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A pilot field-scale study on biotrickling filter treatment of NH_3-containing odorous gases from organic waste composting plants*

Nian-tao XUE[1], Qun-hui WANG[†‡1], Chuan-fu WU[1], Xiao-hong SUN[2], Wei-min XIE[3]

([1]*Department of Environmental Engineering, School of Civil and Environmental Engineering, University of Science and Technology Beijing, Beijing 100083, China*)

([2]*Beijing Agricultural Biotechnology Centre, Beijing Academy of Agriculture and Forestry Sciences, Beijing 100089, China*)

([3]*Japan TOHIN Company Group Profile, Tokyo 346-0028, Japan*)

[†]E-mail: wangqh59@sina.com

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Abstract: The use of a biotrickling filter was investigated for a pilot field-scale elimination of NH_3 gas and other odorous gases from a composting plant in Tongzhou District, Beijing. The inlet gas flow rate was 3500 m^3/h and NH_3 concentration fluctuated between 2.76–27.84 mg/m^3, while the average outlet concentration was 1.06 mg/m^3 with an average of 94.9% removal. Critical volumetric loading (removal efficiency=100%) was 11.22 g-N/(m^3·h). The odor concentration removal was 86.7%. NH_3 removal efficiency decreased as the free ammonia (FA) in the trickling liquid increased. The pressure drop was maintained at about 50 Pa/m and was never more than 55 Pa/m. During the experiment, there was neither backflushing required nor any indication of clogging. Overall, the biotrickling filter was highly efficient and cost-effective for the simultaneous biodegradation of NH_3 and other odorous gases from composting, suggesting the possibility of treating odorous gases at the industrial level.

Key words: Ammonia, Biotrickling filter, Odor, Organic waste composting, Pilot field-scale study, Trickling liquid
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1 Introduction

Odor control has been a growing challenge because odorous gases are common industrial emissions, and can have significant impacts on environmental health and life quality. Composting facilities have numerous odor sources, including the receiving and handling of materials, active composting, and storage piles, etc. (Park *et al.*, 2001). Being a major odor component, NH_3 gas is one of the key contributors to air pollution, which leads to the deterioration of the atmospheric environment (Ho *et al.*, 2008; Nisola *et al.*, 2009). It is a colorless, toxic, reactive, and corrosive gas, and is emitted in several industrial and agricultural processes, including petrochemical refining and livestock farming, as well as composting facilities (Chung *et al.*, 2005). NH_3 has received much attention, as it can be easily identified from other composting odors, often representing the main nitrogen gas emitted during composting, and because it may be released in large amounts (Pagans *et al.*, 2005).

Biological waste air treatment has been proven to be an efficient, environmentally friendly, and cost-effective alternative for waste gas treatment (Chang and Lu, 2003; Fortuny *et al.*, 2008). At present, biofilters, bioscrubbers, and biotrickling filters are the main biological waste gas treatment technologies. Biofilters are more popular than biotrickling filters in waste gas treatment. They require low maintenance, with appropriate applicability for the large gas flows of complex, yet easily degradable, compounds, producing harmless by-products with greater than 90% removal efficiency (Galera *et al.*,

‡ Corresponding author
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2007). Most of the current studies on the biological treatment of NH_3 concern biofilters with organic supports, while very few studies have yet been conducted with biotrickling filters (Chou and Wang, 2007).

While biotrickling filters are more complex and often more expensive to operate than biofilters, they often exhibit higher performance than biofilters. This is because they allow better control of environmental conditions and because they rely on growing organisms rather than on resting organisms, as is the case for biofilters (Fortin and Deshusses, 1999). The contaminants are induced to diffuse from the gaseous phase through the wet biofilm, and are consequently catabolized aerobically to carbon dioxide, water vapor, and/or inorganic salts (Zhang *et al*., 2009). Biotrickling filters have been successfully used to treat single gases, such as H_2S (Zhang *et al*., 2008) and NH_3 (Sakuma *et al*., 2008) or mixed gases, such as combinations of H_2S and NH_3 (Chung *et al*., 2005; Yu *et al*., 2007), odorous volatile fatty acids (Tsang *et al*., 2008), and mixtures of butyl acetate, *n*-butyl alcohol, and phenyl acetic acid from pharmaceutical factory (Wang *et al*., 2007). Biotrickling filters were better at eliminating the concentrations of odor, NH_3, amines, S-compounds, and volatile organic compounds (VOCs) from a food waste composting plant than chemical scrubbers and biofilters (Mao *et al*., 2006).

