The acceleration degradation processes of different aged refuses with the forced aeration for landfill reclamation

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Abstract

Forced aeration is one of the promising ways to accelerate landfill reclamation, and understanding the relation between aeration rates and waste properties is the prerequisite to implementing forced aeration under the target of energy saving and carbon reduction. In this work, landfill reclamation processes with forced aeration were simulated using aged refuses (ARs) of 1, 4, 7, 10, and 13 disposal years, and the potential of field application was also investigated based on a field project, to identify the degradation rate of organic components, the O_2 consumption efficiency and their correlations to microbes. It was found that the removal rate of organic matters declined from 20.3% (AR_1) to 12.6% (AR_{13}), and that biodegradable matter (BDM) decreased from 5.2–2.4% at the set aeration rate of 0.12 L O_2/kg DM/day, respectively. A linear relationship between degradation rate constant (K) of BDM and disposal ages (x) was established: K=-0.0002193x + 0.0091 (R^2 = 0.854), suggesting that BDM might be a suitable indicator to reflect the stabilization of ARs. The cellulose/ lignin ratio decrease rate for AR_1 (18.3%) was much higher than AR_{13} (3.1%), while the corresponding humic-acid/ fulvic-acid ratio increased from 1.44 to 2.16. The dominant bacteria shift from *Corynebacterium* (9.2%), *Acinetobacter* (6.6%), and *Fermentimonas* (6.5%), genes related to the decompose of biodegradable organics, to *Stenotrophomonas* (10.2%) and *Clostridiales* (3.7%), which were associated with the humification. The aeration efficiencies of lab-scale tests were at the range of 5.4–11.8 g BDM/ L O_2 for ARs with disposal ages of 1–13 years, and in situ landfill reclamation, ARs with disposal age of 10–18 years were around 1.9–8.8 g BDM/ L O_2, as the disposal age decreased. The increased discrepancy was observed in ARs of lab-scale and field scale, indicating that the forced aeration rate should be adjusted based on aged refuses and the unit compartment combined, to reduce the operation cost.

1. Introduction

Landfills were still the predominant waste disposal process in China, with 0.17 billion tons in 2019 (Guo et al., 2021), and huge amounts of wastes were presented in 27 thousand old dumping sites and 1955 landfills registered in the Ministry of Construction and Ministry of Ecological Environment (Zhao and Lou, 2016). To reduce the secondary pollution and reuse these lands with the rapid urbanization process, landfill reclamation has been implemented to accelerate the landfill stabilization process, including aeration (Omar and Rohani, 2015), leachate circulation (Bae et al., 2019), mining (Faitli et al., 2019), etc. Among which, forced aeration was considered as an indispensable means to transform traditional anaerobic landfills into a biologically stable state (Erses et al., 2008), while the aeration process was really complex due to the heterogeneous of waste and the disposal years, especially for the wastes with high organic matters, water contents and low-value materials (Zhao and Lou, 2016).

Forced aeration was one of the potential manual interventions for the acceleration of landfill stabilization (Ritzkowski and Stegmann, 2012), and has been applied in many landfills, such as Kuhstedt Landfill (Ritzkowski et al., 2016) and Milmersdorf Landfill (Gamperling et al., 2011) in Germany, Mannersdorf Landfill (Hrad and Huber-Humer, 2017) in Austria, and Columbia Landfill and Live Oak landfill (Read et al.,

2001) in the United States. Heishitou Landfill in China also tested the aerobic bioreactor in 2009 (Han et al., 2016). The operation processes were totally different and mainly relied on their operation experiences, since the disposal years, the waste composition and even the landfill locations, all of them will influence the selection of the aeration volume, the layout of the aeration pipes. The Modena landfill with 20 years closure was treated for 6 months at an aeration rate of 0.02 L/kg DM/day in Italy, and the respiration index of the waste was only 33% of the initial value (Raga et al., 2015). The same aeration rate of 0.08 L/kg DM/day was applied for two different landfills of Black Stone Landfill and Jinkou Landfill in China, with the average disposal period of 12 and 18 years, the OM content decreased from 18.5–6.2% in the previous after two years' operation, while that in the Jinkou Landfill decreased from 12.4–9.1% after one-year aeration (Liu et al., 2018). It is necessary to establish the relationship between the landfill properties, the aeration conditions, and the disposal ages for the aeration process.

The degradation processes could be regarded as large-scale composting for landfill aeration, to create an aerobic environment in a heap through aeration (Ritzkowski and Stegmann, 2012; Nanda and Berruti, 2020), and some fresh wastes or waste models were simulated to reflect the aerobic aeration in various countries (Hashisho and El-Fadel, 2014), including the aeration frequency (Nag et al., 2016), aeration rate (Slezak et al., 2010), leachate re-circulation frequency (Luo et al., 2019), compaction density (Qiu et al., 2019), and exogenous aerobic bacteria (Ge et al., 2016). It is no doubt to find that all of these results were different or even conflict, and there were still some gaps for the practical landfill reclamation projects, since almost all of the aeration should be implemented in landfills disposed of several years, instead of fresh waste. The wastes of different ages (we called aged refuses here, ARs) should be used to simulate the landfill reclamation to identify the potential operation conditions for aerobic remediation of landfills. Meanwhile, the generation parameters of the landfill stabilization process, such as leachate properties, landfill gas composition, and waste compositions have been used to reflect the degradation of fresh waste (Rooker, 2000). Some special index should be employed for the aged refuses, which contains more humus-like matters.

In this work, ARs with 1, 4, 7, 10, and 13 years were collected from a working landfill, which has been well recorded during the landfill stabilization process. The landfill remediation was simulated with the forced aeration rate under the same operation conditions, and the variations of AR compositions were analyzed to identify the degradation process. The primary purpose was to qualify the acceleration stabilization process with the forced aeration and AR properties and explore the degradation mechanism through the analysis of the AR properties in terms of humus, cellulose/lignin ratio, and the evolution of the microorganism. The aeration efficiency between the lab-scale and field-scale landfill was compared to finally guideline the potential aeration rate selection.

2. Materials And Methods

2.1. Aged refuses samples

AR samples were collected from the different cell compartments with the disposal ages of 1, 4, 7, 10, and 13 years in Shanghai. The quantity and property of AR, the placement time, and the location of corresponding compartments were well recorded by the landfill operator. AR samples were sampled once at 0–3, 3–6, and 6–9 m each, and the large foreign objects such as stones, glass, and plastics were manually removed. After thoroughly mixing samples of different depths, 5 kg of typical samples were collected according to the quarter method. After natural air drying, the samples that passed through a 60-mesh sieve after grinding were collected. The characterization of AR with different disposal times is shown in **Table 1**.

