Front. Environ. Sci. Eng. 2021, 15(3): 48
https://doi.org/10.1007/s11783-021-1421-7

RESEARCH ARTICLE

Removal of odors and VOCs in municipal solid waste comprehensive treatment plants using a novel three-stage integrated biofilter: Performance and bioaerosol emissions

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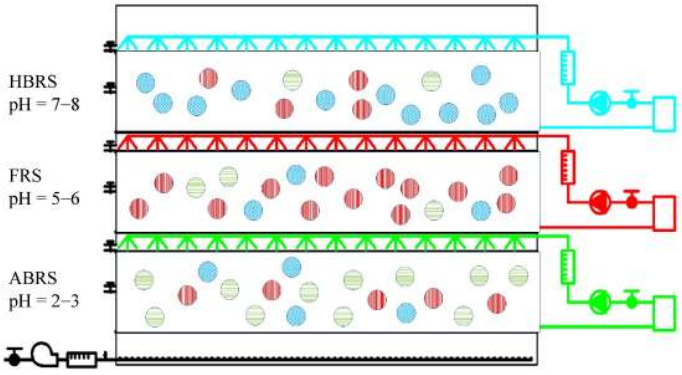
HIGHLIGHTS

- TSIBF was composed of ABRS, FRS and HBRS.
- THIBF can effectively remove various odors, VOCs and bioaerosols.
- Different reaction segments in TSIBF can remove different types of odors and VOCs.
- TSIBF can reduce the emission of bioaerosols through enhanced interception.

GRAPHIC ABSTRACT



ARTICLE INFO

Article history:
Received 17 November 2020
Revised 25 January 2021
Accepted 2 March 2021
Available online 10 May 2021

Keywords:
Biofiltration
Multi-stage biofilter
Volatile organic compounds
Waste gas treatment
Bioaerosol emissions

ABSTRACT

A novel three-stage integrated biofilter (TSIBF) composed of acidophilic bacteria reaction segment (ABRS), fungal reaction segment (FRS) and heterotrophic bacteria reaction segment (HBRS) was constructed for the treatment of odors and volatile organic compounds (VOCs)from municipal solid waste (MSW) comprehensive treatment plants. The performance, counts of predominant microorganisms, and bioaerosol emissions of a full-scale TSIBF system were studied. High and stable removal efficiencies of hydrogen sulfide, ammonia and VOCs could be achieved with the TSIBF system, and the emissions of culturable heterotrophic bacteria, fungi and acidophilic sulfur bacteria were relatively low. The removal efficiencies of different odors and VOCs, emissions of culturable microorganisms, and types of predominant microorganisms were different in the ABRS, FRS and HBRS due to the differences in reaction conditions and mass transfer in each segment. The emissions of bioaerosols from the TSIBF depended on the capture of microorganisms and their volatilization from the packing. The rational segmentation, filling of high-density packings and the accumulation of the predominant functional microorganisms in each segment enhanced the capture effect of the bioaerosols, thus reducing the emissions of microorganisms from the bioreactor.

1 Introduction

With the acceleration of urbanization, the amount of municipal solid waste (MSW) has been increasing rapidly in recent years. MSW treatment technologies include incineration, sanitary landfills and comprehensive treatment technologies based on separation (Wang et al., 2020).

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Special Issue—Bioaerosol, Environment and Health (Responsible Editors: Can Wang, Jungho Hwang, Jingkun Jiang & Maosheng Yao)

Of these technologies, comprehensive treatment technology based on mechanical separation and organic aerobic composting is becoming more widely used in the treatment of MSW due to its great potential for resource utilization (Wang et al., 2020).

Odors and VOCs are produced in the units of unloading, sorting and aerobic composting in MSW comprehensive treatment plants (Wang et al., 2020). Compared with other facilities, such as municipal wastewater treatment plants, the MSW comprehensive treatment plants produce odors and VOCs with higher concentrations and more varied compositions, which include hydrogen sulfide, ammonia, mercaptan, low molecular weight fatty acids, chlorides, linear alkanes and aromatic hydrocarbons (alkanes and aromatics) (Liu et al., 2009). Most of these odors and VOCs are volatile and corrosive irritants, and their concentrations are much higher than those specified in the Emission standards for odor pollutants of China (GB-14544-93). Therefore, these substances have a great impact on human health and atmospheric environment (Han et al., 2020b).

Additionally, microorganisms generated in MSW comprehensive treatment plants during the agitation units of discharging, transporting and separating garbage are very likely to escape and be released to form bioaerosols. Liu et al. (2021) reported that the concentrations of airborne fungi in landfill ranged from 376 to 9318 CFU/m^3. Wikuats et al. (2020) found that the concentrations of bacteria bioaerosol and fungal bioaerosol emitted from a material recycling facility were 1088.8±825.2 and 2738.3±1381.3 CFU/m^3, respectively. The spread of bioaerosols is one of the possible ways to spread diseases, especially in the worldwide COVID-19 pandemic this year, which has made people more aware and vigilant of the generation and harm of bioaerosols (Mutuku et al., 2020). Many researchers have confirmed that harmful microorganisms affect human respiration, the immune system and the blood system (Yang et al., 2019).

MSW treatment facilities should be required to not only achieve quantitative reduction and resource recovery but also dispose of MSW safely. They should also be able to effectively eliminate odors, VOCs and bioaerosols to guarantee ecological environmental protection and public health safety. The treatment of atmospheric pollutants produced by MSW comprehensive treatment plants has become the focus of public attention.

Conventional odors and VOCs treatment technologies include absorption, adsorption, combustion, and biotechnology (Barbusinski et al., 2017). Of these technologies, biofilters have been successfully employed to treat odors and VOCs due to their advantages of low investment and operation costs, convenient maintenance, and facile operation (Han et al., 2020a). However, the odors and VOCs produced in MSW comprehensive treatment processes are different in terms of gas volume and composition, water solubility, pH and biodegradability. The conventional biofilter process is relatively simple, all kinds of odors and VOCs are degraded in a bioreactor, and the type of functional degradation microorganisms is relatively simple. Due to the complexity of the biodegradation reaction and the products of the odors and VOCs, the efficient removal of the odors and VOCs is often affected by the inhibitory or interfering action (Mohammad et al., 2017). Additionally, acid substances accumulate in the packing, reducing the pH and thus affecting the microbial activity and biodegradation efficiency (Estrada et al., 2011). Therefore, it is difficult to effectively remove all kinds of odors and VOCs produced by comprehensive waste separation and treatment plants simultaneously using a biofilter with a single reactor.

