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Application of biological methods in the treatment of gaseous ammonia

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Abstract:

In Vietnam, practical applications of biological methods in air pollution control are highly limited. This study evaluated and compared the ammonia removal performance in air of a cow-manure biofilter, commercial compost biofilter, and biotrickling filter with K3 biomedium cultured with attached microorganisms from activated sludge. The results indicated that with an inlet NH_3 concentration of 65-80 mg/m^3 (95-117 ppm), the treatment efficiency was highly promising with an output concentration in the range of 2-5 mg/m^3 (3.0-7.5 ppm) and elimination capacity of 3-9 gNH_3/m^3.h. With an inlet concentration below 200 mg/m^3, all three experimental models could remove ammonia to meet the emission standard (QCVN 19:2009/BTNMT) of 50 mg/m^3. The study results indicated that the investigated biological technologies have potential for use in removing ammonia and other odorous gases in polluted air.

This study investigated the removal of gaseous ammonia using cow manure, compost, and K3 material as biomedium in biofilters and biotrickling filters.

Keywords: **biological methods, biomedium, gaseous ammonia.**

Classification number: **3.5**

Introduction

Today, air pollution is a serious concern attracting considerable attention from citizens and scientists. Ammonia is one of the most common air pollutants, and is released from various sources such as sewage and wastewater treatment plants, animal-waste decay on livestock farms, organic decomposition in composting processes, as well as many industries such as petrochemical, food, paper pulp, metal, and textiles. Ammonia emissions could have negative impacts on human in terms of comfort because of its bad smell, as well as on the environment because it increases the nitrogen nutrient and acidifies water [1-3]. Traditional technologies have been applied for gaseous ammonia removal, such as condensation at low temperatures and/or high pressures, absorption using water or diluted acidic solutions, adsorption using porous solid materials, and thermal/catalytic oxidation at high temperatures. However, these methods are not particularly efficient, environmentally friendly, or economical, either because they have high costs or are harmful with secondary pollutants [4].

Recently, biological methods have been widely applied for solid waste, wastewater, and even gas treatment. Biofiltration units have been successfully applied to the removal of odorous and toxic air pollutants. They function efficiently and economically when removing low concentrations of pollutants with low installation and operation costs, low energy and maintenance requirements, long life and high durability, environmentally safe operations, and without generating pollutants [5, 6]. Biofiltration units are microbial systems in which microorganisms develop and grow to form a biofilm on a biomedium surface [7]. When polluted gas passes through the biomedium bed, soluble pollutants transfer into the

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liquid phase, and biodegradable pollutants are decomposed by microorganisms in the biofilm. Many industrial and domestic air pollution sources have successfully applied biofiltration to control odours and other air pollutants with high removal efficiencies of >90% and end products of CO_2, water, and microbial biomass [6]. Because of biofiltration's several advantages in terms of cost and the environment, it has become a preferred choice for air pollution control in practical applications. Ammonia emission control through biofiltration with different microorganism media, such as multicultural microbial load on peat and inorganic media [8], compost [9], and agricultural residue media [10] has been investigated by numerous researchers [11, 12].

Biological treatment systems include biofilters (BFs), biotrickling filters (BTFs), bioscrubbers (BSs), and membrane bioreactors (MBRs) [13]. BFs work with biomedia such as compost, activated carbon, peat, perlite, and soil. This traditional technology is widely used and has a long history of development and application. However, it has disadvantages such as media compaction, difficulty in pH and moisture control, biomedia degradation, acidic accumulation, and applicability for low pollutant concentrations. BTFs comprise inert packing materials such as plastic, ceramics, gravel, and wood. This technology works through the recirculation of an aqueous solution distributed from top to bottom of the packed column. The biofilm on packing material surface is the key component in the gas treatment. BTFs have the advantage of liquid phase control, which could provide required nutrients/components and remove acidic/toxic compounds [14]. Therefore, BTFs can avoid the drawbacks associated with BFs and are considered more effective for treating gaseous pollutants. Regarding the other two technologies, BSs are rarely used and MBRs are mainly employed in lab-scale studies. In the biofiltration removal process, ammonia is first converted to nitrite by nitrosomonas, and this nitrite is then converted to nitrate by nitrobacteria [15]. A denitrifying process also exists, where nitrate is converted to nitrogen gas by pseudomonas and clostridium under an anoxic condition.