Table 1

The details of the initial characterization of the AR with different disposal times

Disposal times (year)	1	4	7	10	13
Moisture (%)	50.9 ± 5.3	43.9 ± 5.6	37.1 ± 3.9	39.1 ± 4.8	36.0 ± 2.2
pH	7.93 ± 0.02	8.05 ± 0.06	8.08 ± 0.16	8.12 ± 0.16	8.24 ± 0.21
VS (% dry weight)	29.3 ± 0.7	28.5 ± 0.4	27.4 ± 0.4	27.0 ± 0.6	23.1 ± 0.4
BDM (% dry weight)	20.0 ± 0.2	19.1 ± 0.7	18.1 ± 0.6	16.6 ± 0.5	13.8 ± 0.6
Cellulose (% dry weight)	5.1 ± 0.2	4.8 ± 0.3	4.2 ± 0.5	4.4 ± 0.4	3.8 ± 0.3
Lignin (% dry weight)	8.4 ± 0.3	8.0 ± 0.8	7.2 ± 0.1	8.3 ± 0.5	7.4 ± 0.4

2.2. Start-up of stabilization process

2.2.1. Lab-scale lysimeters

The batch experiment of 3 parallel controlled tests was carried out in 500 mL triangle flasks with a 250 g sample (dry weight), with the initial moisture content of 50% set. The temperature and aeration rate were kept at 30°C and 0.12 L O_2 /kg DM/day in a water bath. The gas was humidified through the scrubber to reduce the evaporation loss. A total of 9 samplings were carried out on days 0, 1, 3, 5, 8, 12, 17, 24, and 35th. The ARs in the triangle flask were evenly mixed before each sampling. The leachate was collected at a designed leaching process with a sample and distilled water at a ratio of 1:10 (w/v, dry weight) in a horizontal shaker at 110 ± 10 rpm, 25°C for 8 h, and leaving for 16 h and then filtering by 0.45 μm.

2.2.2. Field-scale landfills

Based on the lab-scale results, field-scale aeration was performed in a landfill of about 19.65 hectares, located in a city in Zhejiang Province, China. About 2.38×10^4 m^3 of municipal solid waste was landfilled at the site between 8–21 years, with a bulk density of about 650 kg/m^3 and average moisture of 40.3%. A low-pressure aeration system was employed for active aeration and tail gas extraction, with 394 gas wells set up. Four Roots fans with a flow rate of 50 m^3/min were used to aerate the landfill intermittently of 3 hours cycles. The total aeration rate was approximately 10.8 L O_2/kg DM. S1, S2, and S3, with disposal ages of about 18, 14, and 10 years (Fig. 3), were selected as field monitoring points, and AR_{18-f}, AR_{14-f} and AR_{10-f} in these three points were collected and analyzed before and after aeration for 9 months.

2.3. Analytical methods

2.3.1. Physical and chemical properties analysis

BDM content of the AR was determined by oxidation with potassium dichromate at 25°C under strong acid conditions and conversion based on the amount of oxidizer consumed (Han et al., 2013). Cellulose and lignin were determined by Fan's detergent fiber analysis method (Qu et al., 2005). pH and ORP were measured by a portable pH meter (Five Go, METTLER TOLEDO, Switzerland). The AR extracts processes were referred to our previous works (Lou et al., 2011). The total organic carbon (TOC) was evaluated by a Multi N/C 3100 Analyzer (Analyti Jena, Germany). All index analysis was performed three times and then the average value was taken.

2.3.2. Humus extraction and EEM spectra

Humus was extracted and determined. AR samples were extracted with 0.1 M $Na_4P_2O_7$ and 0.1 M NaOH at a ratio of 1:20 (w/v, dry weight) (Wu et al., 2020). After standing for 24 h, the extract was centrifuged at a speed of 10,000 rpm and then filtered by membrane filter (0.45 μm). One part of the mixed combined supernatants were used to determine the content of soluble humus (total humic acid and fulvic acid content) and EEM spectroscopy. Another part was adjusted to pH 1 with 1 M H_2SO_4 in a water bath at 80°C. After standing at 25 ℃ for 24 h, the supernatant containing fulvic acid (FA) was centrifuged at 10,000 rpm, and the precipitate was collected and dissolved with 0.05 M NaOH to determine the humic acid (HA) content (Zhou et al., 2014).

EEM spectroscopy of filtrate was measured by spectrophotometer (Model F-7000, Hitachi, Japan). Both the excitation (Ex) and emission (Em) wavelengths were set at 200–600 nm. The slit bandwidths of Ex and Em were set to 10 nm. Spectra were scanned and recorded at a rate of 40 nm/s. The EEMs data were analyzed by MATLAB R2018b software and DOMFluor toolbox to perform PARAFAC model analysis (Liu et al., 2020), and the number of fluorescence components was obtained through half-analysis. The maximum fluorescence intensity (F_{max}) output of the model was used to express the relative concentration of a specific fluorescent component in Raman units (R.U.) (Stedmon and Bro, 2008).

2.3.3. Microbial community structure analysis

AR samples with 1, 7 and 13 years were collected and used for the microbial community structure analysis, and DNA was extracted with a soil DNA extraction kit (BioTeke, Beijing, China). The DNA solution was stored at − 80 ℃ before being used. The 338F/806R primers were used for bacteria. DNA was amplified by PCR on ABI GeneAmp® 9700 (Thermo, USA). The characterization of AR extracts was based on previous methods (Yang and Wang, 2021). The sequencing was carried out by Majorbio Company (Shanghai, China). Library preparation and sequencing reaction depended on TruSeqTM DNA Sample Preparation Kit and Illumina Miseq (Illumina, USA).

2.4. Data analysis

First-order kinetics was developed to describe the degradation process of MSW in landfill, and BDM was used as an indicator for AR biodegradation.

$$\frac{d\left(BDM\right)}{dt}=-K\bullet\left(BDM\right)$$

1

BDM content was simulated as the following equation:

$$BDM_t=BDM_0e^{-K\bullet t}$$

2

Where, BDM_t refer to BDM content after t days reaction; BDM_0 was the initial value; K was the degradation rate constant; t was the aeration period.

