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Simultaneous removal of bioaerosols, odors and volatile organic compounds from a wastewater treatment plant by a full-scale integrated reactor

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ARTICLE INFO

Article history:
Received 28 March 2020
Received in revised form 2 June 2020
Accepted 2 July 2020
Available online 4 July 2020

Keywords:
Odors
VOCs
Bioaerosols
Two-phase biofilter
Adsorption
Risk assessment

ABSTRACT

Biological control of odors and bioaerosols in wastewater treatment plants (WWTPs) have gained more attention in recent years. The simultaneous removal of odors, volatile organic compounds (VOCs) and bioaerosols in each unit of a full-scale integrated-reactor (FIR) in a sludge dewatering room was investigated. The average removal efficiencies (REs) of odors, VOCs and bioaerosols were recorded as 98.5 %, 94.7 % and 86.4 %, respectively, at an inlet flow rate of 5760 m³/h. The RE of each unit decreased, and the activated carbon adsorption zone (AZ) played a more important role as the inlet flow rate increased. The REs of hydrophobic compounds were higher than those of hydrophilic compounds. For bioaerosols, roughly 35 % of airborne heterotrophic bacteria (HB) was removed in the low-pH zone (LPZ) while over 30 % of total fungi (TF) was removed in the neutral-pH zone (NPZ). Most bioaerosols removed by the biofilter (BF) had a particle size larger than 4.7 μm while bioaerosols with small particle size were apt to be adsorbed by AZ. The microbial community in the BF changed significantly at different units. Health risks were found to be associated with H2S rather than with bioaerosols at the FIR outlet.

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1. Introduction

With the increasing population and the shortage of land resources, wastewater treatment plants (WWTPs) once positioned on the outskirts of cities are now surrounded by new residential areas and gradually forming new urban centers (Sun et al., 2019). WWTPs as potential malodor sources are a growing problem, leading to a rise in public grievances against the occurrence of odorous compounds in areas adjacent to these plants (Lebrero et al., 2011). Thus, demand for air pollution control systems that provide breathable air nuisance free is increasing.

Bioaerosols are viable, air-suspended particles containing microorganisms (e.g., bacteria, fungi, and viruses), endotoxins, and mycotoxins, another important air pollutant produced by WWTPs (Noh et al., 2019). Prolonged exposure to malodors and bioaerosols poses a direct threat to human health, causing emotional stress to physical symptoms (Sironi et al., 2010). The world is currently facing the COVID-19 pandemic caused by virus SARS-CoV-2. Liu et al. (2020) found many air samples collected from the air of a newly built hospital appeared to be positive with SARS-CoV-2. It has been demonstrated that the SARS-CoV-2 virus remains viable in aerosols for at least 3 h and remains stable on plastic and stainless steel for up to 72 h (Correia et al., 2020). While the capacity for pure aerosol transmission is denied by the World Health Organization, there is growing evidence of aerosol-driven infection (Correia et al., 2020; Liu et al., 2020; Patients et al., 2020; Santarpi et al., 2020; Tada et al., 2019; Yao et al., 2020). A variety of bioaerosols, odors and VOCs volatilize from WWTPs during biological metabolism in wastewater treatment processes involving raw wastewater collection, aeration and sludge treatment facilities, such as pump houses, screen rooms, grit chambers, aeration tanks and sludge dewatering rooms (Noh et al., 2019; Sánchez-Monedero et al., 2008; Yang et al., 2019b). The most significant air pollutants emitted by WWTPs are nitrogen compounds including ammonia (NH3); sulfur compounds including hydrogen sulfide (H2S) and mercaptans, bioaerosols, organic acids, chloride, alkanes and aromatics (Kasperczyk et al., 2019; Yang et al., 2019a). Generally, odorous compounds in WWTPs are mostly volatile, corrosive and irritating malodorous nuisances with very low odor thresholds (Alinezhad et al., 2019). Therefore, as

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https://doi.org/10.1016/j.psep.2020.07.003
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Nomenclatures

| Abbreviation | Description                                      |
|--------------|--------------------------------------------------|
| WWTP         | Wastewater treatment plant                       |
| VOCs         | Volatile organic compounds                       |
| FIR          | Full-scale integrated-reactor                    |
| RE           | Removal efficiency                               |
| HB           | Heterotrophic bacterial                          |
| TF           | Total fungi                                      |
| SP           | Suspended phase                                  |
| IP           | Immobilized phase                                |
| LPZ          | Low-pH zone                                     |
| NPZ          | Neutral-pH zone                                  |
| BF           | Biofilter                                        |
| AZ           | Adsorption zone                                  |
| EBRT         | Empty bed retention time                         |
| PU           | Polyurethane                                     |
| VR           | Volcanic rock                                    |
| ACF          | Activated carbon fiber                           |
| NAT          | Non-acidophilic Thiobacillus                    |
| AT           | Acidophilic Thiobacillus                         |
| AOB          | Ammonia oxidizing bacteria                        |
| NOB          | Nitrite oxidizing bacteria                        |

sites of pathogenic microorganism accumulation, WWTPs should not only meet conventional requirements for removing pollutants in wastewater, but must also be able to effectively eliminate air pollution for public health and environmental protection. Air pollutants produced by WWTPs have thus become a central public concern.