In view of the poor ability of conventional biofilters to treat complex odors and VOCs, some novel integrated biotreatment technologies and bioreactors have been developed. López et al. (2017) studied the removal of a mixed waste gas consisting of methanol, hydrogen sulfide and pinene with a biotrickling filter-biofilter combined bioreactor and showed that methanol and hydrogen sulfide were effectively removed by the biotrickling filter, whereas the removal rate of pinene by the biofilter was high. Torretta et al. (2015) used a pilot-scale two-stage bioreactor to study the removal of waste gas from the drying process during solid fuel recovery, indicated that the average removal rates of odors and VOCs could reach 95.4% and 81%, respectively, after 2 weeks of bacterial acclimation. In general, these integrated bioreactors can provide the optimal conditions for degrading different target pollutants and alleviate antagonistic interactions between substrates and the toxic inhibition of target pollutant decomposition. However, they must be rationally designed based on the characteristics of the waste gas (Torretta et al., 2015).

A three-stage integrated biofilter (TSIBF) system was developed and used to treat odors and VOCs in an MSW comprehensive treatment plant. The THIBF system, which uses an integrated multi-stage biological process to remove odors and VOCs, is based on the characteristics of the complex gas components and on the different types and properties of the pollutants in the MSW comprehensive treatment plant. The THIBF system consists of an acidophilic bacteria reaction segment (ABRS), fungal reaction segment (FRS) and heterotrophic bacteria reaction segment (HBRS). By selecting the inoculum and controlling the reaction conditions in the different reaction segments, microorganisms with different physiological characteristics can live in their own suitable environment and degrade different types of odors and VOCs. Due to the synergistic degradation of microorganisms and adsorbents in the different segments, the THIBF system can effectively remove various odors, VOCs and bioaerosols produced by MSW comprehensive treatment plants.

Previous studies have shown that biofilters are a potential source of bioaerosols; however, bioaerosols in

the inlet air are simultaneously subject to capture effects. The efficiency of emission and capture is related to the type of packing and the process parameters of the reactors (Yang et al., 2019; Liu et al., 2020a; 2020b). Compared with the concentration and composition of bioaerosols in conventional biofilters, those in TSIBFs are more complex because of their different reaction segments. Therefore, it is necessary to study the emission and capture characteristics of bioaerosols in the TSIBF.

In this study, the removal effects of a full-scale THIBF system on the odors and VOCs in an MSW comprehensive treatment plant and the stability of its long-term operation were investigated, and the performance of the staged removal of different types of pollutants was studied. In addition, the characteristics of the packings and outgassing microorganisms in the different reaction segments of the THIBF system were investigated. Finally, the mechanisms of outgassing microorganism emissions and capture in the THIBF system were discussed. The results of this study can provide a scientific basis and practical experience for the application of this technology in the treatment of odors and VOCs.

2 Material and methods

2.1 Outline of the MSW comprehensive treatment plant

The MSW comprehensive waste treatment plant covers an area of 35000 m^2. The designed treatment scale is 300 t/d, the actual treatment scale is 270 t/d, and the service population is 250000 people. The facility includes comprehensive sorting workshop, aerobic composting workshop, fertilizer warehouse and auxiliary workshop. The main MSW treatment unit involves a mechanical sorting-organic aerobic composting unit; that is, after domestic waste is comprehensively separated by wind separation, drum screening and magnetic separation, recyclable components such as metal, paper, plastic and glass are recycled, and the organic components are made into organic fertilizer after aerobic composting.

The MSW treated in the comprehensive treatment plant is primary and unclassified solid waste. Exhaust gas containing odors, VOCs and bioaerosol will be generated under the decomposition and fermentation of the organic waste in the process of waste unloading workshop, temporary storage workshop, and comprehensive sorting facility.

2.2 Composition of the THIBF system

The full-scale THIBF system is composed of gas collection device, transmission pipeline and main treatment equipment. Waste gas is first collected by the gas collection device. Then, with the use of a fan, the gas enters the THIBF system through the transmission pipeline and degrades into harmless products such as CO_2, H_2O and SO_4^{2-} due to the function of enzymes produced by microorganisms, which are emitted into the atmosphere after purification. The diagram of the THIBF is shown in Fig. 1.

Each segment of the THIBF system is a stationary phase filled with high-density packing material. The ABRS was filled with an inert and acid-resistant polyurethane (PU) packing, and the pH was controlled at 2.0–3.0 to form acidophilic sulfur bacteria, The FRS was also filled with PU packing, and the pH was controlled at 5.0–6.0, which allowed fungi to be the predominant microorganisms in this segment. The HBRS was filled with ceramsite (CM) packing, and the pH was controlled at 7.0–8.0, at which heterotrophic bacteria were predominant. The differences in the pH values of the packings in the ABRS, FRS and HBRS are generated by changing the pH of the circulating liquid.

The THIBF is made of glass fiber-reinforced plastics, and its shape is cuboid. The length, width and height of the bioreactor are 6.0, 5.0 and 4.5 m, respectively. The total and effective volumes of the THIBF are 135 m^3 and 90 m^3,

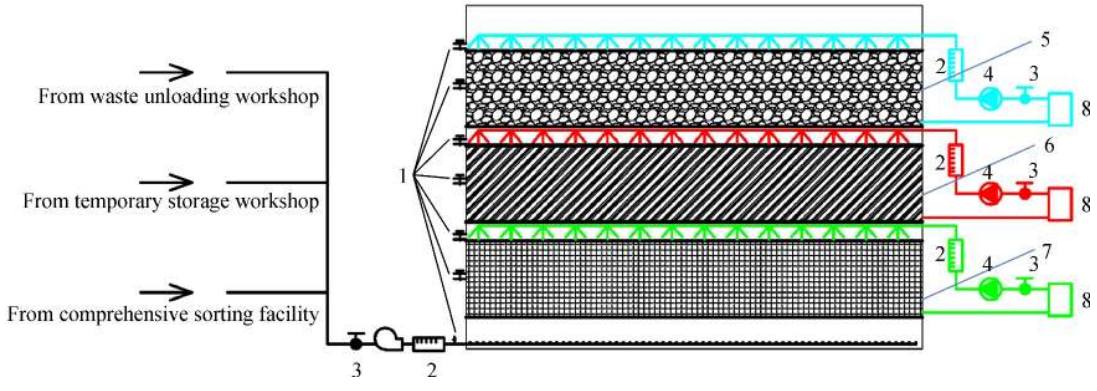

Fig. 1 The TSIBF diagram. 1. Sampling port, 2. Flowmeter, 3. Valve, 4. Pump of liquid circulation, 5. Heterotrophic bacteria reaction segment, 6. Fungal reaction segment, 7. Acidophilic bacteria reaction segment, 8. Nutrient solution tank.

respectively. The treatment capacity is 9000 m^3/h, and the total empty bed retention time (EBRT) is 36 s. The specific process design parameters are listed in Table 1.

