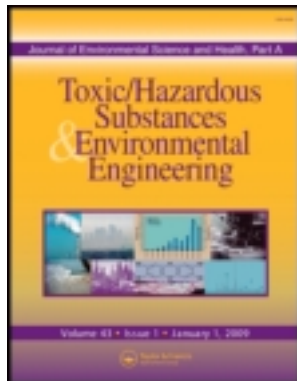


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# Large-scale modular biofiltration system for effective odor removal in a composting facility

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Several different foul odors such as nitrogen-containing groups, sulfur-containing groups, and short-chain fatty-acids commonly emitted from composting facilities. In this study, an experimental laboratory-scale bioreactor was scaled up to build a large-scale modular biofiltration system that can process  $34 \text{ m}^3 \text{ min}^{-1}$  waste gases. This modular reactor system was proven effective in eliminating odors, with a 97% removal efficiency for 96 ppm ammonia, a 98% removal efficiency for 220 ppm amines, and a 100% removal efficiency of other odorous substances. The results of operational parameters indicate that this modular biofiltration system offers long-term operational stability. Specifically, a low pressure drop ( $<45 \text{ mmH}_2\text{O m}^{-1}$ ) was observed, indicating that the packing carrier in bioreactor units does not require frequent replacement. Thus, this modular biofiltration system can be used in field applications to eliminate various odors with compact working volume.

**Keywords:** Odor elimination, large-scale modular system, ammonia, amines, biofiltration.

## Introduction

Composting treatment is considered as the best approach for managing manure waste and food residues. Different agricultural wastes including organic matters are utilized with bulking agents in composting process. However, some common odorous substances, such as ammonia ( $\text{NH}_3$ ), amines ( $\text{R-NH}_2$ ), hydrogen sulfide ( $\text{H}_2\text{S}$ ), organic-sulfur ( $\text{R=S}$  or  $\text{R-SH}$ ) and volatile short-chain fatty-acids (SCFAs, C2–C6) are produced during composting process of livestock farming (animal manure).<sup>[1,2]</sup> Ammonia and amines are the two major compounds among these odorous gases.<sup>[3–5]</sup> Ammonia is colorless with a strong pungent odor, whereas gaseous amines possess a characteristic ammonia smell. Hydrogen sulfide is a very poisonous, flammable gas with the characteristic foul odor of expired eggs. The odors of thiols, particularly those of low molecular weight, are often strong and repulsive. Most short-chain fatty-acids, a subgroup of fatty acids with aliphatic tails of less than six carbons, have a distinctive sour taste and pungent smell.<sup>[4,6–8]</sup>

Odor elimination could be carried out using physical (condensation, adsorption), chemical (chemical scrubbers,

thermal oxidation, catalytic oxidation, ozonation) and biological (biofilters, bioscrubbers, biotrickling filters) methods.<sup>[9]</sup> The selection of an appropriate odor treatment is dependent on the target compounds, site characteristics, odorous substances loading, the properties of odorous air stream, and pumping air flow rates.<sup>[10]</sup> However, the application of physical and chemical methods is limited by the production of secondary pollutants. Therefore, biological techniques for air pollution control may serve as environmentally friendly treatment alternatives for contaminated air streams.<sup>[11]</sup>

Previous studies demonstrated that biofiltration with immobilized cells for odor elimination is significantly effective in the development of a pilot-scale biofiltration system.<sup>[7,12,13]</sup> Some studies reported that a biofiltration system could effectively eliminate high concentrations of  $\text{NH}_3$  and  $\text{H}_2\text{S}$  ranging from 60 ppm to 150 ppm and 120 ppm to 300 ppm for complete removal, respectively.<sup>[4,13–15]</sup>

Biofiltration is considered as the best technique for treating waste air streams containing one or two major contaminants. However, the mixed exhaust treatment is regarded as a hard challenge. Little is known regarding the use of large-scale biofilters for the elimination of different kinds of mixed odors, such as nitrogen (N)-containing or sulfur (S)-containing groups.<sup>[8,16–18]</sup> In this study, five isolates were screened and identified in the laboratory for elimination of odors from N-containing groups, S-containing groups and SCFAs. These isolates were applied in a large-scale biofiltration system with a total working volume of  $11.2 \text{ m}^3$

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to estimate the system's removal efficiencies for odorous substances in a composting facility. To evaluate further the performance of this system, various gas residence times (GRTs) ranging from 20 to 45 s were employed. This modified modular biofiltration system was found to be feasible, showing 98% and 97% removal efficiencies for 220 ppm amines and 96 ppm  $\text{NH}_3$ , respectively, and 100% removal efficiency for t-butyl mercaptan  $[(\text{CH}_3)_3\text{CSH}]$ , ethyl mercaptan ( $\text{C}_2\text{H}_5\text{SH}$ ), acetic acid with various inlet concentrations. The monitoring of operational parameters demonstrated that this system can represent stable operation with low pressure drop and easy maintenance.

## Materials and methods

### Lab-scale biofilter

Granular activated carbon (GAC) was provided as the bacterial supporting material. The removal capacities of the system for odorous substances were estimated using by five isolates: *Arthrobacter* sp. CP1 (for  $\text{NH}_3$  removal), *Paracoccus* sp. CP2 (for amines removal), *Paracoccus* sp. CP3 (for SCFAs removal), *Cellulomonas* sp. CP4 (for Org-S removal), and *Pseudomonas* sp. CP5 (for  $\text{H}_2\text{S}$  removal). Operational parameters with various inlet concentrations and capacities of odorous substances were investigated.

### Microorganism cultivation and medium preparation

All isolated strains inoculated in this study were enriched in nutrient broth (5 g  $\text{L}^{-1}$  yeast extract, 10 g  $\text{L}^{-1}$  tryptone, and 2 g  $\text{L}^{-1}$  dextrose) at 26°C. In long-term experiments, a liquid inflow medium containing 1.0 g  $\text{L}^{-1}$  glucose, 5.4 g  $\text{L}^{-1}$   $\text{KH}_2\text{PO}_4$ , 10.5 g  $\text{L}^{-1}$   $\text{K}_2\text{HPO}_4$ , 0.2 g  $\text{L}^{-1}$   $\text{MgCl}_2 \cdot 6\text{H}_2\text{O}$  and 0.01 g  $\text{L}^{-1}$  Fe(III)-citrate was used. The phosphate buffer capacity was calculated as 54.6 mM per pH.

### Immobilization procedure

In this study, GAC with a uniform size of 4.5 mm and a bulking density of 0.48 g  $\text{cm}^{-3}$  was used as the packing carrier in all bioreactor units. Isolated strains were grown in a 10 L inflow medium for 24 h. The inoculates were placed in a 100 L polyvinyl chloride (PVC) tank containing 90 L of nutrient broth for bacterial growth. When the bacterial count in PVC tanks reached  $10^8$  colony forming units (CFU)  $\text{mL}^{-1}$ , the inflow medium was placed into each bioreactor unit. Approximately 1.34 ton of GAC was packed with the above solution for bacterial growth and pumped with an exhausted fan in each bioreactor unit. It passed an air filter to leach nonessential impurities and then flowed upward through the bottom of each bioreactor unit. Fresh medium was added every 3 d until the number

reached approximately  $10^8$  CFU  $\text{g}^{-1}$ -GAC in each unit. In addition, the medium was confirmed to be fungi free.

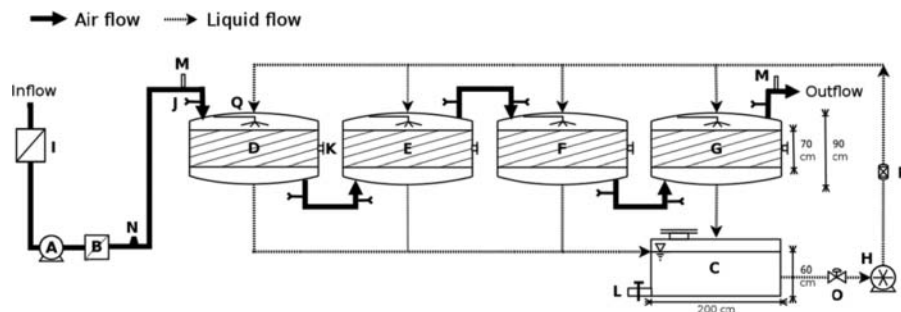
### Source of odorous substances

This biofiltration system was installed at a composting facility in Miao-li County, Taiwan. The facility (approximately 5,300  $\text{m}^2$ ) comprised three storage zones for raw materials, fermentation intermediates and composting products. This study focused on the elimination of odorous substances emitted from the storage zone for raw materials that significantly contribute to odor production, and the odorous substances were pumped through this biofiltration system. The amount of raw materials utilized in the composting process is approximately 24,000 tons per year. The composition of raw materials consisted of 50% animal manure (cattle and swine), 30% agricultural wastes, 10% food-residues, and 10% other organic matters. Before the continuous large-scale biofiltration experiments, the dominant odorous substances in the gas stream were determined as  $\text{NH}_3$ , amines, and SCFAs.

