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The effects of selected parameters on the nitric oxide removal by biofilter

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Abstract

A bench-scale biofilter was used to demonstrate the treatability of off-gas containing nitric oxide (NO) by examining selected operational parameters. After 6 days of operation, the biofilter reached to a steady state and NO reduction was significant, reducing from 200 ppm to 95 and 40 ppm after 6 and 40 days of continuous operation. The oxygen concentrations in the inlet would affect NO removal performance significantly; as oxygen content decreasing from 6% to 0%, the NO removal efficiency increased from 55% to 99%, indicating that oxygen inhibited the progress of denitrification. NO removal was inversely proportional to inlet NO concentration, removal efficiency decreased from 88% to 40 % as NO concentration increasing from 60 to 500 ppm. Column height would significant effect on the NO removal efficiency, under column height = 6.5 m and O_2 = 6% conditions, 90% of removal efficiency was achievable. The effect of glucose added into biofilter would significantly enhance the NO removal efficiencies for both anaerobic and aerobic conditions of which 99% and 55%, respectively.

Keywords: Biofilter nitric oxide (NO); Removal efficiency; Column height; External carbon source

1. Introduction

Nitrogen oxides (NOx) are emitted from various industrial processes and transportation activities. NOx consist of about 95% nitric oxide and about 5% nitrogen dioxide, both of which are hazardous air pollutants and cause serious environmental problems [1,2]. Stationary sources account for about 44% of total NOx emissions in Taiwan, with the power industry being the major source [3]. It is difficult to collect NOx once it becomes dispersed in air, so NOx can be removed effectively only before emission [4].

In the past, traditional control technologies, such as selective catalytic reduction (SCR) and selective non-catalytic reduction (SNCR), were applied to control NOx emissions in some industries. However, these two processes required high temperatures and the use of catalysts, involving high installation and operation costs as well as generating a large quantity of secondary waste for which manufacturers had to pay cleanup and disposal costs [5]. Economic and technical constraints in SCR and SNCR methods motivated researchers to develop new, cost-effective processes to removal NOx from flue gas. Biological NOx treatment has been deemed a promising alternative to eliminate industrial waste generation and enable compliance with emission standards [6]. Biological treatment systems can be operated under ambient temperature with the use of inexpensive microbial inocula. One such system, the biofilter, is a kind of biochemical fixed bed reactor wherein microorganisms settle on the surface of the filter medium material and form a biofilm; the airborne substances are absorbed and utilized by microorganisms [7]. Biofilter treatment has been proven effective in treating odors and volatile organic compounds (VOC), such as benzene [8], styrene [9], phenols [10], and alkenes [11].

Applying a denitrification mechanism to remove NOx relies on the activity of denitrifying organisms that are ubiquitous in nature; denitrifiers have the ability to reduce NOx under limited-oxygen conditions. The reductive process occurs in the following simplified order [12]: $NO_3^- \rightarrow NO_2^- \rightarrow NO \rightarrow N_2O \rightarrow N_2$. In denitrification processes, organic carbons serve as the electron donors, which can include acetic acid, methanol, and domestic sewage [13].

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The aims of this work were to demonstrate the feasibility of the biofilter in a bench-scale installation by examining selected parameters. The effects of various operational parameters (NO and O_2 concentration, column height, flow rate, external carbon source) on NO removal efficiency were examined. The overall treatment performance was also investigated.

2. Materials and Methods

2.1. Filter medium and inoculum

The filter medium contained wood chips and compost. The compost was provided by the experimental animal farm at National Taiwan University (Taipei, Taiwan) and was sieved before use to prevent filter blocking. Cyatheaceae wood chips were added to the compost to serve as a bulking agent that could increase free space, reduce compaction, and enhance ventilation in the system. The inoculum cultures were prepared by mixing the wastewater from the animal farm with the denitrifying medium [14].