In previous reports on biotrickling filters, single compounds or compounds made by several pure substances were used as study objects. Nonetheless, it is very important and useful for the researchers and the industrial practitioners to understand the practical removal principles for gases (Wang *et al*., 2007). These types of conditions are highly unusual at wastewater treatment plants and some other facilities; although, waste gas treatment has been widely reported with biotrickling filters (Mathur and Majumder, 2008; Ramirez *et al*., 2009). The present study is different because its approach addresses the practical treatment of waste gas from a composting plant.

The problems of odor emission in composting plants require immediate attention if composting is to become a viable option for the industrial scale recycling of manure. In this study, a biotrickling filter was chosen because of better degradation by microorganisms and better removal efficiency compared with traditional biofilters. The objectives of the present work were: (1) to determine the NH_3 emissions in the composting of dairy cattle manure; (2) to evaluate the performance of a biotrickling filter packed with zx02-type packing materials (patented product) in terms of variable inlet NH_3 concentrations and loading conditions, trickling flow rate, pressure drop, water quality of the trickling liquid, and microorganisms, etc.; (3) to introduce a cost-effective and environmentally friendly method for the treatment of composting exhaust gas; and (4) to lay a good foundation for the optimal design, operation, and industrial generation of a full-scale biotrickling filter to treat odorous air, since biotrickling filters are newer than biofilters with industrial application being still relatively uncommon (Tian *et al*., 2007). In this research, a pilot field experiment, with a gas flow of 3500 m^3/h, was built in a cattle manure composting plant in Tongzhou District, Beijing, China. It was used to directly remove NH_3-containing odorous gases from a waste gas stream coming from workshops.

2 Materials and method

2.1 Biotrickling filter system

A schematic representation of the biotrickling filter is shown in Fig. 1.

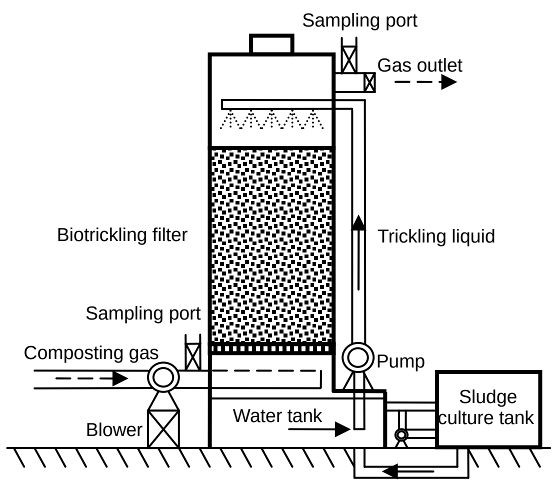


Fig. 1 Schematic of the pilot field-scale biotrickling filter

The pilot field-scale biotrickling filter with a cylinder shape was provided by the Best Environmental Engineering Co. (Shanghai, China). The biotrickling filter system included a blower, a biological treatment unit (biotrickling filter), a pump, and trickling liquid, etc. The bed height and internal diameter were 3.7 and 2.4 m, respectively. The depth of the effective packing materials was 2.0 m. The biotrickling filter consisted of zx02-type packing materials with specific superficial area of 510–680 m^2/m^3, void space of 48%–50%, and diameter of 6 cm. Collecting pipes for composting gas were set at the bottom of the composting bath and congregated at a trunk pipe. Odorous gas was introduced to the bottom of the biotrickling filter through the trunk pipe using a blower at a flow rate of 3500 m^3/h and retention time of 9.3 s. The maximum inlet NH_3 concentration and loading were 28 mg/m^3 and 29.5 g-N/(m^3·h), respectively.