2.3 THIBF inoculation and operation

The THIBF system was installed in July 2017 and operated in August 2017, with a total operation period of 216 days. The operation mode of the reactor is gas-liquid counter-current, and the gas enters from the bottom and is purified when it contacts the biofilm attached to the packing surface in the rising process. The purified gas is discharged from the top of the reactor. The moisture content of the packing and the pH of the circulating liquid are controlled by an automatic control system.

To shorten the start-up time of the bioreactor, the supernatant from the secondary sedimentation tank of a wastewater treatment plant was mixed with *Thiobacillus thiooxidans*, fungi and heterotrophic bacteria and inoculated into the ABRS, FRS and HBRS, respectively. The three inoculated microorganisms were all obtained from previous laboratory-scale devices and enriched in the laboratory before inoculation.

Then, nutrient solutions with different components were regularly sprayed into each reaction segments 4 times a day at a flow rate of 20 m^3/h for 3 min, which not only provided the necessary nutrients for the growth of microorganisms, but also controlled the fillers in the different reaction segments to maintain the suitable pH. The components of the nutrient solutions suitable for the ABRS, FRS and HBRS are given in Table 2. The excess nutrient solution was returned to the circulating liquid tank from the bottom of the reactor.

2.4 Sampling and analysis

2.4.1 Odors and VOCs

Hydrogen sulfide was collected in a PLV gas sampling bag with a Thermogreen® LB-2 septa (1 L, Tedlar) and analyzed in triplicate. The concentration of hydrogen sulfide was measured by a gas chromatograph (Agilent Technologies, GC 6890N, USA) equipped with a capillary column (HP-5, 30 m × 0.25 mm × 0.25 μm, Hewlett Packard, USA) and a flame photometric detector (FPD).

Ammonia is absorbed in a 5 mm sulfuric acid solution using a gas bubble sampler and then determined by Nessler's reagent method. The TVOCs were determined by a TVOC analyzer. The specific VOC substances were first determined by GC-MS (GC6890N-MSD5973) and then quantitatively determined by gas chromatography (Agilent Technologies, GC 6890N, USA). Additionally, the gas velocity is measured by a gas flowmeter.

2.4.2 Bioaerosols

The inlet and outlet bioaerosols were sampled by a six-stage microorganism FA-1 cascade impactor (Westech, UK). The operating conditions of the sampler were described in detail in our previous studies (Liu et al., 2020b).

Glass petri dishes (9 cm) were used for bioaerosol sampling after sterilization at 121°C for 15 min. The heterotrophic bacterial samples were cultured in a beef extract peptone medium at 37°C for 48 h. The fungal samples were cultured in Martin medium at 28°C for 5 days. The acidophilic sulfur bacteria samples were cultured in Waksman medium at 30°C for 21 d.

Table 1 Specific process design parameters for the three-stage integrated biofilter

Parameter	ABRS	FRS	HBRS
Length × width (m)	6.0 × 5.0	6.0 × 5.0	6.0 × 5.0
Effective height (m)	1.0	1.0	1.0
Effective volume (m^3)	30	30	30
Packing material	PU	PU	CM
EBRT (s)	12	12	12
pH of circulating liquid solutions	2.0–3.0	5.0–6.0	7.0–8.0
Moisture content of packing material (%)	40–60	30–50	40–60

Table 2 Composition of nutrient solution suitable for each reaction segments

Reaction segment	Composition
ABRS	2.00 g/L $Na_2S_2O_3 \cdot 5H_2O$, 0.50 g/L beef extract, 2.50 g/L NH_4Cl, 4.50 g/L KH_2PO_4,0.10 g/L $Fe_2SO_4 \cdot 7H_2O$, 0,10 g/L $Mg_2SO_4 \cdot 7H_2O$ (pH = 2.0–3.0, adjusted by 1 mg/L H_2SO_4 or NaOH)
FRS	2.00 g/L $NaNO_3$, 1.00 g/L K_2HPO_4, 0.50 g/L KCl, 0.19 g/L $MgCl_2$. (pH = 5.0–6.0, adjusted by 1 mg/L H_2SO_4 or NaOH)
HBRS	2.00 g/L KH_2PO_4, 2.00 g/L K_2HPO_4, 0.40 g/L NH_4Cl; 0.20 g/L $MgCl_2 \cdot 6H_2O$, 0.01 g/L $Fe_2SO_4 \cdot 7H_2O$ (pH = 7.0–8.0, adjusted by 1M H_2SO_4 or NaOH)

When the number of microorganism particles passing through each stage of the sieve exceeds a certain number, the bacterial particles impact the same point and overlap. Therefore, the number of active biological particles collected at all levels is corrected according to Eq. (1):

$$P_r = N \times \left(\frac{1}{N} + \frac{1}{N-1} + \frac{1}{N-2} + ... + \frac{1}{N-r+1}\right), \quad (1)$$

where P_r is the corrected number of active microorganism particles, N is the number of sampling holes at all levels, and r is the measured number of active microorganism colonies.

The microbial particle concentration is calculated according to Eq. (2):

$$C = \frac{N_1 + N_2 + ... + N_6}{Q \cdot T} \times 1000, \quad (2)$$

where C is the concentration of microorganisms, CFU/m^3; N_1–N_6 is the corrected number of colonies on a scale from 1 to 6 (geometric average of two repeated measurements); Q is the sampling airflow, L/min; and T is the sampling time, min.

The plate count technique was also used to measure the concentration of fungal bioaerosols, which was expressed as CFU/m^3 (Liu et al., 2020b).

2.4.3 Microorganisms in the packing

A microbial count of the packing in the three reaction segments was conducted, and wet packing samples of 1.0 g were taken from each sampling port when the THIBF was in stable operation. Then, these samples were mixed with 100 mL sterile water and stirred for 10 min.

