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Removal of high concentration of NH_3 and coexistent H_2S by biological activated carbon (BAC) biotrickling filter

Ying-Chien Chung [a], Yu-Yen Lin [b], Ching-Ping Tseng [b,*]

[a] Department of Biological Science and Technology, China Institute of Technology Taipei 115, Taiwan, ROC
[b] Department of Biological Science and Technology, National Chiao Tung University, Hsinchu 300, Taiwan, ROC

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Abstract

High efficiency of NH_3 and H_2S removal from waste gases was achieved by the biotrickling filter. Granular activated carbon (GAC), inoculated with *Arthrobacter oxydans* CH8 for NH_3 removal and *Pseudomonas putida* CH11 for H_2S removal, was used as packing material. Under conditions in which 100% H_2S was removed, extensive tests to eliminate high concentrations of NH_3 emission—including removal characteristics, removal efficiency, and removal capacity of the system—were performed. The results of the Bed Depth Service Time (BDST) experiment suggested that physical adsorption of NH_3 gas by GAC was responsible for the first 10 days, after which NH_3 gas was biodegraded by inoculated microorganisms. The dynamic steady state between physical adsorption and biodegradation was about two weeks. After the system achieved equilibrium, the BAC biotrickling filter exhibited high adaptation to shock loading, elevated temperature, and flow rate. Greater than 96% removal efficiency for NH_3 was achieved during the 140-day operating period when inlet H_2S loading was maintained at 6.25 g-S/m^3/h. During the operating period, the pH varied between 6.5 and 8.0 after the physical adsorption stage, and no acidification or alkalinity was observed. The results also demonstrated that NH_3 removal was not affected by the coexistence of H_2S while gas retention time was the key factor in system performance. The retention time of at least 65 s is required to obtain a greater than 95% NH_3 removal efficiency. The critical loading of NH_3 for the system was 4.2 g-N/m^3/h, and the maximal loading was 16.2 g-N/m^3/h. The results of this study could be used as a guide for further design and operation of industrial-scale systems.

Keywords: Biotrickling filter; Activated carbon; Hydrogen sulfide; Ammonia

1. Introduction

H_2S and NH_3 are usually simultaneously emitted from various industries or processes, including food processing, rubber processing, fish processing, animal husbandry, leather manufacturing, compost plants, livestock crematoria, and wastewater treatment plants (Eikum and Storhang, 1986; Ryer-Power, 1991; Yang and Allen, 1994; Chung et al., 1996a; Devinny et al., 1999a). However, the mixed gases containing concentrated NH_3 (80–120 ppm) and diluted H_2S (10–60 ppm) exist in industries like livestock cremation, compost production, and fish processing (Devinny et al., 1999a). The technology of waste gas treatment is often dictated by economic and ecological constraints. Thermal treatments are most economical for high concentration pollutants, but this method becomes increasingly expensive for diluted waste streams because additional fuel is necessary. Activated carbon or zeolite is an efficient adsorbent for treatment of low concentration vapors; however, once the adsorbent has reached capacity, it must be removed and often treated as hazardous waste; therefore it just transfers pollutant from

* Corresponding author. Tel.: +886 3 5712121; fax: +886 3 5729288.
E-mail address: cpts@cc.nctu.edu.tw (C.-P. Tseng).

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a gas phase to a liquid gas, increasing the operating costs of the system as well as the environmental risk (Medina et al., 1995).

Biofilter and biotrickling filter have recently been regarded as the best available control technology (BACT) for odor treatment, if the treatment concentration, physiochemical properties, and treatment cost of the pollutant are considered (Leson and Winer, 1991). Typical waste gas biotreatment consists of two steps: first, the pollutant is removed from the air stream by transfer to liquid film and adsorption on a solid support; then, the pollutant is degraded by microbes living in the liquid phase or the packing material. Hence, the operating condition, support material, and inoculated microbe are important parameters for the removal performance of the biotrickling filter. Gas-phase bioreactors utilize microbial metabolic reactions to remove contaminated air. This biological treatment has proven effective and economical for low concentrations of contaminant in large quantities of air (Leson and Winer, 1991). Two key components of the bioreactor are the support material and inoculated microorganisms. The support material must adsorb the pollutant and provide microbial attachment. Other attributes of a good bioreactor medium are high water-holding capacity, good airflow characteristics, high pH buffer capacity and good mechanical properties (Leson and Winer, 1991). Soils are first used as medium; however, their tendencies to short-circuit and clog limit their effectiveness (Carlson and Leisner, 1966). Although compost has good water retention properties, a large density of microorganisms, and a suitable organic content, it suffers from aging effects that create short-circuiting of the biofilter and further decrease its effectiveness (Langenhove et al., 1992). Using fibrous peat as a packing material has been demonstrated to be preferable to soil or compost, but it is naturally hydrophobic, and moisture of the peat beads is difficult to control (Leson and Winer, 1991). Activated carbon, widely applied in wastewater treatments, has excellent structural properties and good resistance to crushing (Medina et al., 1995). It has substantial water-holding capacity and provides a good surface for microbial attachment. Hence, it is a good candidate to be the biofilter support medium to remove waste gases. In addition, gases with low molecular weight, highly soluble compounds, and simple chemical structures are good targets for removal by biotrickling filter. According to these principles, the best control technology for biotreatment of NH_3 and H_2S gas mixtures will be a BAC biotrickling filter.