In Vietnam, such biological methods for air pollutant control have not been widely applied because of limited research and experience. Therefore, the present study investigated the efficiency of two different BFs and a BTF in NH_3 removal. In the lab-scale setup, both BFs and the BTF used local growth microorganisms.

Materials and methods

Materials

NH_3 solution at a concentration of 25% (w/w) was purchased from Xilong (Guangdong, China) and used as the ammonia source in this study. NH_3 removal experiments were conducted using lab-scale BF and BTF models. These models represented improvements over our previous study on the removal of H_2S, which was designed with parameters taken from relevant literature [16, 17]. Table 1 summarises the present study's detailed design and operational parameters. The following three types of biomedia were used: compost and cow manure for the two BF models and K3 inert media for the BTF model (Fig. 1).

Table 1. Configuration and operational parameters of the three models.

Parameters	Unit	BTF	CM-BF	CP-BF
Height×length×width	mm	1100×140×140	1000×110×110	540×150×150
Packing height	mm	400	270	170
Packing volume	l	7.8	3.3	3.8
Gas flowrate	l/min	7.5	7.5	7.5
Empty bed retention time (EBRT)	sec	63	26	30
Liquid flowrate	l/min	0.24	-	-

Cow-manure BF (CM-BF) model: cow manure was incubated for approximately 2 months and then dried under sunlight before being stored in a household in the Mekong delta (Vietnam). This process was to kill weed seeds and insect germs, pathogenic bacteria, and mould, as well as promote organic decomposition and accelerate mineralisation. The dried cow manure was refined and supplied with water before being incubated under an anaerobic condition for approximately 1 month. Its pH after incubation was 7.72 and moisture content was 72.1%. The manure contains humus content and other ingredients that could provide in-situ sources of carbon as well as macro- and micro-nutrients for the microorganisms.

Compost BF (CP-BF) model: this study used commercially available compost (organic fertiliser Agrimartin) in the market (Ho Chi Minh city, Vietnam). Compost-based media are widely used in BFs because of

Fig. 1. Biomaterials for NH_3 treatment. (A) cow manure, **(B)** commercial compost, and **(C)** K3 biomedium.

their low cost and abundant microbial communities that are ready to decompose various pollutants. Because it would divert the local microbial population, the addition of external microorganisms and enzymes is usually not necessary. The quantitative component of this compost comprised 72% organic (dried) matter, 3.5% N, 2.5% P_2O_5, and 2.5% K_2O.

BTF model: in this model, K3 medium was used to support microorganism growth. It was made of high density polyethylene in a round shape with a honeycomb structure inside, providing a high surface area through numerous folded wrinkles. First, the K3 medium was placed in an activated sludge wastewater tank under aeration to provide dissolved oxygen and nutrients at a chemical oxygen demand (COD) concentration of 500 mg/l as well as a small amount of NH_4Cl for microorganism adaptation. The concentration of ammonia in the wastewater was initially maintained at 1 mg/l for adaptation and then increased to 10 mg/l for microbial growth. The high organic loading and microorganism content accelerated the development of aerobic and anaerobic microorganisms on the surface of the K3 medium. Subsequently, K3 biomedia (i.e., K3 medium with a biofilm) was placed into the BTF model and operated with wastewater (i.e., domestic wastewater from a student dormitory) containing molasses as carbon sources and nutrients for microorganism growth on the K3 medium.