Various technologies have been proposed and applied for the treatment of odorous components and can be classified into physical/chemical systems (e.g., chemical scrubbers, incinerators, activated carbon adsorption, and ozone oxidation) and biological systems (e.g., biofiltration, biotrickling filtration, bioscrubbing, and activated sludge diffusion technologies) (Arellano-García et al., 2018; Gabriel et al., 2013; Qiu and Deshusses, 2017). Each of these technologies have advantages and disadvantages and specific ranges of economic application (Mudilar et al., 2010). Compared to physisochemical treatments, biological methods are less costly, no secondary waste generation, involve mild process conditions and are ecologically clean (Bindra et al., 2015; Rene et al., 2006). Among them, BFs have proved to be some of the most promising tools for odorous compounds treatment in WWTPs due to being compact and moderately priced and well suited to applications requiring operational flexibility (Alinezhad et al., 2019; Lebrero et al., 2011; Lewkowska et al., 2016). Moreover, BFs have been evaluated as means to control airborne microorganisms released in WWTPs (Sanchez-Monedero et al., 2003), as when filled with high density surface area filler they may intercept microorganisms in air inlets. Sanchez-Monedero et al. (2003) found BFs to reduce concentrations of Aspergillus fumigatus and mesophilic bacteria by more than 90 % and 39 % on average, respectively. Due to its high purification efficiency for some recalcitrant substrates and provision of large surface areas for microorganisms to attach to, activated carbon adsorption is considered a reliable technology for air pollutant removal with several advantages for WWTP application (Bansode et al., 2003; Lillo-Ródenas et al., 2005). Li et al. (2011b) found that over 85 % of airborne bacteria and fungi emitted from an oxidation ditch were taken up by activated carbon within 80 h of contact time.

However, while the removal performance of BFs is generally satisfactory under moderate conditions, it is reduced significantly when BFs are applied to undiluted recalcitrant waste gases at large quantities. The complex mixture of odorous compounds in WWTPs poses a special challenge to the use of BFs. The accumulation of sulfuric acid during the oxidation of H2S processes reduces both pH levels and microbial activity, and ultimately affects treatment efficiency. Therefore, it is difficult to remove pollutants with different properties by using a single conventional biological or physical/chemical approach. To overcome these shortcomings, new technologies such as two-stage BF, integrated-bioreactors, and trickle-bed bioreactors have been developed. Chung et al. (2007) reported that a two-stage BF can indeed improve NH3 removal from waste gases containing high H2S concentrations. As multiple pollutants are concurrently degraded in a BF, antagonistic, neutral, and synergistic interactions may occur between pollutants (Mohammad et al., 2017; Strauss et al., 2004). Two-phase or two-stage BFs are considered as means to enhance removal performance and alleviate antagonistic substrate interactions (Yang et al., 2018a).

Despite numerous studies conducted on odors and VOCs, few studies have addressed the simultaneous removal of bioaerosols, odors and VOCs in contaminated gas streams. On this basis, a FIR is used in this study to simultaneously remove multiple pollutants from the sludge dewatering room of a WWTP in Beijing. The FIR includes a two-phase BF and an activated carbon AZ. The first BF phase is suspended phase (SP), which includes suspended microorganisms that remove some hydrophilic compounds and humidify the air stream. The second phase is the immobilized phase (IP), which is divided into the LPZ and NPZ, which are filled with polyurethane (PU) and volcanic rock (VR) forming a fixed biofilm, respectively. The LPZ was operated at pH 4–5 and dominated by acidophilic microorganisms mainly to remove hydrophobic compounds. Different species of microbes live in suitable environments to degrade target pollutants in each unit. Due to a large number of microorganisms attached to the packing medium, BF bioaerosols removal performance is affected by an increasing of air flow rate. The activated carbon AZ is set after the BF for the thorough removal of bioaerosols and recalcitrant substrates and to maintain stable FIR removal. Based on emission and composition characteristics of exhaust in the WWTP, the treatment performance, long-term operational stability, and microbial community in the FIR for the simultaneous removal of multiple pollutants were investigated.

2. Materials and methods

2.1. FIR setup

The FIR was constructed to treat sludge dewatering exhaust in a WWTP in Beijing (Fig. 1), China. Nearly 2.0 × 10^5 m^3/day of domestic wastewater was treated in the WWTP based on an anaerobic–anoxic–oxic process. The exhaust control system includes a gas collection unit, transmission pipeline and FIR. Sludge dewatering room exhaust was collected by pipes and introduced into the FIR treatment system through a blower. The two-phase BF composited a fiberglass cuboid with a height of 5.6 m, base area of 12 m, and total packed volume of 52.8 m³. The empty bed retention time (EBRT) was set to 24–48 s by changing the feeding air stream from 4320 to 8640 m³/h. Inlet air was humidified by passing through SP. The two zones of the IP were filled with PU and VR medium, respectively. The AZ includes a pretreatment unit, adsorption unit and desorption unit. The adsorption unit was packed with modified activated carbon fiber (ACF) filler. The temperature and humidity of the system were measured using a Dewpoint Thermohygrometer (WD-35612, OAKTON, USA). The FIR parameters are shown in Table 1.
2.2. Operating conditions

The FIR was operated for 307 d to treat sludge dewatering exhaust. The two-phase BF was initially inoculated with activated sludge supernatant obtained from the WWTP and with odor-degrading microorganisms from a BF in our laboratory for a fast start-up. A mineral nutrient solution with 0.54 g/L KH₂PO₄, 1 g/L NaCl, 0.025 g/L MgSO₄, 0.02 g/L CaCl₂, 0.005 g/L FeSO₄·4H₂O, 0.000088 g/L MnSO₄·H₂O and 0.2 g/L glucose was sprayed onto the packing material at a flow rate of 5 m³/h and 12 times a day for 1 min at a time to supply nutrients. The pH of the nutrient solution was adjusted via acid or alkali addition. Exhaust passes through the two-phase BF in an upflow direction through the air distribution device and then enters the AZ. The pretreatment unit of the AZ is mainly designed to reduce moisture in the air stream. Three sets of adsorption units form the main module of the AZ; two work online and the other operates in a regeneration state during operation. Steam was used in the desorption unit for desorption. Temperatures were kept at 150–180 °C by controlling the steam flow. Pollutants adsorbed in the ACF were desorbed and discharged with the steam. The desorption process was complete when temperatures fell below 60 °C and when the adsorption capacity of activated carbon fiber had recovered. The stages of FIR operation are shown in Table 2.