2.2. Nutrient supplement

Nutrient supplement was provided to the biofilter as the source of carbon (glucose), inorganic nutrients, and moisture. The medium was delivered from the top of biofilter at the rate of 40 mL/day. The nutrient solution (pH 8.0) contained the following components (in 1 L of H_2O): K_2HPO_4 (0.4 g), KH_2PO_4 (0.15 g), NH_4Cl (0.3 g), $MgSO_4{\cdot}7H_2O$, (0.4 g), sodium acetate 2.93 g, and 2 mL of trace element solution, which contained (in 1 L of H_2O): EDTA (50.0 g), $ZnSO_4{\cdot}7H_2O$ (2.2 g), $CaCl_2{\cdot}2H_2O$ (5.5 g), $MnCl_2{\cdot}4H_2O$ (5.06 g), $FeSO_4{\cdot}7H_2O$ (5.0 g), $(NH_4)_6Mo_7O_{24}{\cdot}2H_2O$ (1.1 g), $CuSO_4{\cdot}5H_2O$ (1.57 g) and $CoCl_2{\cdot}H_2O$ (1.61 g). These inorganic materials were selected because they had previously been used to grow aerobic nitrifying bacteria [15].

2.3. 2.3 Chemicals

Nitrous oxide (NO), pure nitrogen, and air were obtained from Sanford Chemical Co. (Taoyuan, Taiwan), and the standard solutions for nitrate and nitrite were provided by Merck (New Jersey, USA).

2.4. Biofilter design and equipments

Fig. 1 shows the schematic diagram of the biofilter column. The packed bed was divided into six 35 cm-high sections with an inner diameter of 8 cm. A perforated stainless steel plate at the bottom of each section supported the packing material. Each packed section was separated by a plenum to redistribute the gas flow. Gas sampling ports were located in each of the plenum at the 35, 70, 105 cm-high positions as well as at the inlet (140 cm) and outlet (0 cm) ports to the column. The gaseous samples were conducted into a NO_x analyzer (Ishibashi Science Industries, Tokyo, Japan) to analyze the NO concentration, and the on-off switch control was computer programmed.

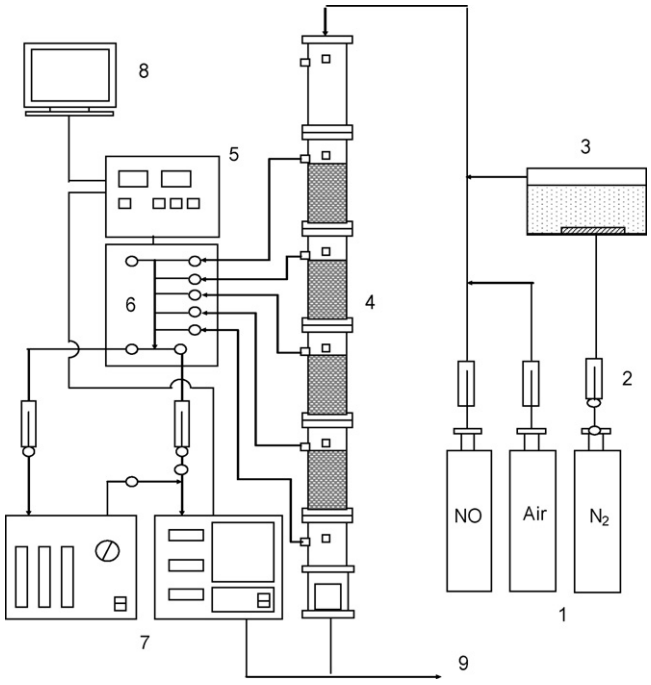

Fig. 1. Biofilter design. Component: 1, gas supply; 2, flow control; 3, humidifier; 4, biofilter column; 5, control pannel; 6, multi-switch sampling ports; 7, nitrous oxide analyzer; 8, data acquisition device; 9, ventilation.

Experimental gases were injected from the column top, and the flow rate was regulated by a set of precision flow controllers. The off gas was discharged to the atmosphere via ventilation. Real-time monitoring data from the NO_x analyzer was collected through signal processors and modules (Adventech, Taipei, Taiwan) and transmitted to a computer. The oxygen content in the gas flow was measured by an oxygen analyzer (Liston Scientific, California, USA). All connecting tubes in this study were made of stainless steel to avoid corrosion problems. A scanning electron microscope (Hitachi, Tokyo, Japan) was used to observe the microbial effect on the filter medium. An element analyzer (Heraeus, Hanau, Germany) was applied to the analysis of C, N, H, and an ion analyzer (Dionex, California, USA) was applied to analyze nitrate concentration.