### Scheme of the large-scale biofiltration system

The biofilter system comprised an air compressor (A), an air prefilter (B), a 2.8  $\text{m}^3$  nutrient tank (C), four bioreactor units (D-G), each having a 3.6  $\text{m}^3$  total volume packed with a 2.8  $\text{m}^3$  granular activated carbon (GAC), a peristaltic pump (H), an automatic control system (I), two gas sampling ports (J) and a medium sampling port (K) for each bioreactor unit, a liquid sampling port (L), two pressure drop sampling ports for the influent and effluent (M), a sampling port for flow velocity (N), a valve for controlling the flow rate of recirculated water (O), a flow meter for monitoring the liquid fluent (P), and a spray nozzle for maintaining the moisture content of the biofilter carrier packed in each bioreactor unit (Q) (Fig. 1).

The flow rate of gas inflow could be adjusted by the automatic control panel. The air prefilter was designed for the removal of particles in the gas stream to prevent contamination and obstacles in the bioreactor units. The nutrient tank with peristaltic pump can maintain the proper nutrient and moisture contents in the biofilter media for bacterial growth. To obtain a biofiltration system with ideal odor removal efficiency, the order of bioreactor units should be as presented as below. A great amount of  $\text{NH}_3$  and amines are expected to be eliminated in the former units. *Arthrobacter* sp. CP1 and *Paracoccus* sp. CP2 were inoculated in bioreactor unit 1 for the removal of  $\text{NH}_3$  and amines, respectively. *Paracoccus* sp. CP2 and CP3 were immobilized in bioreactor 2 for the removal of amines and SCFAs, respectively. *Cellulomonas* sp. CP4 was immobilized in bioreactor 3 for the removal of organic sulfur, and *Pseudomonas* sp. CP5 was inoculated for the removal of  $\text{H}_2\text{S}$ .



**Fig. 1.** Schematic diagram of the biofiltration system: (A) air compressor; (B) air prefilter; (C) nutrient tank; (D) bioreactor unit 1 inoculated with *Arthrobacter* sp. CP1 and *Paracoccus* sp. CP2; (E) bioreactor unit 2 inoculated with *Paracoccus* sp. CP2 and *Paracoccus* sp. CP3; (F) bioreactor unit 3 inoculated with *Cellulomonas* sp. CP4; (G) bioreactor unit 4 inoculated with *Pseudomonas* sp. CP5; (H) peristaltic pump; (I) automatic control system; (J) gas sampling port; (K) medium sampling port; (L) liquid sampling port; (M) pressure drop sampling port; (N) sampling port for flow velocity; (O) valve; (P) flow meter; (Q) spray nozzle. The GAC was packed as the square fulfilled with slashes in each bioreactor unit.

### Analytical methods

The inlet and outlet concentrations of SCFA were analyzed using a gas chromatography (Perkin-Elmer Clarus 500, Waltham, MA, USA) equipped with a Stabilwax-DB column (Restek, Bellefonte, PA, USA) and a flame-ionization detector. The concentration of ammonia gas was measured with a portable ammonia-monitoring device (Dräger, Lübeck, Germany). Amines and  $H_2S$  were detected by gas detector tubes (Kitagawa, Kawasaki, Japan). The pH value in the leachates was determined using a pH meter (Hanna pH-211, Woonsocket, RI, USA). The concentration of organic nitrogen was determined using the Kjeldahl method.<sup>[19]</sup>

## Results

### Lab-Scale biofiltration for the removal of odorous substances

To study the removal efficiency of the biofiltration system for odorous substances, five isolates were screened from the sludge of a swine wastewater treatment system. The target compounds were  $NH_3$ , amines [methylamine (MA), dimethylamine (DMA), trimethylamine (TMA)], methanethiol (MT), acetic acid, propionic acid and butyric acid. The removal efficiencies were determined within a 7-d time course experiment.

The results showed that *Paracoccus* sp. CP2 and *Arthrobacter* sp. CP1 can effectively remove 250 ppm TMA and 50 ppm  $NH_3$  within a GRT of 60 s. The co-culture of *Paracoccus* sp. CP2 and *Arthrobacter* sp. CP1 showed 99% and 96% removal efficiencies for the mixed inlet of 50 ppm  $NH_3$  and 250 ppm TMA, respectively, within a GRT of 60 s (Fig. 2). A previous study reported that *Pseudomonas* sp. can remove  $H_2S$  inlet concentrations ranging from 30 ppm to 180 ppm.<sup>[14]</sup> *Cellulomonas* sp. CP4 showed >92% removal efficiency for an MT inlet concentration range of 50 ppm to 300 ppm within a GRT of 45 s (Fig.

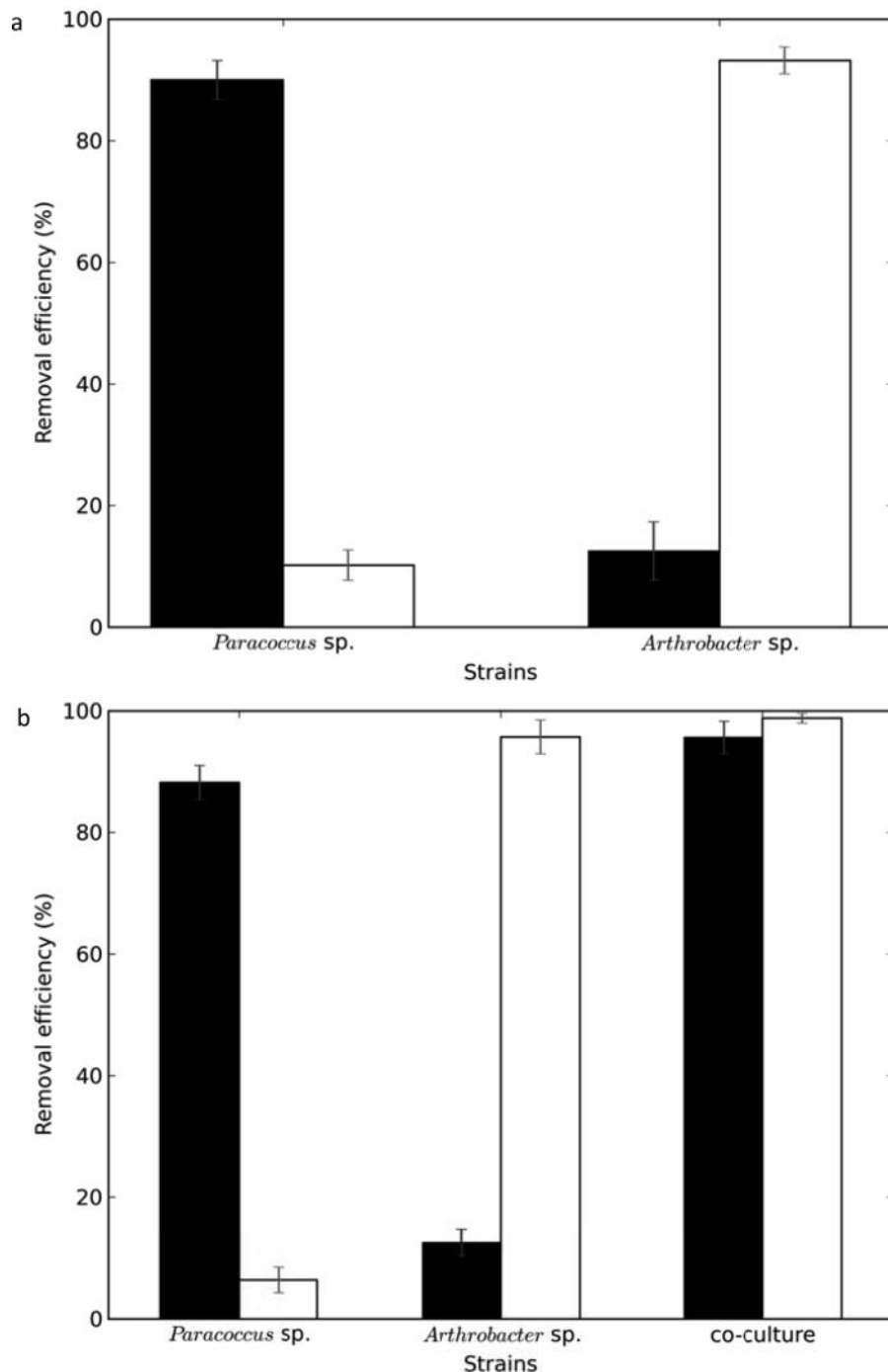
3a). *Paracoccus* sp. CP3 exhibited >90% and >86% removal efficiencies for an SCFA (acetic acid) inlet concentration range of 20 ppm to 500 ppm within GRTs of 45 s and 30 s, respectively. *Paracoccus* sp. CP3 demonstrated >82% removal efficiency for 25 to 500 ppm propionic acid within a GRT of 45 s. These results suggest that the five bacterial isolates can effectively remove different odors emitted from the studied composting facility.

