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Biodegradation of Ammonia in Biofiltration Systems: Changes of Metabolic Products and Microbial Communities

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Abstract

In the first stage, the feasibility of using the waste materials from coal power plants (i.e., coal slag) and landscapes (i.e., wood chip and compost) as packing media in various biofiltration systems for ammonia (NH_3) removal was investigated. In the second stage, the optimized biotrickling system packed with coal slag was employed to investigate the effects of inlet concentration on NH_3 treatment performance. A complete NH_3 removal was achieved at concentrations of up to 250 ppm at an empty bed retention time of as low as 8 s, which is shorter than most previously reported biofiltration systems. Results of metabolic product analysis indicated that half of introduced NH_3 was oxidized to nitrate and the rest was converted to ammonium ion at low loadings, while nitrite and ammonium ions predominate at high loadings. A bacterial community shift was observed with regard to the loading rates and pH conditions. In addition, there were no common operating problems, such as clogging and compaction, in the operation for more than 1 year.

Keywords: biofilter, biotrickling filter, ammonia removal, nitrogen mass balance, microbial community

1. Introduction

Ammonia is characterized as a colorless, toxic, reactive, and corrosive gaseous pollutant with a strong and repulsive smell [1]. NH_3 is emitted as a by-product of different industrial processes, such as wastewater treatment, composting, livestock production, and petrochemical refining [2, 3]. Its emission causes significant odor nuisance, health impacts, and environmental problems. It has been reported that exposure to NH_3 above 1 ppm could cause nausea,

headaches, bronchial tract irritation, and burning sensation in the eyes and skin [4, 5]. It is crucial to control NH_3 emission to protect public health and the environment.

The conventional ammonia treatment methods are based on physical and chemical processes, such as adsorption, scrubbing, and chemical oxidation. Unfortunately, these are expensive and produce secondary waste that may require further treatment or disposal, thereby creating additional environmental problems [6, 7]. Biofiltration is an emerging technology for the control of odor and ammonia from contaminated air streams [8–10]. Studies suggest that compost-based biofilters are the most cost-effective for low-concentration ammonia removal in the agricultural industry due to their low operating and maintenance costs. Other supplementary materials (e.g., wood chips) are commonly added to reduce pressure drop and provide a solid-phase buffer [8]. The primary mechanism of biofiltration is the heterogeneous biochemical process controlled by either mass transfer or biochemical reaction or both. Pollutants are transferred from the air to the water layer or to the biofilm attached on the packing media by adsorption or absorption. The sorbed contaminants in the biofilm are degraded by microorganisms into carbon dioxide, water, biomass, and energy [6, 7].

The main functions of the packing media are to provide contact between the gaseous contaminants and the active biofilm and to distribute water and nutrients on the packing surface [8, 11]. Biofilter performance and operating cost are affected by the media characteristics, such as surface area, mechanical properties, buffer capacity, nutrient availability, porosity, and water retention capacity, hence providing an ideal environment for microbial growth [10, 11]. Therefore, the selection of suitable supporting materials and operating mode is an important aspect of a successful biofiltration process.

There are three general classifications of media, namely, natural, inert, and synthetic. Natural materials, including peat, soil, and compost, are generally chosen as biofilter media because they are inexpensive and have a wide diversity of indigenous microorganisms [11, 12]. In addition, several research studies have revealed that natural packing materials provide superior performance in ammonia treatment [8, 13]. Nonetheless, natural-based biofilters are often plagued by common operating problems, such as compaction and decomposition, hence resulting in high pressure drop and air channeling. Common inert materials used in biofiltration include glass beads, perlite, and porous ceramics. Inert materials are difficult to compact. Moreover, they maintain a stable composition during long-term operation. Consequently, they could be used as an alternative to natural media [11]. However, their wide application is stifled due to high material costs and nutrient deficiency.

Different natural and inert packing materials have been successfully applied in biofiltration systems [10, 14, 15]. Likewise, extensive studies have focused on the selection of filter materials and on the optimization of reactor design and operating criteria to obtain efficient ammonia removal in biofiltration systems. However, it is difficult to evaluate the efficiency of various filter materials, because the simultaneous comparison of natural and inert packing media has not been clearly determined under the same conditions. In addition, there are only limited studies on inert packing materials and trickling operations. In our previous study, an attached growth bioreactor packed with coal slag was successfully utilized for domestic wastewater treatment both in lab- and pilot-scale experiments, indicating that coal slag is

a viable supporting material for biofilm attachment and long-term operation [16–18]. The relatively high adsorption capacity of coal slag is also an advantage in the biotrickling filter.

The aim of this study is to determine the feasibility of using recycled wastes as packing media in biofiltration systems for ammonia removal. Potential packing materials were characterized and selected for further investigation, and different operating modes of reactors, namely, biofilter and biotrickling reactor, were also evaluated in terms of ammonia elimination capacities. The removal mechanisms and the inhibitory effects were also investigated through the mass balance analysis.

2. Materials and methods

2.1. Experimental setup

The biofiltration system consisted of four conventional biofilters and one biotrickling reactor (Figure 1a and b), which were packed with selected waste materials (Table 1), and could be operated independently to one another or as replicates. Reactors 1–5 were constructed with cylindrical PVC plastic buckets with a sealable lid. Each reactor has a effective volume of 14.8 l ($D = 305$ mm; effective height = 204 mm). Perforated plastic plates were installed as support for the packing materials and to provide a good gas and liquid distribution. The biotrickling filter (Reactor 5) consisted of a recirculation system (10 l nutrient tank + peristaltic pump (Masterflex, 7518–10) with an anti-clogging sprinkler), a pH controller (model 8156, Orion pH electrode), and a NaOH feeding tank. The recirculation liquid was regularly replaced after each 7-day operation, in order to prevent accumulation of toxic by-products in the reactor. The flow rate of odorous gas stream entering each reactor was controlled by five adjustable panel mounted rotameters (CZ-3246-24, Cole-Parmer) and a mass flow controller. The sampling system monitored the inlet and outlet ammonia concentrations, ambient temperature, and temperature of each reactor and regulated the mass flow controller to provide steady and desirable ammonia concentrations from a pure NH_3 gas cylinder (S.J. Smith Co., USA). Throughout all the experimental runs, the reactors were operated at room temperature of about 25°C. In Reactors 1–3, biofilm development was employed by natural selection without an external inoculum. In Reactor 4, diluted activated sludge solution was completely mixed with wood chips and coal slag as a seed. For Reactor 5 (biotrickling filter), the acclimated microbial broth was added into the fresh mineral medium without $(\text{NH}_4)_2\text{SO}_4$. The solution was sprayed on the top of the filter bed with a flow rate of 100 ml min^{-1} . Thereafter, the synthetic odorous gas with a relatively low NH_3 concentration (10–20 ppm) was introduced into the reactor for the start-up process.