Ammonia treatment experiments

Figure 2 illustrates the experimental setup for the biological removal of NH_3 in air. The experimental model consisted of three lab-scale models (BTF, CM-BF, and CP-BF) made of acrylic resin. Ammonia-laden air with a high (almost saturated) concentration of ammonia was prepared by passing a clean air flow through a vessel containing 25% ammonia solution for ammonia evaporation. The ammonia-laden air was then mixed with fresh air at certain ratios to prepare a gaseous mixture with the desired concentrations of ammonia. The mixed gas was then flowed through the three models using a three-way connector split into two lines. One line flowed through an impinger for inlet gas sampling and ammonia concentration analysis, whereas the other line was divided into three lines that flowed directly into the BTF and BF models. In the BTF model, recirculated wastewater was irrigated from top to bottom to provide substrates and nutrients for the microorganisms. Initially, all three models were begun with low ammonia concentrations of 15-30 mg/m^3 for 20 days of adaptation. Subsequently, the ammonia concentration was increased to the required concentration of 50-80 mg/m^3. During the experiment, the BF models were supplied with water to maintain moisture above 50%. Before sampling, the models were operated and controlled stably for 1 to 2 h. Ammonia gas samples from the inlet and outlet of each model were taken for 1 min at a flow rate of 7 l/min and then sent for concentration analysis using the indophenol method. Ammonia samples were taken and analysed three times/day and the average results were reported.

The ammonia removal efficiency (RE, %) and elimination capacity (EC, amount of ammonia removal per unit volume of biomedium per unit of time, gNH_3/m^3.h) were calculated as follows:

$$RE = \frac{C_{in} - C_{out}}{C_{in}} \times 100\% \quad (1)$$

$$EC = \frac{(C_{in} - C_{out})}{V} \times Q \quad (2)$$

where C_{in} and C_{out} (mg/m^3) are the inlet and outlet ammonia concentrations, respectively, and Q (m^3/h) and V (l) are the gas flowrate and packing volume (i.e., cow manure, compost, and K3 biomedium), respectively.

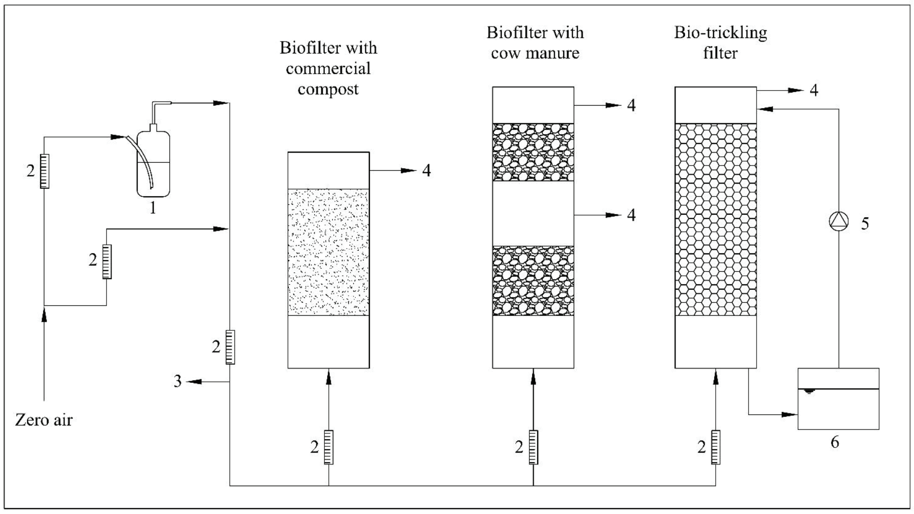

Fig. 2. Diagram of the experimental models: (1) NH_3 solution, (2) flowmeter, (3) inlet sampling location, (4) outlet sampling location, (5) circulation pump, and (6) wastewater tank.

Results and discussion

Evaluation of the ammonia RE of the three models

After 20 days of adaptive operation with low ammonia concentrations, ammonia treatment of the three models was investigated at inlet concentrations of approximately 60 mg/m^3 for 30 days. The treatment efficiency was calculated by measuring the inlet and outlet ammonia concentrations. The results in Fig. 3 show that the performance of the CM-BF model was unstable during the first 12 days, which might have been because the microorganisms in cow manure take longer to adapt and stabilise. The RE of this model was stable at 92% after 26 days of operation and increased to 96% at the end of experiment. A similar trend was observed for the CP-BF model with an RE of 94% after 30 days of operation. The RE of the BTF model was unstable, possibly because of the characteristics of the model and variation in wastewater recirculation. However, this model could still achieve an ammonia RE of 94% under the experimental conditions after 30 days of operation.