### Elimination of odorous substances in the large-scale biofiltration system

GAC has been proven to be a good adsorbent in many studies.<sup>[10]</sup> Thus, biofilters packed with a structured carrier such as GAC could provide a good support material for odor-biodegrading isolates. According to previous studies, the saturation of physical adsorption of GAC was evaluated. The adsorption capacity ( $N_o$ ) and the adsorption rate constant ( $K$ ) of  $NH_3$ , amines,  $H_2S$ , t-butyl mercaptan, and ethyl mercaptan were 280.87 mg L<sup>-1</sup> and 1.71 L mg<sup>-1</sup> h<sup>-1</sup>, 186 g L<sup>-1</sup> and 0.001 L mg<sup>-1</sup> h<sup>-1</sup>, 5057 mg L<sup>-1</sup> and 70.29 L mg<sup>-1</sup> h<sup>-1</sup>, 5273 mg L<sup>-1</sup> and 0.017 L mg<sup>-1</sup> h<sup>-1</sup>, 23 g L<sup>-1</sup> and 0.004 L mg<sup>-1</sup> h<sup>-1</sup>, respectively (unpublished data). Therefore, the breakthrough time of  $NH_3$ ,  $H_2S$ , t-butyl mercaptan, ethyl mercaptan, and acetic acid were 18, 16, 18, 25, and 44 d, respectively, when odor was continuously introduced into the biofilter without isolates on GAC at 20 ppm to 100 ppm and 60 L h<sup>-1</sup>.

To verify the removal of odorous substances, the removal efficiency could be calculated by detecting the inlet and outlet concentrations of each substance. In this study, GRT was set from 45 s to 20 s to change different inflow rates from 15 m<sup>3</sup> min<sup>-1</sup> to 34 m<sup>3</sup> min<sup>-1</sup>.

The data showed the relationship between the removal efficiencies and inlet concentrations of  $NH_3$  after 9 months of operation (Fig. 4a). When the inlet and outlet concentrations of  $NH_3$  were 96 ppm and 3 ppm, respectively, a 97% removal efficiency was achieved. Similarly, another data showed the relationship between the removal efficiencies

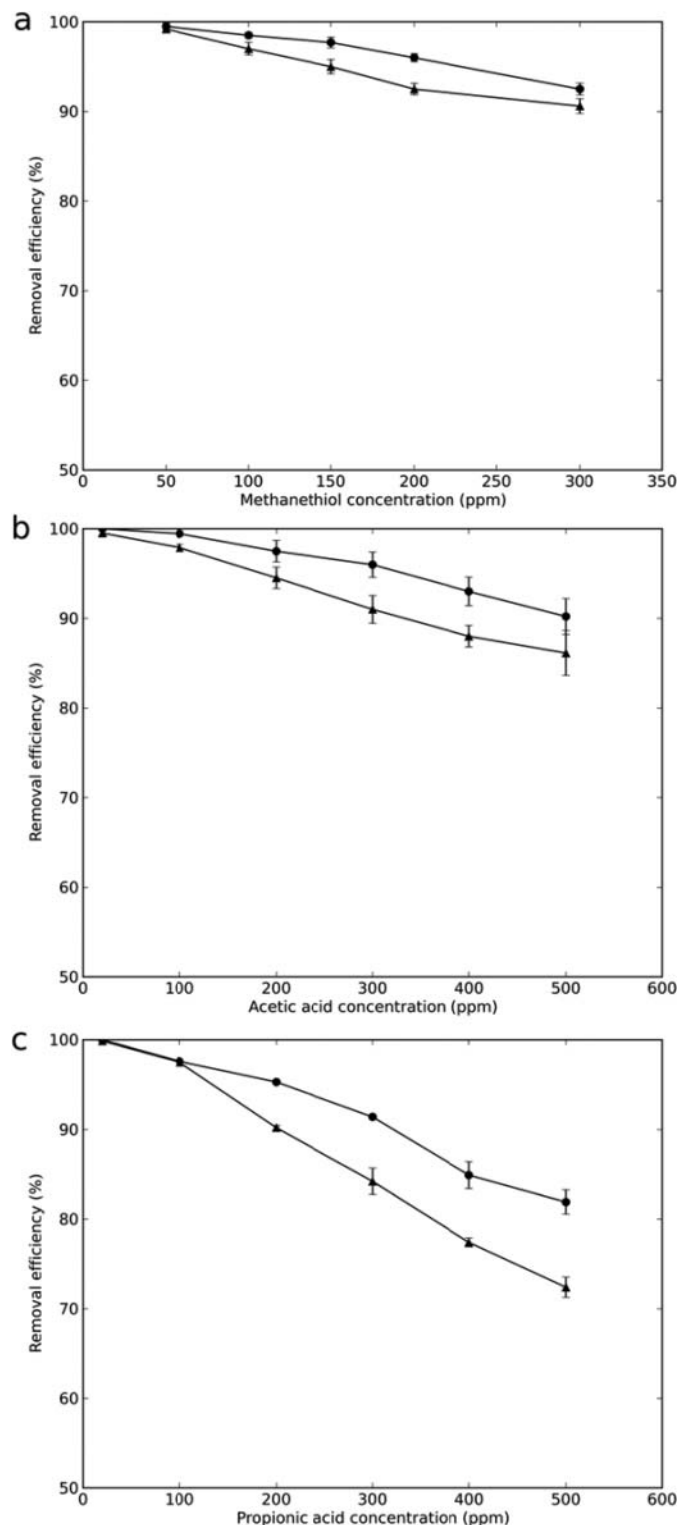


**Fig. 2.** Removal efficiencies for single and combination inlets of TMA and NH<sub>3</sub> with a laboratory reactor test. (a) Black bar marker: 250 ppm TMA, white bar marker: 50 ppm NH<sub>3</sub>; (b), the same legend as (a).

and inlet concentrations of amines (Fig. 4b). When the inlet and outlet concentrations of amines were 220 and 3.5 ppm, respectively, a 98% removal efficiency was achieved. In addition, H<sub>2</sub>S, t-butyl mercaptan, ethyl mercaptan, and acetic acid were also detected in this biofiltration system. In the operational period, the inlet concentrations of t-butyl mercaptan were lower than 13 ppm, and no outlet concentrations were detected (Fig. 5b). The inlet concentrations of both H<sub>2</sub>S and acetic acid were <1 ppm, whereas

their outlet concentrations were not observed (Figs. 5a and 5c). Moreover, propionic acid and other organic sulfur compounds were not detected during the operational period.

Monitoring of operational parameters could provide references for automatic control to maintain a stable operation of this biofiltration system. In a continuous operational period, the pH in the nutrient tank ranged from 6.8 to 7.8 (Fig. 6a). The result showed that the neutral pH was



**Fig. 3.** Removal efficiencies for methanethiol, acetic acid, and propionic acid with a laboratory reactor test. (a) Circle marker: GRT 45 s, triangle marker: GRT 30 s; (b and c), the same legend as (a).

maintained by the buffer system. Moreover, the pressure drop between the inlet and the outlet was determined from  $25 \text{ mmH}_2\text{O m}^{-1}$  to  $45 \text{ mmH}_2\text{O m}^{-1}$  during the operational period (Fig. 6b). To confirm the effectiveness of this biofiltration system with stable operation, the bacterial count of inoculated microflora was monitored by plating. The bacterial count of the inoculated microflora was kept at  $>10^7 \text{ CFU g}^{-1}\text{-GAC}$  throughout the operational period (Fig. 6c).