2.2. Waste materials

Six waste materials, including shredded hardwood mulch, wood mulch fine compost, chipped hardwood mulch, mushroom compost, landscape wood chips, and coal slag, were selected to determine the feasibility of using these materials as packing media for ammonia removal. The physicochemical characteristics of the waste materials were analyzed. In the continuous

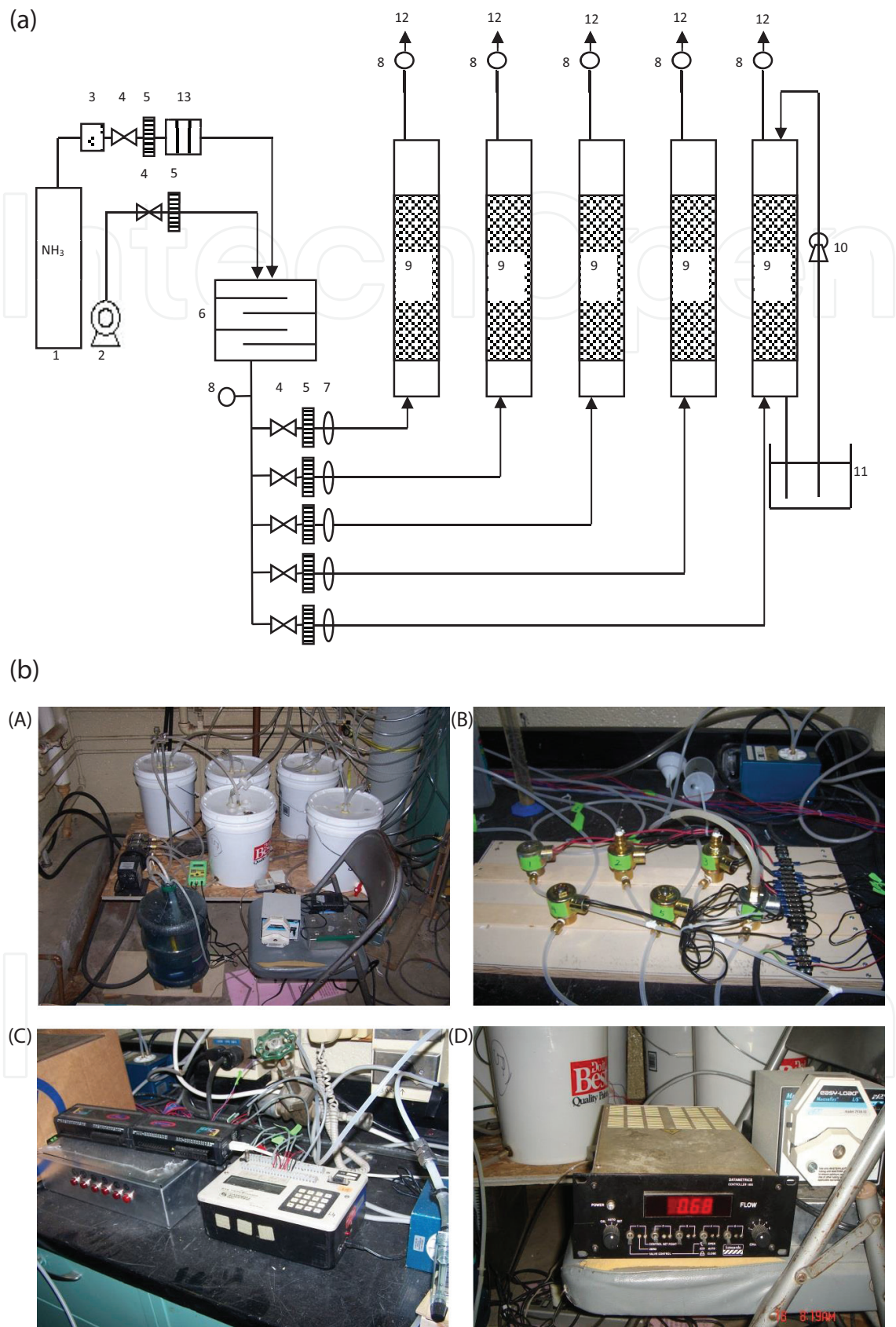


Figure 1. (a) Schematic diagram of the biofiltration system: (1) gas cylinder, (2) compressed air, (3) pressure regulator, (4) valve, (5) flowmeter, (6) gas mixing chamber, (7) air filter, (8) sampling points, (9) filter medium, (10) peristaltic pump, (11) recirculation tank, (12) gas outlet, and (13) mass flow controller. (b) Photos of the biofiltration system: (A) reactors, (B) sampling systems, (C) datalog/control system, and (D) mass flow controller.

	Supporting materials	Operating mode	Inoculation	Feeding solution	Spraying frequency
Reactor 1	Wood chips (100%)	Conventional	No	Water	20 min d ⁻¹
Reactor 2	Wood chips (50%) + compost (50%)	Conventional	No	Water	20 min d ⁻¹
Reactor 3	Wood chips (50%) + coal slag (50%)	Conventional	No	Water	20 min d ⁻¹
Reactor 4	Wood chips (50%) + coal slag (50%)	Conventional	Mixed with sludge	Water	20 min d ⁻¹
Reactor 5	Coal slag (100%)	Trickling	Inoculated with sludge	Mineral nutrient	Continuous spraying

Table 1. Operating conditions used in the preliminary study for ammonia removal.

experiment for ammonia removal, three potential materials were selected from six wastes as packing media based on their desirable properties, namely, landscape wood chips, mushroom compost, and coal slag.