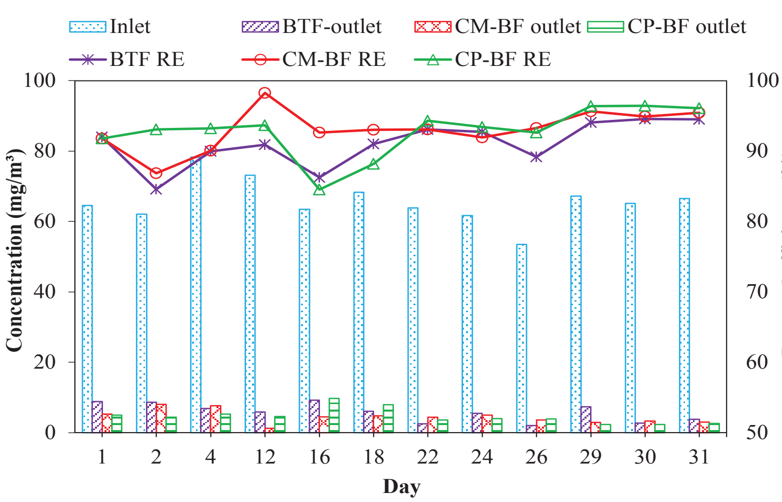

Fig. 3. Comparison of the treatment efficiency of the three models.

Because the treatment efficiency depended on various factors such as inlet concentration and gas flow rate on the biological bed, this study calculated the ECs of ammonia (gNH_3/m^3.h) and the results of which are presented in Fig. 4. The highest EC was achieved by the CM-BF in the range of 6.9-10.0 gNH_3/m^3.h. By contrast, the CP-BF model achieved a lower EC in the range of 5.9-8.8 gNH_3/m^3.h. These results are comparable to relevant studies that have used municipal compost inoculated with thickened municipal activated sludge with ECs of 9.85 gNH_3/m^3.h (three-stage BF) and 8.08 gNH_3/m^3.h (one-stage BF) [18], co-immobilised cells with an EC of 6.8 gNH_3/m^3.h (164 ppm NH_3) [19], and agricultural residue BF medium with ECs of 14 gNH_3/m^3.h (500 ppm NH_3) and 23.5 gNH_3/m^3.h (1000 ppm NH_3) [10].

In this experiment, the BTF had an EC of 3-4 gNH_3/m^3.h, which is rather low compared with the ECs of the BFs. It was also low compared with that reported in a study that used polyurethane foam (0.9-21.7 gNH_3/m^3.h; 60-1600 ppm NH_3, EBRT of 150 s) [20]. In terms of stability, the BTF model was more stable than the two BF models, possibly because of the recirculation of liquid and stable attached microorganisms. In addition, the EC for NH_3 in the BTF depended on gas flow rates and bed lengths [21]; thus, further investigation is required to optimise the operation.

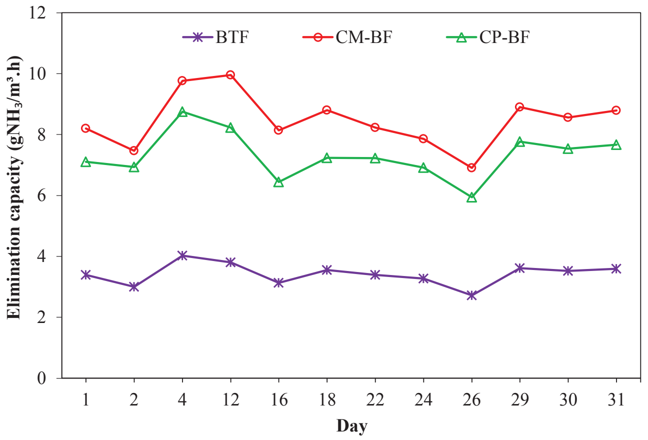

Fig. 4. Comparison of the elimination capacities of the three models.

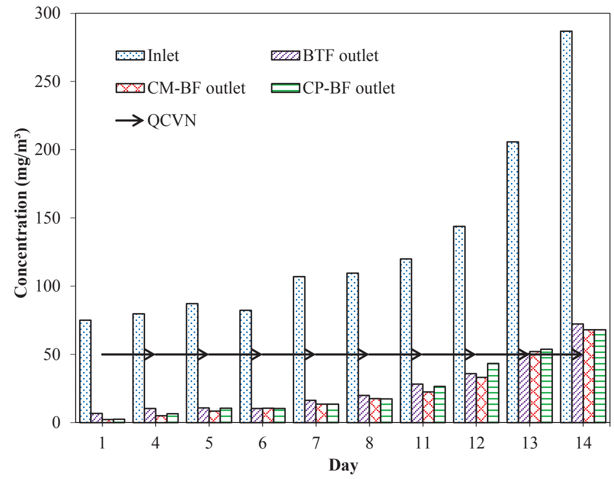

Fig. 5. Inlet and outlet concentrations of the three models.