Although the biological process removal of H_2S or NH_3 alone has been reported and with high removal efficiencies (Chung et al., 1996a, 1997), only a few studies have dealt with the biological treatment of both NH_3 and H_2S in a single air stream (Chung et al., 2001). Even when some treatment proposals for gas mixtures containing NH_3 are presented, unsatisfactory removal efficiencies or short experimental periods are their drawbacks (Lee and Shoda, 1989; Kapahi and Gross, 1995). Ca-alginate was used as carrier for simultaneous treatment of NH_3 and H_2S in a previous study, but violent pH changes and slight acidification proved to be fatal drawbacks for the system (Chung et al., 2001). In addition, microbial nitrification, the main mechanism in ammonia oxidation, is often inhibited by sulfur compounds, reducing the efficiency of NH_3 biooxidation (Julitte et al., 1993). Hence, high concentration of NH_3 removal characteristics in the coexistence of H_2S gas needs to be studied in detail. In previous studies, *Arthrobacter oxydans* CH8 was effective in removing NH_3 and *Pseudomonas putida* CH11 in eliminating H_2S (Chung et al., 1996b, 1997). Therefore, these two species were chosen for inoculation in the BAC biotrickling filter. In this study, the biotrickling filter on a pilot scale was simultaneously fed high concentrations of NH_3 and low concentrations of H_2S gas mixtures to evaluate the removal capacity of the system for 140 days. Since the removal efficiency of H_2S was maintained at 100%, the detailed removal characteristics and appropriate operation conditions for removal of high NH_3 concentrations in the H_2S coexistence were studied. The results showed that the BAC biotrickling filter was very effective at removing both NH_3 and H_2S.

2. Methods

2.1. Organism cultivation and medium preparation

The original pure-culture strains of heterotrophic ammonia oxidizer, *A. oxydans* CH8, and heterotrophic sulfur oxidizer, *P. putida* CH11, were isolated from swine wastewater (Chung et al., 1996b, 1997). These bacteria were purified by repeatedly transferring the cells to fresh medium. Stock cultures were both enriched in nutrient broth at 30 °C. The nutrient broth contained (in grams per liter) yeast extracts 5, tryptone 10, and dextrose 2. In continuous-treatment experiments, the inflow medium was used and stored in the nutrient tank. The inflow medium contained (in grams per liter) glucose 10, KH_2PO_4 4.08, K_2HPO_4 5.22, NH_4Cl 0.4, $MgCl_2 \cdot 6H_2O$ 0.2, and Fe(III)–citrate 0.01. The final pH of the culture was adjusted to 7 using 2 N NaOH or HCl. The buffer capacity (i.e. the ratio of the increment of strong base or strong acid to the change in pH) in the inflow medium was calculated as 0.033 (M/pH).

2.2. Immobilization procedure

Granular activated carbon with 4.5-mm diameter obtained from Cherng Tay Corporation Ltd. in Taiwan

was used as the packing material for the biotrickling filter. The main characteristics of the material, including bulking density, specific surface area and pH value, were 0.48 g/cm^3, 1250 m^2/g and 9, respectively. *A. oxydans* CH8 or *P. putida* CH11, each grown in 1000 mL nutrient broth, were harvested by centrifugation ($8000 \times g$ for 10 min). The precipitates were drawn and mixed with 7 L nutrient broth in a 10-L PVC tank for enriched growth. About 2.7 kg of the granular activated carbon (GAC) were separately mixed with above solution to develop immobilized cells' biofilm. During the cultivated period, the nutrient broth was replaced every three days until the cell numbers of *A. oxydans* CH8 and *P. putida* CH11 were up to about 10^{10} CFU/g-dry GAC. After 28 days, the cell-laden GAC medium in the tank was transferred into the biotrickling filter.

2.3. Apparatus and gas removal for continuous operation

To investigate the adsorption capacity of GAC to NH_3, three glass columns (12 cm $\phi \times 7$ cm working length) connected in series were packed by GAC without microbes to conduct the Bed Depth Service Time (BDST) experiment. One hundred and ten parts per million NH_3 was continuously supplied to these glass columns at 500 L/h. Outlet NH_3 concentrations in each column were separately measured every 2 h. The desired concentration of NH_3 at the breakthrough was defined as 24 ppm (i.e.: $C_e/C_0 = 0.4$, C_e and C_0 were the desired and inlet NH_3 concentrations, respectively). The experimental results could serve as background values for continuous operation of a BAC biotrickling filter.

A schematic of the pilot-scale BAC biotrickling filter is shown in Fig. 1. Two glass columns (12 cm $\phi \times 40$ cm of working height) connected in series were packed with cell-laden GAC (called BAC) supported by a perforated sieve plate at the bottom of the column to allow the circulating liquid to flow out. The initial packed volume, dry weight of GAC, and number of cells in the biotrickling filter were 9.05 L, 4.34 kg, and 3.8×10^{10} CFU/g-dry GAC, respectively. The column wall contained four sampling ports, 25 cm apart, for sampling or measuring NH_3 and H_2S concentrations during the experiments. The flow meter and valve were used for monitoring and controlling the gas flow through the reactor. The $NH_{3(g)}$ and $H_2S_{(g)}$, supplied from separate gas cylinders, were first diluted with compressed air, which passed through an air filter (pore size 0.2 μm, LIDA 3000-06, made in USA). They then flowed downward through the biotrickling filter at the top. An inflow medium (see medium preparation) stored in the nutrient tank was intermittently re-circulated by a peristaltic pump at 10 L/min for 6 min once every 4 h to maintain the moisture of the biotrickling filter and supply nutrient to the attached cells. The peristaltic pump was connected to a spray nozzle to uniformly spay the medium

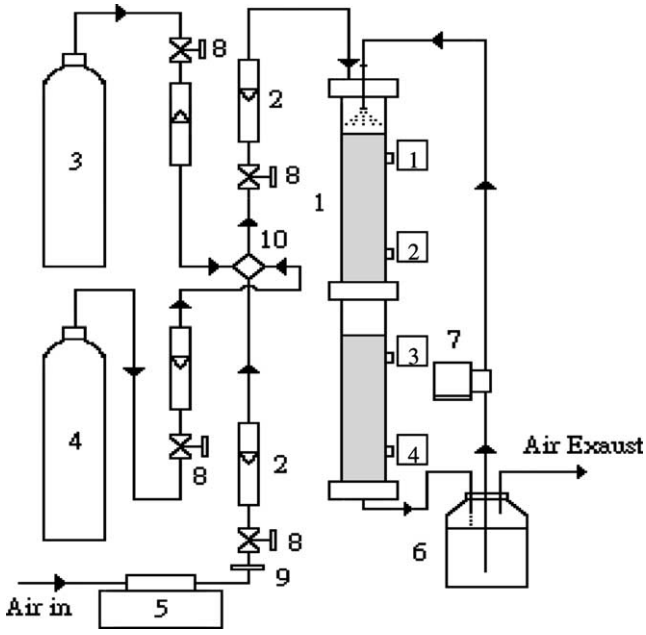

Fig. 1. Schematic of the pilot scale BAC biotrickling filter. 1. glass column; 2. flow meter; 3. NH_3 gas cylinder; 4. H_2S gas cylinder; 5. air compressor; 6. nutrient tank; 7. pump; 8. regulator; 9. air filter; 10. 4-way connector.