2.3. Microorganisms and inocula

The sludge was obtained from the return activated sludge stream at a local wastewater treatment plant in Urbana, Illinois, USA. The autotrophic ammonia-oxidizing bacteria (AOB) used in the study of the biotrickling reactor were prepared by acclimating activated sludge with selective ammonia medium (**Table 2**) and sufficient aeration (CO₂ source) for a week [19, 20]. Subsequently, the selected microbial strains were transferred to fresh media. After 3–4 transfers (about 3 weeks of acclimation), the inoculum was ready for inoculation into the biotrickling filter. During the acclimation and operation periods, the mineral nutrient that had no supply of nitrogen source for bacterial growth was used for the recirculation liquid.

2.4. Analytical methods

The inlet and outlet ammonia concentrations of each reactor were monitored using a chemiluminescence NH₃ analyzer (Model 17C, Thermo Electron Corporation) with a sampling system composed of six solenoid valves (Parker, C3A) and a data log (Personal Daq/56) and control (Campbell Scientific Inc., 21X) system. Prior to the study, the analyzer was calibrated using

Chemicals	Concentration
(NH ₄) ₂ SO ₄	8 g l ⁻¹
KH ₂ PO ₄	2 g l ⁻¹
K ₂ HPO ₄	2 g l ⁻¹
MgCl ₂ ·6H ₂ O	0.4 g l ⁻¹
CaCl ₂ ·2H ₂ O	0.05 g l ⁻¹

Table 2. Composition of ammonia medium.

standard NH₃ gas with a range of zero to 100 ppm. Ammonia, nitrate, and nitrite in water and solid phases were measured using the colorimetric method (Technicon AA II Continuous-flow AutoAnalyzer) according to the standard methods [21].

2.5. Microbial community analysis

The total DNA from inoculated coal slag and recirculation liquid was extracted by UltraClean Soil DNA kits with inclusion of PCR inhibition removal solution (Mo Bio Laboratories, Solana Beach, CA), followed by the manufacturer’s protocol. The intact DNA was confirmed on a 1% agarose gel electrophoresis. 16S rRNA genes of DNA extracted from the samples were amplified by the PCR using the eubacterial primers 338f and 1492r (Table 3) [22]. The 5’ end of the forward primer was labeled with 6-carboxyfluorescein (FAM) for terminal restriction fragment length polymorphism (T-RFLP) analysis. The PCR reaction mixture contained 1 x PCR buffer, 2 mM MgCl₂, 0.2 mM dNTP, 0.2 μM of each forward and reverse primers, 2 μl of DNA template, and 2.5 U of Ex Taq DNA polymerase (TaKaRa Biomedicals, Otsu, Shiga, Japan) in a 50 ml final volume. The PCR was performed in a thermal cycler (PTC-200 DNA Engine, MJ Research Inc., Reno, NV, USA). Meanwhile, the amplification was done with one denaturing step at 94°C for 5 min, followed by 30 cycles of denaturing at 94°C for 1 min, annealing at 55°C for 1 min, and extension at 72°C for 1.5 min with a final extension step at 72°C for 10 min.

Terminal restriction fragment length polymorphism (T-RFLP) based on the 16S rRNA gene for microbial community analysis was applied to the samples collected from the reactor at various conditions and operation periods. The PCR reactions were carried out in triplicate and were pooled. Following the confirmation of a successful PCR reaction by agarose gel electrophoresis, the PCR products were purified with a PCR purification kit (UltraClean PCR Clean-Up Kit, Mo Bio Laboratories Inc., Carlsbad, CA, USA). The purified fluorescently-labeled PCR products were digested with restriction enzymes: (1) MspI (TaKaRa, China) and (2) RsaI (New England Biolabs, USA) for 3 h at 37°C, followed by an enzyme inactivation step at 65°C for 20 min, and (3) TaqI (TaKaRa, China) for 3 h at 65°C followed by an enzyme inactivation step at room temperature for 20 min. The digested samples were treated by ethanol precipitation to remove excess salt, and the entire 10 μl digested product was analyzed using the ABI Prism 3100 system (Applied Biosystems, USA). The fluorescently-labeled terminal restriction fragments (T-RFs) were separated by capillary electrophoresis to determine the number and size of T-RFs and subsequently compared to the mobility of size standard fragments. Meanwhile, the fragment analysis was conducted using the GeneMapper™ Version 3.7 software (Applied Biosystems, USA). In both cases, the peak areas (related directly to peak fluorescence) of each T-RF relative to the total peak areas were used to determine the relative abundance of individual

Primer	Sequence (5’-3’)	Specificity	References
Eub338f	ACTCCTACGGGAGGCAGC	Bacteria 16S rRNA gene	Amann et al., [34]
1492r	TACCTTGTTACGACTT	Bacteria 16S rRNA gene	Lin and Stahl [35]

Table 3. PCR primers.

microorganisms within the community being examined. The T-RFLP profiles were analyzed using the TAP T-RFLP program at the Ribosomal Database Project II (RDP) website (<http://rdp8.cme.msu.edu/html/TAP-trflp.html#introduction>).

3. Results and discussion

3.1. Physicochemical properties

The common properties of the waste materials, such as density, porosity, moisture content (MC), and C/N ratio, were determined to establish the background information for process design (**Table 4**). In general, the pH of various materials was relatively neutral, with the range of 6.3–7.8; hence, these are suitable conditions for most bacterial activities. Based on their physicochemical properties, three wastes were selected to study the performance of the continuous treatment of ammonia. Landscape wood chip and mushroom compost were selected according to higher yield, higher moisture content, and more favorable pH. Coal slag was selected for comparison purpose and due to high availability of the materials from commonly used power stations.