2.5. Experimental procedures

In order to maintain a suitable environment for the growth of denitrification microorganisms, a buffer solution was added to the filter medium to maintain the pH and relative humidity (RH) at around 6.5–7.0 and $80 \pm 10\%$, respectively. To avoid drying of the filter medium, the inlet gas was split into two streams; NO gas was injected into one stream by flow-controlled gauges (AAlobrg, New York, USA), which were pre-adjusted before the experiments; the other stream supplied the nutrient-contained droplets by an aerosol generator (Heart Nebulizer, Arizona, USA). The median diameter of the droplet was approximately 15 μm. The characteristics of the filter medium were investigated before the experiments started, so the results could serve as the adjustment baseline of microbial growth.

To compare NO adsorption by the filter medium, a separate biofilter column (control set) was set up, and NO was directed into the column at various flow rates (from 20 to 200 ppm) to determine the amount of adsorption at equilibrium status (0.4 μg NO/g filter medium).

The NO gas concentration was pre-adjusted to fit the experimental design; then, the gas was directed into the humidifier to increase the humidity, which could provide the moisture for microbial communities in the biofilter. Moisture-rich (RH = 80 ± 10%) NO gas was fed from the top of the column at various concentrations; the gas flowed through the column and was discharged from the bottom to the vent. The NO concentration in the off-gas was monitored by a NO_x analyzer, which was pre-adjusted and calibrated before the experiments were conducted. The oxygen content in the inflow gas stream was sampled using a stack sampler and analyzed by an oxygen analyzer (Liston Scientific, California, USA). All measured data was transmitted and stored in a computer.

3. Results and discussion

3.1. NO removal performance

To investigate the performance of the biofilter column, some controlling factors were set, i.e., the inflow concentration, the oxygen content in the gas stream, and the amount of added carbon source (glucose). Before the experiments were conducted, a NO removal test was performed to evaluate the microbial communities. Time courses of the NO concentration variation by a biofilter under the conditions of flow rate = 30 L/h, NO concentration at the inlet = 200 ppm, oxygen content = 6%, and the addition of glucose at 1 g/day are shown in Fig. 2. After 6 days of continuous operation, the NO concentration in the effluent gas was stepwise decreased to about 95 ppm, while the O_2 concentration remained constant around 6%; the decreasing trend reached a relatively steady condition at around 40 ppm after 40 days of operation, suggesting that the microbial communities and their functions were stable. Fig. 3 presents the microbial communities on filter medium materials before inoculation and after 6 days of operation. It was observed that the microbial communities were established after 6 days of operation; however, the species were not identified in this study. A comparison of Fig. 3(a) and (b) shows clearly that the microorganism count in (b) is far greater than that in (a), suggesting that the microorganisms can grow on the filter medium and use NO as electron acceptors to promote metabolism. In the following section, the effects of selected parameters on NO removal are discussed and presented.

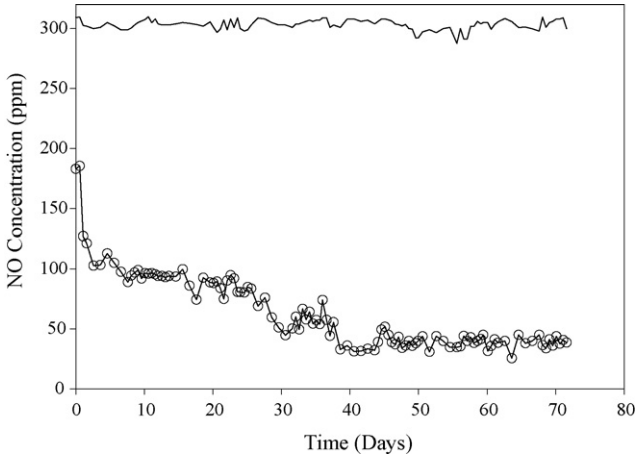

Fig. 2. Time course of NO concentration variation. Flow rate = 30 L/h, NO = 200 ppm, O_2 = 6%, and the external carbon source (glucose) = 1 g/day. Symbol: (—) inlet; (◯) exist.