### Kinetics of ammonia removal

Analysis of the kinetics of  $\text{NH}_3$  removal showed the efficiency and capacity of the biofiltration system. The related data were sorted by regression (Fig. 7) according to the Michaelis–Menten equation<sup>[20]</sup>

$$\frac{1}{R} = \frac{K_s}{V_m} \times \left( \frac{1}{C_{ln}} \right) + \frac{1}{V_m}$$

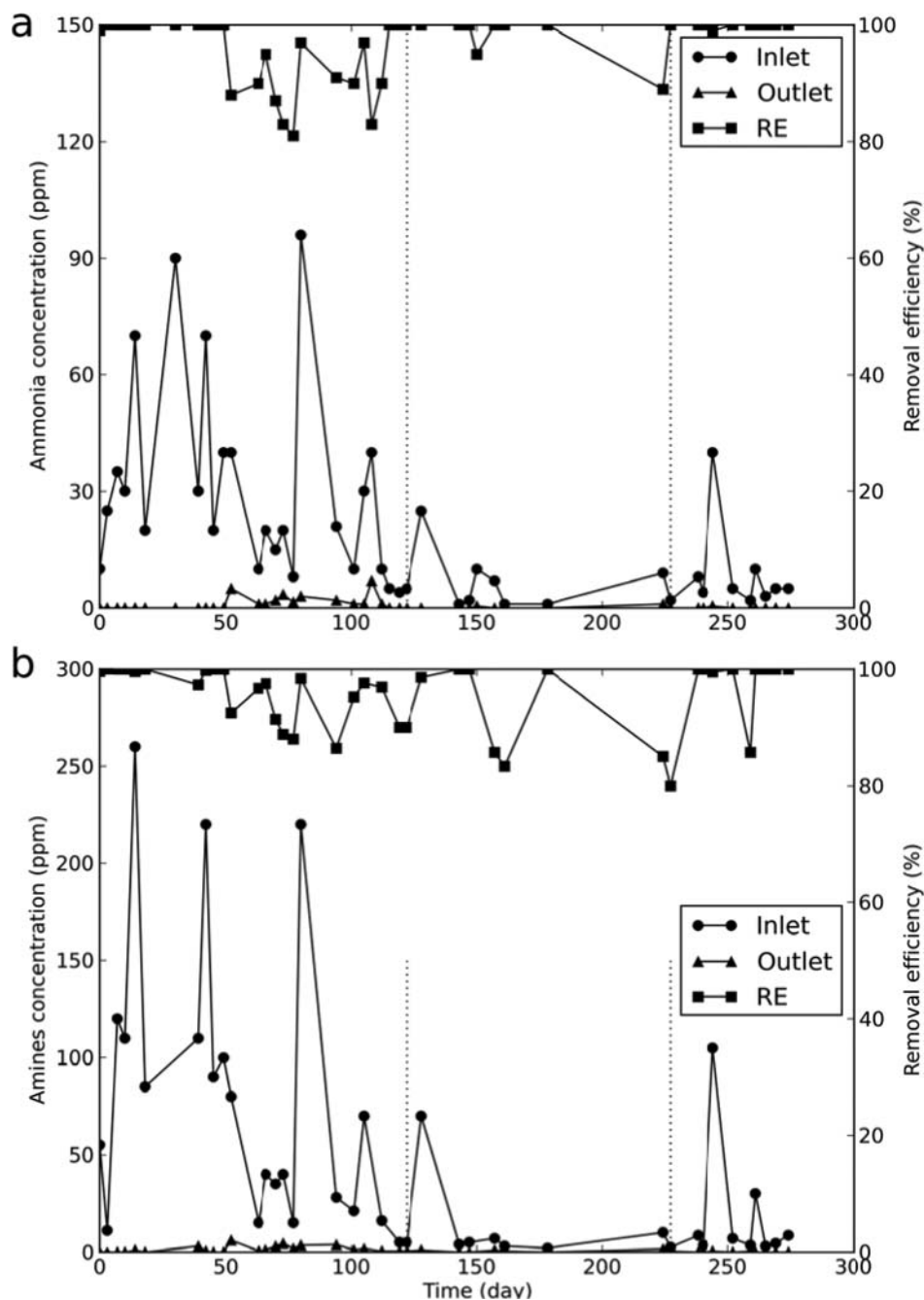
In this equation,  $R$  ( $\text{g N day}^{-1} \text{ kg GAC}^{-1}$ ) is the apparent removal rate calculated as  $(Q \times C_o \times R\%)/W$ , where  $Q$  is the gas flow rate,  $C_o$  is the inlet concentration,  $R\%$  is the gas removal efficiency, and  $W$  is the weight of GAC;  $K_s$  is the apparent half-saturation constant;  $V_m$  is the maximum apparent removal rate; and  $C_{ln}$  (ppmv) is the logarithmic mean concentration of  $\text{NH}_3$  at the inlet and outlet of the second-stage biofilter calculated as  $(C_o - C_e)/\ln(C_o/C_e)$ . The slope and intercept were calculated to obtain the values,  $K_s$  ( $14.178 \text{ mg L}^{-1}$ ) and  $V_m$  ( $6.135 \text{ g-NH}_3 \text{ m}^{-3} \text{ h}^{-1}$ ), respectively.

### Analysis of metabolic products

To study the inhibition of metabolic products, several N-containing compounds such as ammonium cation ( $\text{NH}_4^+$ ), organic nitrogen (Kjeldahl nitrogen), nitrite ( $\text{NO}_2^-$ ), and nitrate ( $\text{NO}_3^-$ ) were detected. N-containing compounds were not only utilized by microorganisms but were also adsorbed in water. Analysis of these N-containing compounds could avoid high nitrogen inhibition for bacterial growth and biochemical activities.<sup>[21,22]</sup> The concentrations of the related N-containing compounds in liquid medium was  $0 \text{ mg L}^{-1}$  Org-N,  $560 \text{ mg L}^{-1} \text{ NH}_4^+$ ,  $10 \text{ mg L}^{-1} \text{ NO}_2^-$ ,  $20 \text{ mg L}^{-1} \text{ NO}_3^-$ , respectively. After the entire operational period, low concentrations of organic nitrogen ( $<6 \text{ mg L}^{-1}$ ) and nitrite ( $<120 \text{ mg L}^{-1}$ ) were detected. However, high concentrations of ammonium cation ( $3,672 \text{ mg L}^{-1}$  on 100 d and  $1,514 \text{ mg L}^{-1}$  on 200 d) and nitrate ( $423 \text{ mg L}^{-1}$  on 100 d and  $2,130 \text{ mg L}^{-1}$  on 200 d) were determined (Fig. 8).

### Discussion

Little work has been devoted on odor elimination from laboratory to full-scale biofiltration in the composting process. In this study, a large-scale bioreactor was designed in