3.2. Start-up

All reactors started with an inlet loading of about $1.5 \text{ g m}^{-3} \text{ h}^{-1}$, corresponding to the NH_3 concentration of 20 ppm and the EBRT of 30 s. After 14-day operation, the stable removal efficiencies of each reactor were achieved with a range of 17–80%. It was found that the immobilization period of Reactor 5 (biotrickling filter) was completed within 6-day operation. This is comparatively shorter than other traditional biotrickling filters packed with inert or inorganic materials; a 1- to 2-week (up to 4 weeks) operation was required for the completion of start-up [23, 24]. The biodegradation does not occur immediately because some species that are capable of degrading the contaminants may not be initially present [25]. Therefore, the poor performance in Reactors 1, 3, and 4 was observed even at the low loading rates during the start-up.

	Density (kg m^{-3})	Porosity	pH	MC (%)	C/N
Shredded hardwood mulch (1)	0.16 ± 0.01	0.68 ± 0.01	6.58 ± 0.09	7.1 ± 0.7	87.3 ± 0.4
Wood mulch fine compost (2)	0.53 ± 0.02	0.49 ± 0.01	7.33 ± 0.17	18.3 ± 1.4	12.8 ± 0.5
Chipped hardwood mulch (3)	0.23 ± 0.01	0.63 ± 0.02	7.85 ± 0.09	8.3 ± 1.4	33.2 ± 0.3
Mushroom compost (4)	0.21 ± 0.01	0.48 ± 0.02	7.96 ± 0.11	23.7 ± 1.3	20.4 ± 0.3
Landscape wood chip (5)	0.21 ± 0.01	0.65 ± 0.02	7.06 ± 0.06	14.9 ± 0.7	61.3 ± 0.2
Coal slag (6)	1.43 ± 0.04	0.68 ± 0.02	6.32 ± 0.10	3.5 ± 0.8	62.0 ± 0.4

Table 4. Physicochemical properties of natural materials and coal slag (n = 5).

The removal of NH_3 in the biotrickling reactor occurred immediately, primarily because of the adsorption on the coal slag and the absorption in the water layer. From Days 3 to 10, the pH of the recirculation liquid decreased from 7.7 to 5.4, indicating that the biofilm had begun to develop and the NH_3 was mainly removed by microbial activities. Nitrite, nitrate, and hydrogen ions were produced during the nitrification process. As a result, the removal rate subsequently increased to more than 80% by Day 6. This was an indication that the bacteria had been acclimated to NH_3 .

From **Figure 2**, Reactors 1, 3, and 4 had poor treatment performance (17–30% removal). The compost-based biofilter (Reactor 2) had the best treatment performance among the four biofilters. The pH decreased from 9 to 6.64 during the start-up period, indicating that the inherent source of bacteria was important for the biofilter operation mode (**Figure 3**). In Reactors 1, 3, and 4, the pH values fluctuated between 8 and 9.5. Therefore, adsorption and absorption processes were the main removal mechanisms during the 2-week operation, leading to the poor elimination capacities. Furthermore, due to an insufficient amount of indigenous bacteria in the landscape wood chips, it was found that either there was no developed biofilm or it was inactive in these biofilters.

3.3. Continuous operation under stepwise increase of NH_3 concentration

After the 14-day operation, continuous experiments with a stepwise increase of NH_3 concentrations from 20 to 70 ppm at a constant flow rate of 28.3 l min^{-1} (EBRT = 30 s) were carried out for a period of 5 months to evaluate the performance of the reactors packed with different materials. Ammonia in biofilters is partly retained by adsorption onto the packing media and by absorption into the water fraction of the carrier materials [26] and partly achieved through nitrification by the autotrophic ammonia-oxidizing bacteria (AOB) and nitrite-oxidizing bacteria

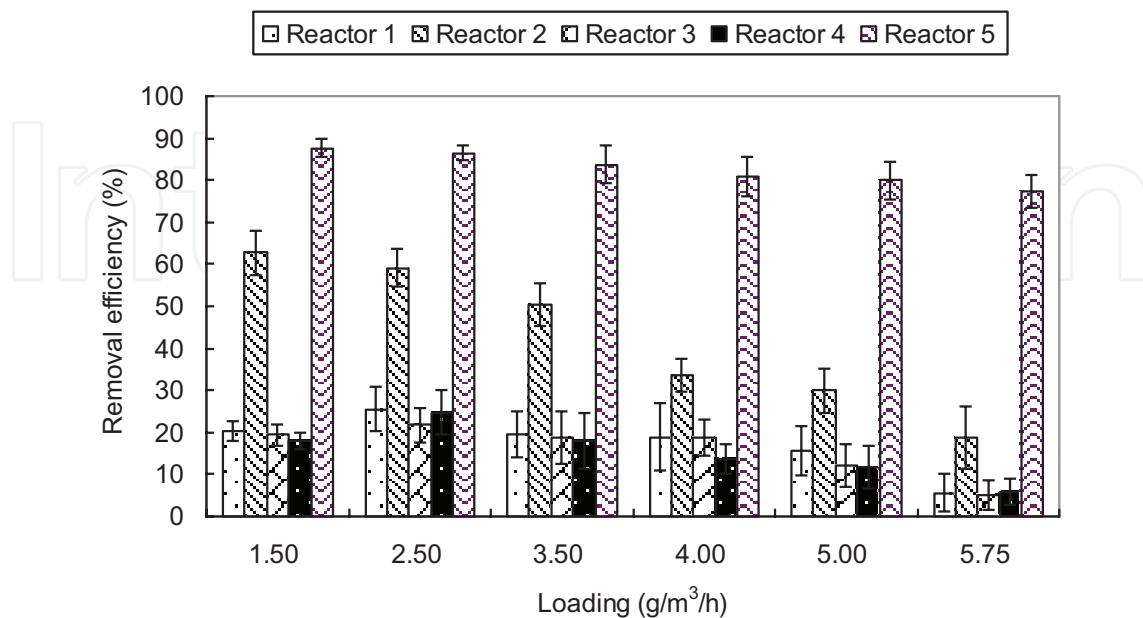


Figure 2. NH_3 removal efficiencies under various loading rates.

