0273-2289/06/129–132/1077–1088/$30.00

Application of Two-Stage Biofilter System for the Removal of Odorous Compounds

Gwi-Taek Jeong,[1,2] Don-Hee Park,*[,2–6] Gwang-Yeon Lee,[7] and Jin-Myeong Cha[8]

[1]Engineering Research Institute, [2]School of Biological Sciences and Technology, [3]Faculty of Applied Chemical Engineering, [4]Biotechnology Research Institute, [5]Research Institute for Catalysis, [6]Institute of Bioindustrial Technology, Chonnam National University, Gwangju, 500-757, Korea, E-mail: dhpark@chonnam.ac.kr; [7]Department of Ophthalmic Optics, Dong-A College, Jeonnam, 526-872; and [8]B & E Tech Co., Ltd., Jeonnam, 519-831, Korea

Abstract

Biofiltration is a biological process which is considered to be one of the more successful examples of biotechnological applications to environmental engineering, and is most commonly used in the removal of odoriferous compounds. In this study, we have attempted to assess the efficiency with which both single and complex odoriferous compounds could be removed, using one- or two-stage biofiltration systems. The tested single odor gases, limonene, α-pinene, and iso-butyl alcohol, were separately evaluated in the biofilters. Both limonene and α-pinene were removed by 90% or more *EC* (elimination capacity), 364 $g/m^3/h$ and 321 $g/m^3/h$, respectively, at an input concentration of 50 ppm and a retention time of 30 s. The iso-butyl alcohol was maintained with an effective removal yield of more than 90% (*EC* 375 $g/m^3/h$) at an input concentration of 100 ppm. The complex gas removal scheme was applied with a 200 ppm inlet concentration of ethanol, 70 ppm of acetaldehyde, and 70 ppm of toluene with residence time of 45 s in a one- or two-stage biofiltration system. The removal yield of toluene was determined to be lower than that of the other gases in the one-stage biofilter. Otherwise, the complex gases were sufficiently eliminated by the two-stage biofiltration system.

Index Entries: Biofilm; biofilter; complex odor; two-stages; VOCs.

Introduction

Emissions of volatile organic compounds (VOCs) can be controlled using a variety of chemical, physical, or biological technologies, including incineration, adsorption, chemical scrubbing, bioscrubbing, and biofiltration *(1)*. Within the two last decades, biological treatment protocols

*Author to whom all correspondence and reprint requests should be addressed.

including bioscrubbers, trickling beds, and biofilters have been employed successfully in the control of both VOCs and odors (2).

Biofiltration is a biological process which is considered to be one of the more successful examples of biotechnological techniques applied to environmental engineering, and has been most commonly employed in the removal of odoriferous compounds and VOCs. This biological process has been shown to be competitive in applications which involve the treatment of large volumes of air that contain low concentrations of odors. These processes tend to be associated with low operating costs, and are quite effective in the treatment of large volumes of moist air containing low concentrations of biodegradable compounds. One advantage of such biological processes is that they are typically of lower cost than other processes, including incineration, absorption, adsorption, and condensation. In general, biological processes tend to be both ecologically and economically desirable, particularly regarding off-gases at concentrations of up to 1–5 g/m^3 (1–3).

The biofiltration process utilizes microorganisms, which have been attached to porous support media, to breakdown both VOCs and odoriferous compounds. It has also been found to have potential applications in the control of emissions from industrial plants (3). A biofilter typically consists of a container filled with packing material and populated with microbes, through which the odor-containing air is passed, normally in an upward direction (4).

The contaminants are transferred from the air to the biofilm, in which they are subsequently biodegraded into carbon dioxide and water. Odor and VOCs are similarly transferred from the air into a biofilm (bio-active layer) which surrounds the organic or inorganic packing material in the biofilter. The odorous gases are then degraded into a variety of end products, or subsumed into the biomass. The end products appear to depend on the nature of the odors (1,2,4).

The packing material in the biofilter also performs as a carrier for the microbes, nutrients, and water. The packing material must possess a number of characteristics in order to ensure high deodorizing performance, such as, large surface area for gas contact, high levels of microbe immobilization, high water retention capacity, and easy removal of deodorization wastes. Also, the carrier is required to be extremely durable, with no clogging/blocking and a low pressure drop in the packed bed during its operation. Currently, a variety of carriers which fulfill these criteria are actually in use (2,5–7).

The air which is introduced into the biofilter may be prehumidified in order to maintain adequate moisture in the biofilm. Alternatively, or in addition, water may be sprinkled over the biofilm's surface and allowed to trickle downwards, counter to the flow of odorous air. This water must then contain nutrients required for the growth of the microbes (4).

