Journal of Environmental Science and Engineering B 13 (2024) 97-104
doi:10.17265/2162-5263/2024.03.001

Biomass Control via UV Lamp Application and Nutrient Supply Restriction in the Biofiltration of Gaseous VOCs

Takashi Higuchi[1] and Kokoro Hashizume[2]

1. School of Science and Engineering, Ritsumeikan University, 1-1-1 Noji-higashi, Kusatsu, Shiga 525-8577, Japan

2. Graduate School of Science and Engineering, Ritsumeikan University, 1-1-1 Noji-higashi, Kusatsu, Shiga 525-8577, Japan

Abstract: Biofiltration may have clogging problems owing to excess biomass growth during the treatment of gaseous pollutants. In this study, we employed an UV (Ultraviolet) lamp and controlled the nutrient supply to conduct a biofiltration process for treating 2-butanone (MEK: Methyl Ethyl Ketone) and toluene in a gas stream. Two methods of UV lamp usage (direct and indirect irradiation) and several nutrient supply methods were tested. However, no clear effect was observed with either UV usage. Under the optimal conditions, 97% of the MEK and 69% of the toluene gases were removed after 29 s of EBRT (Empty Bed Retention Time). The inlet loads were 18 and 19 mg/(m^3 ћ) for MEK and toluene, respectively. Under these conditions, 23 g-N/(m^3 day) of nitrate-nitrogen was consumed. Excess biomass growth occurred during simultaneous excess nutrient supply and a persistent irrigation schedule. In this study, we demonstrated the effective use of a dense nitrate solution to deliver an appropriate amount of nutrients and moisture, and the optimal irrigation frequency was four times per week.

Key words: Biofiltration, gaseous VOCs (Volatile Organic Compounds), nutrient supply, UV lamp, ozone.

1. Introduction

Biofiltration is considered a more sustainable and economical air pollution control technology compared with conventional physical or chemical treatment methods. Biofiltration typically involves a packed bed structure where gaseous pollutants are removed from the airflow. The performance of biofiltration is directly influenced by the activity of the microbial community that inhabits the surface of the packing material. Therefore, maintaining the appropriate conditions for microbes in a packed bed is indispensable for biofiltration operations [1-3].

The moisture content of the packed bed is a key parameter affecting biofiltration performance, which has been highlighted in numerous previous studies. Inadequate moisture content levels, both too low and too high, decrease microbial activity and the contact area between the airflow and the surface of the packing materials, respectively. Every material has an optimal range of moisture content, which is generally recommended to be 30%-60% by weight [4, 5].

Another important factor is the amount of biomass attached to the surface of the packing material. Similar to the influence of moisture content, excessive biomass in the packed bed disturbs the contact between the airflow and the surface of the packing materials, thereby reducing the removal performance of gaseous pollutants in the airflow. Therefore, controlling the biomass during biofiltration is essential, and several methods have been proposed. These methods can be broadly categorized into physical, chemical, and biological methods and modifications of the system design [6, 7].

Chemical treatment is a useful biomass control method; however, it usually requires additional care owing to safety and incurs additional operational costs. The characteristics of the liquid phase in biofiltration affect not only the biomass growth but also the microbial

Corresponding author: Takashi Higuchi, Doctor of engineering, professor, research field: air pollution/malodor control.

flora. For example, acidic conditions dominate the growth of fungi and suppress bacterial growth [8]. Conversely, many chemical substances are indispensable for microorganisms to maintain their activity, with nitrogen being the most important element because of its high consumption rate [9].

Ozone usage is an option for biomass control in biofiltration, and it offers the advantage of being relatively easy to handle compared to chemical methods [10-12]. Ozone generation via UV (Ultraviolet) lamps is a relatively straightforward and cost-effective method compared to other methods, such as electric discharge. Moreover, UV rays can be used for biomass control by reducing microbial activity.

When treating binary or multiple target compounds in a gas stream, biofiltration can lead to complex phenomena, such as competitive metabolism [13, 14]. The primary focus for the application of biofiltration is the influence of the coexistence of compounds with different physicochemical characteristics upon removal.

In line with various previous studies on biomass control in biofiltration processes, we used a UV lamp as the ozone generator in this study. Two typical VOCs (Volatile Organic Compounds), 2-butanone (MEK: Methyl Ethyl Ketone) and toluene, were adopted as the target coexistent gaseous components treated by the biofiltration experiment. These compounds were selected based on their widespread use and their varying levels of water solubility. In addition to ozone, several nutrient supply methods were explored to identify effective biomass control measures.