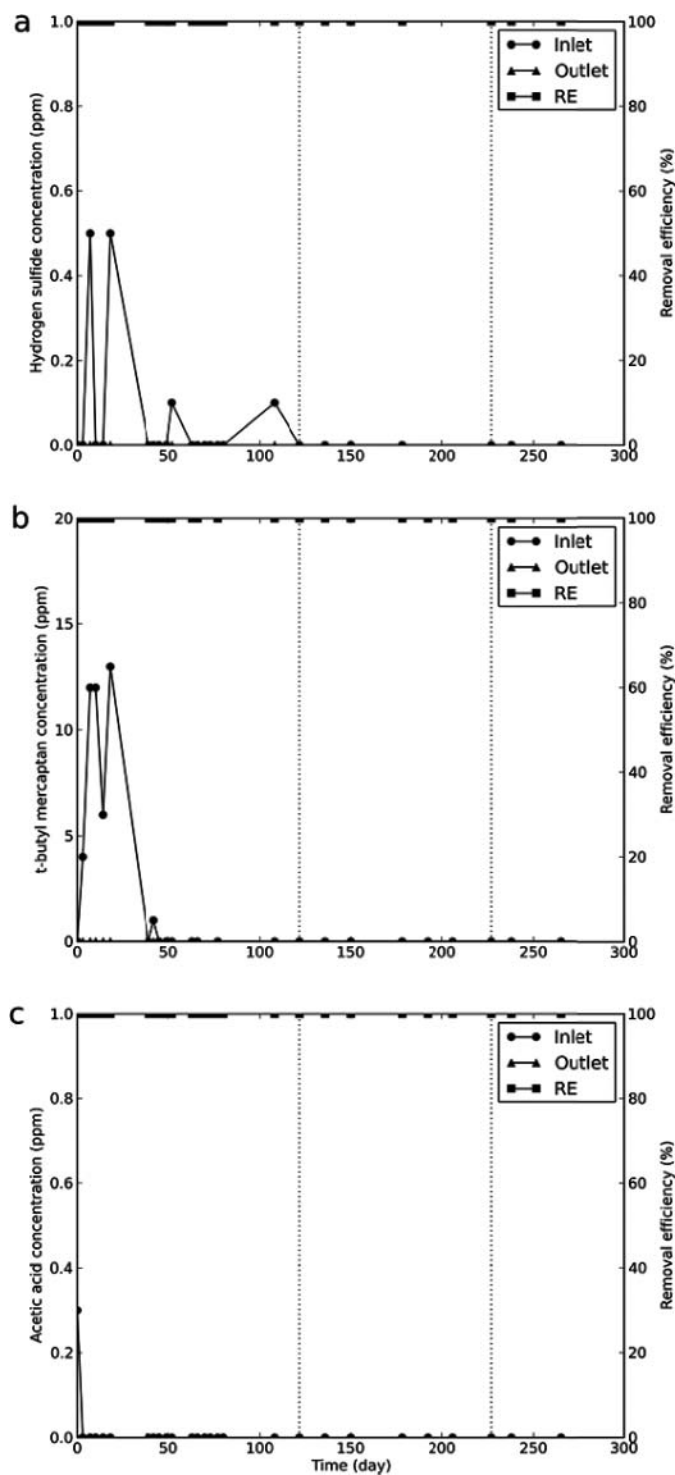


**Fig. 4.** Long-term operational test with the large-scale biofiltration system for dominant odor removal. (a) ammonia ( $\text{NH}_3$ ); (b) amines ( $\text{R-NH}_2$ ). Circle and triangle markers: inlet and outlet concentrations of odorous substances, respectively; square marker: removal efficiencies. Different GRTs: 0 d to 122 d (45 s), 122 d to 227 d (30 s), and 227 d to 274 d (20 s).

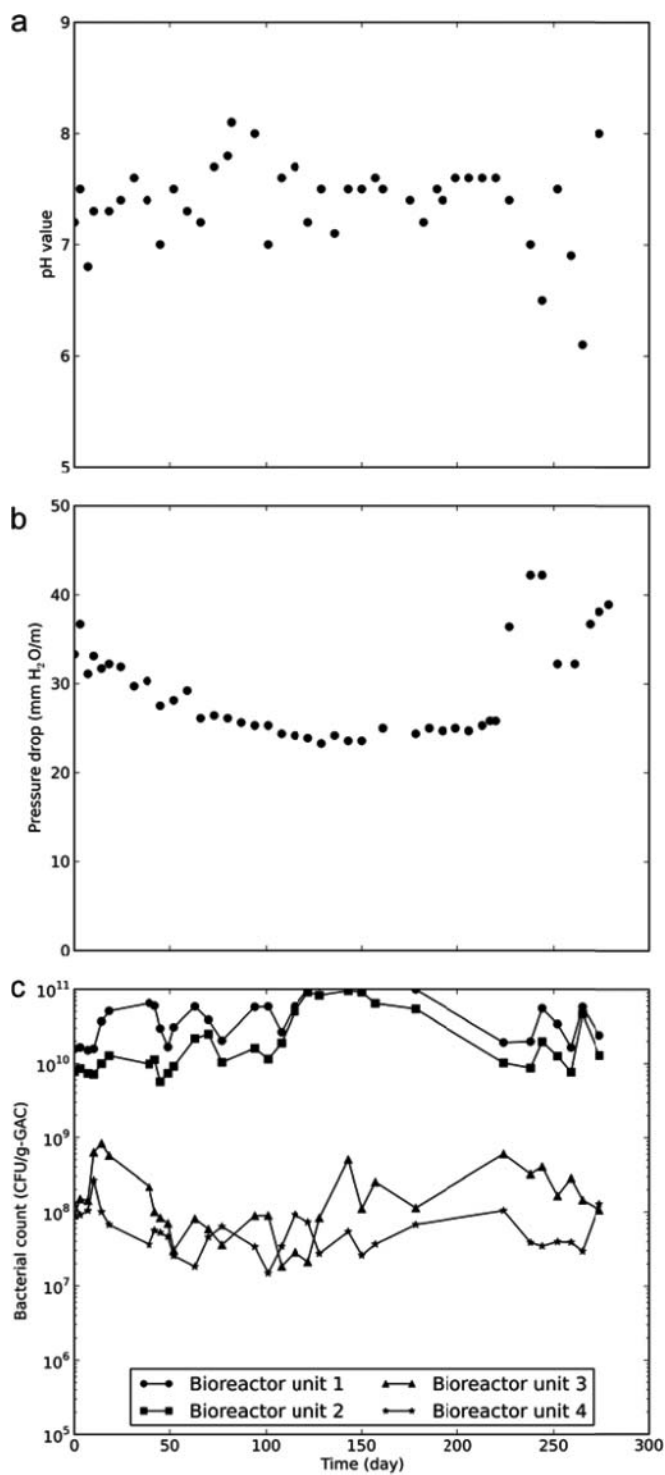
cuboid (Fig. 1). The modular reactor units could respond to field demands, adapting to proper functional bacteria. Several advantages including assembly and disassembly of these units were done easily. In addition, the transportation and installation costs for all components are reduced. Hence, the biofilter of modular reactor units in cuboid provides easy manufacture and assembly.

In the present study,  $\text{NH}_3$  and amines were the dominant substances in the composting facility. *Arthrobacter* sp. CP1

and *Paracoccus* sp. CP2 inoculated in bioreactor unit 1 for the removal of  $\text{NH}_3$  and amines. As a result, the accumulation of high concentrations of  $\text{NH}_3$  and amines in the liquid medium was prevented, causing a rapid change in the pH and growth inhibition of the bacteria inoculated in bioreactor units.<sup>[4,7,9,16]</sup> The low outlet concentrations of  $\text{NH}_3$  and amines during the operational period suggest that these odorous substances could be eliminated effectively by the inoculated aerobic hetero ammonia- and amines-oxidizing

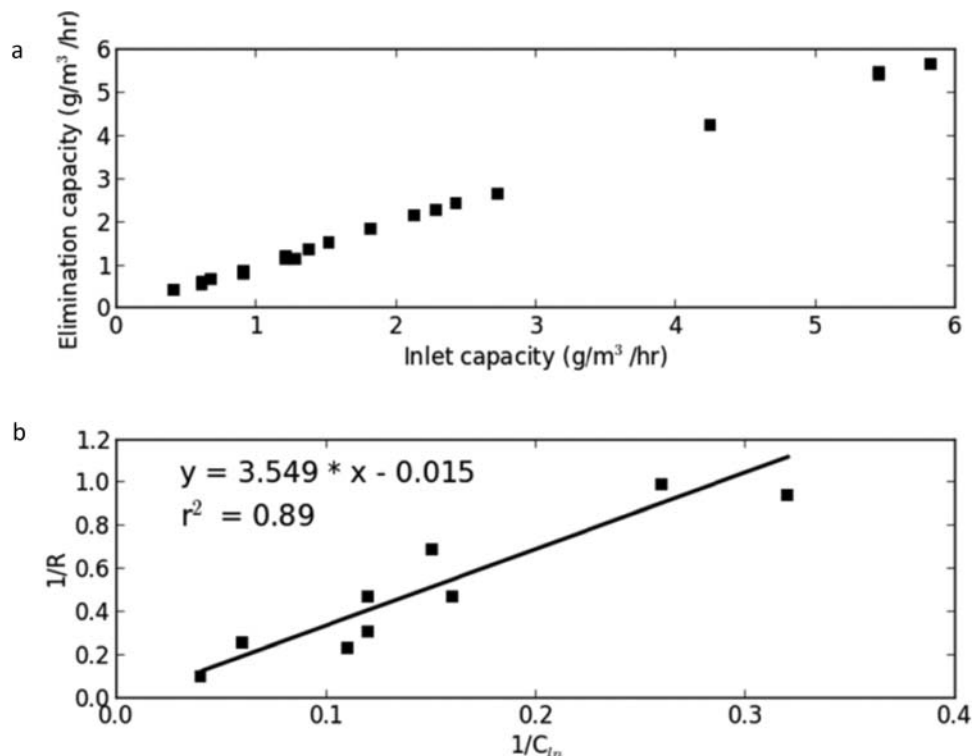


**Fig. 5.** Long-term operational test with the large-scale biofiltration system for the removal of other odorous substances. (a) hydrogen sulfide (H<sub>2</sub>S); (b) t-butyl mercaptan [(CH<sub>3</sub>)<sub>3</sub>CSH]; (c) acetic acid (C<sub>2</sub>H<sub>5</sub>COOH). Circle marker and triangle markers: inlet and outlet concentrations of odorous substances, respectively; square marker: removal efficiencies. Different GRTs: 0 d to 122 d (45 s), 122 d to 227 d (30 s), 227 d to 274 d (20 s).