The trickling liquid in water tank was supernatant of a cattle manure solution and 250–300 L/d water was supplied as make-up. It was pumped to the top of the biotrickling filter and dispersed through a water distributor, then trickled to the packing materials. On the last day of composting, the trickling liquid was drained and supernatant of a cattle manure solution was added to the water tank for the next composting period. The pH was manually regulated using hydrated lime to 7.0–8.0, and the alkalinity was simultaneously regulated.

2.2 Composting process

The main composting material was cattle manure from a nearby cattle farm. Auxiliary materials, such as mushroom bran and urea, were added to regulate moisture content to about 50%–55% with a C/N ratio of 25:1–30:1. Aerobic static composting was applied, which produced the NH_3-containing odorous gas.

2.3 Sampling and analytical methods

The inlet and outlet NH_3 concentrations were measured using a water-proof single gas detector (model GAXT-A, 0–100×10^{-6}, BW, Canada). Ammonia-nitrogen (NH_4^++NH_3) in the trickling liquid was determined by the Nessler reagent method (GB 7479-87), nitrite-nitrogen (NO_2^--N) by the *N*-(1-Naphthyl) ethylene diamine dihydrochloride spectrophotometric method (GB 7493-87), nitrate-nitrogen (NO_3^--N) by the ultraviolet spectrophotometric method (HJ/T 346-2007), chemical oxygen demand (COD) by the fast digestion-spectrophotometric method (HJ/T 399-2007), and pH by a pH meter (model pHS-3C, Leici Instrument Plant, Shanghai, China). Pressure drop of the biotrickling filter was monitored daily using a U-shaped tube.

The free ammonia (FA) (NH_3) and free nitrous acid (FNA) (HNO_2) in the trickling liquid were calculated using Eqs. (1) and (2), respectively (Anthonisen *et al*., 1976):

$$C_{\mathrm{FA}}=\frac{17}{14}\frac{C_{\text{Ammonia-nitrogen}}\times 10^{\mathrm{pH}}}{\exp(6334/(273+T))+10^{\mathrm{pH}}}, \quad (1)$$

$$C_{\mathrm{FNA}}=\frac{47}{14}\frac{C_{\text{Nitrite-nitrogen}}}{\exp(-2330/(273+T))\times 10^{\mathrm{pH}}+1}, \quad (2)$$

where C_{FA}, C_{FNA}, $C_{\text{Ammonia-nitrogen}}$, and $C_{\text{Nitrite-nitrogen}}$ are the concentrations of FA, FNA, ammonia-nitrogen, and nitrite-nitrogen, mg/L, respectively.

Odor concentration is defined as an index to quantify the smell by the olfactory organ experimental method, i.e., the dilution multiple when odor sample is continuously diluted with odor-free clean air up to the panel's odor threshold value. This was determined by the odor-triangle odor bag method (GB 14675-93). Two odor-free bags are inflated with odor-free air. Another odor-free bag is inflated with odor gas sample and odor-free air to dilute. If the panels could identify the bag with odor gas, the odor gas sample was diluted again until odor concentration was lower than the odor threshold values of the panels. Odor concentration was calculated in terms of the personal and average odor threshold values of the panels.

2.4 Enrichment culture of the nitrifying bacteria in the biofilm

Several packings were taken out of the biotrickling filter, and the biofilm on the packings was peeled off into a sterilized beaker to be the initial consortia for the enrichment culture of nitrifying bacteria. The enrichment culture medium was composed of the followings (per liter): $NaHCO_3$, 2.00 g; $NaNO_2$, 2.36 g; Na_2CO_3, 0.37 g; NaCl, 0.34 g; KH_2PO_4, 0.05 g; $MgSO_4·7H_2O$, 0.05 g; $FeSO_4·7H_2O$, 0.03 g; $K_2HPO_4·3H_2O$, 0.75 g; and $MnSO_4·4H_2O$, 0.01 g.