Heterotrophic bacteria, fungi, and acidophilic sulfur bacteria were counted by the culture counting method. The media used were nutrient agar medium, Martin's medium and Waksman medium. The final value was the average of three parallel samples at a given time, and the number of microorganisms is expressed in CFU/g packing.

3 Results and discussion

3.1 Performance of the THIBF system

The performance of the THIBF system was investigated within the operation time of 216 d. The inlet concentrations, outlet concentrations and removal efficiencies (REs) of hydrogen sulfide, ammonia and TVOCs in the TSIBF were monitored and analyzed, and the results are shown in Figs. 2(a)–2(c).

During the start-up periods of the TSIBF, the REs of hydrogen sulfide, ammonia and TVOCs were low and unstable, and the outlet concentrations of these pollutants fluctuated to a certain extent. The time required for hydrogen sulfide, ammonia and TVOCs to reach high and stable removal efficiencies of 98.75%, 98.55% and 97.55% were 31 d, 15 d and 23 d, respectively, indicating that the corresponding degrading microorganisms had grown and adapted to the environment of the reactor. Generally, the start-up time of the TSIBF is shorter than that of conventional biofilters for the treatment of complex gases containing hydrogen sulfide, ammonia and TVOCs due to the inoculation of each reaction segment of the reactor with the predominant high-efficiency functional microorganisms (Liu et al., 2009).

After the start-up period of the TSIBF, the numbers and activities of different types of functional microorganisms increased continuously and began to play an important role. The REs of hydrogen sulfide, ammonia and TVOCs tended to be stable and relatively high. During the stable operation period from 31 d to 216 d, the inlet concentrations of hydrogen sulfide, ammonia and TVOCs were 3.45–36.73 mg/m^3, 4.33–29.53 mg/m^3 and 10.21–82.28 mg/m^3, respectively, whereas the effluent concentrations were 0–0.97 mg/m^3, 0–0.30 mg/m^3 and 0.16–2.66 mg/m^3, respectively. The REs were 97.52%–100%, 99.04%–100% and 96.77%–99.29%, respectively. Although the concentrations of hydrogen sulfide, ammonia and TVOCs fluctuated to a certain extent during the stable operation period, the REs of these pollutants were not significantly affected and remained relatively high.

Compared with the study of Han et al. (2020b) of the treatment of ammonia, hydrogen sulfide and TVOCs by a biofilter process, the TSIBF demonstrated relatively higher REs. The results can be explained from two aspects. Firstly, there were high-efficiency acidophilic sulfur bacteria, fungi and heterotrophic bacteria, which were determined to degrade sulfur-containing compounds, hydrophobic pollutants and hydrophilic pollutants, respectively, in the inlet gases in the ABRS, FRS and HBRS, respectively, of the TSIBF, due to rational segmentation. Different types of microorganisms exhibited suitable growth, and the metabolic environmental conditions in each segment were optimized to achieve the highest activity. Additionally, the segmented removal of various components could also reduce inhibitory effects on each component. Hydrogen sulfide, ammonia and TVOCs were removed efficiently and stably by synergistic process of chemical, physicochemical and biodegradation which occurred in different segments, respectively.

3.2 Removal characteristics of the main odors and VOC substances

Odors and VOCs with relatively high concentrations were identified by mass spectrum retrieval. Nineteen kinds of odors and VOCs, including ammonia, hydrogen sulfide, methyl mercaptan, methyl sulfide, ethyl mercaptan, *etc.*, were identified which are similar to those of Liu et al. (2009). During the stable operation of the THIBF system,