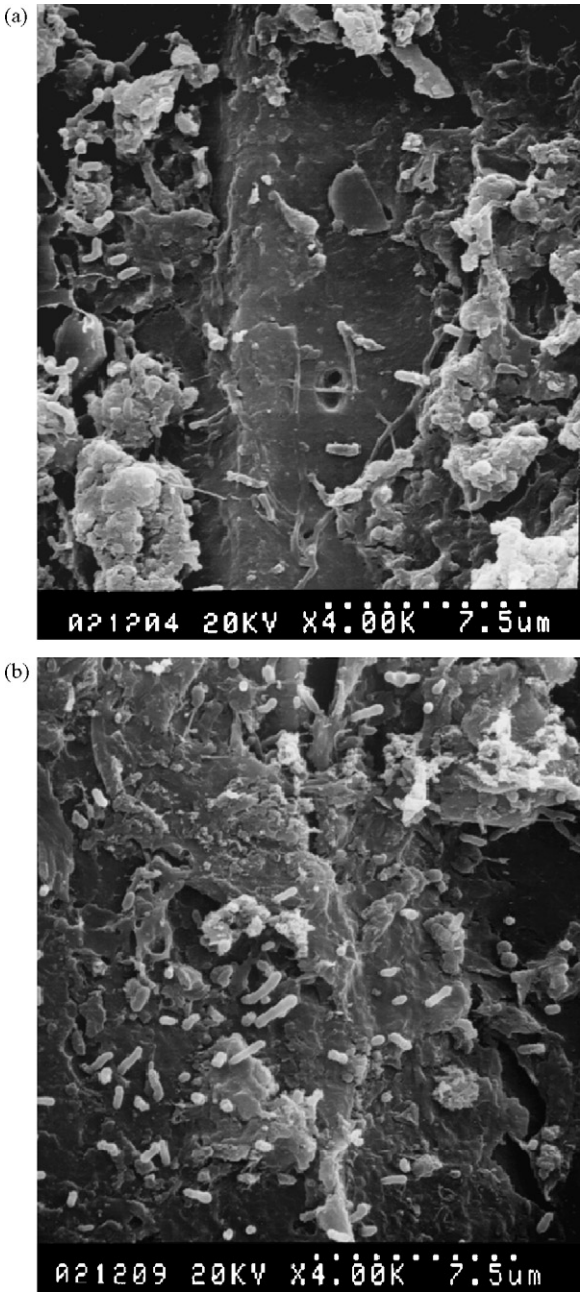

Fig. 3. In situ detection of microbial communities (a) before and (b) after NO treatment.

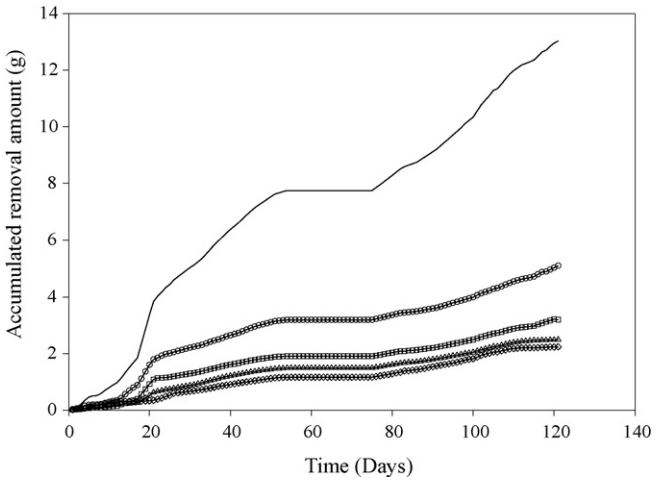

Fig. 4. Accumulated NO removal amount at five different positions. Flow rate = 30 L/h, NO = 200 ppm, anaerobic, and glucose = 1 g/day. Symbols: removal amount at height = 0 m: (◊); 0.35 m: (Δ); 0.7 m: (□); 1.05 m: (◯); total removal amount: (—).

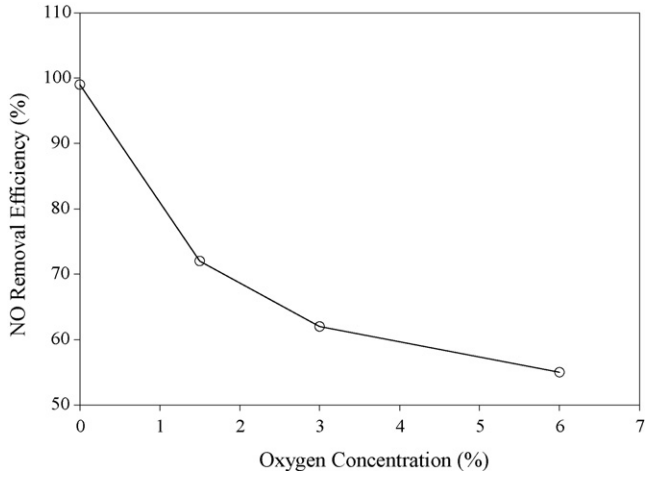

Fig. 5. Effect of oxygen concentration on the NO removal efficiency. Flow rate = 30 L/h, NO = 200 ppm, and glucose = 1 g/day.