on the surface of the filter bed in a direction counter to the influent gas. Ten grams per liter of glucose was added once every two weeks. In the 140-day treatment periods, various NH_3 concentrations (80–120 ppm) under constant inlet H_2S loading (6.25 g-S/m^3/h) were supplied to the BAC biotrickling filter at various flow rates (180–1440 L/h) to evaluate the system performance.

2.4. Experimental design

Response surface methodology (RSM), described first by Box et al. (1978), is an effective experimental design to search the appropriate operation conditions for a multivariable system. The combined effect of inlet concentration and gas flow rate on NH_3 removal was investigated applying response surface methodology. In the present work, a central composite design (CCD) for two variables was used to examine the response pattern. The full-factorial design, consisting of a two-factor-two-level pattern with eleven design points, included nine combinations with three replications of the center points. A multiple regression analysis was performed to obtain the coefficients, and an equation was used predict the response using the STATISTCA program Version 5.0 (StatSoft Inc., Tulsa, OK).

2.5. Analytical methods

Inlet NH_3 gas concentrations in the reactor were periodically measured by gas detector tubes (Model 3LA,

Kitagawa, Japan) in the range of 2.5–200 ppm. Outlet concentrations were continuously measured using a Single Point Monitor (MDA Scientific, USA) in the range of 0.01–0.2 ppm or periodically measured by gas detector tubes (Model 3L, Kitagawa, Japan) in the range of 0.2–20 ppm. In all continuous experiments, NH_3 concentrations were recorded as the variation of NH_3 concentrations within ±5% in 2 h. The total 12 data were recorded, and the average was taken to be the NH_3 outlet concentration. To determine the moisture content in the GAC, about 5 g were withdrawn, weighed, and dried for a 24-h period at 103 ± 0.5 °C. To measure the pH value, 0.5 g of GAC was withdrawn through the appropriate sampling port and mixed with 5 mL of distilled water. The sample was vortexed for 3 min, and the pH value was then determined using a pH meter (Model SP-2200, Suntex, USA). To understand the changes of pH in the leachate, 20 mL of circulating liquid was withdrawn from the nutrient tank at regular intervals to measure the pH values. For microbial analyses, 0.5 g of GAC was separately taken from sampling ports at different depths and mixed with 5 mL of sterile water. The sample was vortexed for 3 min, and the cell numbers were enumerated by traditional plate-counting methods. In this case, the LB (Luria-Bertani) medium for heterotrophic microbial cultivation, the Hagedorn and Holt selective medium for *A. oxydans* spp. (Hagedorn and Holt, 1975), and the cetrimide selective medium for *P. putida* CH11 were used (Mossel and Indacochea, 1971). The inoculated plates grew for 2 days in an incubator at 26 °C.

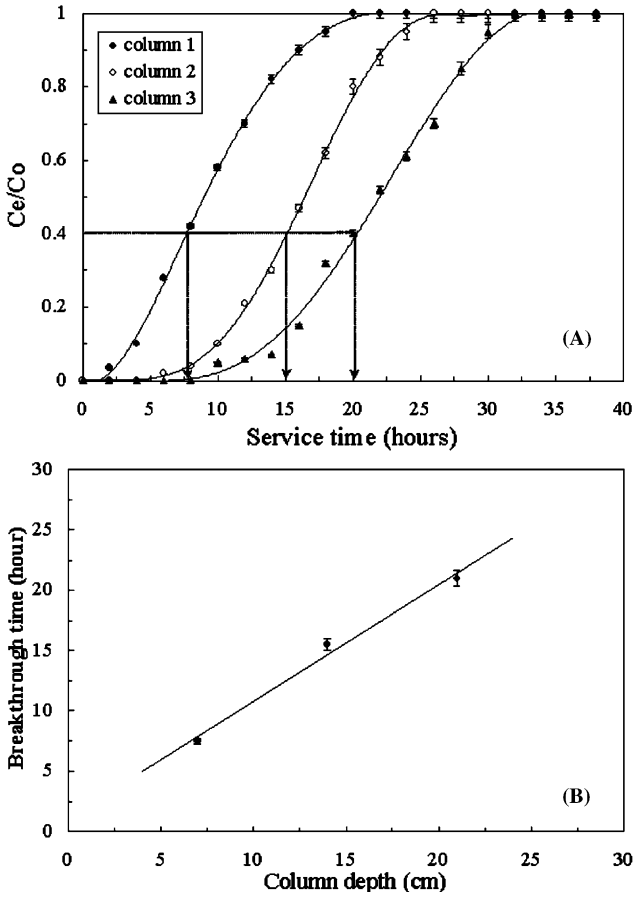

Fig. 2. Adsorption capacity of GAC to NH_3 evaluated by same three GAC columns connected in series. (A) Breakthrough curve by BDST method and (B) linear correlation between packing height and breakthrough time according to the calculation of equation by using breakthrough data.