2.3. Sampling of bioaerosols, gas and packing medium

2.3.1. Bioaerosol sampling

A 6-stage, 400-hole Andersen impactor with aerodynamic cut-size diameters of 7.0, 4.7, 3.3, 2.1, 1.1, and 0.65 mm was applied for bioaerosol particle collection. The air sample was pumped by a Quick Take 30 Sample Pump at a constant flow of 28.3 L/min for 2–5 min. Parallel samples were collected at each sampling site. Airborne HB were incubated in nutrient agar (BR, Aoboxing Biotech Co., China) at 37 °C for 48 h. Airborne TF were incubated in Martin Broth medium (BR, Aoboxing Biotech Co., China) at 28 °C for 5 days.
2.3.2. Gas and packing medium sampling

The gases were sampled periodically at a sample port in 10 L Tedlar bags. For microbial enumeration, 1.0 mL of liquor in SP, 1.0 g of PU (cut into pieces) and 1.0 mL of leakage from the VR filter were taken from the sampling ports, mixed with 100 mL of sterile water and agitated for 10 min. The HB was incubated in nutrient agar (BR, Aoboxing Biotech, Co., China) at 37 °C for 48 h. TF were incubated in Martin Broth medium (BR, Aoboxing Biotech Co., China) at 28 °C for 5 days. Thiosulfate and modified Waksman media were used to culture non-acidophilic *Thiobacillus* (NAT) and acidophilic *Thiobacillus* (AT), respectively (Cho et al., 1991). Inoculation was conducted in triplicate, and the average value for each sample was calculated. The distinct colonies were sub-cultured and purified by streaking on fresh agar plates after incubation. The population sizes of ammonia oxidizing bacteria (AOB) and nitrite oxidizing bacteria (NOB) of packing mediums were determined by the MPN method. The basal medium and procedures used were selected based on a previous study (Yasuda et al., 2010). Counts of AOB and NOB were recorded as MPN g⁻¹ of dry packing medium or MPN mL⁻¹ of liquor.

2.4. Analytical methods

2.4.1. Chemical analysis

Gas chromatography (GC, Agilent 6890N, USA) equipped with a flame photometric detector (FPD) and HP-5 capillary column (30 m × 0.32 mm × 0.25 μm, Hewlett Packard, USA) using nitrogen as a carrier gas was applied to measure H₂S. Before sampling, all canisters were cleaned and then vacuumed using humid N₂ pure gas (99.999 %). Oven, injection and detector temperatures were set to 45 °C, 100 °C and 200 °C, respectively. A spectrophotometer (Xinyue-T6) was used to analyze NH₃ in gas samples via Nessler reagent spectrophotometry. The measurement of nitrate, nitrite, and sulfate concentrations in the liquid phase was described in our previous studies (Liu et al., 2011, 2017). VOCs were analyzed using a gas chromatograph (GC, Agilent 6890N, USA) equipped with a DB-5MS column (60 m × 0.32 mm × 1.0 μm) and a mass spectrometer (MS, Agilent 5973MSD, USA). The United States Environmental Protection Agency’s (US EPA’s) Method TO-15 was adopted. The sample air was first passed through a stainless steel cryotrap (1/8 in × 8 cm) packed with fine glass beads and cooled with liquid nitrogen at −170 °C. Several standard gases were used to calibrate VOCs, and Helium 5.0 was used as a carrier gas. The GC oven was first set to 35 °C for 5 min, to 150 °C at 5 °C /min, to 220 °C at 15 °C /min, and then kept at this temperature for 7 min. This process took 40 min, and the temperature of injection port was 100 °C. Full scan and selected ion monitoring (SIM) modes were utilized in MS for quantitative determination at a scan range 15–300 amu and scan rate of 0.2 scan/s. Electron impact ionization at 70 eV was used for each measurement. Ion source, quadrupole and transfer line temperatures were maintained at 230 °C, 150 °C and 280 °C, respectively. Each compound was identified based on its retention time and mass spectrum. For more details, refer to Zhu et al. (2016).

2.4.2. Bioaerosol assay

Positive-hole correction was used to determine colony count concentrations. The results were calculated as the geometric mean of the replicates and expressed as colony-forming units per cubic meter of air (CFU/m³).

\[
C = \frac{N_1 + N_2 + N_3 + N_4 + N_5 + N_6}{Q \times t} \times 1000
\]

where C denotes microbe concentrations in CFU/m³, N is the total number of microbes in each disk colony in CFU, t is sampling time in minutes, and Q is the gas flow rate during sampling in L/min.

2.5. Health risk assessment

The AERSCREEN model recommended by the US EPA was used to estimate maximum ground-level concentrations of air pollutants emitted from different sources under worst-case meteorological conditions (Asadi et al., 2014). The AERSCREEN model was applied to estimate concentrations of microorganism and gaseous compounds (NH₃, benzene, H₂S, ethylbenzene, and xylene) 50 m–1000 m downwind the FIR outlet (Table S1). The FIR outlet was considered as a point source of emissions; software input parameters employed are shown in Table S2.

The inhalation of microorganism and gaseous compounds was the main exposure pathway considered. Most microorganism and gaseous compounds released from the FIR pose noncarcinogenic risks. Exposure concentrations and noncarcinogenic risks of gaseous compounds were estimated using Eqs. (1) and (2), and those of bioaerosols were estimated using Eqs. (3) and (4).

\[
EC = \frac{C_1 \times ET \times EF \times ED}{AT \times 365 \times 24}
\]

\[
HQ_1 = \frac{EC}{RfD}
\]

\[
ADD = C_2 \times IR \times EF \times ED
\]

\[
HQ_2 = \frac{ADD}{RfD}
\]

where EC is exposure concentration (μg/m³), C₁ is air pollutant concentration (μg/m³), C₂ is bioaerosols concentration (CFU/m³), ET is inhalation exposure time (8 h/d), EF is exposure frequency (250 d/year), ED is exposure duration (25 year), AT is average lifespan (77 years), IR is inhalation rate (17.7 m³ d⁻¹ for an adult), BW is body weight (66.1 kg for an adult), HQ is the hazard quotient, and RfC is the reference concentration (mg/m³). The RfC values of these compounds were obtained from the Integrated Risk Information System. The World Health Organization has established a bioaerosol exposure limit for ambient air of 300 colony forming units (CFU) per cubic meter (Esquivel-Gonzalez et al., 2017; Soret et al., 2018) employing commonly used culture dependent techniques for bioaerosol quantification. Thus, the value of 300 CFU/m³ was employed in this study. When HQ < 1, non carcinogenic risks can be disregarded. However, when HQ > 1, potential adverse health risks are of concern (Chen and Carter, 2017; Liu et al., 2018).