After four cycles of enrichment culture, the bacteria were spread-plate cultured in Petri dish.

3 Results and discussion

3.1 Startup

Activated sludge was collected from the dehydrated sludge cake of the Qinghe Municipal Wastewater Treatment Plant in Beijing, China. Tap water was added to dissolve the cake in an activated sludge culture tank and this solution was then pumped to the top of biotrickling filter. At the same time, a low concentration NH_3-containing odor gas from composting was passed through the packing materials to culture microorganisms. The pH of the trickling liquid decreased rapidly from the reactor startup, from 8.0 to 5.5, as a consequence of the accumulation of soluble products from the NH_3 conversion over this period. The inlet and outlet NH_3 concentrations are shown in Fig. 2.

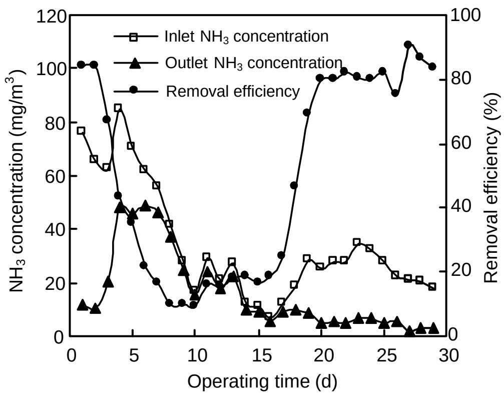


Fig. 2 Inlet and outlet NH_3 concentrations during startup

During the first two days, the inlet concentration was high. However, the removal efficiency was more than 80%. Two days later, NH_3 removal efficiency sharply decreased, and 9.1% efficiency was observed at the outlet of the filter on the 10th day because FA concentrations in the trickling liquid were much higher than that during the first two days. During days 11–16 removal efficiency fluctuated between 15.5%–18.6%, rapidly increased afterward, and the highest efficiency reached 90.2% on the 27th day. Thus, a four-week acclimation period, or lag phase, was required for biological NH_3 removal. This was similar to that of Chou and Wang (2007)'s results indicating that a time of 30 d was required for the development of biofilm for nitrification of the absorbed ammonia from the gas. Microorganisms in the packing materials became increasingly abundant. The removal efficiency was approximately 85% during the late startup stage.

3.2 Effect of inlet NH_3 concentration on NH_3 removal

The effect of inlet NH_3 concentration on outlet concentration is shown in Fig. 3.

The influent NH_3 fluctuated between 2.76 (18.5 °C, 100.6 kPa)–27.84 mg/m^3 (42.6 °C, 100.6 kPa) (i.e., 2.97–32.42 mg/m^3 at standard state), while the removal efficiency fluctuated from 70.2% to 100%. The average removal efficiency was 94.9% with an average effluent NH_3 of 1.06 mg/m^3. This conformed to the Chinese National Standard for occupational exposure limits for hazardous agents in the workplace (GBZ 2.1-2007) ($NH_3 \leq 20$ mg/m^3). The NH_3 concentration at the plant boundary was 0.46 mg/m^3, which also met with Chinese National Standards for emission of odor pollutants (GB 14554-93) ($NH_3 <$ 1.0 mg/m^3).

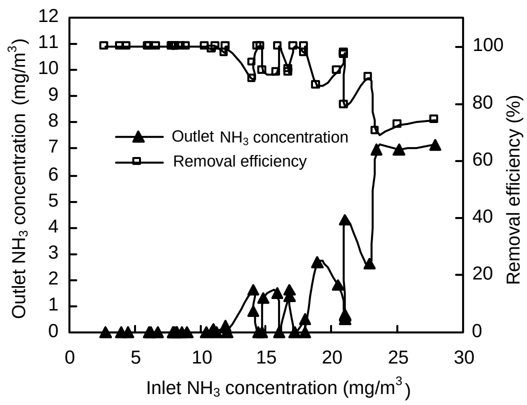


Fig. 3 Effect of inlet NH_3 concentration on removal efficiency

3.3 Impact of volumetric loading rate on NH_3 removal

The loading rate is an important consideration in the system design, and is dependent on the physicochemical properties of NH_3, the inlet concentration, and gas retention time of the system. Fig. 4 demonstrates the relationship between elimination capacity and loading rate.