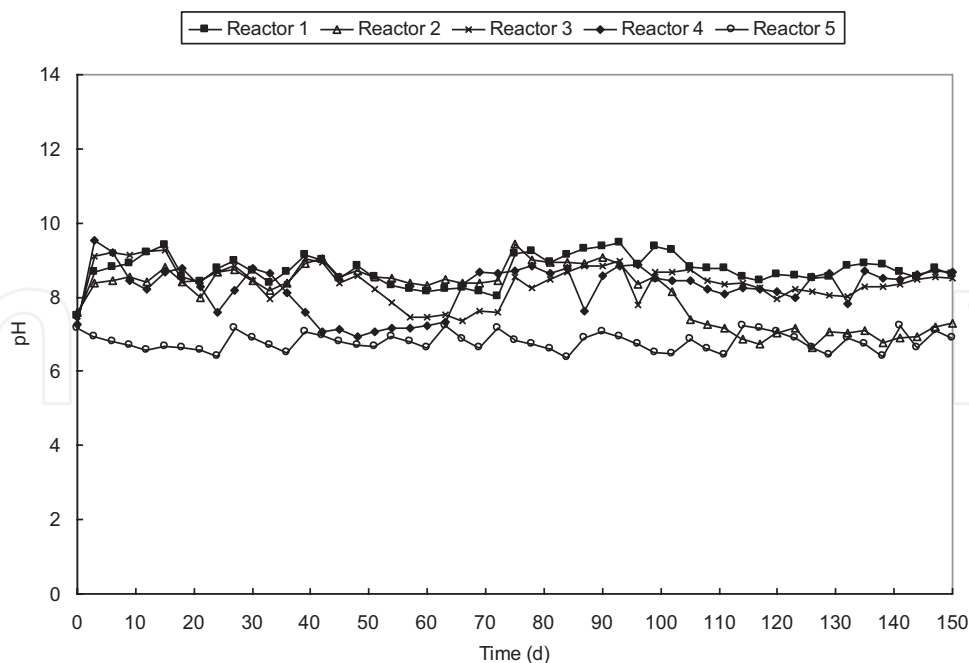


Figure 3. pH profiles during the 150-day continuous treatment.

(NOB) [27]. The results indicated that good bacteria activities with steady and consistent removal (75–88%) were achieved under different loading rates in Reactor 5 only (**Figure 2**). It was found that the pH of Reactor 5 reduced from 7.2 to 6.4 during each replacement of the recirculation liquid, indicating that the ammonia was oxidized by nitrifying bacteria (**Figure 3**). During the change of inlet concentrations, it was observed that various reactors required different adaptation periods to achieve new steady-state conditions. On Day 76, the inlet concentration increased from 40 to 50 ppm, and the removal efficiency significantly decreased from 87 to 70% but was followed by a stable removal efficiency of 80%. The reactor required a longer period to achieve the steady performance at higher NH_3 loading rates.

The mass balance analysis of ammonia oxidation process in Reactor 5 is shown in **Figure 4**. As the loading rate increased up to around $4.5 \text{ g m}^{-3} \text{ h}^{-1}$, the removal efficiency of Reactor 5 decreased from 87 to 77%. This may be attributed to the nitrifying bacteria activities in the reactor that reached the critical loading rate, whereas the relative abundance of NH_4^+ significantly increased from 28 to 45%. This showed that absorption became the dominant mechanism for NH_3 removal under higher loadings. It was also observed that the relative abundance of NO_2^- increased from 0.3 to 18% and that of NO_3^- decreased from 46 to 1%. Both phenomena indicated that the NOB activities were inhibited under high loadings (**Figure 4**). NH_4^+ and NO_2^- , which are the undissociated forms of NH_3 and HNO_2 , are common inhibitors of nitrifying bacteria. The results in this study are consistent with other findings [28, 29]. NH_3 removal in a biotrickling filter is deteriorated due to the accumulation of NH_4^+ and NO_2^- which induces inhibitory effects on both AOB (*Nitrosomonas* sp.) and NOB (*Nitrobacter* sp.).

Among the four biofilters, the highest removal capacity (65%) was achieved in Reactor 2 (50% wood chips + 50% compost) under low loading rate (**Figure 2**). This is also demonstrated

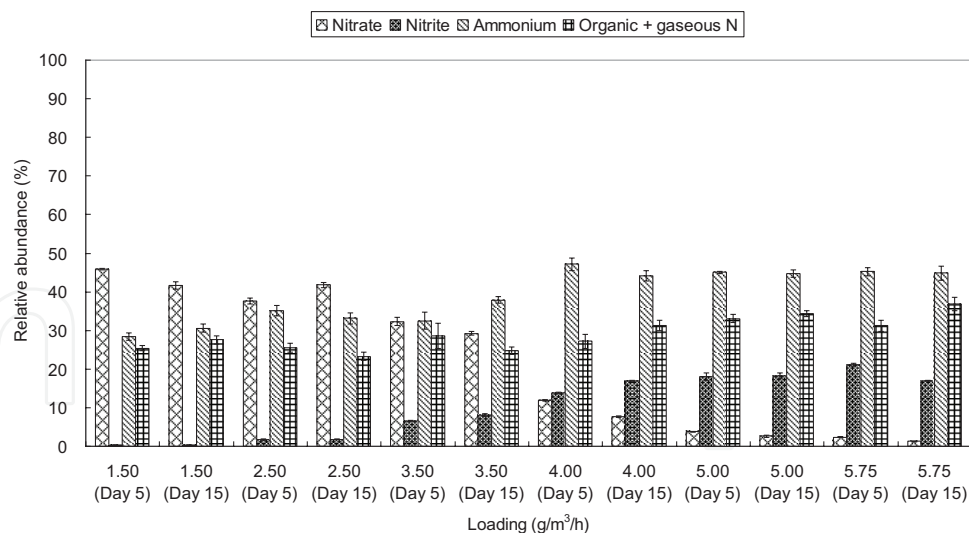


Figure 4. Mass balance analysis of metabolic products in Reactor 5.