2. Materials and Methods

2.1 Experimental Setup

The setup of bioreactors (packed-column-type biofilters) for the treatment of VOC-contaminated air closely resembled the methodology used in the authors' previous study [15]. Three reactors (BF1, BF2, and BF3) with the same dimensions were prepared. The diameter and packing height of each reactor were 0.1 m and 0.3 m, respectively. The gaseous VOCs generated from a toluene-MEK liquid mixture using a VOC gas generator were diluted with indoor air and introduced into each reactor at a flow rate of 5 L/min (resulting in a residence time of 28.8 s). A schematic of the experimental setup is shown in Fig. 1. The packing material, made of polyvinyl formal resin (Be-Fine LL prototype, AION Co., Ltd.), was the same as that used in a previous study [15]. To seed the reactors, packing materials containing

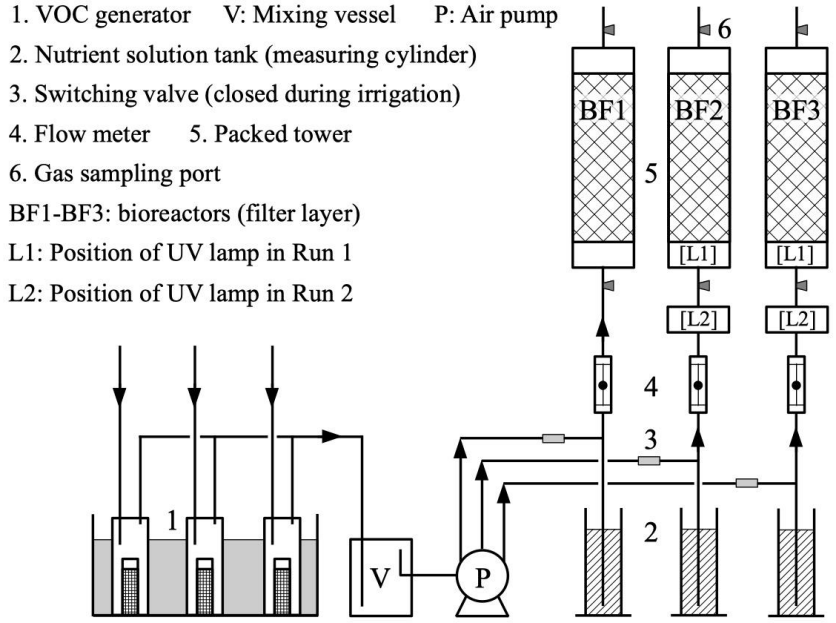

Fig. 1 Experimental setup for bioreactors.

MEK-toluene-degrading microorganisms were mixed with the new materials at a ratio of 1:2 and packed into three reactors.

2.2 Operating Conditions

We conducted two experiments, Runs 1 and 2. The operating conditions of the reactors and the composition of the nutrient solution used for irrigation are listed in Tables 1 and 2, respectively. The nutrient solution from each reactor was circulatively used for irrigation, which was conducted by spraying the solution using a watering pot at approximately the same velocity (approximately 10 mL/s for each reactor on average).

UV lamps were placed at the bottom of the packed columns of BF2 and BF3 in Run 1. The purpose was to assess the effect of UV irradiation on biomass control, in addition to the effect of ozone. In Run 1, an initial volume of 3,000 mL of the nutrient solution was used as the irrigation liquid for each reactor. Subsequently, the volume decreased with each successive irrigation. The solution in all three reactors was replaced with a fresh nutrient solution whenever the liquid volume of one of the reactors fell below 1,000 mL. The frequency of irrigation and concentration of the fresh nutrient solution were adjusted according to the conditions of moisture and biomass growth in the reactors, as shown in Table 3.

UV lamps were positioned along the path of the contaminated gas entering the packed columns of BF2 and BF3 in Run 2. This was specifically done to isolate and utilize the effect of ozone on biomass control. In Run 2, the nutrient solution, outlined in Table 1, was diluted by a factor of five and used for irrigation. The initial liquid volume in each reactor was 2,000 mL. The liquids for BF1 and BF2 were replaced with fresh nutrient solutions every two weeks, whereas the liquid for BF3 was replaced every week. Furthermore, a KNO_3 solution with the concentration and volume specified in Table 4 was added to BF1 and BF2. The total volume of the irrigation liquid was then adjusted to 2,000 mL using distilled water prior to each irrigation.

Table 1 Operating conditions of the bioreactors.