3.2. NO removal amount

The accumulated NO removal amount could be estimated by measuring the average inlet and outlet NO concentrations multiplied by the flow rate; the results are presented in Fig. 4 Under 120 days of continuous anaerobic operation, the total NO removal amount could reach about 13.0 g. It was also observed that the individual NO removal amounts for four different sampling positions were 5.1 (at height = 1.4 m), 3.2 (1.05 m), 2.5 (0.7 m), and 2.2 g (0.35 m), respectively. The results indicated that the amount of NO removed was dependent on the height of the material in the column, suggesting that a higher column position could achieve better removal performance. It could be explained that the more NO gas came in contact with inoculated denitrification microorganisms on the filter medium, the more NO was removed. As NO traveled down through the biofilter column, NO was utilized and the concentration was reduced; therefore, less NO remained in the gas stream for chemoorganoheterotrophic denitrification.

3.3. Effect of oxygen concentration

The effect of the O_2 concentration on NO removal was investigated, and the results are presented in Fig. 5. The O_2 concentrations varied from 0% to 6% with NO = 200 ppm and flow rate = 30 L/h; the inlet gas was injected from the top of the column, and the samples were collected at five sampling ports. The results revealed that the amounts of NO removed were inversely proportional to the inlet O_2 concentration. It was observed that the best NO removal efficiency was achieved under anaerobic conditions, which could reach about 99%. The NO removal efficiency was defined as

$$\text{NO removal efficiency}\,(\%) = (1 - [\text{NO}]_{\text{out}}/[\text{NO}]_{\text{in}}) \times 100\% \quad (1)$$

where $[\text{NO}]_{\text{in}}$ and $[\text{NO}]_{\text{out}}$ are the measured NO concentrations in the inlet and exit gas.

As O_2 concentration increased from 0% to 6%, the removal efficiency stepwise decreased from 99% to 55%, suggesting that increasing O_2 concentration would not enhance the efficiency but inhibit the NO removal performance.

The O_2 consumption was investigated in the presented study that could involve in both chemical and biological reactions, and the chemical reaction of O_2 and NO in the gas and liquid phases could be described as follows:

In the gas phase [16]:

$$2\text{NO} + \text{O}_2 \rightarrow 2\text{NO}_2 \quad (2)$$

The proposed reaction rate was

$$-\frac{\text{d}[\text{NO}]_{\text{g}}}{\text{d}t} = 2k_1[\text{NO}]_{\text{g}}^2[\text{O}_2]_{\text{g}} \quad (3)$$

where the rate constant k_1 was $7.5 \times 10^3\ \text{L}^2/(\text{mol}^2\ \text{s})$ at 25 °C.

According to this value, only 0.23 ppm of NO reacted with 3% of O_2 in the gas phase under the NO = 200 ppm condition, indicating that the amount of O_2 consumption in the gas phase was negligible in the conducted experiments.

Awad and Stanbury (1993) [17] reported the reaction of NO and O_2 in the liquid phase as follows:

$$4\text{NO} + \text{O}_2 + 2\text{H}_2\text{O} \rightarrow 4\text{NO}^-{}_2 + 4\text{H}^+ \quad (4)$$

$$-\frac{\text{d}[\text{NO}]_1}{\text{d}t} = 4k_2[\text{NO}]_1^2[\text{O}_2]_1 \quad (5)$$

where the rate constant k_2 was $2.1 \times 10^6\ \text{L}^2/(\text{mol}^2\ \text{s})$ at 25 °C. A comparison of the reaction rate constants, k_1 and k_2, showed that the oxidation of NO in the liquid phase was more rapid than that in the gas phase. Typically, denitrifiers could use their own enzyme system to consume NO that could be found under anaerobic growth conditions with N-oxides as electron acceptors [18]. Furthermore, in the case of the flue gas containing NO, O_2 concentration was often around 3%–8%, which would increase the dissolution of oxygen in water, thus likely inhibiting denitrification [19]. O_2 content would inhibit the NO utilization significantly and served as an important factor in NO removal process.