3. Results And Discussion

3.1. Biodegradation kinetics of AR with different disposal ages

Variations of AR properties with the elapsed time are shown in Fig. 4. The initial pH values were 7.93–8.24, and then dropped by 0.2–0.3 after aeration, meaning that pH range was within an optimal period for aeration (Han et al., 2019). A significant difference of pH was observed between AR_{13} and other samples. pH in AR_{13} increased significantly from 7.86 to 8.59 after 5 days of aeration, and then kept almost stable. pH in the other samples declined until 12 days of operation, since more organic matters were presented in young ARs. The decline of pH in ARs was mainly due to the generation of organic acids and the conservation of NH_4^+ into NO_3^- during the aeration process (Omar and Rohani, 2015), as shown in equation (Nag et al., 2018). Similar results were observed in ORP. Lower OPR range of -30 and − 60 mv was in young ARs with high organic matter content, which resulted in active degradation reactions with the forced aeration.

$$NH_4^+ + O_2 + H_2O \rightarrow NO_3^- + 5H^+ + 4e^-$$

3

$$NH_4^+ \rightarrow NH_3 + H^+$$

4

BDM contents of ARs were 19.6%, 18.8%, 18.0%, 16.9% and 14.0% as the disposal age increased. The highest BDM content decrease of 5.2% was observed in AR_1, from 19.6–14.4%, while others were around 2.4%-4.7%. More ready biodegradable matters were presented in ARs with low disposal years, and presented a fast degradation process. 50%-56% of BDM degraded in the first 8 days in AR_{1-10}, while 12 days was necessary for AR_{13}. The aeration efficiencies were 11.8, 10.7, 10.0, 9.1 and 5.4 g BDM/L O_2, respectively, indicating the O_2 consumption rate should be adjusted according to the BDM content in AR. The degradation constant K of all samples were summarised and shown in Fig. 4d, as 8.72×10^{-3} (R^2 = 0.858), 8.12×10 − 3 (R^2 = 0.897), 7.83×10^{-3} (R^2 = 0.926), 7.38×10^{-3} (R^2 = 0.856) and 5.80×10^{-3} (R^2 = 0.915), respectively, which followed a positive relationship with the disposal time (Eq. (5)), indicating that higher BDM content resulted in faster the degradation rate (Song et al., 2015).

$y = -0.0002193x + 0.0091\ (R^2 = 0.854)$ (5)

The relationship between BDM content in AR and the disposal age x was as follows:

$$BDM_t = BDM_0 e^{(-0.0002193x+0.0091)\bullet t}$$

6

3.2. Effects of aeration on the properties of ARs

3.2.1. Variations of ARs components

Variations between AR properties, including VS, C/L and HA/FA, and the aeration time are shown in Fig. 5. VS content decreased around 5.9% in AR_1, from 29.0–23.1%, and only 2.9% of VS was degraded in AR_{13}, meaning that most of organic content in old ARs were non-biodegradable matters. The cellulose and lignin contents were around 3.8%-5.1% and 7.2%-8.4%, which belongs to hard degradable organic matters. C/L ratio decreased around 3.1% as the aeration time extended, from 49.5–46.4% in AR_{13}, while more than 10% were found in those in AR_{1-7}, and C/L of all ARs kept between 44%-48% (with an average value of 46%), meaning that C/L could be employed as the indicator to assess the degradable potential of ARs. With regards to HA/FA ratio, AR with 1, 7 and 13 years increased from 1.27, 0.73 and 0.15 to 1.44, 1.55 and 2.16, respectively, meaning that more HA was generated as the aeration time increased (Wang et al., 2013).

EEM spectra of ARs were employed to identify humus properties, as shown in Fig. 5d. Ex/Em = 320/420 (protein-like substances) decreased by 2.4% and 8.25% of AR_7 and AR_{13}, while Ex/Em of 290/440 (humic-like substances) increased 2.3% and 3.2%. The protein-like and humic-like substances had a gradient relationship with the disposal ages, suggesting that during the aeration process, the overall composition of young ARs maintains a dynamic balance due to the rapid degradation of organic matter. In ARs with more disposal ages, most organic substances tend to be converted to humic acid.

3.2.2. Characteristics of leachate pollutants

Leachate collected and their properties are presented in Fig. 6. COD showed a rapid decline from 2280 to 127 mg/L after 35 days of aeration in AR_{13}, and those of AR_{1-7} showed a similar slow decline, with an average decline of 677 mg/L. More substances could be leaching from ARs, since COD concentration increased greatly in the first 4 days, and then decreased in the following days, indicating that more dissolved matters of the organic substance were presented in ARs after aeration. TOC/COD was around 35–42%, and then decreased slightly to 36.6%, 36.5%, 32.6%, 23.1% and 21.4%, respectively, suggesting that most biodegradable components were degraded.

TN showed a declining trend under the text periods, decreasing by 236–368 mg/L, and the removal rates of AR_{1-13} were 50.4%, 62.6%, 76.8%, 83.1% and 91.3%, respectively. For NH_3-N/TN of AR_{1-10}, it increased firstly in the first days and the largest for AR_7, 70.7%, since more NH_3-N were generated in the initial period, although fluctuations occurred in the following days, NH_3-N continued to decrease due to the substantial decrease in TN. The NH_3-N/TN of AR_{13} continued to drop from 27.0–6.6%, NH_3-N concentration dropped from 109 to 2 mg/L, and the degradation rate was 97.9%, indicating that aeration can thoroughly remove NH_3-H from old ARs.

3.2.3. Evolutions of microbial community

The bacterial community at the genus level are shown in Fig. 7, after 35 days of aeration, and the dominant species were *Corynebacterium* 9.2%, *Acinetobacter* 6.6%, *Fermentimonas* 6.5%, in AR_1, followed by the proteolytic bacteria (*Proteiniclasticum* 10.7%, *Proteiniphilum* 3.2%), and anaerobic humic bacteria (*Stenotrophomonas* 10.2%, *Clostridiales* 3.7%, etc.) were the main contents in AR_{13}. With regards to AR_7, the proportion of *Aquabacterium* occupied 9.8%, which could be used to degrade complex organic compounds, such as gasoline and diesel (Yamashita et al., 2022). The degradation substances in ARs conversed from the readily degradable organics in young ARs (Wu et al., 2022) to humus-like organics in old ARs (Li et al., 2019; Wang et al., 2022). The aerobic denitrifying bacteria *Diaphorobacter* was around 3.5% in AR_7, meaning that the denitrification process was still active (Xia et al., 2022).