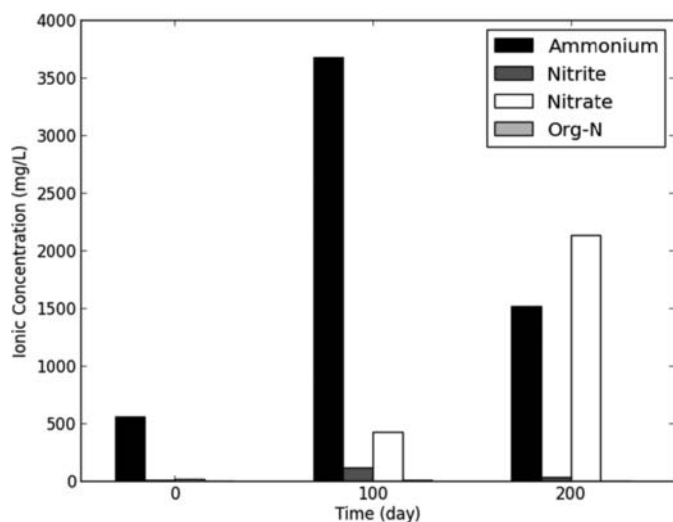
**Fig. 6.** Operational parameters of the large-scale biofiltration system. Monitoring of (a) pH change, (b) pressure drop, and (c) total bacterial count in each bioreactor unit.





**Fig. 7.** Kinetics analysis for NH<sub>3</sub> removal. (a) Relationship between the inlet capacity and elimination capacity for NH<sub>3</sub> removal; (b) regression with 1/R and 1/C<sub>in</sub> for the calculation of  $V_m$  and  $K_s$ .

bacteria in this biofiltration system. Furthermore, these two bioreactor units showed  $\geq 90\%$  removal efficiencies for NH<sub>3</sub> and amines. This finding is consistent with the expected arranged order of bioreactor units with proper inoculates (Fig. 1).



**Fig. 8.** Metabolic products of N-containing compounds. Ammonium (NH<sub>4</sub><sup>+</sup>), nitrite (NO<sub>2</sub><sup>-</sup>), nitrate (NO<sub>3</sub><sup>-</sup>), and Org-N (Kjeldahl nitrogen) were monitored.

Ammonia could be oxidized by the autotrophic nitrifiers *Nitrosomonas* sp. and *Nitrosospira* sp.; however, in the nitrification process, they would lose superiority because of their low growth rates.<sup>[13,14,23]</sup> By contrast, the heterotrophic nitrifier *Arthrobacter* sp. is a promising potential microorganism for biofiltration because its biomass concentration is 10<sup>4</sup> to 10<sup>5</sup> times greater than those of the autotrophs.<sup>[24]</sup>

In the composting facility, the major nitrogen-containing waste gases include TMA, DMA, and MA. Only a few reports focused on the metabolic pathways of TMA. Previous studies showed that TMA is initially oxidized to DMA by *Paracoccus* sp. and that the metabolic intermediates of TMA, DMA, TMA N-oxide are further oxidized to MA. Eventually, MA would be completely oxidized to NH<sub>3</sub> under aerobic conditions.<sup>[8,25]</sup> Other studies reported that a lab-scale biofiltration system immobilized with *Arthrobacter* sp. is more effective (>99%) than other biofiltration systems with similar conditions for NH<sub>3</sub> removal.<sup>[19,26,27]</sup> Hence, the co-immobilization of *Paracoccus* sp. and *Arthrobacter* sp. was considered effective for the complete removal of amines and for reducing NH<sub>3</sub> emissions.<sup>[8]</sup> This finding is consistent with the results of previous studies.

Aside from the high inlet concentrations of NH<sub>3</sub> and amines, some other odorous substances were detected in the gas stream. No outlet concentration of H<sub>2</sub>S was detected, indicating that the immobilized hetero sulfur-oxidizing

bacteria *Pseudomonas* sp. could completely remove H<sub>2</sub>S. Moreover, large-scale biofiltration experiments showed 100% removal efficiencies for t-butyl mercaptan and acetic acid (Figs. 5b and 5c). In treating a gas mixture with various compounds, the performance of a biofiltration system is usually difficult to predict because of the different active microbial species involved in biochemical mechanisms.<sup>[11]</sup> However, interspecific inhibition (production of toxic or acidifying metabolites) and interspecific competition (for available space, substrates, oxygen, and nutrients) within different active microflora were not observed in this study.

The design of biofilters for treating gas mixtures should be optimized. The GRT is believed to be a critical factor directly related to the reaction time for the microbial degradation of pollutants. Theoretically, the GRT should be greater than the required diffusion time of pollution in the reactor. The empty-bed retention time (EBRT) varies with the inflow loading of odorous substances and the type of reactor system used. EBRTs of 30 s to 60 s are common operational conditions for most closed reactors packing with organic media.<sup>[28]</sup> In this study, a large-scale bioreactor with a modular reactor unit can perform the effective odors removal within a GRT of 20 s, thereby reducing the capital cost and equipment size of biofiltration system.

In waste gases treatment, the conventional biofilters are often affected by acidification or alkalization. Treatments of odors from H<sub>2</sub>S, NH<sub>3</sub> and amines may result in the accumulation of acidic end products, such as SO<sub>4</sub><sup>2-</sup>, NO<sub>3</sub><sup>-</sup> or NO<sub>2</sub><sup>-</sup>, which could decline the odor removal efficiency of biofiltration systems.<sup>[11]</sup> However, the monitoring results showed that the pH of the biofiltration system could be controlled in neutral (6–8) for a long operational period, and neither acidification nor alkalization occurred in this system (Fig. 6a). In this study, high concentrations of NH<sub>3</sub> and amines could be dissolved in liquid medium, which may increase the pH of the biofiltration system. Thus, the system provided a conducive environment for the growth of bacteria.

Pressure drop is an extremely important operational parameter related to the water spray volume and biofilm thickness in the system.<sup>[11]</sup> Changes in pressure drop directly affect flow rates (loading) and operational costs of the biofiltration system. In this study, monitoring of pressure drop showed that no significant cloggings occurred in this biofiltration system (25 mmH<sub>2</sub>O m<sup>-1</sup> to 45 mmH<sub>2</sub>O m<sup>-1</sup>) (Fig. 6b). These results are consistent with the reports of previous studies, in which the biofiltration system is suggested to perform a stable operation with a pressure drop of <250 mmH<sub>2</sub>O m<sup>-1</sup>.<sup>[29]</sup> After 270 d of operation, the pressure drop of the modular reactors with GAC was better than those of previously reported reactors packed with ceramics (73 mmH<sub>2</sub>O m<sup>-1</sup>), fuyolite (74 mmH<sub>2</sub>O m<sup>-1</sup>), rock wool (78 mmH<sub>2</sub>O m<sup>-1</sup>), and peat (82 mmH<sub>2</sub>O m<sup>-1</sup>) as carrier under similar operational conditions with a long operation time.<sup>[30]</sup> The results of the biofiltration system developed in this study were significantly lower than the refer-

ence value, suggesting a reduction in the actual operational cost.

During the operational period, monitoring of bacterial count showed that the total microflora could be maintained at >10<sup>7</sup> CFU g<sup>-1</sup>-GAC and that >90% odor removal efficiencies could be achieved for various inlet concentrations. The bacterial counts in the former two reactor units were higher than those in the latter ones, which may be attributed to the abundant inlet concentrations of odorous substances. The high bacterial count of total microflora might indicate an association between stable removal efficiencies and total microflora quantities. The maintenance of a conducive environment (neutral pH) for the growth of microflora is essential in a successful biofiltration operation. Mixed cultures often originating from wastewater treatment plants or similar sources were isolated as inoculates.<sup>[31,32]</sup> The advantage of mixed cultures is the ability to work in a wavering environment and the presence of various microorganisms with a wide degradable range. However, the acclimation period may be long, and the degradation of some compounds may be difficult to accomplish. Inoculation using specific microbial species can reduce the acclimation and enhance overall odor removal efficiencies of biofiltration system.<sup>[33]</sup>

Nitrogen plays an important role in microorganisms, which limits microbial growth under low concentrations.<sup>[8,13,16]</sup> Based on the monitoring of N-containing metabolic products, the data showed that the concentration of NH<sub>4</sub><sup>+</sup> in the former operational period was high (560 mg L<sup>-1</sup>) (Fig. 8). An increase in NO<sub>3</sub><sup>-</sup> concentrations with time were detected (423 mg L<sup>-1</sup> after 100 d and 2,130 mg L<sup>-1</sup> after 200 d). This finding indicates that nitrification is controlling by ammonia-oxidizing bacteria, whereas the liquid phase of NH<sub>3</sub> and amines could be completely oxidized by microbial metabolism.