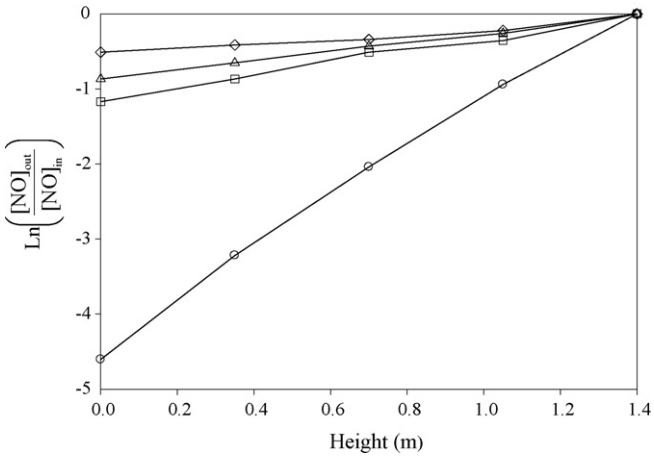

Fig. 6. Effect of column height on NO removal. Flow rate = 30 L/h, NO = 200 ppm, and glucose = 1 g/day. Symbols: (◯) 0%, (□) 1.5%, (Δ) 3%, (◇) 6%.

3.4. Effect of column height

The inlet gas containing NO was injected from the top of the column; therefore, the NO and filter medium contact time was proportional to the distance from the column top. Thus, the effect of the column height on NO removal could be investigated by measuring the NO concentration at five sampling ports, with the heights set at 0, 0.35, 0.7, 1.05, and 1.4 m. The results are presented in Fig. 6 which shows a linear relationship between the column height (h) and $\ln([NO_{out}]/([NO_{in}])$, suggesting that the removal of NO in the filter medium obeyed first-order kinetics with respect to the inlet NO concentration. The figure reveals that the longer contact time resulted in better NO removal efficiency, suggesting that the higher biofilter column achieved higher removal performance. According to the results obtained in this study, a simple way to enhance NO removal in this system is to increase the column height. Because NO removal obeyed first-order kinetics with respect to the inlet NO concentration, by calculation, 90% removal efficiency could be reached by increasing the column height to 6.5 m under 6% O_2 conditions.

3.5. Effect of inlet NO concentration

The relationship between the removal efficiency and the inlet concentration is illustrated in Fig. 7 under the following conditions: anaerobic, inlet NO concentration of 60–500 ppm in the biofilter, and no glucose added. It was found that the NO removal was inversely proportional to the NO concentration in the inlet gas; removal efficiency decreased from 88% to 40% with an increase in inlet concentration from 60 to 500 ppm, which might be due to insufficient biomass. With an increase of NO concentration in the inlet gas, the amount of NO removed by microorganisms was proportional to the NO concentration in the inlet gas, which increased from 0.1 to 5.1 μmol/min, suggesting that more NO would enhance microorganism growth under sufficient nutrient conditions.

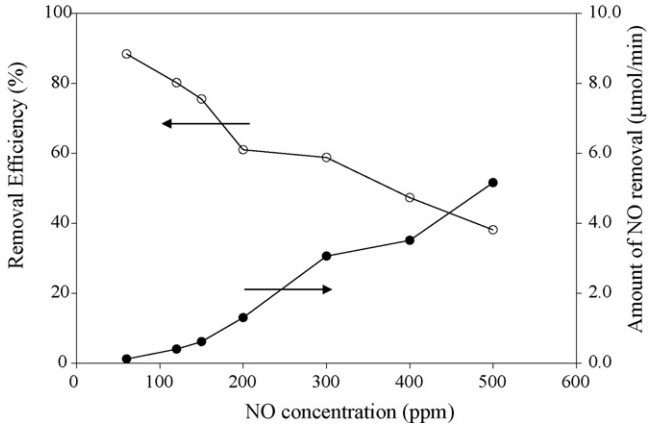

Fig. 7. Relationship between removal efficiency and inlet concentration in biofilter. Flow rate = 30 L/h, anaerobic. Symbol: (◯) removal efficiency, (●) removal amount.