3. Results and discussion

3.1. NH_3 adsorption by GAC

Before NH_3 biooxidation by microorganisms, the physical adsorption of NH_3 by GAC begins immediately as waste gases are introduced and the results of adsorption are shown in Fig. 2A and B. The adsorption process is described by the equation below (Hutchins, 1973):

$$t = \left(\frac{N_0}{C_0 V}\right)X - \frac{1}{KC_0}\ln\left(\frac{C_0}{C_e} - 1\right)$$

where C_0 = inlet NH_3 concentration (mg/L); C_e = desired NH_3 concentration at breakthrough (mg/L); V = hydraulic loading (cm/h); t = service time (h); X = bed depth (cm); N_0 = adsorptive capacity (mg/L); K = adsorption rate constant (L/mg/h).

The equation constants were obtained from Fig. 2B by regression method, and the values of slope and interception were 0.83 and 3.09, respectively. Hence, the adsorptive capacity (N_0) and the adsorption rate constant (K) were calculated as 280.9 mg/L and 1.71 L/mg/h. The adsorptive capacity expressed as dry weight of GAC was about 0.6 mg-N/g-dry GAC. The NH_3 adsorption capacity of pure GAC can provide a reference value for the removal capacity of cell-laden GAC. In this study, GAC would be physically saturated on the tenth day in the continuous experiment when the actual operating conditions were introduced to the adsorption equation.

3.2. High concentration of NH_3 removal from mixed H_2S in continuous operation

The NH_3 removal efficiency profile in the BAC biotrickling filter during the 140-day operation is indicated in Fig. 3. In this study, NH_3 concentrations (80–120 ppm) and gas flow rates (180–1440 L/h) were varied while inlet H_2S loading was controlled at 6.25 g-S/m^3/h and 100% removal efficiency was maintained. Our goal was to examine the performance of the system for NH_3 removal under these conditions. The theoretical saturation time for NH_3 adsorption was about 10 days by the estimation of the adsorption equation of Fig. 2.

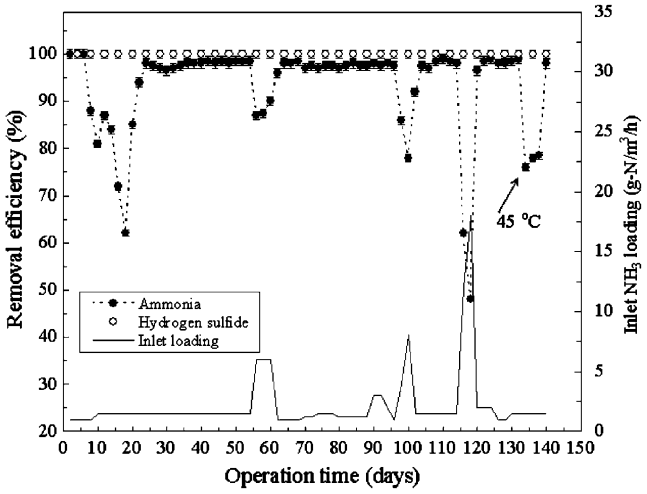

Fig. 3. NH_3 and H_2S removal efficiency as well as inlet NH_3 loading profiles in the BAC biotrickling filter under constant inlet H_2S loading (6.25 g-S/m^3/h) during 140 days operation. Experimental temperature was controlled at 26 °C during the whole experimental period, except days 134–138, operated at 45 °C.

However, the adsorption period was shortened because of H_2S coexistence and competitive adsorption. Hence, physical adsorption of GAC was responsible for 100% of NH_3 removal in the first 7 days as shown in Fig. 3. After 7 days, NH_3 removal efficiency gradually decreased, and 62% efficiency was observed at the outlet of the filter bed on the 15th day. Afterward, removal efficiency gradually increased, and the whole removal efficiency of the BAC biotrickling filter reached 98% on the 23rd day. Thus, a two-week acclimation period or lag phase was required for biological NH_3 removal. To test the adaptability of the BAC biotrickling filter to shock loading, 1.5 g-N/m^3/h of the NH_3 loading was elevated 4-fold (6 g-N/m^3/h) and 12-fold (18 g-N/m^3/h) by days 56 and 118, respectively. The high removal efficiency (>98%) of NH_3 immediately fell to 87% and 48% under those different shock-loading conditions, respectively. However, once the original inlet NH_3 level was restored, high removal efficiency was obtained within 2 or 3 days. Therefore, the system was able to adjust to shock loadings and remained useful even in the face of the severe fluctuations in NH_3 loading that might occur during real industrial operations. Over the four-month study, an average of 96% NH_3 removal was obtained. In comparison to other systems (compost biofilter, peat biofilter, wood bark biofilter, and GAC biofilter) under similar operating conditions, the BAC biotrickling filter inoculated with *A. oxydans* CH8 and *P. putida* CH11 exhibited higher efficiencies. Removal ability of the other systems fell between 85% and 93% (Lee and Shoda, 1989; Kapahi and Gross, 1995; Hartikainen et al., 1996; Yani et al., 1998). Although the removal efficiency of the BAC biotrickling filter was a little lower than that of the immobilized cell biofilter using Ca-alginate as carrier (Chung et al., 1997), it exhibited a high removal efficiency during strongly fluctuating NH_3 inputs in the long-term experiment. These results suggest that the BAC biotrickling filter is more appropriate than the Ca-alginate biofilter for waste gas treatment in the field. Fast physical adsorption and effective biooxidation resulted in successful NH_3 removal by the BAC biotrickling filter. Simultaneous bioregeneration of the carbon was an additional economic advantage of the system.