3.3. Aeration efficiency comparation in field-scale landfill reclamation

BDM contents of AR_{18-f}, AR_{14-f} and AR_{10-f} decreased from 10.1%, 12.3% and 16.3–8.1%, 7.3% and 7.1% after 9 months of operation, respectively, as shown in Fig. 8. Based on Eq. (6), BDM contents of AR_{18-f}, AR_{14-f} and AR_{10-f} should be around 6.4%, 7.1%, and 8.8%, respectively, under the same aeration rate, meaning that the oxygen consumption efficiency for ARs decreased significantly with increasing disposal ages in both field scales, from 8.8 (AR_{10-f}) to 1.90 (AR_{18-f}) g BDM/L O_2. Taken S1 as an example, the practical value of BDM of AR_{18-f} was 1.7% higher than the predicted value, which occupied around 26.6% higher, the difference between field scale and lab-scale was due to the difference in the operation conditions, especially for the landfill unit compartments, and lab-scale lysimeters created a more favorable environment to promote the degradation of refractory ARs in the disposal of overaged AR (Grisey and Aleya, 2016). For ARs with a lower disposal age, predictions of BDM deviate relatively less from practical. The practical value of BDM of AR_{14-f} was in good agreement with the predicted value, with an error of 2.8% and the error of AR_{10-f} was 18.1%, indicating that the BDM degradation model could be one of the promising methods to optimize the aeration parameters according to the disposal age of the AR.

4. Conclusions

ARs with different ages were collected and employed for the landfill aeration, to identify the relationship between aged refuse property and the forced aeration. pH was in the suitable aeration range, and the degradation of BDM in different stable stages were 5.2%, 4.7%, 4.4%, 4.0% and 2.4%, respectively, at 0.12 L O_2 /kg DM/day with the intermittent aeration of 3 hours cycles in 35 aeration days. The BDM degradation rate constants decreased from 8.72×10^{-3} (R^2 = 0.858) in AR of 1 year to 5.80×10^{-3} (R^2 = 0.915) in that of 13 years. With the increases of disposal year, the ratios of cellulose/lignin (C/L) decreased by 18.3–3.1% and finally stabilized at around 46%, while HA/FA gradually increased, means that the degradation target of AR changes from ready biodegradation to complex organic matter. The increase in HA/FA of AR with 13 disposal years was the most, from 0.15 to 2.16 and *Stenotrophomonas*, *Clostridiales* (humus related) of 13.9%, indicating that in this stabilization stage, aerobic treatment mainly promotes the formation of humic and enhanced humification. The degradation rate of ARs in the field landfill was lower than that of the lab-scale landfill, especially for the ARs with longer disposal age indicating that the aerobic frequency and volume should be adjusted based on the disposal ages and BDM contents together for landfill reclamation.

Declarations

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Author Contributions

Ziyang Lou: conceptualization, validation and funding acquisition; Yihang Liu: investigation and writing-original draft; Weihua Cao, Hui Liu and Jia Song: resources; Qiujie Huang, Changfu Yang and Zhaowen Cheng: validation; Luochun Wang: supervision; Xianghui Wang: field-scale support. All authors read and approved the final manuscript.

Ethics approval

Not applicable.

Consent to participate

All authors have given consent to their contribution.

Consent for publication

All authors have agreed with the content and all have given explicit consent to publish.

Competing interests

The authors declare no competing interests.

Availability of data and materials

Data are available on request to the authors.

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Figures

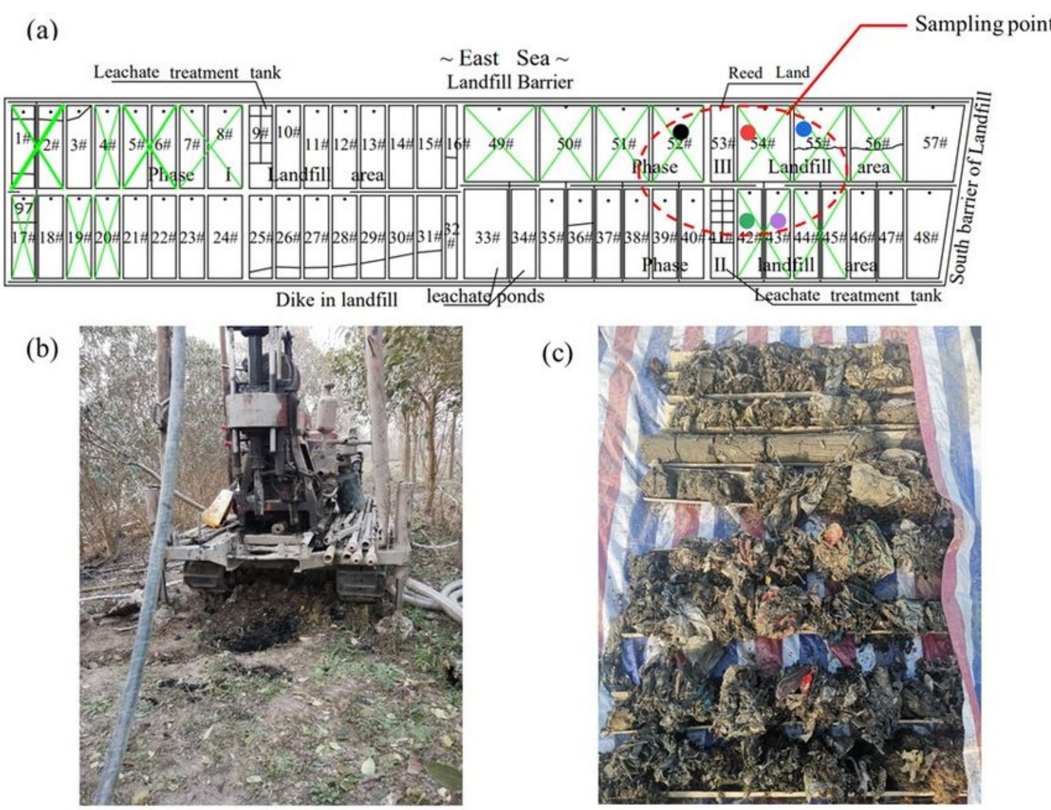

Figure 1

Schematic diagram of sampling points in Landfill (a), ARs collected (b) and ARs (c)