The main metabolic product of the S-containing compounds was SO<sub>4</sub><sup>2-</sup> (data not shown). The data indicated that S-containing compounds such as H<sub>2</sub>S could be completely oxidized by the inoculated microorganism in this biofiltration system. The pH of the liquid effluent ranged from 6 to 8 over the operational period, which would be a stable buffer and effectively prevent acidification and alkalization in the system.<sup>[19]</sup>

Previous studies on similar odor elimination were compared. In this study, a 97% removal efficiency was achieved for NH<sub>3</sub> with an inlet concentration of 96 ppm, reaching a maximum elimination rate of up to 6.135 g-NH<sub>3</sub> m<sup>-3</sup> h<sup>-1</sup> (147.24 g-NH<sub>3</sub> m<sup>-3</sup> d<sup>-1</sup>). Weckhuysen et al.<sup>[34]</sup> reported a 83% removal efficiency for NH<sub>3</sub> with low concentrations (4 ppm to 6 ppm) at mass loading rates between 6.8 and 27.2 g m<sup>-3</sup> d<sup>-1</sup>. In an inoculated peat biofilter, an elimination capacity of up to 41 g m<sup>-3</sup> d<sup>-1</sup> was achieved for NH<sub>3</sub> with an inlet concentration of 20 ppm.<sup>[35]</sup> In an inoculated perlite biofilter with a 100 d operation, a 95% removal efficiency was achieved for NH<sub>3</sub> with a concentration of 50 ppm and loading rates between 8.6 and 21.5 g

**Table 1.** Comparison of various biofiltration systems for odors elimination.

Target compounds	Working volume	Loading capacity	Mass inflow	Removal efficiency (%)	Reference
Ammonia	116 m <sup>3</sup>	1.09 g m <sup>-3</sup> h <sup>-1</sup>	126.44 g h <sup>-1</sup>	100	Yasuda et al. <sup>[37]</sup>
	11.2 m <sup>3</sup>	5.75 g m <sup>-3</sup> h <sup>-1</sup>	64.40 g h <sup>-1</sup>	97	This study
	8.5 L	6.00 g m <sup>-3</sup> h <sup>-1</sup>	0.05 g h <sup>-1</sup>	100	Kim et al. <sup>[13]</sup>
	7.2 L	6.70 g m <sup>-3</sup> h <sup>-1</sup>	0.05 g h <sup>-1</sup>	96	Pagans et al. <sup>[16]</sup>
Amines	11.2 m <sup>3</sup>	34.88 g m <sup>-3</sup> h <sup>-1</sup>	390.66 g h <sup>-1</sup>	98	This study
	Amines(TMA)	217 L	1.30 g m <sup>-3</sup> h <sup>-1</sup>	0.28 g h <sup>-1</sup>	97
Hydrogen sulfide	7.9 L	10.40 g m <sup>-3</sup> h <sup>-1</sup>	0.08 g h <sup>-1</sup>	100	Ding et al. <sup>[38]</sup>
	28.3 L	14.13 g m <sup>-3</sup> h <sup>-1</sup>	0.40 g h <sup>-1</sup>	90	Chang et al. <sup>[6]</sup>
	11.2 m <sup>3</sup>	0.06 g m <sup>-3</sup> h <sup>-1</sup>	0.67 g h <sup>-1</sup>	100	This study
t-Butyl mercaptan	7.9 m <sup>3</sup>	0.03 mg m <sup>-3</sup> h <sup>-1</sup>	0.24 mg h <sup>-1</sup>	95	Gabriel et al. <sup>[39]</sup>
	11.2 m <sup>3</sup>	4.12 g m <sup>-3</sup> h <sup>-1</sup>	46.14 g h <sup>-1</sup>	100	This study

m<sup>-3</sup> d<sup>-1</sup>.<sup>[36]</sup> In recent years, a few studies have explored large-scale biofiltration systems for odor removal.

Only pilot-scale studies are conducted, generally targeting one for few odorous substances. Compared with the findings of previous studies on NH<sub>3</sub> removal, the study could achieve a removal efficiency of up to 97%. Kim et al.<sup>[13]</sup> and Pagans et al.<sup>[16]</sup> treated a high loading capacity of NH<sub>3</sub> with less working volumes (8.5 and 7.2 L, respectively). However, the mass flow that can be treated in this study (64.40 g h<sup>-1</sup>) is much higher than those in previous reports (0.05 g h<sup>-1</sup>).

Yasuda et al.<sup>[37]</sup> reported a large working volume (116 m<sup>3</sup>) for a high mass flow of NH<sub>3</sub> (126.44 g h<sup>-1</sup>). In this study, a tenth of working volume (11.2 m<sup>3</sup>) could treat for a half of mass flow (64.40 g h<sup>-1</sup>) compared with above report. The loading capacity in this study (5.75 g m<sup>-3</sup> h<sup>-1</sup>) was five times than that in Yasuda's study (1.09 g m<sup>-3</sup> h<sup>-1</sup>). Therefore, the biofiltration system used in this study can treat high mass flow with compact working volume. Some studies<sup>[6,8,38]</sup> treated TMA with a lower working volume (<220 L). However, the mass flow and loading capacity in this study (390.66 g h<sup>-1</sup>) were much higher than those in previous studies (0.08 g h<sup>-1</sup> to 0.40 g h<sup>-1</sup>).

Moreover, the biofiltration system in this study can exhibit complete removal of S-containing compound such as H<sub>2</sub>S and t-butyl mercaptan (0.06 and 4.12 g m<sup>-3</sup> h<sup>-1</sup>, respectively). Previous studies on large-scale bioreactors for similar odor elimination were compared (Table 1). In the present study, we used multiple modular reactor units under various field odor conditions and found that abundant inoculants can effectively remove various odors. The assembly design enhances the stable operation of the biofiltration system.

## Conclusions

The modular biofiltration system developed in this study was scaled-up by 2,800-fold, which increased the flow rates

from 0.005 m<sup>3</sup> min<sup>-1</sup> to 34 m<sup>3</sup> min<sup>-1</sup>. The biofiltration system used in this study can achieve high removal efficiencies for various odorous substances (97% for 96 ppm NH<sub>3</sub>, 98% for 220 ppm amines, and 100% for the other odorous substances with low concentrations). All bioreactor operational parameters remained stable during a long operation period with a low pressure drop (<45 mmH<sub>2</sub>O m<sup>-1</sup>). The inoculated microflora of each reactor unit remained at >10<sup>7</sup> CFU g<sup>-1</sup>-GAC, thus providing a stable removal efficiency. Therefore, we verified comprehensively the high odor removal efficiency of the biofiltration system for N-containing groups, S-containing groups, and SCFAs in composting facilities. Overall, the results of this study clearly demonstrated the effectiveness of large-scale modular reactors in odor elimination.

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## References

- [1] Ge, X.; Wexler, A.S.; Clegg, S.L. Atmospheric amines – Part I. A review. *Atmos. Environ.* **2011**, *45* (3), 524–546.
- [2] Feilberg, A.; Liu, D.; Adamsen, A.P.; Hansen, M.J.; Jonassen, K.E. Odorant emissions from intensive pig production measured by on-line proton-transfer-reaction mass spectrometry. *Environ. Sci. Technol.* **2010**, *44* (15), 5894–5900.
- [3] Ekland, Y.; Sundberg, C.; Smårs, S.; Steger, K.; Sundh, I.; Kirchmann, H.; Jönsson, H. Carbon turnover and ammonia emissions during composting of biowaste at different temperatures. *J. Environ. Qual.* **2007**, *36* (5), 1512–1520.
- [4] Hong, J.H.; Park, K.J. Compost biofiltration of ammonia gas from bin composting. *Bioresour. Technol.* **2005**, *96* (6), 741–745.
- [5] Komilis, D.P.; Ham, R.K. Carbon dioxide and ammonia emissions during composting of mixed paper, yard waste and food waste. *Waste Manag.* **2006**, *26* (1), 62–70.
- [6] Chang, C.T.; Chen, B.Y.; Shiu, I.S.; Jeng, F.T. Biofiltration of trimethylamine-containing waste gas by entrapped mixed microbial cells. *Chemosphere* **2004**, *55* (5), 751–756.