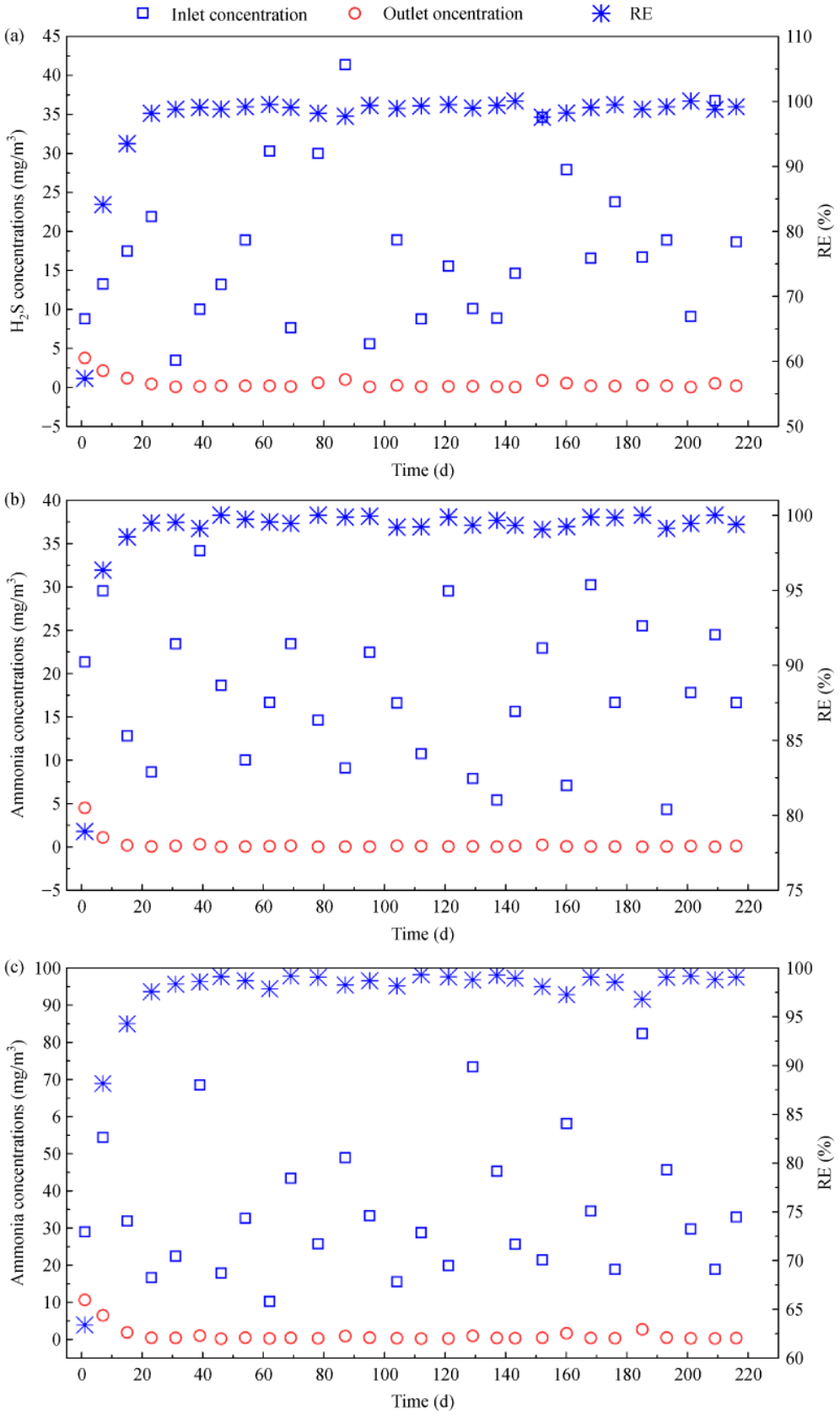

Fig. 2 (a) Inlet concentrations, outlet concentrations and REs for hydrogen sulfide; (b) Inlet concentrations, outlet concentrations and REs for ammonia and (c) Inlet concentrations, outlet concentrations and REs for TVOCs.

the removal characteristics of the main odorous and VOC substances were studied, which are shown in Table 3.

Table 3 demonstrates that TSIBF can efficiently remove 19 kinds of odors and VOCs simultaneously, and the REs of all the pollutants were above 94.39%. However, the total REs of different substances were different. The REs of dimethyl disulfide, p-xylene, α-pinene and cyclohexane, which have been proven to be four hydrophobic and non-biodegradable pollutants, were 96.47±6.48%, 97.17±6.15%, 95.86±4.94% and 94.39±6.61%, respectively. The REs of the other odor and VOC components were all above 98.0%.

Additionally, the REs of the odor and VOCs in the THIBF system were significantly different in each reaction segments. The REs of ammonia, hydrogen sulfide, methyl mercaptan, methyl sulfide, ethyl mercaptan and dimethyl disulfide in the ABRS were higher, with values of 65.31±3.63%, 74.87±3.92%, 70.25±3.43%, 71.08±3.78%, 76.22±3.73% and 73.59±3.66%, respectively. Except that the ammonia might be chemically neutralized by acidic substances in the ABRS, the removal of sulfur-containing pollutants mainly relies on its efficient degradation by acidophilic sulfur bacteria (Wang et al., 2019; Zhan et al., 2019).

The hydrophobic pollutants could be effectively removed in the FRS. The REs were 78.26±5.05% for benzene, 76.33±4.78% for toluene, 81.58±5.44% for ethylbenzene, 78.81±4.97% for styrene, 79.32±5.16% for *p*-xylene, 81.44±4.93% for *α*-pinene and 79.04±5.07% for cyclohexane. These pollutants are hydrophobic and nonbiodegradable. However, the mycelium of the fungi grown in the FRS was well developed, and the enzyme activity was high, resulting in a certain advantage for treating refractory substances (Khatami et al., 2019; Wang et al., 2019). Other hydrophilic and biodegradable odors and VOCs had higher REs in the HBRS: 78.18±4.26% for methanol, 74.23±5.05% for ethanol, 75.34±5.84% for acetic acid, 76.02±5.59% for acetone, 71.35±4.86% for ethyl acetate and 70.4±4.75% for butyl acetate.

The significant differences in the odors and VOCs removal characteristics in the different reaction segments can be attributed to the differences in environmental conditions and the resulting differences in number and activities of dominant microorganisms. Previous studies have shown that the mass transfer of pollutants and the formation of efficient microbial populations are closely related to the variations in removal characteristics (Sun et al., 2019; Zhang et al., 2019). The gas-liquid mass transfer and biodegradation processes in the different segments of the TSIBF are relatively independent. The environmental conditions in each reaction segment are controlled to make strongly acidic, weakly acidic, and neutral in the ABRS, FRS and HBRS, respectively. The gas-liquid mass transfer of pollutants with different acid-base properties and

Table 3 Removal characteristics of main odor and VOC substances

Pollutants	Total removal efficiency (%)	Removal efficiency of ABRS (%)	Removal efficiency of FRS (%)	Removal efficiency of HBRS (%)	Average removal efficiency (%)
Ammonia	99.57±5.82	65.31±3.63	23.06±1.64	11.63±0.85	99.57
Hydrogen sulfide	98.96±6.17	74.87±3.92	9.76±0.81	14.39±1.03	98.96
Methylmercaptan	99.32±6.34	70.25±3.43	13.45±1.15	15.62±1.27	99.32
Dimethyl sulfide	99.05±6.51	71.08±3.78	11.22±0.96	16.75±1.33	99.05
Ethanethiol	98.75±6.22	76.22±3.73	13.15±1.04	9.38±0.93	98.75
Dimethyl disulfide	96.47±6.48	73.59±3.66	12.06±0.98	10.82±0.95	96.47
Methanol	99.92±6.78	6.75±0.52	14.99±1.05	78.18±4.26	99.92
Ethanol	99.84±6.61	7.87±0.63	17.74±1.33	74.23±5.05	99.84
Acetic acid	99.67±7.35	8.05±0.77	16.28±1.29	75.34±5.84	99.67
Acetone	99.21±7.14	10.14±0.95	13.05±1.41	76.02±5.59	99.21
Ethyl acetate	99.48±6.82	9.76±0.85	18.37±1.33	71.35±4.86	99.48
Butyl acetate	98.93±6.90	11.38±1.05	17.15±1.56	70.4±4.75	98.93
Benzene	99.12±5.87	4.81±0.25	78.26±5.05	16.05±1.21	99.12
Toluene	98.76±6.22	5.26±0.34	76.33±4.78	17.17±0.81	98.76
Ethyl benzene	98.05±5.17	5.84±0.31	81.58±5.44	10.63±0.69	96.25
Styrene	98.38±5.92	6.67±0.38	78.81±4.97	12.9±1.14	97.88
P-xylene	97.17±6.15	4.22±0.19	79.32±5.16	13.63±1.23	96.17
α-pinene	95.86±4.94	4.55±0.30	81.44±4.93	9.87±0.73	95.86
Cyclohexane	94.39±6.61	5.54±0.42	79.04±5.07	9.81±0.72	94.39

solubilities is quite different, and the number and activity of microorganisms also varies.

The solubility and biodegradability of various odors and VOCs as well as the types of microorganisms suitable for degradation in the different segments are quite different. Therefore, the different reaction segments exhibit different removal performances for the odors and VOCs. Due to the complementarity of the three segments, the simultaneous and efficient removal of all kinds of pollutants can be achieved in the THIBF system.