From the profile of the heterotrophic bacterial numbers, we found that initially inoculated cells were about 3.8×10^{10} CFU/g-dry GAC. The cell numbers were then dropped to 3×10^9 CFU/g-dry GAC on the third day because of scrubbing by the circulating solution (data not shown). During the 140-day experiment, except for the first several days, the variation in the cell number ranged between 10^9 and 10^{10} CFU/g-dry GAC in the samples obtained from different sampling ports. Besides, the profile of the cell numbers did not show gradient change along column length, and the higher cell numbers occurred in the middle zone of the biotrickling filter (data not shown). Because NH_3 removal by heterotrophic bacteria was regarded as a detoxification process, high NH_3 concentrations would inhibit microbial activity in the inlet zone of the biotrickling filter (Martin et al., 1996). The data of Table 1 shows the cell numbers and distribution ratios of the inoculated cells (*A. oxydans* and *P. putida*) and other heterotrophic bacteria. The results indicated that ammonia oxidizer *A. oxydans* was the dominant bacteria, accounting for 87.9%, 93.1% and 94.2% of the total community on days 45, 120, and 138. The sulfur oxidizer *P. putida* accounted for 12.1%, 6.9%, and 5.8%, respectively, on the same days. Although the distribution ratios of sulfur oxidizer decreased slightly, complete H_2S removal was still

Table 1
Cell numbers and distribution ratios of inoculated cells and other heterotrophic bacteria in the middle zone of BAC biotrickling filter

Strain*	Day			
	0	45	120	138
A. oxydans	2.5×10^{10} (65.8%)	3.8×10^9 (87.9%)	8.4×10^9 (93.1%)	7.2×10^9 (94.2%)
P. putida	1.3×10^{10} (34.2%)	5.2×10^8 (12.1%)	6.2×10^8 (6.9%)	4.4×10^8 (5.8%)
Other	0 (0%**)	$4.8 \times 10^{5**}$	$3.7 \times 10^{6**}$	$2.5 \times 10^{6**}$

* Indicates the unit as CFU/g-dry GAC.
** Indicated the distribution ratio lower than 0.05%.

achieved during the operational period. The results also demonstrated that the inoculated cells of *A. oxydans* maintained high distribution ratios and cell numbers in the bacterial community during shock loading at day 120 as well as during temperature increases on day 138. This would be another advantage of the BAC biotrickling filter with further application in the field.

3.3. Effect of temperature change on NH_3 removal

Temperature is an important factor affecting the physical adsorption of NH_3 by GAC and biooxidation by microorganisms. To estimate the system response to temperature variation, NH_3 and H_2S removal efficiencies with temperature change are shown in Fig. 4. During the whole experimental period, environmental temperature was controlled at 26 ± 2 °C except on days 134–138. After the 134-day operation, the temperature of system was elevated to 45 °C, and a significant decrease of 23% (99% reduced to 76%) in NH_3 removal efficiency was observed. Since the optimum temperature for *A. oxydans* CH8 growth was 26 °C (Chung et al., 1997), the higher temperature could have resulted in less efficiency. To further understand the system in response to temperature change, NH_3 and H_2S concentrations were respectively controlled at 100 ppm and 30 ppm at a 720 L/h flow rate. The experimental temperature was conducted at 15 °C, 45 °C, and 26 °C with results shown in Fig. 4. When temperature rose from 15 °C to 45 °C in 4 days, the removal efficiency decreased from 94% to 75%. However, 94% removal of inlet NH_3 was achieved within one day after the environmental temperature was returned to 26 °C. In the meantime, H_2S removal maintained at 100% throughout different operational temperatures. Also, the data indicated that temperature change resulted in no decrease of bacterial numbers (Table 1). Therefore, high temperature may cause a low physical adsorption of NH_3 by GAC and result in a decrease of removal efficiency.

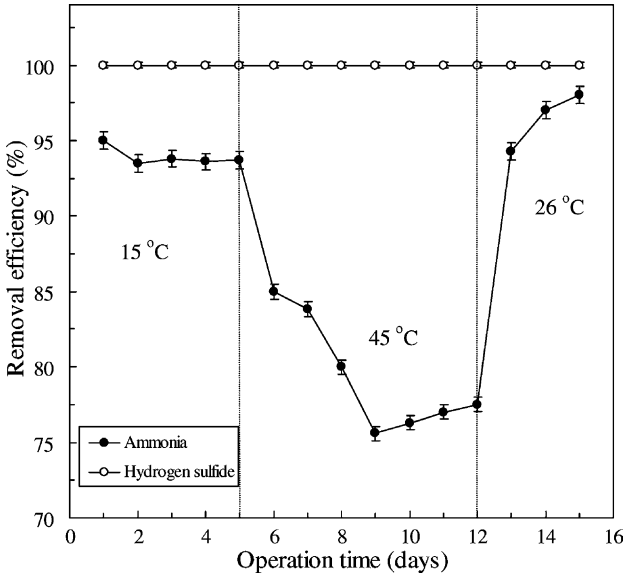

Fig. 4. Effect of temperature change on NH_3 and H_2S removal efficiency. NH_3 concentration was controlled at 100 ppm. H_2S concentrations were in the range of 10–30 ppm, and 100% H_2S removal efficiency was achieved.

3.4. pH and moisture change in the BAC biotrickling filter

The phenomenon of acidification or alkalinity has often been an obstacle to traditional biofilter operation because of the accumulation of inlet gas or oxidized product (Devinny et al., 1999b). Hence, pH monitoring or operation in the filter and leachate was very important for operational stability. During the 140-day treatment, the pH values in the filter were measured once every 4 days, and those in the leachate were measured daily, and these results are shown in Fig. 5A and B. The pH values in the leachate increased gradually in the first 10 days and peaked at 9. Since physical adsorption was responsible for the first 10 days, NH_3 gas would accumulate in the GAC beads and become flushed into the stored tank by circulating solution, increasing the