3. Results and discussion

3.1. Removal performance of odors

During the sludge treatment process, many odors, bioaerosols, and VOCs including sulfur-containing compounds, nitrogen-containing compounds, aromatic compounds, and benzene series are generated and released (Liu et al., 2011a; Lv et al., 2016; Weng et al., 2015). A total of 18 pollutants, including 6 benzenes, 3 ketones, 2 inorganic odor compounds, 2 sulfur organic compounds, 2 esters, an alcohol and a halogenated compounds were detected in this study. The concentration ranges of H₂S, NH₃ and VOCs were recorded as 0.95–41.26 mg/m³, 0.88–8.83 mg/m³, and 0.91–21.37 mg/m³, respectively. Inlet pollutants and bioaerosols concentrations recorded in each stage are shown in Table 2. The FIR operated continuously over four stages for 307 days. Profiles of inlet and outlet concentrations and REs of H₂S and NH₃ are shown in Fig. 2.

During the initial period of adaptation (0–37 d), inlet concentrations of H₂S and NH₃ reached 0.95–15.62 and 0.98–7.55 mg/m³, and outlet concentrations of H₂S and NH₃ were 0.03–1.49 and 0.02–0.59 mg/m³, respectively. The subsequent steady increase in REs indicates that the two-phase BF gradually became rich in
microorganisms, potentially degrading the target contaminants. Microorganisms in the two-phase BF gradually became adapted to highly odors concentrations after the acclimatization period, and the REs of H₂S and NH₃ remained about 95%. These results indicate that 37 days was sufficient for the FIR to successfully start up and reach steady-state conditions. The complete removal of contaminants could be achieved under a steady state. We thus achieved superior odor treatment performance than previous works (Liu et al., 2008, 2017) with the same EBRT, as the FIR was equipped with SP at the air intake port and AZ after the BF enhanced the treatment performance. Fluctuation of REs occurred in stage IV with a continued decrease in EBRT, which can be attributed to the increase in the inlet loading rate and less contact time between contaminants and microorganisms. Past works suggest that odor removal mechanisms are dependent on the adsorption of odorous compounds into the biofilm layer on the media surface where biodegradation takes place, which relies on long residence time (Fletcher et al., 2014). Overall, the REs of H₂S and NH₃ were stabilized at relatively high levels and maintained for nearly 270 days. The contributions of different modules to global performance under a steady state are shown in Table 3.

Removal performance depend on the characteristics of compounds, microbial populations, and operational parameters. From Table 3 we know that NH₃ has a higher RE level in SP than in IP. By contrast, most H₂S was eliminated by the LPZ in IP. Due to differences in hydrophobicity, NH₃ in the inlet stream immediately dissolved in SP through the aeration system and was degraded by microorganisms while less water-soluble compound H₂S was adsorbed by the packing medium and degraded by microorganisms in IP. Although NPZ has a lower intake concentration, NH₃ has a similar RE level in the LPZ and NPZ. This might be ascribed to multisubstrate inhibitions or cross-substrate interactions occurring in a BF (Liu et al., 2008). A high H₂S concentration inhibits nitrification when H₂S and NH₃ are simultaneously treated (Chung et al., 2007). As a less water-soluble compound, H₂S did not affect the removal of NH₃ in SP. Nevertheless, in the NPZ, inhibition between H₂S and NH₃ was relieved with a decrease in H₂S concentrations. Chung et al. (2007) designed a two-stage BF to address inhibits between H₂S and NH₃ and obtained an average 98 % RE for H₂S and 100 % RE for NH₃. The synergistic effects of different reaction conditions in the two-phase BF produce high REs of target pollutants. For the AZ, due to its hydrophobic properties and high surface area, activated carbons are some of the best options for adsorbing molecules with molecular weights of between 45 and 130 (Lillo-Ródenas et al., 2005). The AZ presents good REs of H₂S and NH₃ in low inlet concentrations (0.03–6.71 and 0.01–1.21 mg/m³), playing an important role with increasing air flow rates and ensuring the stable removal performance of odors.
3.2. Removal performance of VOCs

3.2.1. Removal of VOCs

The FIR’s VOCs removal performance is shown in Fig. 3. When VOCs in the inlet flow through the support medium, these pollutants are absorbed by the biofilm and converted into innocuous products such as carbon dioxide, water, and cell mass without generating undesirable byproducts (Moe and Irvine, 2001). As the inlet concentration of VOCs ranged from 0.91 to 21.37 mg/m³, the RE of VOCs achieved over 90 % when FIR was adjusted to 32 days after acclimatization period. When entering a steady state, the average REs of VOCs in stages B, C and D were 96.59 %, 94.72 % and 90.17 %, respectively. Relatively high REs were obtained in the present study relative to other works (Plant et al., 2011; Yang et al., 2019c), which is attributed to synergy between the FIR units. The removal of VOCs slightly fluctuated with an increase in the gas flow rate but quickly recovered. Antagonistic, neutral, and synergistic interactions occur between pollutants when multiple VOCs are concurrently degraded in a BF (Mohammad et al., 2017; Strauss et al., 2004). Two-phase or two-stage BFs are considered as means to enhance the removal performance of VOCs by promoting gas–biofilm transfer rates (Yang et al., 2018a). Using different partition sets in the FIR can efficiently alleviate antagonistic substrate interactions, and microorganisms distributed in different units facilitate degradation under suitable conditions. López et al. (2017) reported that most hydrophilic compounds are assimilated in the first stage and that the highest elimination capacity for hydrophobic substrates is achieved in a second stage when mixtures of VOCs of varying hydrophobicity are biodegraded in a two-stage reactor. In this work, different pH zones were found to be suitable for the removal of VOCs, which were difficult to degrade by conventional biological technologies. Together with the AZ, VOCs can be effectively removed by the FIR.