by the pH profiles of each biofilter (**Figure 3**). At the low loading rate of $1.5 \text{ g m}^{-3} \text{ h}^{-1}$, the treatment performance of other three biofilters (Reactors 1, 3, and 4) was around 17–20%. As the loading rates increased from 2.5 to $5.75 \text{ g m}^{-3} \text{ h}^{-1}$, the trend of different reactor performances was similar to that at the loading rate of $1.5 \text{ g m}^{-3} \text{ h}^{-1}$. The removal efficiency of Reactor 2 dropped significantly from 60 to 18.8%. For Reactors 1, 3 and 4, the removal efficiencies dropped substantially from around 21 to 5.5%.

With the exception of Reactor 2, adsorption and absorption were the main removal mechanisms for the other three biofilters. The dominant form of nitrogen in Reactors 1, 3, and 4 was NH_4^+ . It was above 85% under all operating conditions, indicating that the absorption of NH_3 into the water layer was the main removal process. Therefore, poor treatment performance was achieved in Reactors 1, 3, and 4. In Reactor 2, the relative abundance of NH_4^+ was around 55% and remained stable under all operating conditions. However, the amount of NO_3^- and NO_2^- in Reactor 2 was 25% of the total nitrogen, which was about 3–10 times in Reactors 1, 3, and 4. Therefore, part of the NH_3 in Reactor 2 was removed by the activities of AOB and NOB through the oxidation of NH_3 into NO_2^- and NO_3^- . The quantity of NO_2^- increased from 5% to over 20% when the loading increased from 1.5 to $5.75 \text{ g m}^{-3} \text{ h}^{-1}$, indicating that the relatively poor NH_3 removal capacity of Reactor 2 may be due to the inhibitory effects of high NH_3 concentration on the NOB community.

In Reactors 1, 3, and 4, the high pH values in the effluent implied the accumulation of ammonium and was mainly due to the overloading of ammonia. High pH and ammonium concentration inhibited the nitrification rates, especially for the activities of NOB. This consequently resulted in the significant increase of nitrite concentration [30].

In this study, Reactor 2 had a significantly higher ammonia removal efficiency compared with the other three biofilters, while Reactor 5 had the highest removal efficiency among the five reactors. It could be explained by the presence of a well-developed biofilm. Since Reactor 5 could provide a more favorable environment for bacterial growth, and the recirculation liquid

could remove the toxic by-products, such as nitrite and nitrate, produced by nitrification. At the same time, the water layer outside the biofilm could provide a buffer capacity to absorb part of the ammonia for further bacterial activities. The length of start-up could also be shortened by the suitable inoculation of acclimated activated sludge bacteria in the reactor. Based on the findings in this experiment, the biotrickling operating mode was selected for detailed study.

3.4. Microbial community analysis

Figure 5a to c illustrates the electropherograms of the samples collected from Reactor 2, Reactor 5, and the recirculation of Reactor 5, after a 40-day operation, respectively. The relative diversity of the bacterial community is related to the number of peaks in the electropherogram. Meanwhile, the degree of abundance constituting each bacterial group is correlated to the intensity and area of the specific peak. Generally, the appearance or disappearance of peaks indicated changes in the bacterial community of the treatment system during the operation.