3.3 Changes in the concentrations of microorganisms

The removal of odors and VOCs by biofilter systems is mainly based on biodegradation by microorganisms in the packing. In the different operation periods of the THIBF, the concentrations of culturable heterotrophic bacteria, fungi and acidophilic sulfur bacteria in each reaction segment were analyzed. The results are shown in Table 4.

Table 4 shows that the concentrations of acidophilic sulfur bacteria in the ABRS, fungi in the FRS and heterotrophic bacteria in the HBRS had values of 2.92×10^7 CFU/g packing, 9.14×10^7 CFU/g packing and 8.44×10^7 CFU/g packing, respectively, in the early stage of operation (15 d). This is because a certain amount of acidophilic sulfur bacteria, fungi and heterotrophic bacteria were inoculated in the ABRS, FRS and HBRS, respectively, before the operation of the THIBF. The concentrations of acidophilic sulfur bacteria in the ABRS, fungi in the FRS and heterotrophic bacteria in the HBRS increased during the start-up operation of the reactor. On the 30th day, the concentrations of acidophilic sulfur bacteria in the ABRS, fungi in the FRS and heterotrophic bacteria in the HBRS were 4.95×10^7 CFU/g packing, 2.38×10^8 CFU/g packing and 6.17×10^8 CFU/g packing, respectively.

Additionally, the predominant microorganism types and distribution quantities in the different reaction segments were different when the THIBF system was in stable operation. In the ABRS, acidophilic sulfur bacteria with a high concentration of $10^7 - 10^8$ CFU/g packing were always the predominant species, whereas the concentrations of heterotrophic bacteria and fungi were low ($\times 10^4 - 10^5$ CFU/g packing). The strongly acidic environment in the ABRS makes it difficult for heterotrophic bacteria and fungi to grow, but it is suitable for the growth of acidophilic sulfur bacteria. The FRS had a weakly acidic environment and low packing humidity, which are beneficial to the growth of fungi instead of heterotrophic bacteria and acidophilic sulfur bacteria; therefore, the concentration of fungi in this reaction segment was significantly higher ($\times 10^8$ CFU/g) (Rene et al., 2012). In the HBRS, the concentration of heterotrophic bacteria was 10^8 CFU/g, whereas the concentrations of acidophilic sulfur bacteria and fungi were relatively low ($\times 10^3$ CFU/g and $\times 10^5$ CFU/g, respectively). Combined with the segmented REs of the pollutants, it can be concluded that the variations in the segmented REs of different types of odors and VOCs might be due to differences in the types of the predominant microorganisms, the reaction conditions and the mass transfer. The predominant microorganisms in the ABRS segment are acidophilic sulfur bacteria, and the pollutants removed are mainly hydrogen sulfide and sulfur-containing organic substances. The predominant microorganisms in the FRS are fungi, and the main pollutants removed are hydrophobic pollutants. The predominant microorganisms in the HBRS segment are heterotrophic

Table 4 Change in the concentrations of microorganisms in different segments of the TSIBF

Reaction segment	Time (d)	Heterotrophic bacteria (CFU/g packing)	Fungi (CFU/g packing)	Acidophilic sulfur bacteria (CFU/g packing)
ABRS	15	7.84×10^5	5.01×10^5	2.92×10^7
	30	8.02×10^4	4.33×10^4	4.95×10^7
	90	6.47×10^4	3.94×10^4	7.77×10^7
	150	3.28×10^4	4.21×10^4	1.29×10^8
	210	2.95×10^4	3.77×10^4	6.06×10^7
FRS	15	2.86×10^6	9.14×10^7	3.47×10^6
	30	6.09×10^5	2.38×10^8	2.26×10^6
	90	4.34×10^5	7.34×10^8	4.05×10^6
	150	4.71×10^5	6.86×10^8	5.58×10^6
	210	4.22×10^5	7.40×10^8	4.91×10^6
HBRS	15	8.44×10^7	2.34×10^5	5.49×10^3
	30	6.17×10^8	3.15×10^5	4.78×10^3
	90	7.88×10^8	2.78×10^5	3.55×10^3
	150	5.79×10^8	3.02×10^5	5.73×10^3
	210	6.45×10^8	2.96×10^5	4.99×10^3

bacteria, which mainly remove hydrophilic organic pollutants. The results indicated that the removal of different odors and VOCs was related to the predominant microbial populations formed in the different segments.

Therefore, through reasonable process design and control of reaction conditions, the acidophilic sulfur bacteria, fungi and heterotrophic bacteria that were initially inoculated into the three different reaction segments of the THIBF can always be predominant in terms of quantity. By designing different reaction segments in which different types of microorganisms were predominant, the distribution of different types of microorganisms in ABRS, FRS and HBRS was achieved, enabling the effective removal of different types of odors and VOCs, respectively.

3.4 Bioaerosol emissions

The bioaerosols emitted from the biofiltration system might come from the volatilization of microorganisms in the packing or from microorganisms in the inlet air (Ghanbarian et al., 2020). The variations in the inlet and outlet concentrations and REs of the culturable heterotrophic bacteria, fungi and acidophilic sulfur bacteria in the THIBF during the operation period was studied. The results are shown in Fig. 3.

Figure 3 shows that during 216 d of operation, the concentrations of culturable heterotrophic bacteria, fungi and acidophilic sulfur bacteria in the outlet were 249–1389 CFU/m^3, 277–940 CFU/m^3 and 32–104 CFU/m^3, respectively. Although the concentrations of odors and VOCs in the outlet of the THIBF system are low, certain concentrations of culturable heterotrophic bacteria, fungi and acidophilic sulfur bacteria are emitted from the THIBF. The concentrations of culturable heterotrophic bacteria and fungi in the inlet of the THIBF were 1248–6327 CFU/m^3 and 876–3129 CFU/m^3, respectively. Compared to the inlet concentrations of the THIBF, the concentrations of culturable heterotrophic bacteria and fungi released from the THIBF were reduced by 74.37%–83.45% and 67.77%–78.62%, respectively. As also demonstrated, the concentrations of culturable heterotrophic bacteria and fungi in the outlet of the THIBF fluctuated greatly, but the REs were relatively stable, and no significant differences were observed between the stable operation and start-up period. Additionally, the concentration of culturable acidophilic sulfur bacteria in the inlet was relatively low. The outlet concentration of culturable acidophilic sulfur bacteria in the TSIBF was higher than or close to the inlet concentration, indicating that the efficiency of the THIBF to remove the culturable acidophilic sulfur bacteria is limited.