3.2.2. Individual component removal in different units

Individual component and concentrations of VOCs in inlet gas in the sludge dewatering room were monitored. A total of 16 VOC components were detected, including methyl mercaptan, methyl sulfide, 1,2-dichloroethane, etc. Since an FIR airflow rate of 5760 m³/h is most economically and given sludge dewatering room features, the RE of VOCs in different units in stage III is shown in Table 4.

Overall, the REs of the 16 VOCs exceeded 92.1 % and different in each unit. The characteristics of VOC pollutants, and principally their hydrophobicity, heavily govern their removal performance in BF (Vergara-Fernández et al., 2018). Among them, hydrophilic compounds such as acetic acid, acetone, and methanol have high REs of approximately 98.5 % and reach a significantly higher RE level in SP than hydrophobic compounds. By contrast, most hydrophobic compounds are degraded in SP while recalcitrant substrates with high chain hydrocarbons and most hydrophobic compounds are more amenable to be treated within IP and the AZ. A number of phenomena, including adsorption, absorption, biodegradation and diffusion, are the main processes that cause pollutants to transfer from the gas phase to biofilm (Mohammad et al., 2017). During the simultaneous biofiltration of hydrophilic and hydrophobic VOCs, hydrophilic compounds have a better RE than hydrophobic compounds. Hydrophilic substrates can be more effectively biodegraded in BF because high gas-liquid transfer rates make target pollutants more available to microorganisms (Cheng et al., 2016b). However, the low mass-transfer rates are the main limitations to removal performance for hydrophobic VOCs (Yang et al., 2018a). Ethyl acetate and butyl acetate, which are slightly soluble in water, have similar removal profiles with hydrophilic compounds. This behavior may be related to antagonistic and/or synergistic interactions occurring among multiple contaminants in a BF. Substrate interactions between pollutants can also alter the biodegradation rate of an individual compound. When multiple VOCs are concurrently degraded in a BF, the elimination capacity of each compound is generally suppressed relative to a single compound. Chan and Wu (2012) found the biodegradation rate of toluene to be inversely proportional to the loading rate of a hydrophilic methyl ethyl ketone.

Furthermore, Table 4 shows that higher REs from hydrophobic compounds were obtained in the LPZ. In addition to the characteristics of VOC compounds, microbial populations also play an important role in pollutants degradation. Mohammad et al. (2017) found o-xylene to be more efficiently degraded under all loads applied in a thermophilic gas–phase BF than p-xylene and m-xylene due to the varying affinity of degrading microorganisms towards each xylene isomer. Fungi species have been used in BF for hydrophobic organic compounds. Previous studies show that fungi can take up hydrophobic compounds faster than bacteria due to their aerial mycelia, which form a very large surface area and may come into direct contact with gas flowing through a bioreactor (Liu et al., 2008; Van Groenestijn and Liu, 2002). Due to the presence of higher concentrations of fungi (discussed in section 3.4), the LPZ achieved better hydrophobic VOCs removal performance than the NPZ. After the degradation of SP and LPZ, concentrations of many components decreased significantly. The competition and antagonistic interactions among multiple pollutants is alleviated, which is beneficial to the further degradation of pollutants in NPZ. The AZ improved the REs of recalcitrant substrates even under low VOCs intake concentration conditions. According to Lillo-Rodenas et al. (2005), the porosity and content of surface oxygen groups affect
benzene and toluene adsorption capacity. A combination of high micropore volumes (size <0.7 nm) and an activated carbon surface with low levels of surface oxygen groups is desired for activated carbon with high VOCs adsorption capacities at low concentrations. The results support the concept that compounds with different characteristics are removed thoroughly in different units due to the functionalized partitioning of microbial and reaction modules division of FIR.

3.3. Removal performance of bioaerosols

3.3.1. Removal of airborne HB and TF

Large quantities of bioaerosols containing pathogenic microbes are emitted from sludge dewatering rooms of WWTPs (Han et al., 2018; Li et al., 2011a; Szymak-Szydłowski et al., 2016). To reduce the environmental pollutants and protect the health of workers, it is necessary to reduce levels of air pollution generated by bioaerosols. Fig. 4 shows the REs and concentrations of airborne HB and TF in the FIR inlet and outlet.

Inlet HB concentrations ranged from 868 to 6327 CFU/m³ where the FIR achieved a RE of 69.8–98.2 % and outlet concentrations of 52–1239 CFU/m³. Similarly, the RE of TF reached 75.5–99.8 % with inlet concentrations of 534–2623 CFU/m³ and outlet concentrations of 4–479 CFU/m³. HB concentrations in the inlet were far higher than those of TF, conforming to past research (Li et al., 2011a). Unlike odors and VOCs, the REs of airborne HB and TF directly reached 78.74 % and 85.76 % on average, respectively, in stage I without an adaptation period. To evaluate FIR bioaerosol emissions and retention, differences between outlet and inlet bioaerosol concentrations observed across all operation stages are shown in Fig. 5.