(a) Run 1

		BF1	BF2	BF3
Average room temperature	(°C)		19.8	
Average inlet MEK concentration	(ppm)	57.3	58.4	58.6
Average inlet toluene concentration	(ppm)	50.9	49.6	46.8
Average inlet load of MEK	(g/(m^3 h))	21.9	22.3	22.4
Average inlet load of toluene	(g/(m^3 h))	24.8	24.2	22.9

(b) Run 2

		BF1	BF2	BF3
Average room temperature	(°C)		21.1	
Average inlet MEK concentration	(ppm)	47.4	46.6	46.5
Average inlet toluene concentration	(ppm)	39.8	39.0	38.5
Average inlet load of MEK	(g/(m^3 h))	18.0	17.7	17.7
Average inlet load of toluene	(g/(m^3 h))	19.3	19.0	18.7

Table 2 Composition of the nutrient solution used for irrigation.

Nutrient components	Concentration (mg/L)
KNO_3	4,000
NH_4Cl	2,100
$Na_2HPO_4 \cdot 12H_2O$	7,200
KH_2PO_4	2,700
$MgSO_4 \cdot 7H_2O$	2,100
$CaCl_2 \cdot 2H_2O$	170
$ZnSO_4 \cdot 7H_2O$	8
$Na_2Mo_4 \cdot 2H_2O$	43
$CoCl_2 \cdot 6H_2O$	65
$MnCl_2 \cdot 4H_2O$	670
$FeSO_4 \cdot 7H_2O$	73

Table 3 Conditions of irrigation in Run 1.

Phase	Period	Frequency of irrigation	Dilution of irrigation liquid
Acclimation	Start-Day 21	Once/day	No dilution
Phase 1	Day 22-Day 42	Twice/week	10 times dilution
Phase 2	Day 43-Day 69	Twice/week	No dilution
Phase 3	Day 70-Day 88	Once/day	No dilution

* Composition of irrigation liquid is the same as Table 2 in case of “no dilution”.

Table 4 Concentration and volume of KNO_3 solution added to the irrigation liquid of BF1 and BF2 in Run 2.

Phase	Period	Concentration of KNO_3 aq. (mg/L)	Volume of KNO_3 aq. (mL)
Acclimation	Start-Day 22	2,000	15
Phase 1	Day 23-Day 36	4,000	15
Phase 2	Day 37-Day 64	4,000	45

Table 5 Operating conditions of UV lamps.

Run, Phase	BF2	BF3
Run 1 Acclimation	Turn on the lamp 15 min with one third of full output, before every irrigation	
Run 1 Phase 1-Phase 3	Turn on the lamp 30 min ×3 times/day by regular interval with full output	Turn on the lamp 3 h ×3 times/day by regular interval with full output
Run 2 All phases	Turn on the lamp 3 h ×3 times/day by regular interval with full output	

* UV lamp was not installed in BF1, in both Run 1 and Run 2.

Irrigation was performed four times per week throughout the experiment.

UV lamps (cold cathode fluorescent lamp CCUV-A1, Nippon Shin Kogen, Japan) were used to control the biomass growth in the reactor. The operating conditions are summarized in Table 5. The ozone concentration in the gas stream was only measured in Run 2 because the inlet ozone concentration could not be observed because of the positioning of the UV lamp.

2.3 Measurement Methods

Prior to collecting gas samples, two (one new and one used) and four (two new and two used) pieces of packing materials were selected from the top and middle of the packing layer, respectively. All the selected materials were then weighed.

Gas samples, consisting of a mixture of MEK and toluene, were collected via the sampling ports located at the inlet and outlet of each reactor using a 1.0 mL syringe (Ito micro syringe, Ito Seisakusho Co., Ltd., Japan). Gas samples were collected prior to irrigation.

The concentrations of MEK and toluene in the gas samples were measured using a gas chromatograph (GC-2014, Shimadzu, Japan) equipped with a flame ionization detector. The measurement conditions were identical to those described in our previous study [15].

The nitrate concentration in the nutrient solution used for irrigation was measured using an ion chromatography system (HIC-20A, Shimadzu, Japan) equipped with a conductivity detector (CDD-10Asp, Shimadzu, Japan) and an anion-exchange resin column (Shim-pak IC-SA4, Shimadzu, Japan). The designated measurement conditions (e.g., eluent and oven temperature) for this column were used for the ion chromatography measurement.