Biofiltration has demonstrated, in many studies, an ability to remove alcohols, toluene, phenol, ketones, petroleum fuel vapors, and a variety

of other VOCs *(2,7,8)*. Most recently, simultaneous treatments of single or complex VOCs have been demonstrated using biofilters *(2,9)*. Many industrial air emissions have been shown to contain VOC mixtures, which exhibit different physical and chemical characteristics which affect their biological treatment. In particular, the effectiveness of biofiltration in the treatment of VOC mixtures is highly dependent on the solubility of the compounds within the liquid layer of the biofilm. For example, methanol, a VOC which is both hydrophilic and readily biodegradable, could be expected to be removed effectively via biofiltration. However, α-pinene removal by biofiltration is more difficult owing to its extreme hydrophobicity, which results in low diffusivity through the biofilm *(3)*. The operating conditions of a given biofilter, including temperature, pH, nutrient concentration, water contents, and relative humidity in the air, are the most relevant factors affecting the removal capacity of a biofilter *(2,7–10)*.

The objective of this research, then, was to determine the effectiveness of biofilter packed with a reticulated polyurethane foam carrier to remove hydrophilic and hydrophobic components from VOC mixtures.

Materials and Methods

Microbes, Nutrient, and Packing Material

All of the biofilters were inoculated with a microbial consortium, primarily composed of a variety of microbes able to degrade ethanol, acetaldehyde, iso-butyl alcohol, limonene, α-pinene, and toluene, developed by enrichment of sludge isolated from compost at composting facilities in Seosan, Chungnam, Korea. The nutrient solution provided for the growth and maintenance of the microbes in the biofilter was a mineral medium consisting of basic mineral salt supplemented with: 1.5 g/L KH_2PO_4, 6 g/L Na_2HPO_4, 3 g/L $(NH_4)_2SO_4$, 0.05 g/L $MgSO_4$, and 0.01 g/L $CaCl_2$. In order to prepare the biofilter packing material, reticulated polyurethane foam (RPF; Yoowonurethane Co., Ltd., Korea) was cut to pieces of $1.9 \times 1\,9 \times 1.9$ cm^3 in size, and these were used in all experiments. The characteristics of the packing material are summarized in Table 1.

Biofilter Experiments

The biofilter (Fig. 1) was constructed from a transparent acrylic tube with an inner diameter of 18 cm. This was then divided into three 80 cm sections, and upper two sections were filled to a volume of 3.1 L (packing ratio 30.5%), with equal amounts of the prepared filter-bed materials (Table 1).

In order to characterize the removal characteristics and yields at several different loading concentrations of limonene, α-pinene, and iso-butyl alcohol, gases containing each of these components were separately introduced to the biofilter. The biofiltration of single gases under continuous flow conditions was conducted at a variety of loading concentrations of limonene, α-pinene,

Table 1
Used Carrier and Characteristics

Bed material	Reticulated polyurethane foam (RPF)
Surface area (m^2/g)	367.5
Absorptance ($g \cdot H_2O/g$)	57
Porosity (%)	80
Bulk volume (L)	10.2 L/reactor
Bed volume (L)	3.1 L/reactor
Packing density (g/cm^3)	0.0195
Packing ratio (%)	30.5