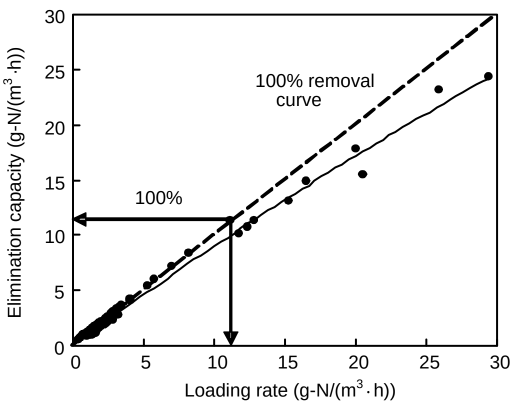


Fig. 4 NH_3 elimination capacity (=(inlet–outlet) concentration×flow/packing material volume) vs. loading rate (=inlet concentration×flow/packing material volume). The dashed diagonal line represents 100% removal

Complete gas removal (i.e., removal efficiency=100%) can be achieved only when the inlet loading is less than the critical loading. If this critical loading is exceeded, the target gas will be detected at the system outlet. Hence, finding an optimum or a maximum inlet loading of NH_3 during the operation is important (Chung *et al*., 2005). As shown in Fig. 4, the relationship curve first rapidly rose and then gradually leveled off to a maximum level. Critical loading (removal efficiency=100%) was determined as 11.22 g-N/(m^3·h). Chung *et al*. (2005) reported that a biological activated carbon biotrickling filter was capable of removal of a high NH_3 concentration and coexistent H_2S with critical loading value of 4.2 g-N/(m^3·h). Chou and Wang (2007) reached a value of 7.37 g-N/(m^3·h) (removal efficiency=98%) in a pilot field-scale biotrickling filter for NH_3 treatment. Sakuma *et al*. (2008) obtained a very high critical loading fluctuating from 60 to 120 g-N/(m^3·h). However, a more complex system with three reactors (biotrickling filter, denitrification, and later a post-treatment) were used to hold back biological inhibition by accumulating nitrate and nitrite, and avoiding generation of contaminated water in their study.

3.4 Influence of liquid trickling rate on NH_3 removal

In biotrickling filters, the trickling liquid aids in nutrient addition, contaminant absorption, metabolite removal, biofilm moistening, and control of biofilm thickness (Chou and Huang, 1997). An appropriate liquid trickling rate must also avoid flushing out a large amount of the biofilm. At the liquid trickling rates of 0.25, 0.5, and 2.21 m^3/(m^3 packing material·h), removal efficiency of NH_3 was tested and the results are demonstrated in Fig. 5.