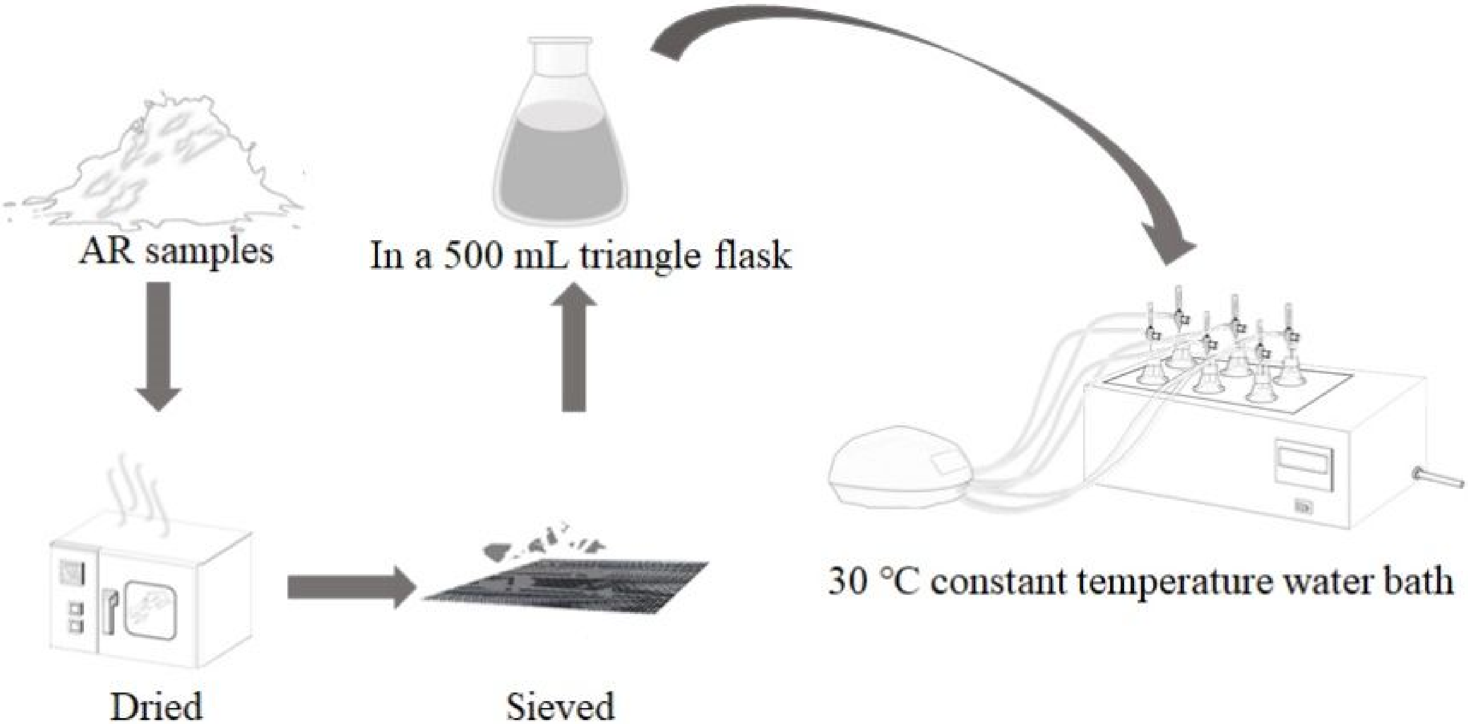

Figure 2

Sample pretreatment process and experimental device

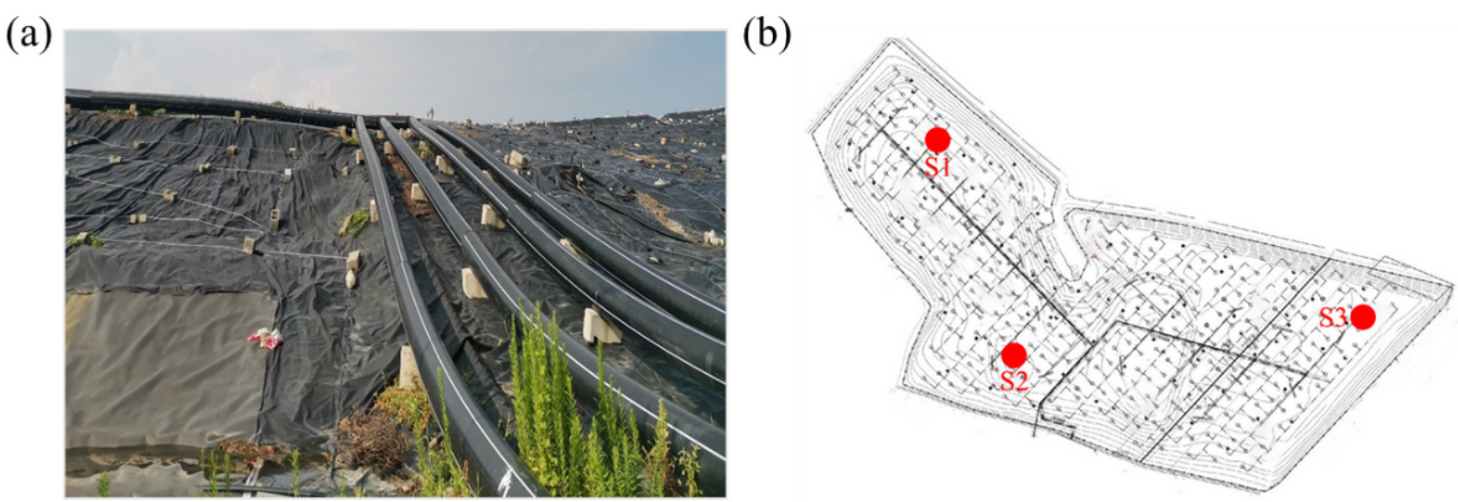

Figure 3

The field-scale landfill (a) and distribution of monitoring sites (b)