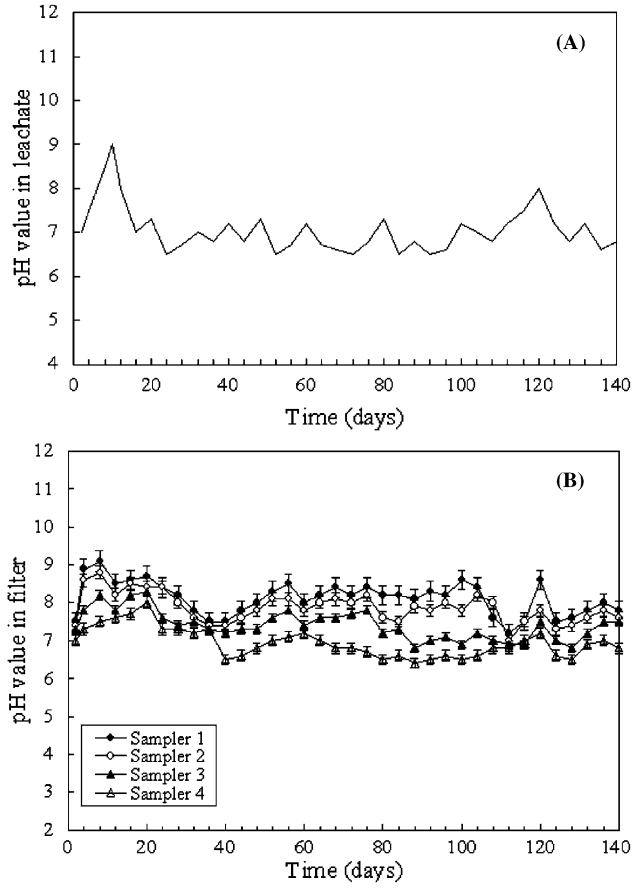

Fig. 5. The pH value profile of operation with BAC biotrickling filter for 140 days. (A) pH value in the leachate and (B) pH value in different sampling ports of the filter.

pH (Fig. 5A). Biodegradation activity was progressively more responsible for NH_3 removal after the physical adsorption period. Ammonia oxidizer *A. oxydans* CH8 converted partial NH_3 to NO_2^- (Chung et al., 1997), and pH value edged down to the neutral range. During the 140-day treatment, the pH of BAC biotrickling filter was not chemically manipulated and pH in the leachate varied between 6.5 and 8.0 except for the initial operating stage. The pH in the biotrickling filter held steady between 6.5 and 9.1, and the profile of the pH showed a gradient change in the axial direction of the filter bed. High pH occurred at the filter inlet because of more basic ammonia accumulation, while low pH was measured at the filter outlet because of more acid product accumulation (e.g. NO_2^-) due to gravity. An acidification or alkalinity phenomenon did not occur in the system due to the effective nitrification or the high alkalinity of GAC itself (Medina et al., 1995).

Gas mass-transfer rate and microbial activity often depended on moisture content in or around the support material (Devinny et al., 1999c). On the other hand, a lower or higher moisture content could lead to some serious problems such as decreased microbial activity and gas diffusion or increased anaerobic area or pressure drop (Ottengraf and Van Den Oever, 1983; Langenhove et al., 1986). Hence, appropriate moisture management in GAC was required to efficiently remove waste gas. In this study, the profile of the moisture content in the system (biotrickling filter) showed a gradient change (from high to low) in the axial direction of the filter bed (Fig. 6). The phenomenon might have resulted from both the force of gravity and the gas flow direction (downward). Because a moisture content above 40% was suggested for the biofilter to maintain biological activity (Sabo, 1993), the average 38.5% moisture content observed in the experiment suggested that the GAC beads had a stable water-holding capacity. Thus, compared with other systems, this BAC biotrickling filter demanded a lower moisture content for microbial growth and might be energy saving.

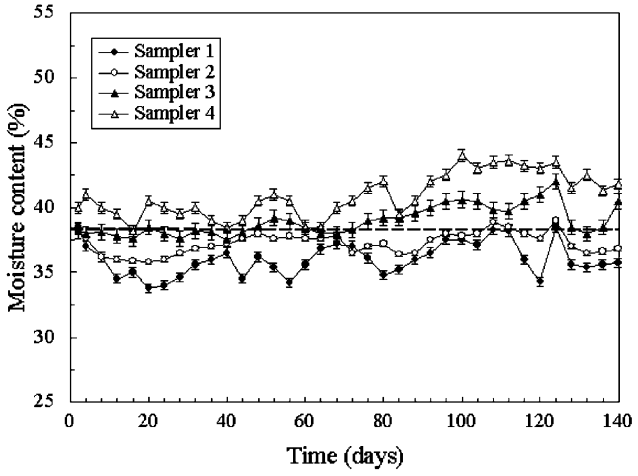

Fig. 6. Moisture content in different sampling ports of the BAC biotrickling filter. Dashes (---) indicates the average value (38.5%) of moisture content.

3.5. Effect of H_2S concentration change on NH_3 removal

A previous study showed that high concentrations of H_2S in the Ca-alginate biofilter would affect NH_3 removal (Chung et al., 2001). To understand the effect of H_2S coexistence on NH_3 removal from mixed waste gases in the BAC biotrickling filter, varied H_2S concentrations (10–120 ppm) were examined at a 360 L/h flow rate when NH_3 inlet was controlled at 100 ppm. Apparently, NH_3 removal was not influenced by H_2S concentrations in this operating system (data not shown). NH_3 removal efficiency exceeded 96% under all operating conditions, and the deviation was within 0.5%. In the previous study, the reason NH_3 removal efficiency declined in the Ca-alginate biofilter system was acid H_2S coexistence (Chung et al., 2001). Therefore, these results suggest that using activated carbon as a packing filter can facilitate the pH stabilization of the system and achieve high NH_3 removal efficiency even with H_2S coexistence.

3.6. Effect of gas retention time on NH_3 removal

A longer retention time sometimes results from the fact that the attached microorganisms can take a longer time to decompose pollutants in the filter. Hence, gas retention time is a key factor in pollutant removal (Chung et al., 1996b). When waste gas containing 80 ppm NH_3 was fed into the bioreactor, removal efficiency decreased with decrease in gas retention time (data not shown). A retention time of 65 s was a critical condition in this study. When retention time was longer, NH_3 removal efficiency was higher than 97%. Removal efficiency dramatically decreased if retention time was less than 65 s. For example, a large difference (22%) was observed when retention time was shortened to 20 s. Therefore, NH_3 removal in the system was dependent on gas retention time. According to Michalis–Menten theory, if enough gas retention time were provided, the biological reaction rate would be the rate-determining step. Conversely, if enough gas retention time were not achieved, the mass-transfer rate would be the rate-determining step. Hence, gas retention time should be maintained beyond 65 s for NH_3 removal when the BAC biotrickling filter system is used to remove H_2S and NH_3 gas mixtures.

