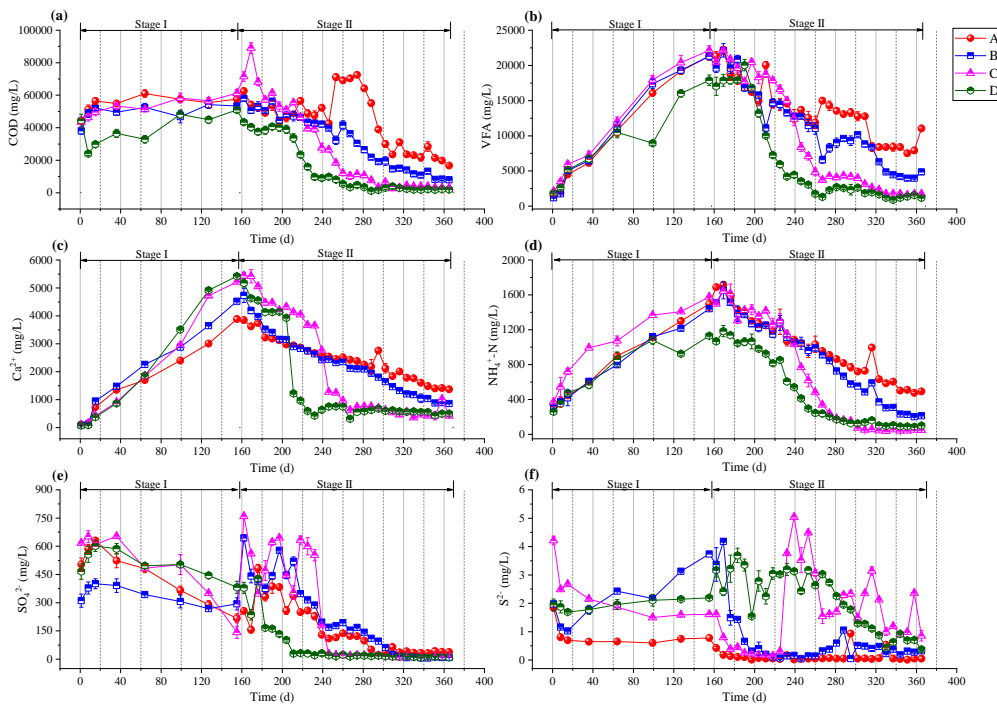


Fig. 2 The changing trends of leachate COD (a), VFA (b), Ca^{2+} (c), $\text{NH}_4^+\text{-N}$ (d), SO_4^{2-} (e), and S^{2-} (f) in each column during the colandfilling of MSW and BA (A, B, C and D represented the columns that loaded with only MSW, 5% BA+MSW, 10% BA+MSW and 20% BA+MSW, respectively).

3.2 Bacterial community structure succession at the phylum and genus levels

(1) Phylum level

The bacterial community structure in each column at the phylum level during the colandfilling process is shown in **Fig. 3**. The structure of the dominant phyla in column A varied slightly during the whole landfilling process. The structure of the

dominant phyla in column B showed obvious differences from that of column A during the initial stage, but it was similar to that of column A during the middle, late and final stages. For columns C and D, the structures of the dominant phyla showed significant differences from those of columns A and B, especially in the initial, late and final stages. These results indicated that BA addition can alter the

microbial community structure, and the higher the addition proportion is, the more significant the change in the microbial community structure in the landfilling process. During the waste degradation process, the predominant phyla were Firmicutes, Proteobacteria and Bacteroides, which was in accordance with previous studies reported by Liu et al. (2019) and Morita et al. (2020). Köchling et al. (2015) and Wang et al. (2021c) reported that Firmicutes and Proteobacteria were the dominant microbial communities associated with lignocellulosic and carbohydrate degradation during the early stage of a landfill, while with increasing landfill age, Bacteroidetes, which can degrade complex refractory organic compounds, began to play a dominant role.

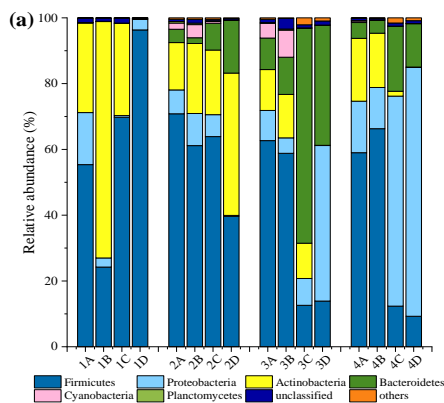


Fig.3 The bacterial community structure at the phylum levels in each column (1, 2, 3 and 4 represent the initial, middle, late and final stages of landfill process, respectively).

(2) Genus level

The changes in the bacterial community at the genus level in each column during different landfill stages are shown in **Fig. 4**. Overall, *Lactobacillus* and *Propionibacterium* were the major genera during the initial degradation stage, while over time, *Prevotella*, *Streptophyta*, and *Acinetobacter* were involved in the landfilling process, and *others* and *unclassified* accounted for high proportions in the late and final stages. Luo et al. (2020) pointed out that *Lactobacillus* is the dominant bacterium that secretes amylase. The presence of *Propionibacterium* can significantly increase the propionic acid concentration

in VFAs (He et al., 2019), and the succession of *Propionibacterium* was greatly related to the variation of VFA (Wei et al., 2021).