The ozone concentration of the gas stream, pH of the liquids, and the ambient temperature were measured using an ozone gas monitor (OZG-EB-01, Applics, Japan), a pH electrode (F-21, Horiba, Japan), and a temperature logger (LR5001, Hioki, Japan), respectively.

3. Results

3.1 Run 1 (Direct UV Lamp Irradiation)

MEK was removed to a greater extent than toluene throughout the experiment. The average ± standard deviation removal efficiencies of MEK in BF1 (without UV lamp), BF2 (low lamp irradiation), and BF3 (high lamp irradiation) were 84.6% ±16.5%, 88.1% ±14.0%, and 84.1% ± 19.4%, respectively. The change in toluene removal efficiency is shown in Fig. 2.

Toluene removal was not only lower than MEK removal but also more unstable. Following the acclimation phase, the toluene removal efficiency gradually decreased over time. In the next phase (Phase 2), a nutrient solution with a ten-fold higher concentration was used for irrigation, and the efficiency gradually recovered.

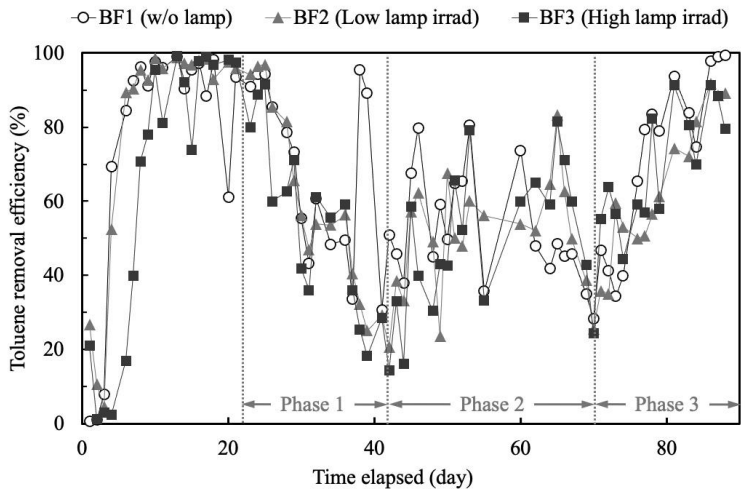

Fig. 2 Change of toluene removal efficiency in Run 1.

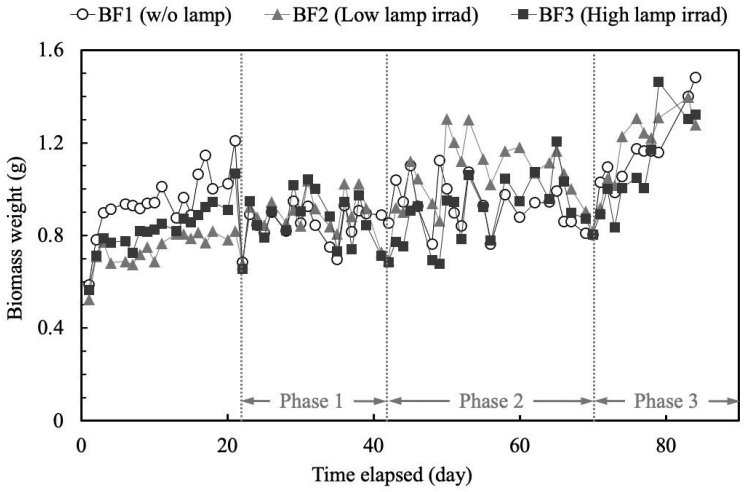

Fig. 3 Change of biomass weight in the packing materials in Run 1.

The frequency of irrigation was increased in the following phase (Phase 3) to improve performance, and the efficiency of toluene increased steadily. However, as depicted in Fig. 3, the biomass in the entire packed column increased, leading to evident clogging that necessitated the termination of the experiment. The average ± standard deviation removal efficiencies of toluene in BF1, BF2, and BF3 were 63.0% ± 21.9%, 57.3% ± 20.5%, and 56.4% ± 21.1%, respectively.

The nitrate concentration and pH trends in the nutrient solution were similar among the three reactors throughout Run 1. Nitrate ions were nearly completely consumed by the end of Phase 1, whereas a certain level of nitrate concentration was maintained in the nutrient solution during Phase 2. Owing to irregular measurements, the precise trend of the nitrate concentration until the end of Phase 2 could not be clarified. In Phase 3, the constant consumption of nitrate was observed by measuring at regular intervals. The daily decrease in nitrate concentration in the nutrient solution was approximately 20 mg N/L. An almost neutral pH was maintained, although a slight increase in pH was observed during Phase 3. The pH range was 6.7-7.9.