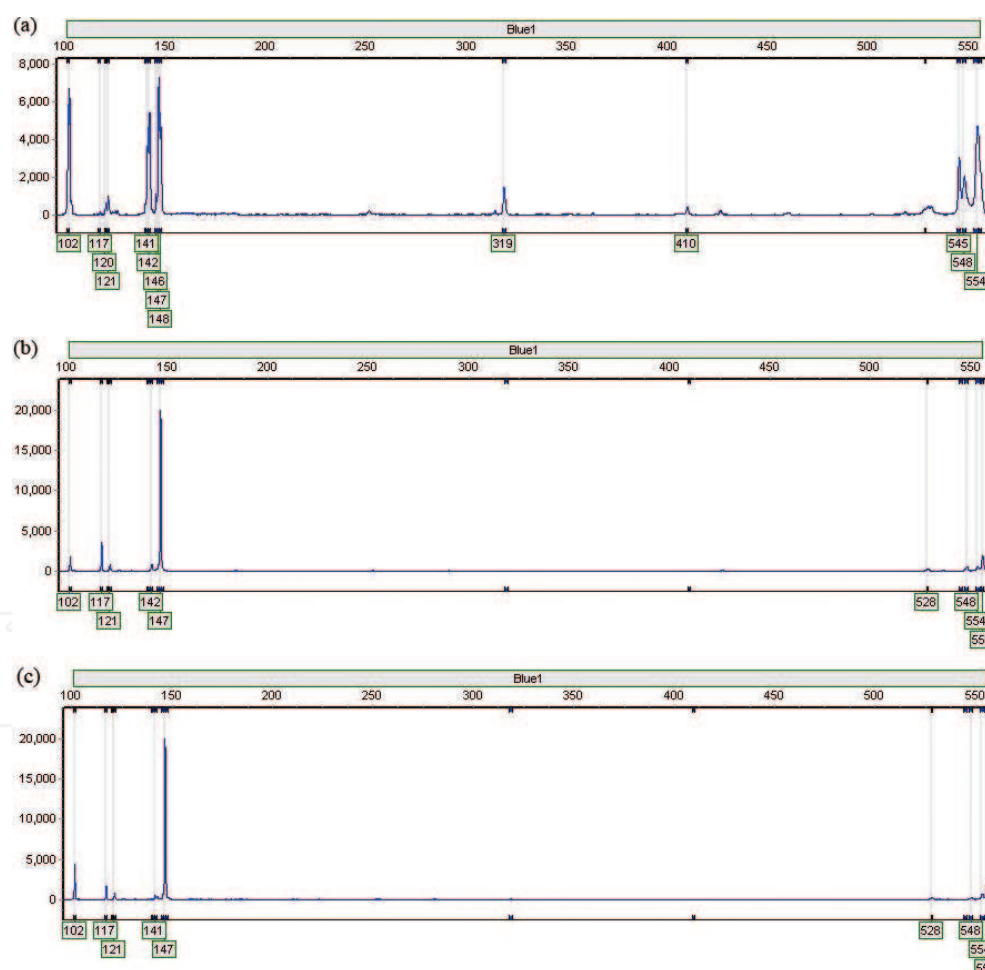


Figure 5. (a) Electropherogram of the sample collected from the compost-based biofilter (Reactor 2) during the preliminary study for the single NH_3 treatment. (b) Electropherogram of the sample collected from the biotrickling filter (Reactor 5) during the preliminary study for the single NH_3 treatment. (c) Electropherogram of the sample collected from the recirculation liquid in the biotrickling filter (Reactor 5) during the preliminary study for the single NH_3 treatment.

The results showed that a higher level of bacterial diversity was found in the compost biofilter (Reactor 2). However, the intensity of its peaks was less than that of the biotrickling reactor (Reactor 5). This indicates that the lower treatment performance of the compost biofilter may be due to the diminutive amount of bacteria developed in the biofilm. Perhaps, another possible reason would be that the compost biofilter was not inoculated with acclimated microorganisms from the activated sludge. Although the compost contains higher bacterial diversity, the indigenous bacteria may not grow well under the operating conditions. This results in a poor treatment performance and a longer start-up period.

Table 5 shows the relative abundance of the bacteria that was determined by the size of terminal restriction length (T-RF). The typical AOB, *Nitrosomonas europaea*, was found in both reactors. In contrast, the results showed a different composition of AOB in Reactors 2 and 5. It may be the reason for the varied treatment performance. A number of common bacteria in the activated sludge were also discovered in the community, including *Pseudomonas* sp. and *Bacillus* sp. Interestingly, a common sulfide-oxidizing bacteria (SOB), *Thiobacillus*, was found in the compost-based biofilter with a relatively high abundance (around 20%). It may be probably due to some H₂S or reduced sulfur in the compost which acted as the nutrients for the growth of SOB.

A similar bacterial population distribution was found in the medium and the recirculation liquid of the biotrickling system. This showed that the analysis of recirculation liquid is satisfactory for the bacterial community analysis of reactor operated in the performance study. This may be useful for the general monitoring because it is difficult to frequently collect the coal slag from the closed treatment system for the microbial study.

TF size (bp)	Possible bacteria	Compost	Coal slag	Recirculation liquid
Nitrifiers		Relative abundance (%)		
147	<i>Nitrosomonas</i> , <i>Nitrospira</i>	21.3	59.3	58.7
554	<i>Nitrosococcus</i> , <i>Nitrosomonas</i>	20.6	3.6	5.0
Other microorganisms		Relative abundance (%)		
105	<i>Pseudomonas</i>	17.1	4.9	12.2
121	<i>Vibrio</i> , <i>Bacillus</i>	1.7	2.5	2.4
319	<i>Actinobacillus</i> , <i>Pseudomonas</i> , <i>Vibrio</i>	4.0	0.0	0.0
410	<i>Actinobacillus</i>	1.0	0.0	0.0
528	<i>Rhodopseudomonas</i>	0.0	1.9	1.6
548	<i>Lactobacillus</i> , <i>Pseudomonas</i> , <i>Thiocapsa</i>	16.4	3.7	2.4
554	<i>Thiobacillus</i>	20.6	3.6	5.0
556	<i>Bacillus</i>	0.0	12.2	12.1

Table 5. Terminal fragments and their corresponding bacteria.

3.5. Performance of ammonia removal

The NH_3 removal efficiency in the biotrickling filter was investigated at the inlet concentrations in the range from 250 to 450 ppm and at a constant flow rate of 5 l min^{-1} for more than 9 months. The corresponding inlet loading rate was from 78.2 to $140.8 \text{ g m}^{-3} \text{ h}^{-1}$ under the operating conditions. Less than 2 days were required to adapt to the new operating condition for NH_3 treatment during each step increase of the mass loadings. Thereafter, the new steady state is also reached, and the pH of recirculation liquid was maintained between 6.5 and 7 in this experiment.

Figure 6 shows the NH_3 removal efficiency as a function of the inlet concentration. The results demonstrated that the bacterial community of AOB in the reactor provided a stable and consistent removal even at the beginning of the experiments. In general, the removal efficiency decreased with the increase of NH_3 concentration. A complete NH_3 removal was achieved during the operation at the inlet concentration of 250 ppm. No NH_3 (below the detection limit = 0.1 ppm) was consistently detected at the outlet. A superior treatment performance, higher than 98%, was achieved at the inlet concentration below 400 ppm. The removal efficiency remained above 99.9% at the concentration of 275 ppm (0.2 ppm detected at the outlet). This slightly decreased to 98.9% at the concentration of 350 ppm (4.6 ppm detected at the outlet). The outlet concentration significantly increased from 4.6 to 75.2 ppm when the NH_3 concentration increased from 350 to 450 ppm (removal efficiency dropped from 98.9 to 83.3%). The complete removal capacity and critical loading rate of the system were $90.5 \text{ g m}^{-3} \text{ h}^{-1}$ and $108.1 \text{ g m}^{-3} \text{ h}^{-1}$, respectively, while the maximum elimination capacity was $118 \text{ g m}^{-3} \text{ h}^{-1}$. **Table 6** shows the comparison of NH_3 removal capacities in various biofiltration systems. The results of this study are relatively high in comparison with other studies [20, 26, 31].