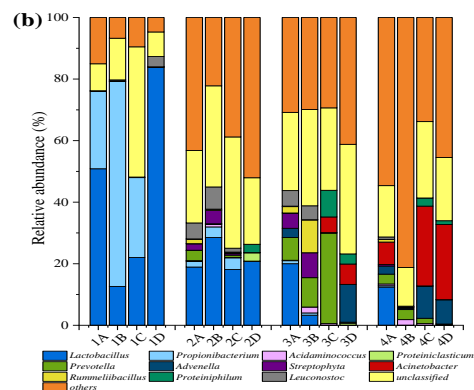


Fig.4 The bacterial community structure at the genus levels in each column (1, 2, 3 and 4 represent the initial, middle, late and final stages of landfill process, respectively).

3.3 Correlations between the bacterial community structure and the landfill environment

The RDA results of the bacteria and environmental factors at the genus level in the mixed landfilling process is shown in **Fig. 5**. The COD and EC are the main environmental factors affecting the microbial community structure in the early stage of waste degradation. The pH and alkalinity have great impacts on the microbial community in the middle and late stages of waste degradation. The COD and alkalinity were the main influencing factors of bacterial community structure in the MSW-alone system and 5%-BA system, while the pH, $E_{300/400}$ and ORP were the main influencing factors of bacterial community structure in the columns with higher BA proportions. The order of influence of the leachate properties on the bacterial community structure was $pH > COD > EC > E_{300/400} > alkalinity > ORP$. Moreover, the genera *Lactobacillus* and *Propionibacterium* had strong positive correlations with COD and EC, while *Pseudomonas*, *Carnobacterium*, *Acinetobacter*, and *Corynebacterium* were positively correlated with pH. Wang et al. (2021c) and Zhao et al. (2021) reported that the effects of pH, organic matters, total Kjeldahl nitrogen

and EC were the major environmental factors that influencing the variation and distribution of bacterial community in landfills.

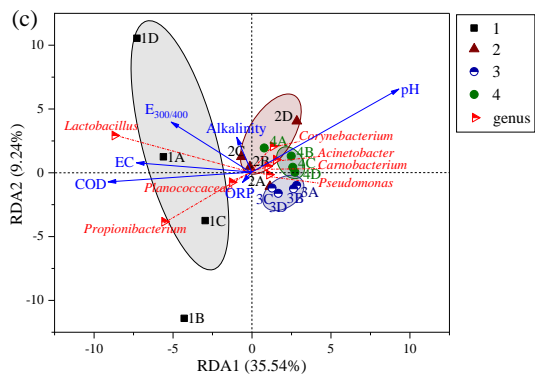
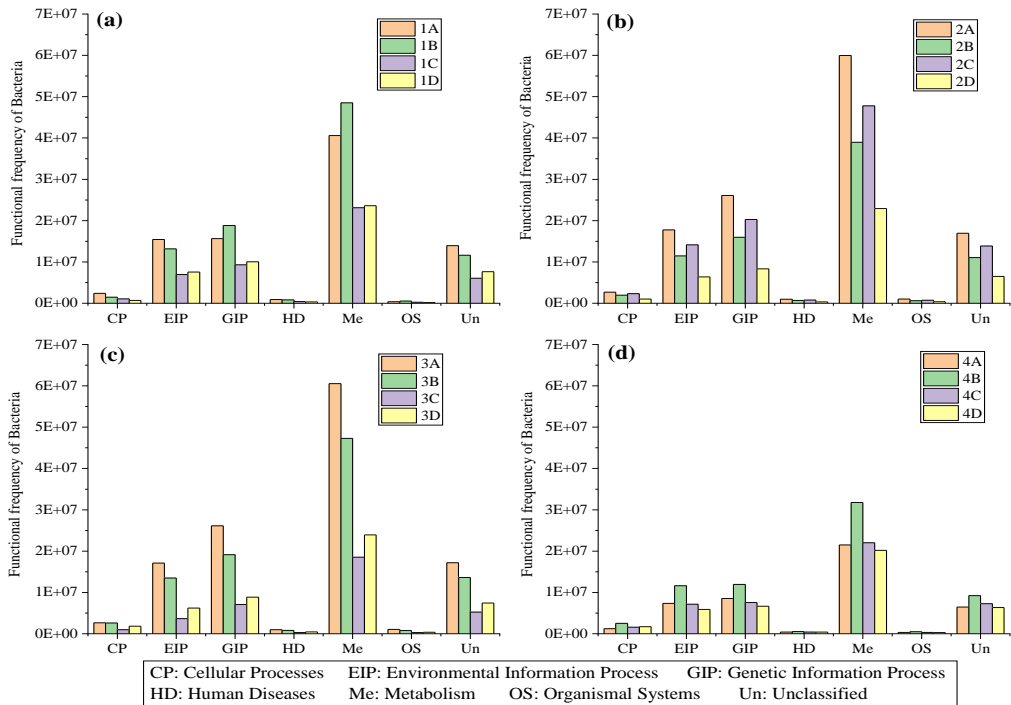


Fig.5 The redundancy analysis of bacteria and environmental factors at the genus level during the co-landfill process (1, 2, 3 and 4 represent the initial, middle, late and final stages of landfill process, respectively).