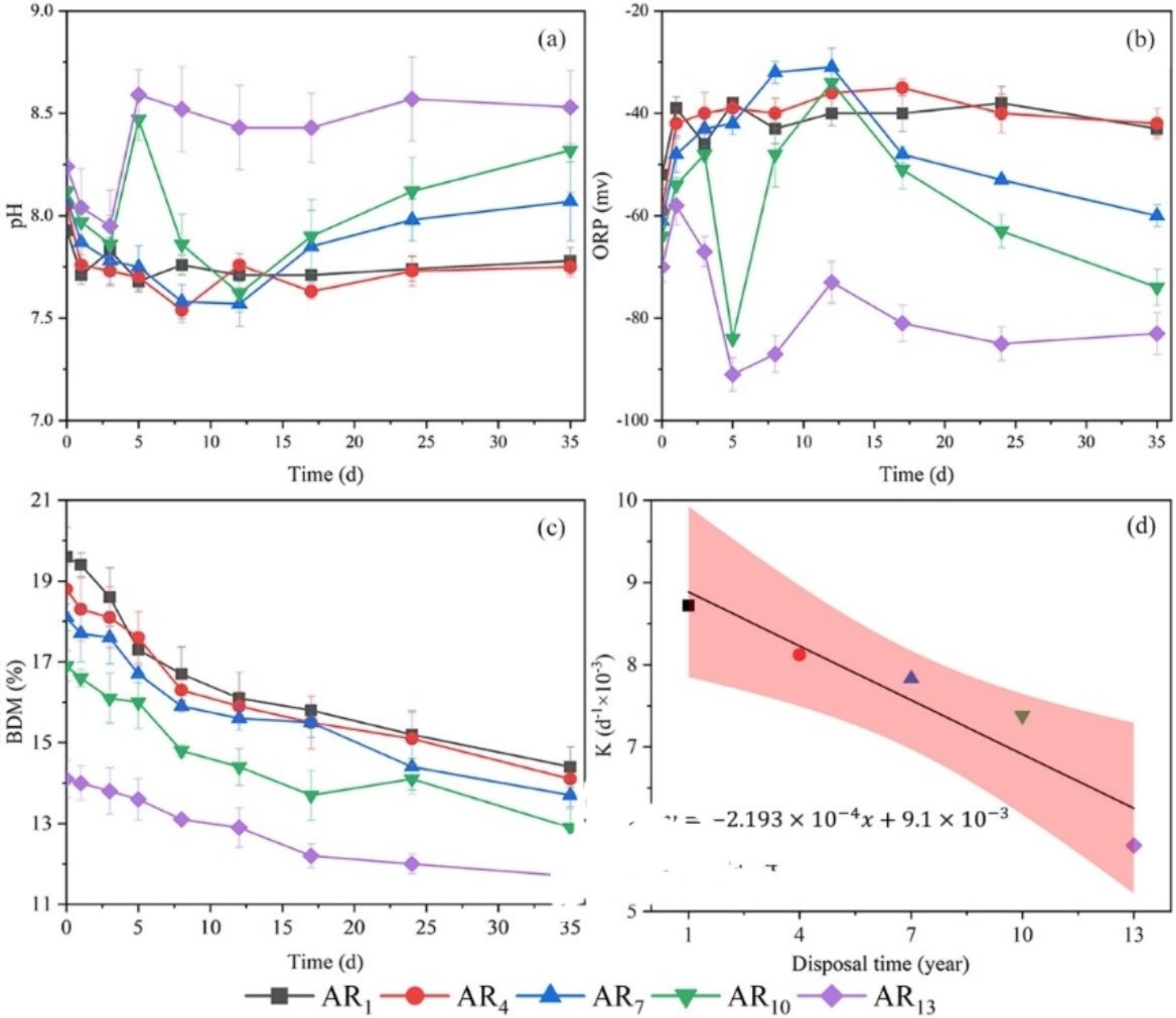

Figure 4

Variations of pH (a) ORP (b), BDM (c) of ARs under aeration conditions and K value (d) with the disposal time

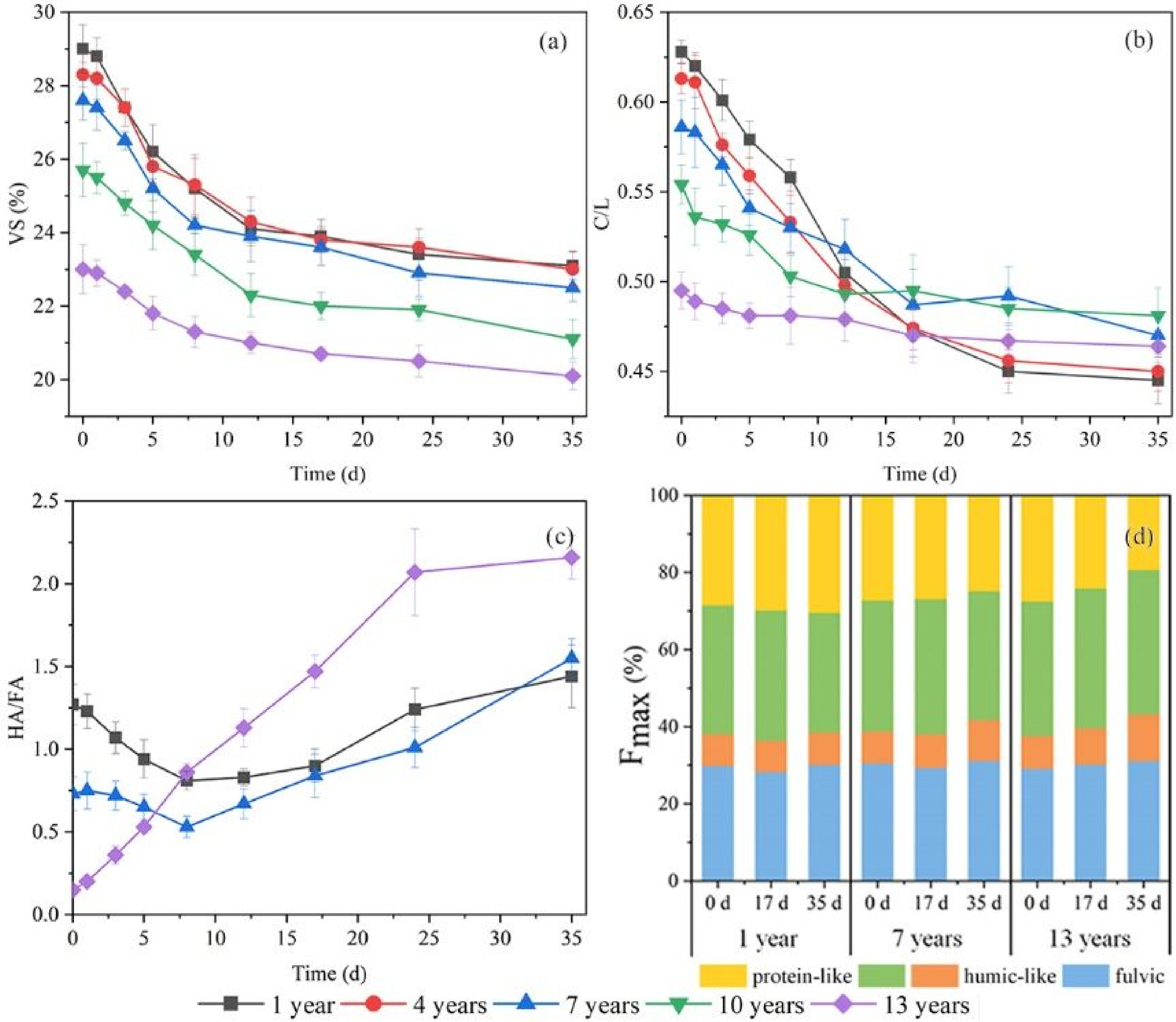

Figure 5

Variations of VS (a), C/L (b), HA/FA (c) and the F_{max} (d) of ARs under the aeration condition