3.2 Run 2 (Indirect UV Lamp Irradiation)

In Run 2, the removal of MEK was even higher and more consistent compared to Run 1. The removal efficiencies in BF1 (without UV lamp and dense nitrate

liquid added), BF2 (with UV lamp irradiation and dense nitrate liquid added), and BF3 (with UV lamp irradiation without dense nitrate liquid addition) were 95.2 ±3.5%, 95.7 ±3.0%, and 97.3 ±1.6%, respectively. Fig. 4 demonstrates that toluene removal in Run 2 was much more stable than that in Run 1. The toluene removal efficiencies in BF1, BF2, and BF3 were 53.1% ±14.7%, 52.9% ±9.7%, and 68.8% ±8.6%, respectively. BF3 achieved the highest efficiency among all the experiments in this study. The biomass weight in Run 2, as depicted in Fig. 5, was also kept stable.

During Phase 1 of this run, no evident change in the nitrate concentration was detected in the irrigation liquid of BF1 and BF2 prior to the addition of the dense nitrate solution. In Phase 2, a decrease in the nitrate concentration in the nutrient solution was observed in all reactors. Table 6 shows a comparison of nitrate reduction among the three reactors. The values gradually decreased over time in BF1 and BF2. This indicates that the nitrate concentration reached a steady state, in which the supplied nitrogen added as a dense nitrate solution was consumed within a specific interval following its addition. All reactors consistently maintained a pH range close to neutral (6.6-7.9) throughout Run 2.

The inlet and outlet ozone concentrations were 28.8 ±9.1 ppm and 10.7 ±6.5 ppm in BF2 as well as 32.9 ± 6.8 ppm and 4.7 ±1.7 ppm in BF3, respectively.

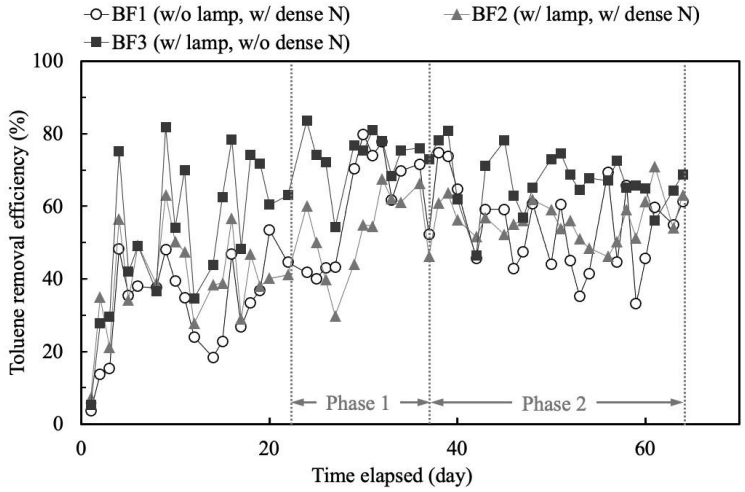

Fig. 4 Change of toluene removal efficiency in Run 2.

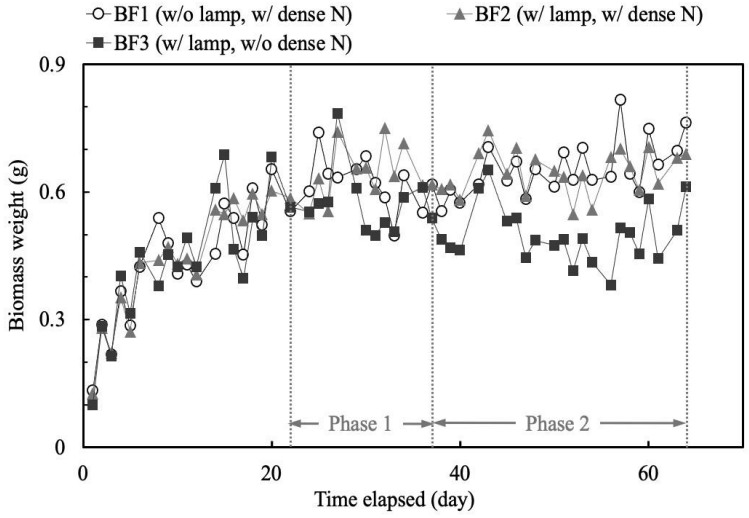

Fig. 5 Change of biomass weight in the packing materials in Run 2.