3.4 Functional characterization of the bacterial community

The functional properties of the bacterial

communities in different landfill systems during waste degradation based on the KEGG pathway at levels I and II are shown in **Fig. 6**. The KEGG level I annotation of the bacterial community indicated that metabolism (Me), genetic information process (GIP) and environmental information process (EIP) were the main metabolic pathways. The KEGG level II annotation of the bacterial community showed that carbohydrate and amino acid metabolism were the dominant metabolic pathways in each system, followed by energy metabolism, coenzyme factor and vitamin metabolism, nucleic acid metabolism, and lipid compound metabolism. Overall, during the initial stage, a low BA addition proportion increased metabolism pathway abundance, but a high BA addition proportion had an inhibitory effect on the metabolism pathway. With refuse degradation, the metabolism abundance in each column increased, but the metabolism abundance in column A was higher than that in columns loaded with BA, which was in accordance with the variation in leachate COD.



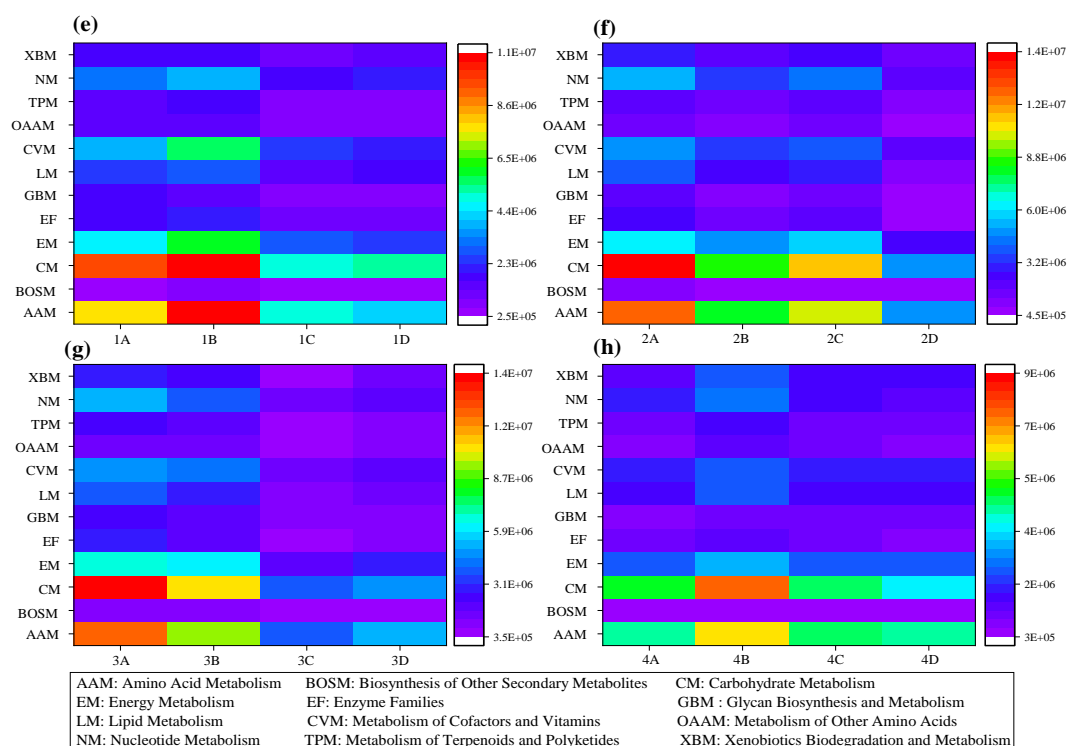


Fig. 6 The bacterial functional abundance based on KEGG level I (a, b, c, d) and level II (e, f, g, h) annotations in each column during the colandfilling of MSW and BA (1, 2, 3 and 4 represent the initial, middle, late and final stages of landfill process, respectively).

4. CONCLUSIONS

When incineration BA was codisposed with MSW during the landfilling process, the leachate pH could quickly enter the neutral stage, the COD, VFA, $\text{NH}_4^+\text{-N}$, Ca^{2+} and SO_4^{2-} presented faster downward trends. BA can quickly increase bacterial diversity, while the long-term selectivity of BA in the middle and late stages of the landfilling process resulted in a certain decrease in bacterial community diversity. *Lactobacillus* and *Propionibacterium* were the major genera during the initial degradation stage, while over time, *Prevotella*, *Streptophyta*, and *Acinetobacter* were involved in the landfilling process. Carbohydrate and amino acid metabolism were the dominant metabolic pathways in each system.

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