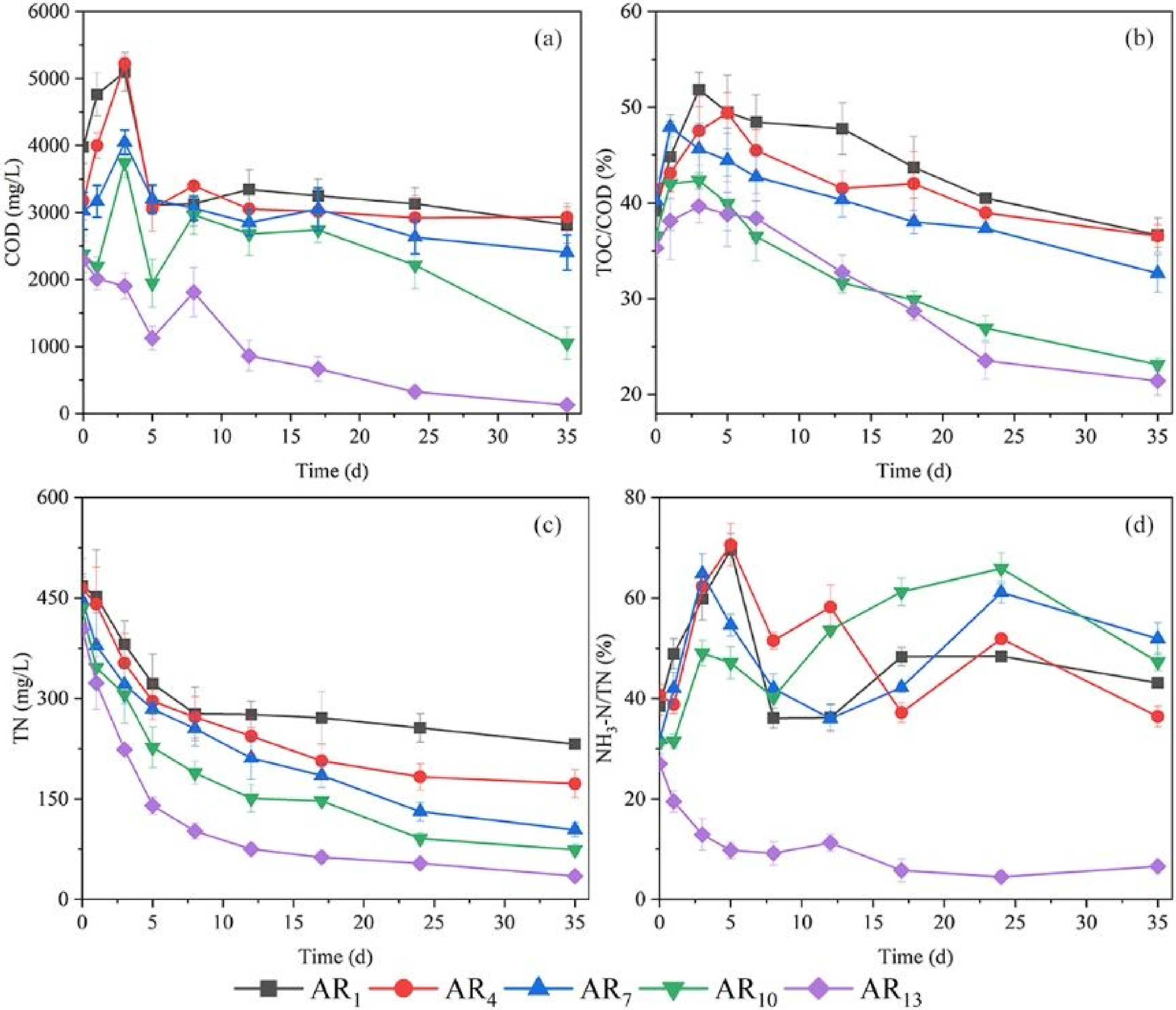

Figure 6

Variations of VS (a), C/L (b), HA/FA (c) and the F_{max} (d) of ARs under the aeration condition