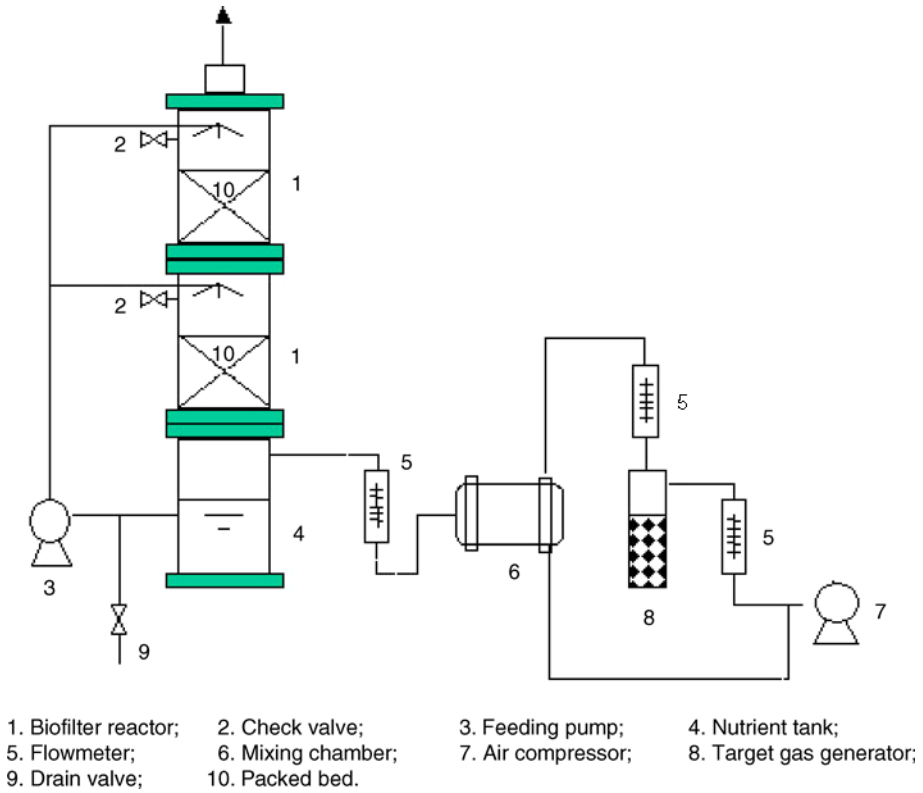

Fig. 1. Scheme of two-stage biofilter for odor removal.

and iso-butyl alcohol, for a period of 1 mo. The gas samples were obtained from both the inlet and outlet streams, and also taken axially along the biofilter. The input gases, limonene, α-pinene, and iso-butyl alcohol in air, were prepared via the evaporation of liquid solutions with air flow, and the input concentrations were adjusted by controlling of flow rates of the prepared gases and air. In order to control the humidity of the input gas, air

was passed though a humidifier before being mixed with the prepared gas, and then the mixed gas was introduced into the bottom portion of the biofilter. The biofilter was inoculated with a microbial consortium consisting of an enriched version of that reported previously *(11)*. In order to feed the microorganisms, four times concentrated BSM nutrient solution was periodically fed into the biofilters via peristaltic pump. Water and nutrient were sprayed from the upper nozzles of each section of the biofilter in order to protect the biofilter media from drying. The operation temperature was set to 30°C, and pH levels were not controlled.

In order to determine the process characteristics and removal yields at a variety of loading concentrations of mixtures of ethanol, acetaldehyde, and toluene, the mixed gases were introduced to the bottom portion of the biofilter at a variety of loading concentrations, under continuous flow conditions for a period of 1 mo. In this study, we applied a two-stage biofilter and compared the results to those obtained using the one-stage biofilter. As for the single VOC gases, the introduced gas mixture of ethanol, acetaldehyde, and toluene was prepared via the evaporation of a liquid solution of this mixture by flowing air, and the input concentrations were adjusted by controlling of flow rates of the prepared gas mixture and air. Experiments concerning the removal of the gas mixtures were conducted as described earlier. The applied ranges of VOC concentrations in the ethanol, acetaldehyde, and toluene mixture were 50–200 ppm, 20–70 ppm, and 10–70 ppm, respectively.

Analytical Method

Both the VOCs and the odoriferous gases were collected at both the inlet and outlet of the biofilter, with a 10 mL Hamiltion gas tight syringe, and then dissolved in 15-mL sealed (septum) glass tubes containing 1 mL of methylene chloride. Samples were then extracted from the methylene chloride solution with a syringe, and 2 μL of each sample was injected onto a gas chromatograph. Different concentrations of VOCs and odoriferous gases were measured with a gas chromatograph (GC-14A, Shimadzu, Japan) which was equipped with a FID. We used a DB-WAX column (30 m × 0.53 mm × 1 mm, J&W Scientific, Folsom, CA) in this phase of the experiment. The GC oven temperature was set to 50°C, and detector temperature was 200°C. Helium was used as the carrier gas, and the column flow rate was set to 8 mL/min. The air flow rate was 140 mL/min, and the hydrogen flow rate was set at 40 mL/min. A portable VOC detector (Multi gas monitor PGM-50, RAE Systems Inc., CA) was also used.

Removal Yield and Elimination Capacity

Removal yield (RY) and elimination capacity (EC) were the variables used to determine the treatment capacity of the biofilter. Removal yield was expressed as the content (%) of odoriferous gas eliminated by the biofilter.

Elimination capacity was expressed as the amount of odoriferous gas removed in the bed volume/unit time.

$$RY\ [\%] = (C_{Gi} - C_{Go})/C_{Gi} \times 100$$

$$EC\ [g/m^3 \cdot hr] = (C_{Gi} - C_{Go}) \times Q/V_f$$

where Q is the gas flow rate (m^3/h), V_f is the volume of the filter bed (m^3) and C_{Gi} and C_{Go} are the inlet and outlet odoriferous gas concentration (ppm; g/m^3).