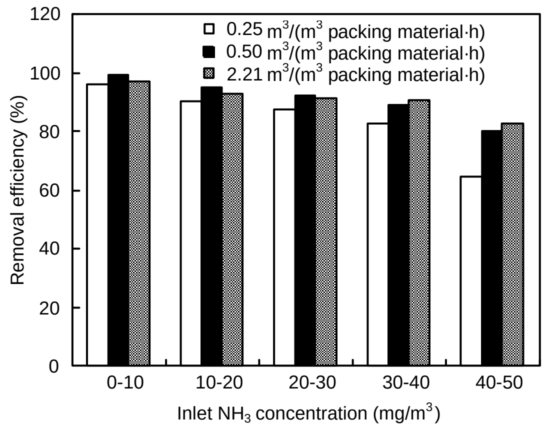


Fig. 5 Relationship between trickling rate and average removal efficiency

At low inlet concentrations (0–30 mg/m^3), a higher trickling rate had little effect on removal efficiency. When the inlet concentration was between 20–30 mg/m^3, removal efficiency was more than 85%, even if trickling rate was only 0.25 m^3/(m^3 packing material·h). At high inlet concentrations (30–50 mg/m^3), a higher removal efficiency could be achieved when trickling rate was increased. NH_3 removal efficiency was more than 80% at a trickling rate of 2.21 m^3/(m^3 packing material·h). It was proportional to the trickling rate at high NH_3 concentrations. The removal efficiency for each trickling rate decreased as the inlet concentration was increased.

NH_3 is readily soluble in water. As a result, the trickling rate increases may benefit pollutant transfer from the gas phase to the liquid phase to achieve a good elimination capacity. However, some gases are not very soluble. If the liquid film on the surface of biofilm is too thick, the mass transfer resistance will increase and eventually gas elimination capacity will decrease. Generally speaking, a higher trickling rate is needed to readily treat soluble gases.

3.5 Water quality of trickling liquid and effect of FA on NH_3 removal

The water quality of the biotrickling filter and the relationship between FA and NH_3 removal efficiency are shown in Fig. 6.

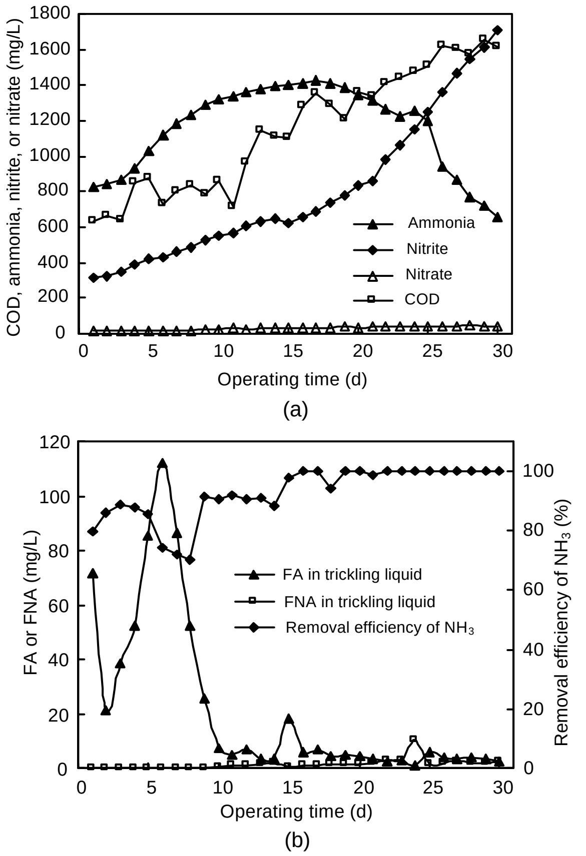


Fig. 6 (a) COD, ammonia, nitrite, and nitrate concentrations in the trickling liquid; (b) Relationship between FA or FNA and removal efficiency of NH_3

During the initial 17 d, ammonia in the trickling liquid was increased because the inlet NH_3 was higher. Then it decreased when the inlet NH_3 was lower and it was biooxidized to nitrite. A small amount of the VOCs during composting was emitted outside the biotrickling filter, while the remainder was dissolved in the trickling liquid (the trickling liquid will be reused through appropriate treatment to avoid emissions. Due to the limited space, this part is omitted). Most dissolved VOCs served as food for microorganisms, while the others resulted in an undulate increase in COD (Fig. 6a).