3.6. Effect of external carbon source

Fig. 8 shows the effect of an external carbon source (glucose) added into the biofilter system at a gas flow rate = 30 L/h and under anaerobic conditions. It was observed that the NO concentration decreased significantly as the glucose was added into the column, which could be observed at column heights = 1.05 and 0.7 m. For the other two column heights (0.35 and 0 m), the effect was not remarkable, which could be explained by the fact that the glucose was utilized quickly by microorganisms in the top layer of the filter medium material due to easy biodegradation of glucose. As glucose was added from the top of the column, microorganisms at the top of the column could be utilized to process NO removal function easily, significantly reducing NO concentration reduction. The effect of added glucose on NO removal was not remarkable as the column height decreased, which was due to the small amount of remaining glucose in the filter medium, indicating that the organic matters in the filter medium was difficult to degrade compared to glucose. Under the same conditions with the exception

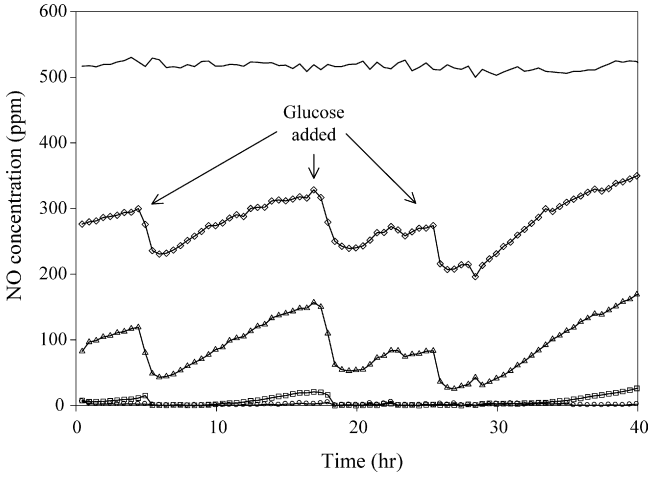

Fig. 8. Effect of glucose added on the variation of NO concentration at different column height. Flow rate = 30 L/h, anaerobic, and NO = 200 ppm. Symbol: (◯) 0 m, (□) 0.35 m,(Δ) 0.7 m, (◇) 1.05 m, (—) 1.4 m (inlet).

Table 1
Effect of external carbon source on the NO removal

	Conditions	Flow rate (ml/min)	External carbon source	Removal efficiency (%)	
				Before added	After added
Apel et al. (1995)	Anaerobic	1000	Molasses (5 ml/day)	20	90
This study	Anaerobic	500	Glucose	21	99
	O_2 = 6%	500	(1 g/day)	47	60

of no added glucose, the NO removal efficiency decreased to 21%, compared to 99% of removal under glucose-added conditions. The results were consistent with the similar experiments conducted by Apel et al. [20] in which molasses was added; both NO removal experiments reached 90% and above of performance, suggesting that an external carbon source could significantly enhance removal. The effect of an external carbon source on NO removal under 6% of O_2 content was investigated, and the results are summarized in Table 1 Under the same NO concentration, the effect of glucose added into the biofilter significantly enhanced the NO removal efficiencies for both anaerobic and aerobic conditions. Furthermore, the effect was remarkable under anaerobic conditions, which could reach 99% of removal; in contrast, under aerobic conditions, only about 60% removal was achieved. A comparison of the external carbon sources showed that molasses and glucose had a similar effect on NO removal, and the difference of removal efficiencies between them was not significant. The results indicated that NO removal efficiency under anaerobic conditions was better than that under aerobic conditions, which is consistent with the denitrifier significant growth under anaerobic conditions reported previously [2].

4. Conclusions

The effect of selected parameters on NO removal by continuous biofilter operation was investigated and discussed in this study, and the results show the feasibility of this method. The results of NO removal in this study suggest the following:

- The NO concentration was reduced from 200 ppm to 95 ppm after 6 days and downed to 40 ppm after 40 days of continuous operation, suggesting that the microbial communities and their functions were stable.
- The total amount of NO removal could reach 13 g after 120 days of continuous anaerobic operation, and the removal amount decreased as column height decreased.
- The removal process was favorable under anaerobic conditions, which could achieve a 99% removal rate, compared with 55% removal under 6% of O_2 content.
- An external carbon source (glucose) added into the biofilter reduced NO concentration significantly, which could be utilized quickly by microorganisms in the top layer of the filter medium material. Under the same conditions with the exception of no glucose added, NO removal efficiency decreased to 21%, compared to 99% of removal under glucose-added conditions.

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