As outlets are located outside, the FIR both retains and emits bioaerosols. It acts as an emission source of outdoor air and a retainer of ambient air in a sludge dewatering room. The observed negative and positive values indicate that the studied biofilter acted as a bioaerosol retainer and emitter, respectively. According to previous research, a BF can act as a net emitter of bioaerosols (the absolute negative value is greater than the positive value), which is a major concern for regulators (Flores-Barbosa et al., 2020; Ibanga et al., 2018). Overall, the FIR served as a retainer for both airborne HB and TF in this study. Bioaerosol capture mechanisms of a BF have been thought to include inertial deposition, diffusional or Brownian deposition and flow line interception (Ottingraf and Konings, 1991), and these combine to affect bioaerosol impingement on solid media material such as bioaerosol-laden air sweeps through the media bed, particles are deposited in the media (Ibanga et al., 2018). However, bioaerosol emissions become apparent in stages III and IV, which is related to an increase in the flow rate. A higher inlet load and thus higher biomass density in a biofilter seems to boost bioaerosol emissions (Chung, 2007; Vergara-Fernández et al., 2012b). With an increase in biofilm thickness, the binding force between the biofilm and packing materials weakens, causing microorganisms to be carried by the airflow and emitted from the biofilter in the form of bioaerosols (Chung, 2007; Vergara-Fernández et al., 2012b; Wang et al., 2018). Yang et al. (2019a) also found bacterial emissions of BF to increase from 449 ± 27 CFU/m³ to 643 ± 46 CFU/m³ as flow rates increased from 1500 m³/h to 3500 m³/h. However, increasing gas velocities first raise bioaerosol emissions and then decrease concentrations due to the shearing force and dilution effects (Hu et al., 2020). Bioaerosols emitted from biofilters include a combination of nonimpacted microorganisms retained in treated process air and those blown off from the surfaces of media particles by the passing airstream, suggesting that the species composition of outlet air may be different from that of inlet air (Martens et al., 2001). The contributions of different units to bioaerosol removal in a steady state are shown in Table 5.

With increasing gas flow rates, the REs of HB and TF decreased in the BF and AZ. Microorganisms attached to BF packing material can be sheared off from the surface of the support media by the gas stream, promoting the emission of microbes in medium (Yang et al., 2018b). Large amounts of airborne HB were removed in the LPZ (up to 38.62 % in stage I) and AZ while most TF removal occurred in the NPZ and AZ. The removal of HB was less effective than that of TF in SP. Factors such as gas flow rates, the moisture content of packing materials, EBRT, particle sizes and shapes, and operating conditions influence the removal performance of bioaerosols (Willeke et al., 1996; Yang et al., 2018b). Due to the different operating conditions of each unit, in this study the influence of microbial characteristics cannot be ignored. In the AZ, inlet concentrations of airborne HB and TF reached 414–2335 and 184–1011 CFU/m³, respectively. It should be noted that the AZ maintained a similar RE for airborne HB and TF and no selectivity was exhibited. The porous structure, large surface area, and hydrophobic surfaces of activated carbon favor the effective attachment of airborne microorganisms (Dunne, 2002; Li et al., 2011b; Lillo-Rödenas et al., 2005). Hydrophobic interactions between cell and absorbent surfaces enable cells to overcome repulsive forces active within a certain distance from adsorbent surfaces and irreversibly attach (Dunne, 2002; Li et al., 2011b). Li et al. (2011b) obtained a 93.75 % adsorption rate for culturable airborne bacteria and a rate of 100 % for fungi over a longer contact.
Fig. 4. Profiles of inlet concentration ($C_{in}$), outlet concentration ($C_{out}$) and removal efficiencies (RE) for (a) HB, (b) TF.

Fig. 5. Bioaerosols emitted or retained during operation (a) HB, (b) TF.

| Stage | Flow rate (m$^3$/h) | Bioaerosols | Two-phase BF | AZ % | Total % |
|-------|---------------------|-------------|--------------|------|---------|
|       |                     |             | SP %         | IP % | LPZ %   | NPZ % |       |       |
| II    | 4320                | HB          | 9.12 ± 0.82  | 37.05 ± 2.83 | 20.76 ± 1.34 | 23.84 ± 1.81 | 90.77 ± 6.35 |       |       |
|       |                     | TF          | 18.33 ± 1.42 | 13.40 ± 1.35 | 36.37 ± 2.31 | 27.76 ± 2.04 | 95.86 ± 3.89 |       |       |
| III   | 5760                | HB          | 7.96 ± 0.62  | 35.80 ± 2.76 | 19.33 ± 1.35 | 22.64 ± 1.67 | 85.73 ± 7.34 |       |       |
|       |                     | TF          | 18.07 ± 1.20 | 12.34 ± 1.37 | 35.15 ± 2.06 | 26.34 ± 1.98 | 91.90 ± 4.89 |       |       |
| IV    | 8640                | HB          | 6.74 ± 0.52  | 33.40 ± 2.43 | 17.25 ± 1.18 | 21.95 ± 1.55 | 79.34 ± 7.38 |       |       |
|       |                     | TF          | 17.35 ± 1.29 | 10.38 ± 0.88 | 33.73 ± 2.14 | 24.28 ± 1.76 | 85.74 ± 6.49 |       |       |

period (8 h) than that used in this work. The AZ plays a crucial role in bioaerosol adsorption, maintaining total REs of bioaerosols at roughly 80%. Additionally, adopting short starvation periods, partial substrate limitation (with the elimination of some pollutants in the case of a gaseous mixture), high moisture levels and the mineral medium frequency of packing materials to avoid stressful conditions (limiting nutrients and drying) may help prevent the generation of bioaerosol emissions during BF operation (Flores-
Barbosa et al., 2020; Vergara-Fernández et al., 2012a, 2012b; Wang et al., 2018).