When the loading rate was higher than the critical value, the biofilm would be completely saturated with NH_3 until a loading rate of $140.8 \text{ g m}^{-3} \text{ h}^{-1}$. Moreover, the mass transfer limitation would inhibit the overall removal capacity at the loading rate less than $108.4 \text{ g m}^{-3} \text{ h}^{-1}$.

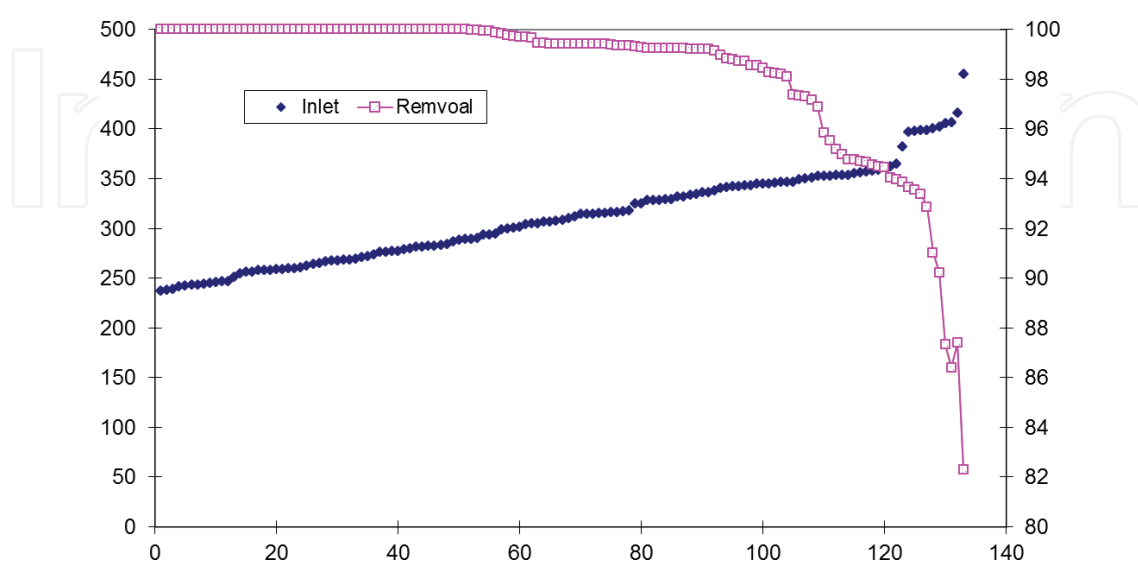


Figure 6. NH_3 removal efficiencies under various concentrations at the flow rate of 5 l min^{-1} .

Packing material	Microorganism	Critical loading (g m ⁻³ h ⁻¹)	Max. elimination capacity (g m ⁻³ h ⁻¹)	References
Biofilter				
Peat	Night soil sludge	30.0	41.7	Kim et al., [31]
Rock wool	Night soil sludge	33.8	50.0	Kim et al., [31]
Fuyolite	Night soil sludge	22.1	28.3	Kim et al., [31]
Ceramics	Night soil sludge	23.8	38.3	Kim et al., [31]
Fuyolite	<i>Vibrio alginolyticus</i>	93.0	114.0	Kim et al., [31]
Granulated sludge	Activated sludge	5.8	20.8	Gracian et al., [36]
Compost + bark + peat	Activated sludge	19.0	22.6	Choi et al., [37]
50% organic + 50% inorganic	Activated sludge	11.8	14.0	Choi et al., [36]
Compost + 20% perlite	Activated sludge	12.0	-	Chen et al., [26]
Sludge + 20% GAC	Activated sludge	10.1	-	Chen et al., [26]
Pall ring	Activated sludge	4.5	5.5	Kim et al., [20]
Biotrickling				
Ceramics	Soil	34.3	49.4	Kanagawa et al., [33]
Coal slag	Activated sludge	108.4	140.8	Present study

Table 6. Comparison of NH₃ removal capacities in various biofiltration systems.

[20, 32]. Authors reported that 4.5 s was required for the biodegradation of NH₃ by nitrifying bacteria [33]. In this study, the system was operated at EBRT of 8 s. This showed that the decrease of removal may be due to the mass transfer limitation from gas to liquid phases or the inhibitory effect of high NH₃ concentration on the oxidation activity of AOB.

At lower loading rates (i.e., 78 to 86 g m⁻³ h⁻¹), the main metabolite in the system was NO₃⁻. This confirmed that the complete removal obtained was mainly contributed by the activities of autotrophic AOB and not by the physical absorption or adsorption. In fact, the organic and gaseous nitrogen was only around 5% under these conditions. The percentages of nitrite and ammonium increased directly proportional to the loading. At the highest loading rate, ammonium became the dominant by-product which accounted for more than 50%, while the removal efficiency was higher than 80%. Although a very high removal efficiency of NH₃ in the system was attained, a complete nitrification in the biofilm was never achieved due to the inhibitory effects of high NH₃ concentration.

4. Conclusions

The results of this study suggest that the biotrickling filter is a viable and effective method for the NH₃ removal. A comparatively short start-up of the system was accomplished within

a 4-day operation. Excellent removal efficiency was achieved below the NH_3 loading rate at $108.4 \text{ g m}^{-3} \text{ h}^{-1}$ (i.e., 350 ppm) in this study, while the maximum elimination capacity was $118 \text{ g m}^{-3} \text{ h}^{-1}$. Product analysis is allowed for the mass balances on nitrogen to identify the biodegradation processes that were active in the system. The main metabolites of NH_3 oxidation were ammonium, nitrate, and nitrite. No clogging and air channeling were observed during a long-term operation. For full-scale applications, odorous gas emitted from different industrial processes always contains high concentration of H_2S and low concentration of NH_3 ; the nitrogen metabolites available in the liquid may enhance the simultaneous treatment of NH_3 and H_2S from the waste gas stream in biotrickling filters. The acidic by-product (SO_4^{2-}) may be neutralized by the unionized ammonia, and a portion of sulfur compounds may act as nutrients for the growth of AOB and other bacteria; thus, the system could be easily maintained at a neutral range for a long-term operation.

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