3.7. Design criteria for scale-up BAC biotrickling filter

Complete gas removal (i.e. 100% removal efficiency) can be achieved only when the inlet loading is less than critical loading. If this critical loading is exceeded, target

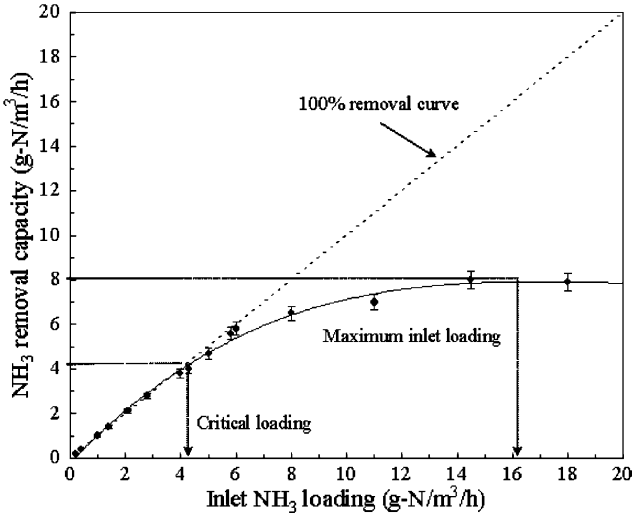

Fig. 7. Relationship between inlet loading and removal capacity of NH_3 gas.

gas will be detected at the system outlet. Hence, finding an optimum or a maximum inlet loading of ammonia during the operation is important. The inlet loading (g-N/m^3/h) is defined as the amount of inlet gas per unit of time and volume of the support. As shown in Fig. 7, the relationship curve first rapidly rose and then gradually leveled off to a maximum level. The critical loading ($R\% = 100\%$) was determined as 4.2 g-N/m^3/h. The extrapolated correlation line suggested the maximum inlet loading was 16.2 g-N/m^3/h (0.81 g-N/kg/day). This value was much higher than those of peat support treatment systems with values of 0.17, 0.18 and 0.38 g-N/kg/day, respectively (Togashi et al., 1986; Hartikainen et al., 1996; Martin et al., 1996). According to Michalis–Menten theory, when low inlet loading is introduced, removal capacity should have a linear relationship with inlet loading (i.e. first kinetic equation). In contrast, when high inlet loading is introduced, removal capacity should be independent of it (i.e. zero kinetic equation) and should be a constant value (i.e. maximum removal capacity). As for middle inlet loading, removal capacity and inlet loading should be in a fractal relationship. In this study, the removal behavior fit in with theory mode. Therefore, high efficiency and acceptable NH_3 removal can be accomplished by adjusting the flow rate and the inlet concentration based on the correlation presented in Fig. 7.

3.8. Comprehensive evaluation of concentration and flow rate for NH_3 removal

The most important operating factors affecting NH_3 removal are inlet NH_3 concentrations and gas flow rates. To evaluate the combined effect of these factors, experiments were performed at different inlet concentrations with various flow rates (Table 2). The suitable combinations for NH_3 emission limits were determined. Accord-

Table 2
NH_3 removal efficiency in response to varying concentrations and flow rates

Flow rate (L/h)	NH_3 (ppm)		
	60 (%)	80 (%)	100 (%)
400	95.9	95.6	95.1
600	93.6	92.7	91.9
800	91.5	90.0	88.2

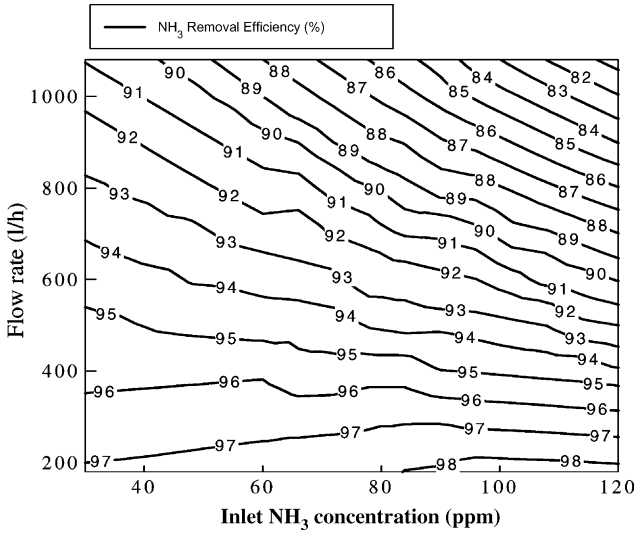

Fig. 8. The Contour plot of NH_3 removal efficiency in response to varying concentrations and flow rates.

ing to the results of experiments, the following equation gives the relative NH_3 removal efficiency, y as a function of inlet concentration, and flow rate was obtained:

$$y = 102.781 - 0.0013x_1 - 0.0061x_2 - 0.00011x_1x_2,$$

where x_1 = NH_3 concentration and x_2 = gas flow rate.

According to the above equation, all the factors have negative effects. The equation also shows that x_2 (gas flow rate) is the most significant factor, with its coefficient effect being the most pronounced. The contour plot (Fig. 8) explains the behavior of the system. If greater than 95% of NH_3 removal efficiency was achieved at inlet NH_3 of 60 ppm, the gas flow rate was required to stay below 500 L/h. Also, 97% NH_3 removal would be obtained when the gas flow rate was controlled below 250 L/h. By the developed equation or contour plot, a set of suitable operating combinations can be determined when the system is operated in the field.