Of particular importance is the inhibitory effect of FA and FNA on nitritation (i.e., the conversion of ammonia to nitrite) and on nitratation (i.e., the conversion of nitrite to nitrate) (Sakuma *et al.*, 2004), because a specific FA concentration controls the activity of both ammonia-oxidizing bacteria (AOB) and nitrite-oxidizing bacteria (NOB) populations (Villaverde *et al.*, 1997). The FA concentrations for the inhibition of *Nitrosomonas* and *Nitrobacters* are 10–150 and 0.1–1.0 mg/L, respectively, while FNA inhibits nitrifying microorganisms at 0.22–2.80 mg/L (Anthonisen *et al.*, 1976). During the first 9 d, FA was between 21.4–112 mg/L and thereafter the FNA was more than 0.59 mg/L, such that the activity of NOB was inhibited. That is, nitrite oxidation is affected more severely than ammonium oxidation by the simultaneous inhibition (Park and Bae, 2009). Nitrate in the trickling liquid never exceeded 45 mg/L.

The relationship between FA and NH_3 removal efficiency was a novel finding. At the beginning, FA was 71.73 mg/L, and NH_3 removal efficiency was 79.47%. FA fluctuated sharply during the first 10 d; therefore, removal efficiency also fluctuated. Subsequently, FA gradually decreased as NH_3 removal efficiency increased. Removal efficiency was close to 100% when FA was less than 5 mg/L.

As also demonstrated in Fig. 6b, there was a lag time of 2–3 d between FA concentration and its effect on NH_3 removal. FA decreased to 21.4 mg/L on the second day, and a fairly high NH_3 removal of 88.64% was achieved on the third day. FA reached the maximum on the 6th and 15th day, and removal efficiency decreased to a minimum after 2 or 3 d, respectively.

3.6 Pressure drop of the biotrickling filter

Another essential parameter for the biological air pollution control technology is the pressure drop across the bed, because this is related to the development of biomass accumulation in the biotrickling filter (Mathur and Majumder, 2008). The pressure drop across the bed plays an important role in determining the amount of energy needed by the blowers to force the contaminated gas through the bed. It should not be too high since this will result in higher energy requirements (Lu *et al.*, 2001). The structural parameters affecting pressure drop are mainly porosity and equivalent diameter of the packing materials. Biomass accumulation caused by packing material compaction and microorganism proliferation, as well as water film thickening caused by excess trickling liquid, gives rise to changes in the packing material structure, and furthermore, gives rise to changes in pressure drop. Within a month, the pressure drop remained at about 50 Pa/m and was never more than 55 Pa/m. During the experiment, there was neither backflushing nor an indication of clogging.

Clogging is one of the major disadvantages that limits the use of biotrickling filters for the removal of pollutants in waste gases. Excessive formation of biomass leads to progressive bed obstruction and is accompanied by an increase in pressure drop and channeling (Iliuta *et al*., 2005). The optimization of the operational parameters to control clogging still requires further research to enhance industrial applications of biotrickling filters.

The excessive accumulation of biomass can be controlled by physical, chemical, or biological methods. Unfortunately, most of the methods tested at the laboratory scale are not easily implemented at the industrial scale (Sempere *et al*., 2008). In this study, no compaction occurred due to the mechanical intensity of packing materials. Therefore, pressure drop was low and clogging was avoided on a pilot-scale.

3.7 Decrease of odor concentration

To assure the accuracy of test results, the State Environmental Protection Key Laboratory of Odor Pollution Control, located in Tianjin, China, was entrusted to test the extent of odor removal. The outlet odor concentration was 412 with a removal efficiency of 86.7%. According to the Chinese National Emission Standards for odor pollutants (GB 14554-93), the odor concentration should be lower than 2000 when the exhaust pipe is 15 m high and the minimum height of an exhaust pipe is 15 m. In this study, the exhaust pipe was 3.70 m in height and did not meet the standard. The odor concentration must be lower than the plant boundary standard (<60). The Beijing Animal Husbandry Environmental Monitoring Station tested the plant boundary odor concentration of <10, which approached the first-order plant boundary standard in GB 14554-93.