### 3.3.2. Particle size distributions of culturable airborne bacteria and fungi

The particle sizes of bioaerosols dictate where aerosolized pathogens deposit in the human respiratory tract, and thereafter the pathogens interact with host tissue, which can cause disease and a host of immunological responses (Thomas, 2013). Larger particles (>8 μm) deposit in the upper respiratory tract in a size dependent manner from the nasal passage to larger bronchioles due to inertia, smaller particles (<1–3 mm) diffuse deep into the lung tissue, and fine particles (<2.5 μm) can be directly inhaled and adhere to the respiratory tract and alveolar region (Thomas, 2013; Wang et al., 2019). In addition, particle size plays a key role in aerosols residence times and transmission distances. Aerosols with small particle sizes have longer residence times and can spread further (Gong et al., 1997; Li et al., 2011a). Therefore, the size associated with exposure to aerosols not only relate to their concentrations and species but also to their particle size distributions (Li et al., 2011b). The particle size distributions of airborne HB and TF during stage III in each sample port are shown in Fig. 6.

Particles of airborne HB and TF in the inlet were 1.1–1.2 μm and larger than 7.0 μm, accounting for 31.3 % and 25.6 % of the HB and for 20.2 % and 31.7 % of the TF, respectively. These results are consistent with the bioaerosols emission characteristics in WWTPs reported by Han et al. (2019) and Li et al. (2011a). The particle sizes of HB and TF did not change after passing SP. This may have occurred because (a) HB and TF were removed in SP as incoming aerosol particle size distributions, leaving the particle size distribution unchanged and/or (b) large amounts of HB and TF were absorbed while many microbes in SP were aerosolized due to mechanical disturbance while the particle size distribution of emitted bioaerosols was similar to that of incoming aerosols. Percentages of particles of individual size changed considerably after particles passed through the LPZ. The proportion of microorganisms with particle sizes of less than 4.7 μm increased rapidly in the LPZ and NPZ by roughly 70 %, implying that the packing in IP better retains large particles. The main purpose of packing media in IP is to provide a surface for microorganism attachment and growth and to retain particles. Particle size distributions vary between IP inlet and outlet air with the outlet including larger proportions of smaller particles with 70 % being < 4.7 μm. This may be a result of the filter bed preferentially trapping larger particles from the gas flow, and/or this may simply be the size range emitted by a BF (Sanchez-Monedero et al., 2003). However, after passing through the AZ, bioaerosols with particle sizes of > 4.7 μm accounted for more than 80 %, showing that HB and TF of large particle sizes were difficult to adsorb while small particles were readily captured by the activated carbon, corroborating previous studies (Li et al., 2011b). Micrographs provided by Li et al. (2011b) demonstrate that activated carbon has a rough surface and includes various pore sizes on particle surfaces. Its small macropore structures thus rendering it effective for the adsorption of small particles.

IP and the AZ better retain and capture airborne HB and TF with large and small particles, respectively. The FIR integrates the advantages of single conventional biological and adsorption processes. The combination of a two-phase BF and AZ can effectively remove bioaerosols with different particle sizes. Therefore, the FIR can reduce bioaerosol levels produced through sludge dewatering and thus reduce worker exposure risk.

### 3.4. Microbial characteristics

Fig. 7 shows the evolution of microorganisms in different stages in each unit where NAT and AT consumed sulfur-containing compounds and AOB and NOB oxidized nitrogen-containing compounds. Start-up A and B samples were collected on the 10th and 37th days, representing microorganism characteristics at inoculation and after acclimatization, respectively. The other three samples were collected on days 90, 172 and 260 under stable FIR conditions.

Performance of FIR mainly relies on the microorganisms, which is composed of biodegradation by microorganisms, dissolution and absorption in the aqueous phase, and adsorption by the packing materials and microorganisms (Yang et al., 2018b, 2019a). In SP, as we know from Fig. 7, the number of HB present was 2–3 orders of magnitude higher than the number of TF present with up to 2.43 × 10^7 CFU/mL found. Microbes in the SP outlet sample port were characterized by aerosolized microbes in liquid phase under the aeration system and by an incomplete removal of inlet bioaerosols. As concentrations of HB are far higher than those of TF in SP, HB are more likely to be emitted during aeration. TF aerosols were absorbed by the liquid while fewer were aerosolized than HB, and thus fewer HB aerosols were removed in SP. A similar phenomenon occurred during the uptake of airborne HB in the LPZ. The numbers of AOB and NOB found in SP is consistent with the removal performance of NH3. In stage III, nitrite, nitrate, and NH4+ concentrations reached 1.05 ± 0.19 mg/L, 6.28 ± 0.29 mg/L, and 11.24 ± 0.49 mg/L, respectively, showing that NH4+ was the main inorganic nitrogen-containing compound present in SP. As a less water-soluble compound, H2S has a poor removal effect of roughly a quarter of NH3, RE, though concentrations of NT remained high in the unit. The main products of sulfur-containing compound biooxidation are elemental sulfur, S2- and sulfate. Concentrations of SO4^2- and S2- reached 1.08 ± 0.06 and 5.12 ± 0.23 mg/L in stage III. Most sulfur-containing compounds were present in the form of S2- in SP.

Contaminants in bulky gas are transferred from the gas phase to the water or biofilm phase by diffusion prior to biodegradation. The mass transfer rate of pollutants from air to water is related to hydrophobicity (Cheng et al., 2016b). As aerial mycelia of fungi form a very large surface area and may come into direct contact with gas flows, fungal biofilters act as an alternative means to enhance hydrophobic compound transfer. Furthermore, fungi are more resistant to acidic and dry conditions than bacteria, which is a helpful property when BF treatment processes produce acid compounds such as SO4^2- (Liu et al., 2013). Fungi became more abundant with operation time and gradually became dominant microorganisms in the LPZ. Less water-soluble or hydrophobic pollutants such as H2S, styrene and ethyl benzene can thus be removed in the LPZ than in SP and the NPZ.