4. Conclusion

The results of this study demonstrate that the BAC biotrickling filter can efficiently remove high concentrations of NH_3 gas from mixed waste gases containing H_2S and NH_3. During the 140-day operating period,

the system achieved an average 96% of NH_3 and 100% of H_2S removal efficiency without pH adjustment. This system also exhibited high adaptability to shock loading and temperature variation. The coexistence H_2S did not inhibit NH_3 removal, and no significant acidification or alkalinity phenomenon occurred in this system. In addition, low moisture demands and high buffer capacity of pH for the system were other advantages. A set of the operating combinations is also established for further application in the field. Thus these results suggest that the BAC biotrickling filter with specific inoculated microorganisms has a significant potential to treat NH_3 and H_2S from mixed waste gases.

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References

Box, G.E.P., Hunter, W.G., Hunter, J.S., 1978. Statistics for Experiments. John Wiley and Sons, New York, pp. 374–418.

Carlson, D.A., Leisner, C.P., 1966. Soil beds for the control of sewage odors. J. Water Pollut. Control Feder. 38 (5), 829–834.

Chung, Y.C., Huang, C., Tseng, C.P., 1996a. Reduction of H_2S/NH_3 production from pig feces by controlling environmental conditions. J. Environ. Sci. Health., Part A 31 (1), 139–155.

Chung, Y.C., Huang, C., Tseng, C.P., 1996b. Biodegradation of hydrogen sulfide by a laboratory-scale immobilized *Pseudomonas putida* CH11 biofilter. Biotechnol. Progr. 12 (6), 773–778.

Chung, Y.C., Huang, C., Tseng, C.P., 1997. Biotreatment of ammonia from air by an immobilized *Arthrobacter oxydans* CH8 biofilter. Biotechnol. Progr. 13 (6), 794–798.

Chung, Y.C., Huang, C., Tseng, C.P., 2001. Biological elimination of H_2S and NH_3 from waste gases by biofilter packed with immobilized heterotrophic bacteria. Chemosphere 43 (8), 1043–1050.

Devinny, J.S., Deshusses, M.A., Webster, T.S., 1999a. Biofiltration for Air Pollutant Control. Lewis Publishers, New York, pp. 7–8.

Devinny, J.S., Deshusses, M.A., Webster, T.S., 1999b. Biofiltration for Air Pollutant Control. Lewis Publishers, New York, pp. 62–65.

Devinny, J.S., Deshusses, M.A., Webster, T.S., 1999c. Biofiltration for Air Pollutant Control. Lewis Publishers, New York, pp. 52–55.

Eikum, A.S., Storhang, R., 1986. Odour Prevention and Control of Organic Sludge and Livestock Farming. Elsevier Applied Science Publishers, London, pp. 12–18.

Hagedorn, C., Holt, J.G., 1975. A nutritional and taxonomic survey of *Arthrobacter* soil isolates. Can. J. Microbiol. 21 (3), 353–361.

Hartikainen, T., Ruuskanen, J., Vanhatalo, M., Martikainen, P.L., 1996. Removal of ammonia from air by a peat biofilter. Environ. Technol. 17 (1), 45–53.

Hutchins, R.A., 1973. New method simplifies design of activated-carbon systems. Chem. Eng. 80 (3), 133–135.

Julitte, L.Y., Michael, R., Daniel, J.A., 1993. Inhibition of ammonia oxidation in *Nitrosomonas europaea* by sulfur compounds. Appl. Environ. Microbiol. 59 (11), 3718–3727.

Kapahi, R., Gross, M., 1995. Biofiltration for VOC and ammonia emissions control. Biocycle 36 (2), 87–88.

Langenhove, H.V., Wuyts, E., Schamp, N., 1986. Elimination of hydrogen sulfide from odorous air by a wood bark biofilter. Water Res. 20 (8), 1471–1476.

Langenhove, H.V., Bendinger, B., Oberthur, R., Schamp, N., 1992. Organic Sulfur Compounds: Persistent Odorants in Biological Treatment of Complex Waste Gases. Elsevier, Amsterdam, The Netherlands, pp. 177–182.

Lee, S.K., Shoda, M., 1989. Biological deodorization using activated carbon fabric as a carrier of microorganism. J. Ferment. Bioeng. 68 (6), 437–444.

Leson, G., Winer, A.M., 1991. Biofiltration: An innovative air pollution control technology for VOC emission. J. Air Waste Manage. 41 (10), 1045–1054.

Martin, G., Lemasle, M., Taha, S., 1996. The control of gaseous nitrogen pollutant removal in a fixed peat bed reactor. J. Biotechnol. 46 (1), 15–21.

Medina, V.F., Webster, T., Devinny, J.S., 1995. Treatment of gasoline residuals by granular activated carbon based biological filtration. J. Environ. Sci. Health, Part A 30 (2), 407–412.

Mossel, D.A.A., Indacochea, L., 1971. A new cetrimide medium for the detection of *Pseudomonas aeruginosa*. J. Med. Microbiol. 4 (3), 380–382.

Ottengraf, S.P.P., Van Den Oever, A.H.C., 1983. Kinetics of organic compound removal from waste gases with a biological filter. Biotechnol. Bioeng. 25 (12), 3089–3102.

Ryer-Power, J.E., 1991. Health effects of ammonia. Plant/Operations Prog. 10 (2), 228–232.

Sabo, F., 1993. Development and testing of high-efficiency biofilters. In: Proceedings of the 86th Annual Meeting of the Air and Waste Management Association, Denver, Colorado.

Togashi, J., Suzuki, M., Hirai, M., Shoda, M., Kubota, H., 1986. Removal of NH_3 by a peat biofilter without and with nitrifer. J. Ferment. Technol. 64 (5), 425–428.

Yang, Y., Allen, E.R., 1994. Biofiltration control of hydrogen sulfide 1. Design and operational parameters. J. Air Waste Manage. 44 (5), 863–868.

Yani, M., Hirai, M., Shoda, M., 1998. Ammonia gas removal characteristics using biofilter with activated carbon fiber as a carrier. Environ. Technol. 19 (6), 709–715.