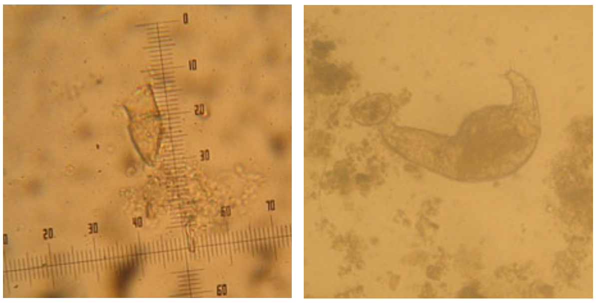
3.8 Microorganisms in biotrickling filter

An experiment was carried out to validate if the enrichment-cultured bacteria could oxidize ammonia or nitrite. Fifty milliliters of a solution containing 2 μg NH_4Cl were prepared. One milliliter bacterial suspension with the contents of ammonia, nitrite, and nitrate of 1000, 500, 50 mg/L, respectively, was added to the flask. Therefore, the ammonia, nitrite, and nitrate in the flask were 3000, 500, and 50 μg, respectively. The flask was put in a constant temperature and humidity incubator for 3 d (Table 1). Nitrite and nitrate were increased by around 4 and 11 times, respectively. This experiment verified that the bacteria were nitrifying bacteria, comprising AOB and NOB.

The initial bacteria consortia were observed in an ordinary optical microscope. Vorticella, a kind of protozoa, and rotifer, a kind of metazoa, were found as shown in Fig. 7.

Table 1 Validation of nitrification reaction by bacteria screened (μg)

	Ammonia	Nitrite	Nitrate
Initial concentration	3000.0	500.0	50.0
Final concentration	394.1	2496.4	617.5



(a) (b)

Fig. 7 Protozoa and metazoa in the biotrickling filter
(a) Vorticella; (b) Rotifer

In the activated sludge process for wastewater treatment, vorticella and rotifers are the bioindicators for a high degree of water self-purification and a good wastewater biological treatment. Bacteria in the biotrickling filter took NH_3, VOCs, and so on from odorous gases during composting as nutrient. The appearance of protozoa and metazoa in the biotrickling filter indicated a long food chain and an enriched microbial morphology, which was conducive to the removal of contaminants in odorous gases during composting. In addition, the existence of protozoa could improve the performance and stability of biotrickling filters, without any loss of pollutant-degradation activity, and a decreased rate of biomass accumulation was obtained (Cox and Deshusses, 1999).

3.9 Cost estimation

The construction and operation costs were 52 240 and 7145 USD, respectively, while the overall treatment costs was 3.53 USD/1000 m^3 gas, which was lower than 8.70 USD/1000 m^3 gas of a pilot/full-scale biotrickling filter (Deshusses and Webster, 2000).

4 Conclusions

1. Inlet NH_3 concentration fluctuated between 2.76–27.84 mg/m^3 (i.e., 2.97–32.42 mg/m^3 at standard state) during cattle manure composting. The average outlet NH_3 concentration of the biotrickling filter was 1.06 mg/m^3 with an average removal efficiency of 94.9%. The critical loading (removal efficiency=100%) was 11.22 g-N/(m^3·h). Odor concentration removal efficiency was 86.7%.

2. The liquid trickling rate had little effect on NH_3 removal efficiency at low inlet concentration (0–30 mg/m^3). At high inlet concentration (30–50 mg/m^3), removal efficiency increased when liquid trickling rate increased.

3. NH_3 removal efficiency decreased as FA in the trickling liquid increased and was close to 100% when FA was less than 5 mg/L. There was a lag time of 2–3 d between the initiation of FA and its effect on NH_3 removal.

4. The pressure drop was maintained at about 50 Pa/m and was never more than 55 Pa/m. There were protozoa and metazoa observed in the biotrickling filter, which indicated a long food chain and an enriched microbial morphology. During the experiment, there was no backflushing required nor any indication of clogging.

In summary, a biotrickling filter was a highly efficient and cost-effective method for simultaneous biodegradation of NH_3 and other odorous gases emitted from composting. The present study lays a good foundation for the optimal design, operation, and generation of a full-scale biotrickling filter to treat odorous waste air at the industrial level.

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