Performance of three models under high-inlet NH_3 concentration

The aim of the experiment was to determine an NH_3 inlet concentration limit that still met the outlet NH_3 emission standard (QCVN 19:2009/BTNMT) of 50 mg/m^3. The ammonia concentration and flowrate were increased gradually each day from the values of 8 l/min and 68.4 mg/m^3, respectively, until the outlet concentration exceeded 50 mg/m^3. As presented in Fig. 5, under an inlet concentration of $\leq$200 mg/m^3 and flowrate of 16 l/min, the three models were still able to remove ammonia to meet the emission standard. Under the same treatment condition, the CM-BF provided superior treatment, although the performance differences between three models were not significant. The minimum gas retention times of the three models were then calculated, and the results were 63 sec for the BTF, 26 sec for the CM-BF, and 30 sec for the CP-BF.

The NH_3 ECs of the three models were calculated, which is depicted in Fig. 6. The ECs can be observed to continuously increase from 4.2 to 67.7 $gNH_3/m^3.h$ as the inlet concentration increased from 75.1 to 286.9 mg/m^3. At an inlet concentration of 206 mg/m^3, the CM-BF, CP-BF, and BTF models had ECs of 47.7, 38.4, and 19.0 $gNH_3/m^3.h$, respectively. These EC values for the CM-BF and CP-BF were remarkably high compared with those reported in the abovementioned studies [10, 18, 19], but that of the BTF was still low [10]. These results confirmed the inefficient operation of the BTF and suggested that BFs seem to be a more suitable choice for ammonia removal in practical applications under the current condition.

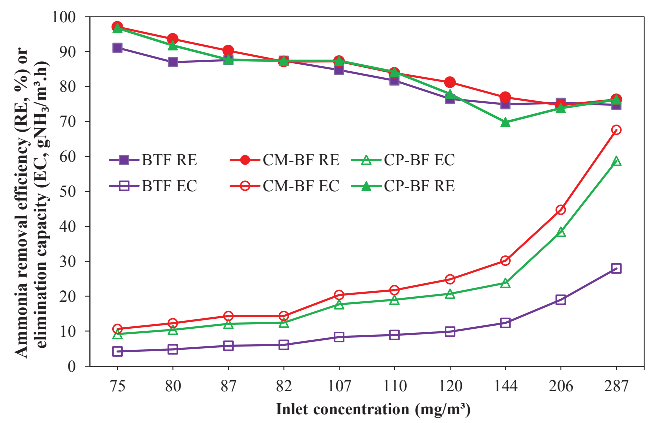

Fig. 6. Removal efficiency (RE) and elimination capacity (EC) of the three models.

Conclusions

The experimental results demonstrated that CM-BFs, CP-BFs, and BTFs with K3 biomedium can be applied to remove ammonia at REs up to 96%, although their optimised conditions have not been investigated. These technologies can remove ammonia in air to meet the National Technical Standard on Industrial Emissions for dust and inorganic substances (QCVN 19:2009/BTNMT) of 50 mg/m^3 if the concentration is below 286 mg/m^3 at a flowrate of 17 l/min. In this study, the stability and efficiency of BFs were higher than those of the BTF, which might have been because of the microorganism attachment and population in the BTF not being well-controlled. This study provides a first attempt at the application of different biological methods to remove ammonia from air. The results indicated that such biological technology could have potential for removing ammonia and other odorous gases from polluted air. Future studies should focus on investigating and optimising operation parameters (e.g., EBRT, concentration, ammonia loading rate, pH, and temperature), determining microbial strains, applying other media that contain superior microbial strains, and nitrogen balance for circulating wastewater used in BTFs.

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The authors declare that there is no conflict of interest regarding the publication of this article.

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