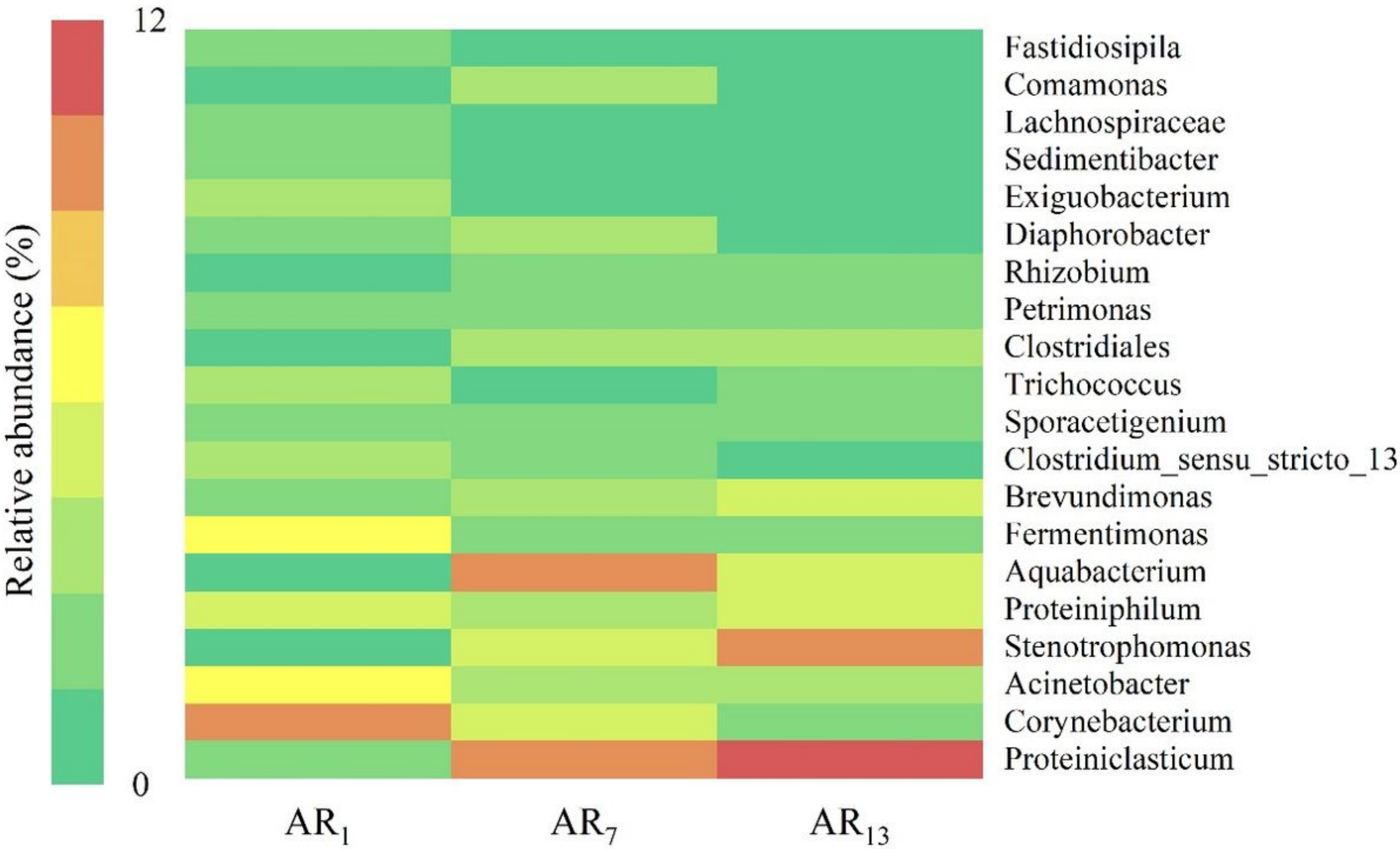

Figure 7

The microbial diversity of ARs at genus levels at the end of the experiment

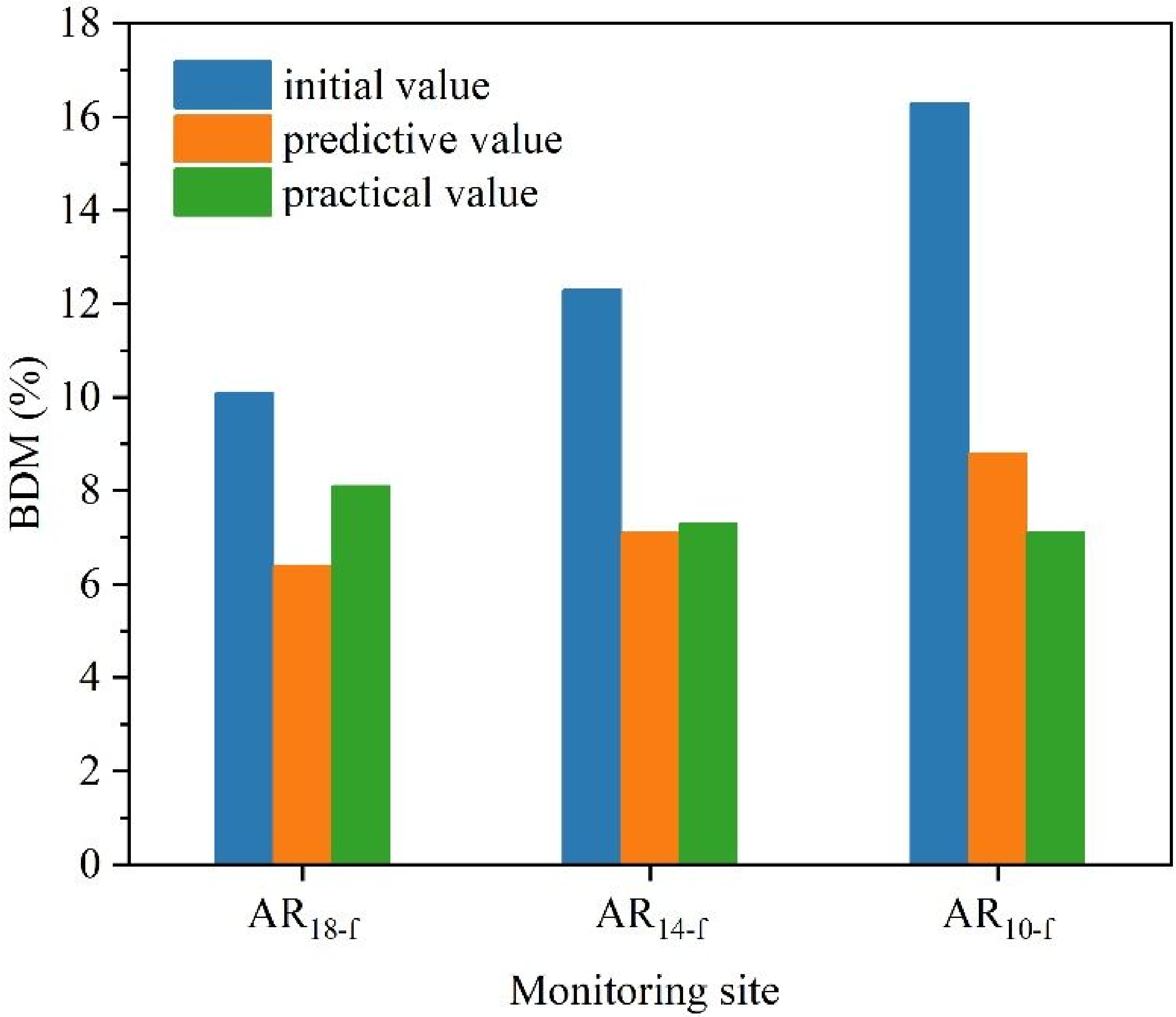

Figure 8

Initial, predicted, and practical values of BDM content at the three monitoring sites

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