- [7] Chung, Y.C.; Lin, Y.Y.; Tseng, C.P. Removal of high concentration of  $\text{NH}_3$  and coexistent  $\text{H}_2\text{S}$  by biological activated carbon (BAC) biotrickling filter. *Bioresour Technol* **2005**, *96* (16), 1812–1820.
- [8] Ho, K.L.; Chung, Y.C.; Lin, Y.H.; Tseng, C.P. Biofiltration of trimethylamine, dimethylamine, and methylamine by immobilized *Paracoccus* sp. CP2 and *Arthrobacter* sp. CP1. *Chemosphere* **2008**, *72* (2), 250–256.
- [9] Yuwono, A.S.; Lammers, P.S.; Boeker, P. Review of research development on environmental odour in the last decade. *J. Appl. Environ. Biol. Sci.* **2012**, *2* (7), 304–319.
- [10] Burgess, J.E.; Parsons, S.A.; Stuetz, R.M. Developments in odour control and waste gas treatment biotechnology: a review. *Biotechnol Adv* **2001**, *19* (1), 35–63.
- [11] Devanny, J.S.; Deshusses, M.A.; Webster, T.S. *Biofiltration for air pollution control*; Lewis Publishers: New York, 1999.
- [12] Kim, J.H.; Rene, E.R.; Park, H.S. Biological oxidation of hydrogen sulfide under steady and transient state conditions in an immobilized cell biofilter. *Bioresour. Technol.* **2008**, *99* (3), 583–588.
- [13] Kim, J.H.; Rene, E.R.; Park, H.S. Performance of an immobilized cell biofilter for ammonia removal from contaminated air stream. *Chemosphere* **2007**, *68* (2), 274–280.
- [14] Chung, Y.C.; Ho, K.L.; Tseng, C.P. Two-stage biofilter for effective  $\text{H}_2\text{S}$  removal from waste gases containing high concentrations of  $\text{H}_2\text{S}$ . *J Air Waste Manag Assoc* **2007**, *57* (3), 337–347.
- [15] Galera, M.M.; Cho, E.; Kim, Y.; Farnazo, D.; Park, S.J.; Oh, Y.S.; Park, J.K.; Chung, W.J. Two-step pilot-scale biofilter system for the abatement of food waste composting emission. *J. Environ. Sci. Health Pt. A* **2008**, *43* (4), 412–418.
- [16] Pagans, E.; Font, X.; Sanchez, A. Biofiltration for ammonia removal from composting exhaust gases. *Chem. Eng. J.* **2005**, *113*, 105–110.
- [17] Hort, C.; Gracy, S.; Platel, V.; Moynault, L. Evaluation of sewage sludge and yard waste compost as a biofilter media for the removal of ammonia and volatile organic sulfur compounds (VOSCs). *Chem. Eng. J.* **2009**, *152* (1), 44–53.
- [18] Serceu, B.; Nunez, D.; Van Langenhove, H.; Aroca, G.; Verstraete, W. Operational and microbiological aspects of a bioaugmented two-stage biotrickling filter removing hydrogen sulfide and dimethyl sulfide. *Biotechnol. Bioeng.* **2005**, *90* (2), 259–269.
- [19] Chung, Y.C.; Lin, Y.Y.; Tseng, C.P. Operational characteristics of effective removal of  $\text{H}_2\text{S}$  and  $\text{NH}_3$  waste gases by activated carbon biofilter. *J. Air Waste Manag. Asso.* **2004**, *54* (4), 450–458.
- [20] Hirai, M.; Ohtake, M.; Shoda, M. Removal kinetics of hydrogen sulfide, methanethiol and dimethyl sulphide by peat biofilters. *J. Ferment. Bioeng.* **1990**, *70* (5), 334–339.
- [21] Smet, E.; Van Langenhove, H.; Maes, K. Abatement of high concentrated ammonia loaded waste gases in compost biofilters. *Water Air Soil Poll.* **2000**, *119*, 177–190.
- [22] Yang, H.; Minuth, B.; Allen, D.G. Effects of nitrogen and oxygen on biofilter performance. *J Air Waste Manag. Assoc.* **2002**, *52* (3), 279–286.
- [23] Galera, M.M.; Cho, E.; Tuuguu, E.; Park, S.J.; Lee, C.; Chung, W.J. Effects of pollutant concentration ratio on the simultaneous removal of  $\text{NH}_3$ ,  $\text{H}_2\text{S}$  and toluene gases using rock wool-compost biofilter. *J. Hazard. Mater.* **2008**, *152* (2), 624–631.
- [24] Prosser, J.I. Autotrophic nitrification in bacteria. *Adv. Microb. Physiol.* **1989**, *30*, 125–181.
- [25] Kim, S.G.; Bae, H.S.; Oh, H.M.; Lee, S.T. Isolation and characterization of novel halotolerant and/or halophilic denitrifying bacteria with versatile metabolic pathways for the degradation of trimethylamine. *FEMS Microbiol. Lett.* **2003**, *225* (2), 263–269.
- [26] Kapahi, R.; Gross, M. Biofiltration for VOC and ammonia emission control. *Biocycle* **1995**, *36* (2), 87–88.
- [27] Yani, M.; Hirai, M.; Shoda, M. Ammonia gas removal characteristics using biofilter with activated carbon fiber as a carrier. *Environ. Technol.* **1998**, *19* (7), 709–715.
- [28] Easter, C.; Quigley, C.; Burrowes, P.; Witherspoon, J.; Apgar, D. Odor and air emissions control using biotechnology for both collection and wastewater treatment systems. *Chem. Eng. J.* **2005**, *113* (2–3), 93–104.
- [29] Malhautier, L.; Gracian, C.; Roux, J.C.; Fanlo, J.L.; Le Cloirec, P. Biological treatment process of air loaded with an ammonia and hydrogen sulfide mixture. *Chemosphere* **2003**, *50* (1), 145–153.
- [30] Lackey, L.W.; Gamble, J.R.; Holt, M.T. Feasibility testing of biofiltration technology for remediating air contaminated by a boat manufacturing facility. *J. Air Waste Manage.* **1998**, *48* (6), 527–536.
- [31] Ergas, S.J.; Schroeder, E.D.; Chang, D.P.Y.; Morton, R.L. Control of volatile organic compound emissions using a compost biofilter. *Water Environ. Res.* **1995**, *67*, 816–821.
- [32] Kong, S.; Allen, E.R. Control of organo-sulfur emissions using biofiltration: dimethyl sulfide. *Air & Waste Management Association's 90th Annual Meeting & Exhibition. A&WMA: Pittsburgh, PA, 1997.*
- [33] Becker, P.; Koster, D.; Popov, M.N. The biodegradation of olive oil and the treatment of lipid-rich wool scouring wastewater under aerobic thermophilic conditions. *Water Res.* **1999**, *33*, 653–660.
- [34] Weckhuysen, B.; Vriens, L.; Verachtert, H. Biotreatment of ammonia- and butanal-containing waste gases. *Appl. Microbiol. Biotechnol.* **1994**, *42* (1), 147–152.
- [35] Hartikainen, T.; Ruuskanen, J.; Vanhatalo, M.; Martikainen, P.J. Removal of ammonia from air by a peat biofilter. *Environ. Technol.* **1996**, *17*, 17–45.
- [36] Joshi, J.A.; Hogan, J.A.; Cowan, R.M.; Strom, P.F.; Finstein, M.S. Biological removal of gaseous ammonia in biofilters: space travel and earth-based applications. *J. Air Waste Manag. Assoc* **2000**, *50* (9), 1647–1654.
- [37] Yasuda, T.; Kuroda, K.; Fukumoto, Y.; Hanajima, D.; Suzuki, K. Evaluation of full-scale biofilter with rockwool mixture treating ammonia gas from livestock manure composting. *Bioresour. Technol.* **2009**, *100* (4), 1568–1572.
- [38] Ding, Y.; Shi, J.Y.; Wu, W.X.; Yin, J.; Chen, Y.X. Trimethylamine (TMA) biofiltration and transformation in biofilters. *J. Hazard. Mater.* **2007**, *143* (1–2), 341–348.
- [39] Gabriel, D.; Deshusses, M.A. Performance of a full-scale biotrickling filter treating  $\text{H}_2\text{S}$  at a gas contact time of 1.6 to 2.2 seconds. *Environ. Prog.* **2003**, *22* (2), 111–118.