Compared with the study of bioaerosol emissions by Agarwal et al. (2016), this study found relatively lower concentrations of emitted culturable heterotrophic bacteria, fungi and acidophilic sulfur bacteria from the THIBF, especially culturable heterotrophic bacteria and fungi. Thus, it can be concluded that the THIBF cannot only effectively remove odors and VOCs with complex components, but also effectively reduce the emissions of bioaerosols from the system, thus reducing the potential risk of bioaerosols. This advantage is related to the capture of bioaerosols by the high-density packings in each segment of the THIBF. Moreover, the segmented structure of the THIBF makes it easier for the predominant functional microorganisms to enrich and degrade specific pollutants in each segment, which also formed a single microbial type in each segment, thereby reducing the emissions of bioaerosols.

Table 5 shows the emissions of culturable heterotrophic bacteria, fungi and acidophilic sulfur bacteria in different segments of the THIBF. It was found that the emission concentrations of culturable heterotrophic bacteria, fungi and acidophilic sulfur bacteria were different in the different segments.

For culturable heterotrophic bacteria, the emission concentration of the TSIBF was only 745±67 CFU/m^3, which was 78.78% lower than that of the inlet. The highest RE occurred in the FRS (50.50%), followed by the ABRS (38.21%), and the lowest RE is observed in the HBRS (30.63%).

For culturable fungi, the emission concentration of the system was only 556±49 CFU/m^3, which was 70.19% lower than that of the inlet. The highest RE was observed in the HBRS (54.72%), whereas the REs in the ABRS and FRS were relatively low (22.10% and 15.49%, respectively).

For culturable acidophilic sulfur bacteria, the concentrations increased from 70±25 CFU/m^3 to 425±56 CFU/m^3 after passing through the ABRS, and the emission concentration increased by nearly 5 times. When passing through the FRS and HBRS, most of the acidophilic sulfur bacteria were captured, with the REs of 48.24% and 57.27%, respectively, which was related to the lower quantities of the culturable acidophilic sulfur bacteria growing in these two segments.

The differences in the emissions and removal of culturable heterotrophic bacteria, fungi and acidophilic sulfur bacteria from the different segments are mainly due to different environmental parameters and types of predominant microorganisms (Melse et al., 2012; Van der Heyden et al., 2019). Acidophilic sulfur bacteria, fungi and heterotrophic bacteria were the predominant bacteria in the ABRS, FRS and HBRS, respectively, and the concentrations of other types of microorganisms were significantly lower than those of the predominant microorganisms. In addition, under the designed reaction conditions of the THIBF, the main function of the packings in each reaction segment on the bioaerosols is emission rather than capture. Therefore, the concentrations of acidophilic sulfur bacteria, fungi and heterotrophic bacteria emitted from the ABRS, FRS and HBRS were higher (Sun et al., 2018; Liu et al., 2020b).

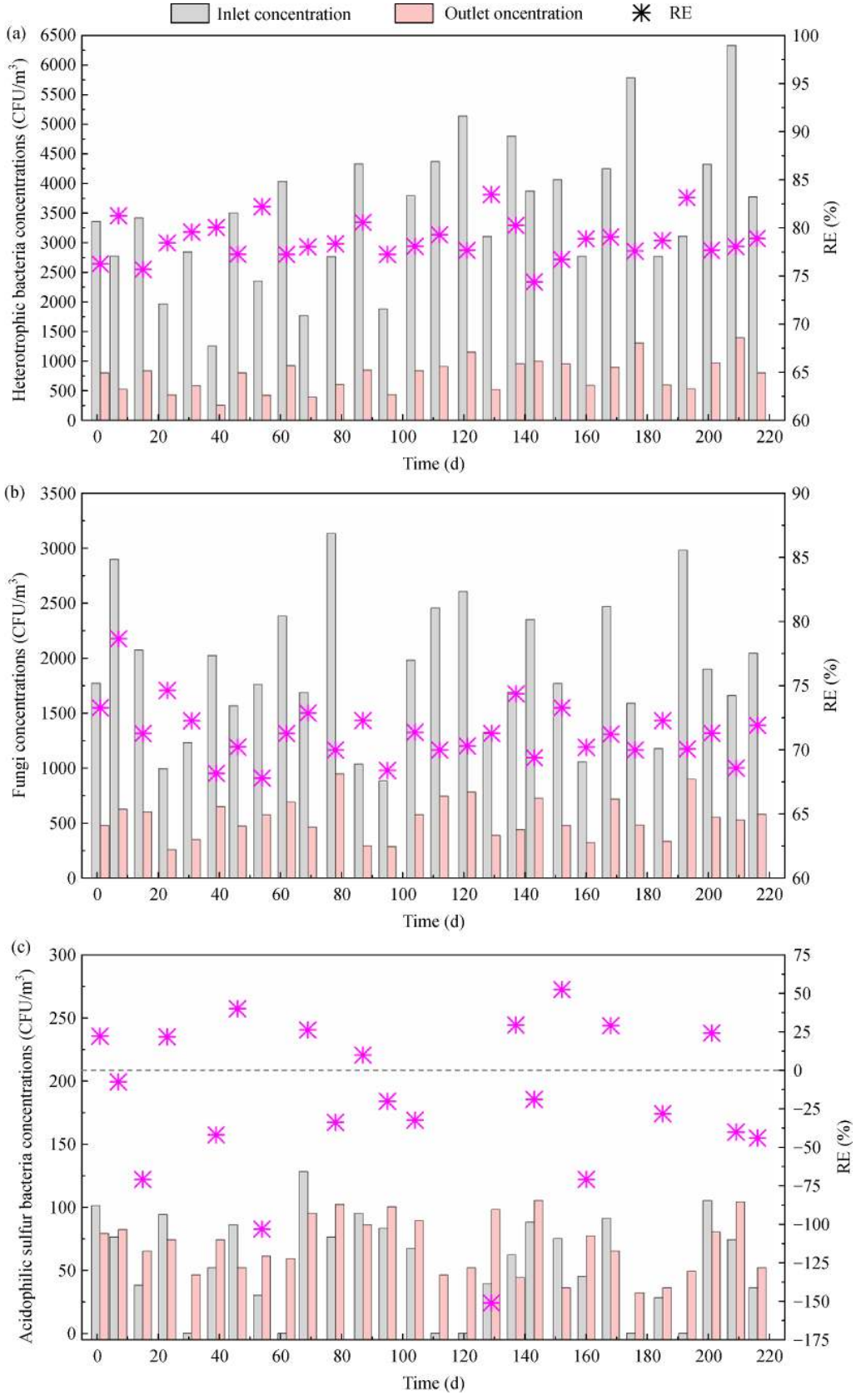

Fig. 3 (a) Inlet concentrations, outlet concentrations and REs of heterotrophic bacteria in the TSIBF; (b) Inlet concentrations, outlet concentrations and REs of fungi in the TSIBF; (c) Inlet concentrations, outlet concentrations and REs of acidophilic sulfur bacteria in the TSIBF.