Results and Discussion

Removal of Single Odorous Compounds

A large quantity of ethanol, aldehydes, and various concentrations of aromatic compounds are emitted from recycling facilities *(11)*. We performed the elimination of limonene, α-pinene, and iso-butyl alcohol separately, in a one-stage biofilter. For the initial adaptation of the microorganisms, we introduced one of the tested single odor gases into the biofilter under low concentration conditions, a maximum concentration of 20 ppm, using a 2 min residence time. After which the outlet concentration was maintained at 5 ppm, the input concentration was increased in a stepwise manner using a 30 s residence time.

The removal characteristics and removal yields of the tested odor gases, limonene, α-pinene, and iso-butyl alcohol, were separately observed in the biofilter (Figs. 2–4). More than 90% (*EC* 364 $g/m^3/h$) of the limonene was removed at a 50 ppm input concentration and a 30 s residence time, but we noted an 85% (*EC* 678 $g/m^3/h$) removal yield at a 100 ppm input concentration and a 30 s residence time (Fig. 2). Nearly 90% (*EC* 321 $g/m^3/h$) of the α-pinene was eliminated at a 50 ppm input concentration and a 30 s residence time, but the removal yield dropped to approx 82% (*EC* 631 $g/m^3/h$) at a 100 ppm input concentration and a 30 s residence time (Fig. 3). However, the biofiltration protocol in this experiment maintained an effective iso-butyl alcohol removal yield of above 90% (*EC* 375 $g/m^3/h$ at 100 ppm), in an input concentration range between 20 and 150 ppm, after the adaptation period (Fig. 4). In general, the capacity of the biofiltration treatment for VOCs is highly dependent on the solubility of the odorous compounds within the liquid layer of the biofilm. α-Pinene is extremely hydrophobic (with a maximum water solubility of less than 5–10 mg/L), which appears to result in a low solubility in the biofilm of the biofilter *(3)*.

Removal of Complex Odorous Compounds

The effectiveness with which a given biofiltration protocol can effect the removal of VOCs appears to be principally dependent on the solubility of the compounds in the biofilm of the biofilter. Hence, the hydrophilic

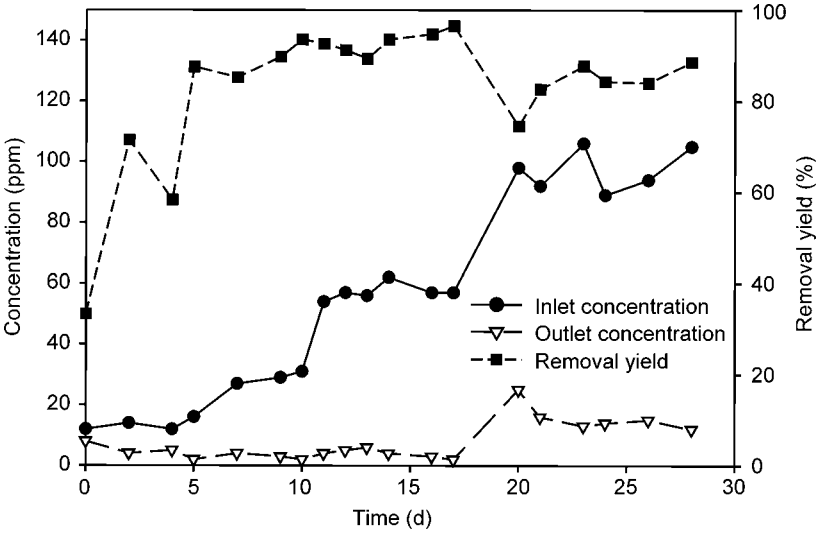

Fig. 2. Removal characteristics of limonene in biofilter.