More AOB and NOB were detected in SP and the NPZ, as they prefer neutral or alkaline environments, resulting in the RE of NH3 being higher in the LPZ. Generally, microbial compositions, richness, and interactions heavily affect FIR performance. Moreover, the RE of contaminants is also related to operational parameters and intake air concentrations. Due to the microbial functionalized partitioning and reaction modules division, the FIR should be capable of simultaneously treating a variety of pollutants. During steady state operations, inlet concentrations of H2S, NH3, and VOCs fluctuated to some extent, but contaminant REs were not affected. The unique configuration, and reaction conditions of FIR provide optimal conditions for the removal of bioaerosols (both large and small particles), odors and VOCs in this system.

### 3.5. Economic analysis

Our cost analysis of the FIR in the sludge dewatering room of a WWTP was divided into three parts: initial investment, operating, and replacement packing material costs. In this work, investment costs include design and construction, activated carbon adsorp-
Fig. 6. Particle size distributions in each sample port of airborne (a) HB and (b) TF.

Fig. 7. Microorganisms in different units (a) suspended phase (b) low-pH zone (c) neutral-pH zone.
tion reactor and other equipment costs. Design and construction costs are estimated at 725,000 CNY. The cost of the activated carbon adsorption reactor with desorption equipment and condenser is estimated at 200,000 CNY. Other equipment costs (e.g., piping, water baths, pumps, draught fans, etc.) are estimated at 90,000 CNY. Thus, the total initial investment cost for a base year is 1,015,000 CNY. Operating costs expressed as yearly costs including electricity, water consumption and labor expenses are estimated at 65,700, 800, and 60,000 CNY/year, respectively, for a maximum airflow rate of 8640 m$^3$/h, resulting in a total operating cost of 126,500 CNY/year. The durability of BF and AZ packing material is estimated at 10, 5 and 2 years, respectively. Packing material replacement costs are estimated at 840, 1800 and 11,000 CNY/year, respectively, with a total cost of 13,640 CNY/year. Total annual costs of the FIR (over 15 years) at an EBRT of 24 s are estimated at 207,807 CNY/year, i.e. 0.0027 CNY/m$^3$. The result is a little bit higher than a BF of 0.00024 €/m$^3$ treating 20,000 m$^3$/h waste gas reported by Prado et al. (2009).

3.6. Risk assessment of bioaerosols and odors compounds

FIR outlet and downwind concentrations of odors compounds were estimated at a gas flow rate of 5760 m$^3$/h. Peak risk was employed to represent the potential high-risk scenario. The HQ values of different contaminants and RIC values are presented in Table 6. The HQ of H$_2$S at the FIR outlet was 77.77, indicating that risks of H$_2$S exposure were significant. NH$_3$, benzene, ethylbenzene and xylenes risk levels were recorded as 2.40 × 10$^{-2}$, 2.47 × 10$^{-2}$, 1.04 × 10$^{-3}$ and 4.07 × 10$^{-3}$, respectively. As these values are less than 1, corresponding health effects on workers are negligible.

For bioaerosols, the highest concentrations of HB and TF emitted from the FIR outlet reached 1202 and 391 CFU/m$^3$, respectively. Risks posed by total bioaerosols (HB and TF) at the FIR outlet were negligible (HQ = 0.32). At 50 m downwind, the risks posed by all contaminants were valued at less than 1. Risks of these compounds decreased exponentially with increasing distance from the emission source of the FIR outlet, showing that no health effects should result 50 m and further downwind from the FIR outlet. Due its strong capacity to remove multiple pollutants, the studied FIR is suitable for urban areas with high population densities. For the bioaerosols of sludge dewatering room control technologies, UV, H$_2$O$_2$, and O$_3$ are applicable given their low energy consumption and high inactivation efficiency. Photocatalysis disinfection is also a very promising technology for controlling bioaerosols, as it generates less secondary pollution and given its high inactivation efficiency (Hu et al., 2020). Proper ventilation and requiring operators to wear face masks are also strongly encouraged.

4. Conclusion

The average REs of odors, VOCs and bioaerosols were recorded as 98.5 %, 94.7 % and 86.4 %, respectively, when the inlet air flow rate was 5760 m$^3$/h. The simultaneous removal performance of each unit worsened with an increase in the inlet air flow rate. Microbial communities in BF changed significantly under different environmental conditions. Most hydrophobic compounds were removed in the LPZ and hydrophilic compounds were degraded in SP and the NPZ. Approximately 35 % of HB aerosols were removed in the LPZ while over 30 % of TF aerosols were removed in the NPZ, which is related to the distribution of microorganisms in the two-phase BF. The combined use of a two-phase BF and AZ can remove bioaerosols with different particle sizes. The AZ can also thoroughly remove residual pollutants at the outlet of a two-phase BF and ensure stable removal capacities of the system. In conclusion, the FIR can simultaneously remove bioaerosols, odors and VOCs effectively through the synergistic degradation of different units. The health risks were negligible 50 m downwind from the FIR outlet.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgments

This work was financially supported by Beijing Municipal Science and Technology Commission (Z181100005518011).

Appendix A. Supplementary data

Supplementary material related to this article can be found, in the online version, at doi: https://doi.org/10.1016/j.jpsef.2020.07.003.

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Table 6

HQ of different contaminants.

| Site | H$_2$S | NH$_3$ | Benzene | EEZ | Xylenes |
|------|--------|--------|---------|-----|---------|
| 0m   | 2 × 10$^{-3}$ | 0.5 | 3 × 10$^{-2}$ | 1   | 0.1     |
| 50m  | 7.77 × 10$^{-3}$ | 2.40 × 10$^{-2}$ | 2.47 × 10$^{-2}$ | 1.04 × 10$^{-3}$ | 4.07 × 10$^{-2}$ |
| 100m | 4.07 × 10$^{-3}$ | 1.21 × 10$^{-6}$ | 1.27 × 10$^{-6}$ | 5.34 × 10$^{-8}$ | 2.10 × 10$^{-6}$ |

EBZ: ethylbenzene.
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