Table 6 Decreasing velocity of nitrate-nitrogen concentration of the irrigation liquid in Run 2.

Period	Decreasing velocity of nitrate-nitrogen concentration (mg/(L day))		
	BF1 w/o lamp w/ dense N	BF2 w/ lamp w/ dense N	BF3 w/ lamp w/o dense N
Earlier week of phase 2 Day 43-Day 50	18.3	22.6	19.0
Last week of phase 2 Day 57-Day 64	9.8	14.8	17.1

4. Discussion

In both Runs 1 and 2, UV lamp usage had minimal influence on MEK and toluene removal efficiency or biomass reduction. In Run 1, the biomass weight in the packed layer was consistent until the end of Phase 2. This was observed not only in BF2 and BF3 (where the lamp was operational) but also in BF1 (where the lamp was not operational). However, in Phase 3, a sudden increase in biomass occurred because more frequent irrigation was conducted. In Run 2, the biomass was well-controlled throughout the operation in all reactors, but there was no difference in biomass weight between BF1 (where the lamp was not operational) and BF2 (where the lamp was operational). Moreover, the UV lamp generated an unknown, highly viscous by-product, possibly resulting from the reaction of ozone or UV with MEK and toluene, which polluted the reactors and their gas piping. These results indicate that the use of this UV lamp is impractical for biomass control during biofiltration.

However, reducing the nutrient supply appears to be effective for biomass control. The biomass weight remained lower in Run 2 than in Run 1, primarily because of the use of a lower concentration of the nutrient solution. In Run 1, the biomass weight did not increase significantly until Phase 2 because of the limited frequency of nutrient supply. In Run 2, nitrate was supplied using a dense nitrate solution to precisely control the amount of nitrate supplied without changing the irrigation volume. In Phase 3 of Run 1, approximately 60 mg-N/day of nitrate-nitrogen consumption (= 20 mg-N/(L day) of nitrate concentration decrease) resulted in excess growth of biomass in the packed layer of the reactors. Therefore, in Run 2, the concentration and volume of dense nitrate solution for additional irrigation were set as shown in Table 4 (34 and 103 mg-N/day in Phases 1 and 2, respectively). Although the addition of nitrate in Phase 2 of Run 2 exceeded the consumption of nitrate in Phase 3 of Run 1, no excess biomass growth was observed in Run 2. This comparison indicates that biomass growth is not solely accelerated by the addition of nutrients such as nitrate.

In Phase 1 of Run 1, the toluene removal efficiency decreased because of the lack of nutrients. However, the efficiency gradually recovered by increasing the concentration of the nutrient solution ten-fold. However, the nutrient supply remained insufficient in Phase 2 of Run 1 because the toluene removal efficiency increased further in Phase 3, during which a higher frequency of irrigation was conducted. In brief, nitrogen supply, namely nitrate, is indispensable for toluene gas removal. However, excess biomass growth occurs when excess irrigation and excess nutrient supply are conducted at the same time. The most notable results were obtained with BF3 in Run 2. This reactor was operated with a diluted nutrient solution, which was frequently replaced to maintain a sufficient nutrient supply. Frequent replacement of the irrigation liquid removes dissolved or suspended microbially derived by-products from the reactor. Consequently, this operation method is likely the most effective in preventing the accumulation of excess biomass. As shown in Table 6, the most stable nitrate consumption was observed for BF3 in Phase 2 of Run 2, with a value of approximately 23 g N/(m^3 day) (18 mg-N/(L day) nitrate concentration decrease = 54 mg-N/day nitrate consumption in this reactor). Because the same amount

of ammonium nitrogen was also supplied in this reactor and approximately 100% of MEK removal would have required a similar amount of nitrogen, this nitrate consumption value was not inconsistent with that in a previous study [16].

5. Conclusions

An attempt was made to control the excess biomass in a biofiltration system designed for the treatment of a gas stream contaminated with MEK and toluene. Direct and indirect UV lamp irradiations were performed on the packed layer, where biomass growth occurs, at the target gas treatment position. However, neither method showed any effect on biomass control. Decreasing the nutrient supply suppressed biomass growth, and the results indicated that the simultaneous existence of excess moisture and nutrients in the packed layer accelerated needless biomass growth. Relatively low but steady nitrate consumption was observed when the biomass was well-controlled and the target gases were extensively treated. More information is required to estimate the ideal amount of nutrient supply for different biofiltration conditions. However, employing a small amount of dense nutrient solution is effective for an accurate and optimal nutrient supply through an appropriate volume of irrigation.

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