Table 5 Segmented emissions of bioaerosol in the THIBF

Total RE (%)	Microorganisms	Inlet concentration (CFU/m^3)	Outlet concentration in ABRS (CFU/m^3)	Outlet concentration in FRS (CFU/m^3)	Outlet concentration in HBRS (CFU/m^3)
78.78	Heterotrophic bacteria	3512±230	2170±148	1074±85	745±67
70.19	Fungi	1865±142	1453±136	1228±104	556±49
–34.29	Acidophilic sulfur bacteria	70±25	425±56	220±30	94±28

Different types of dominant microorganisms were concentrated in different segments to achieve the segmented and efficient removal of different odors and VOCs in the THIBF. Additionally, the segmenting of emission concentrations and REs of cultivable heterotrophic bacteria and culturable fungi was found in the THIBF because of the segmented dominant microorganisms, thereby reducing the emission concentration of bioaerosols.

3.5 Emission mechanisms of bioaerosols in the THIBF

Previous studies have indicated that there are two mechanisms for the emission of bioaerosols from odor and VOC biofiltration systems: the direct volatilization of microorganisms from the packing surface and the escape of microorganisms that were not captured by the packing.

The surface of the packing is covered with a biofilm consisting of numerous microorganisms. In the THIBF, the biofilm falls off under the joint action of the updraft and the shear force on the surface of the biofilm, causing the microorganisms to be desorbed and escape under the action of the airflow when the gas passes through the packing. The mechanism of biofilm desorbed from biofiltration systems may include the automatic peeling of old membranes due to the loss of nutrients, erosion under gas shear stress, abrasion caused by high concentrations of pollutants or extreme environments, and viral lysis (Rittmann, 1989; Gjaltema et al., 1995; Tijhuis et al., 1995; Nicolella et al., 1996). Therefore, the emission of bioaerosols in the outlet of the biofiltration system is related to the composition and concentration of the inlet gas pollutants, the characteristics of the biofilm and the operating conditions. For the TSIBF, the large quantities of acidophilic sulfur bacteria, fungi and heterotrophic bacteria in the packings of the ABRS, FRS and HBRS, respectively, can be considered to be the persistent source of outlet bioaerosols emissions.

ABRS is the first segment of the system, in which the pH of the packing range from 2.0 to 3.0. In extremely acidic environments, high concentrations of odors and VOCs cause biofilms to erode, resulting in considerable damage, and bioaerosols are volatilized relatively easily (Vanek et al., 2015). In the FRS and HBRS, the erosion and dissipation caused by extreme environments or high pollutant concentrations are relatively low, and the dominant mechanism of bioaerosol emissions might be erosion caused by gas shear stress. Generally, due to the high density and rough surface of packing, dense biofilms tend to form (Liu et al., 2020b). Therefore, the emissions by the TSIBF are lower than those determined by other researchers.

When the bioaerosol in the inlet passes through the packing layer, it would adhere to the packing because of the collision with the packing particles, and is thus captured by the packing. The capture mechanisms of inlet bioaerosols in packing include inertial deposition, gravity effects, Brownian deposition and flow line capture effects (Ibanga et al., 2018). Brownian deposition caused by diffusion collision is relevant to small particles with a size of 0.1–1 μm (Ottengraf and Konings, 1991). The gravity effect has an obvious effect on aerosol particles with sizes greater than 0.5 μm, and the flow line capture effect often occurs under the action of high-speed airflow (Ottengraf and Konings, 1991). Generally, the sizes of bioaerosols produced by MSW are larger than 1 μm (Agarwal et al., 2016). Therefore, it was speculated that TSIBF is probably mainly based on the gravity effect and flow line capture effects to capture bioaerosols.

Generally, in the TSIBF, the capture of heterotrophic bacteria and fungi in the inlet was significantly greater than the volatilization of the microorganisms in the packing. However, the emission of acidophilic sulfur bacteria in the ABRS results in a certain concentration of acidophilic sulfur bacteria in the outlet. Therefore, in the TSIBF, rational segmentation, filling of high-density packings and the accumulation of the predominant functional microorganisms in each segment enhanced the capture effect of the packings on bioaerosols, thus reducing the concentrations emitted from the TSIBF.

Thus, to reduce the emission of bioaerosol and to avoid the excessive microbial escape, the design and operation parameters of the biofiltration system should be optimized under the premise of effective removal of odor and VOCs.

4 Conclusions

The TSIBF system, which is divided into ABRS, FRS and HBRS, can effectively remove hydrogen sulfide, ammonia and VOCs produced in MSW comprehensive treatment plants. Each segment of the TSIBF can effectively remove different types of odors and VOCs. Sulfur-containing pollutants, hydrophobic pollutants and hydrophilic pollu-

tants are mainly removed in the ABRS, FRS and HBRS, respectively. The types and distribution of the predominant microorganisms in the different reaction segments of the TSIBF were different: acidophilic sulfur bacteria in the ABRS, fungi in the FRS and heterotrophic bacteria in the HBRS. The emission concentrations of culturable heterotrophic bacteria, fungi and acidophilic sulfur bacteria were 249–1296 CFU/m^3, 277–940 CFU/m^3 and 36–104 CFU/m^3, respectively. In the TSIBF, the emission of bioaerosols depends on volatilization from the packing and the capture of microorganisms from the inlet, and the extent of capture was greater than that of volatilization from the packings. The rational segmentation of the TSIBF, filling of high-density packings and the accumulation of the predominant functional microorganisms in each segment enhance the capture of bioaerosols and reduce the emission of microorganisms in the outlet.

Acknowledgements This work was financially supported by the key Projects in the National Science & Technology Pillar Program of China during the Twelfth Five-Year Plan Period (No. 2013BAC25B00-004).

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