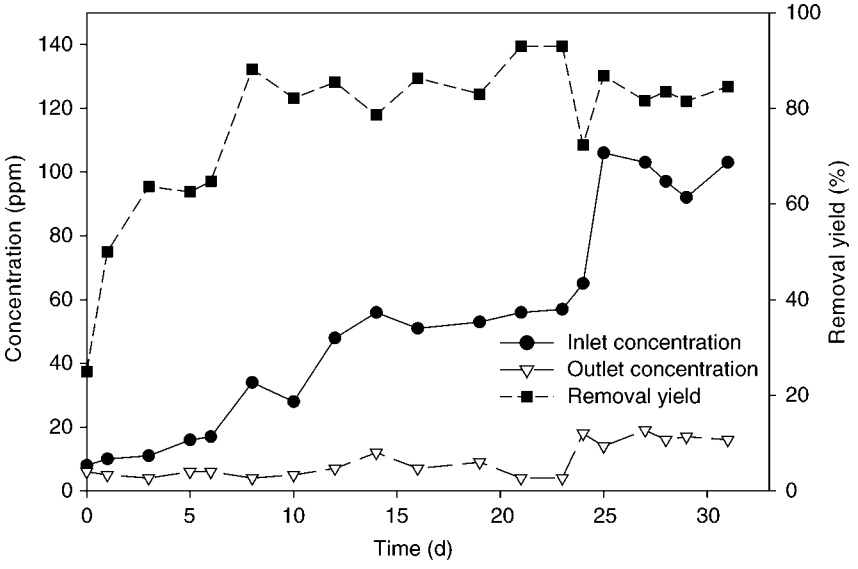

Fig. 3. Removal characteristics of α-pinene in biofilter.

and hydrophobic characteristics of a given pollutant is thought to significantly influence the capacity of a biofilter setup to remove it *(3)*. Mohseni and Allen *(3)* reported that, when hydrophilic and hydrophobic materials were treated simultaneously, the microorganisms involved in the treatment of the hydrophilic materials exhibited overgrowth in the biofilter, and this created an obstacle to the treatment of the hydrophobic materials. Methanol, a VOC which is both hydrophilic and readily biodegradable, is expected to be easily removable via biofiltration. α-Pinene, on the other

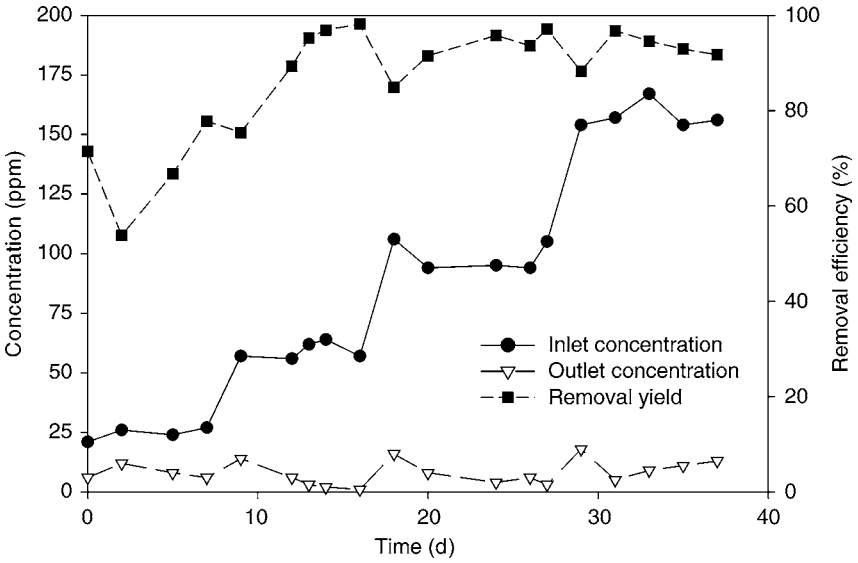

Fig. 4. Removal characteristics of iso-butyl alcohol in biofilter.

hand, is extremely hydrophobic, which may result in it being removed at a lower rate.

Therefore, in this work, we attempted the removal of hydrophilic compounds with a one-stage biofilter, and tried to remove hydrophobic compounds with a two-stage biofilter. We employed ethanol and acetaldehyde as hydrophilic compounds, and used toluene as the model hydrophobic compound. The mixture gas was then applied with the following inlet concentrations, ethanol 200 ppm, acetaldehyde 70 ppm, and toluene 70 ppm, with a residence time of 45 s. This part of the procedure was the same in the single- and double-stage biofiltration systems. Two identical 1-stage bench-scale biofilters (Reactor I and II) were operated in parallel, in order to characterize the influence of step loads on the system's efficacy for removing toluene, a hydrophobic VOC, and ethanol and acetaldehyde, both hydrophilic VOCs. The ethanol/acetaldehyde and toluene were introduced and stabilized in Reactors I and II, respectively. In order to stabilize the biofilter, 20 ppm of ethanol, and 10 ppm of acetaldehyde were introduced into the system (residence time of 1 min). The toluene input concentration was controlled at 10 ppm and stabilized (residence time of 2 min). After completion of the adaptation period, Reactors I and II were connected in order to simultaneously remove the three odoriferous gases in a combined two-stage biofilter. The ethanol, acetaldehyde, and toluene were then applied to the bottom portion of Reactor I, then passed through Reactor II.

The process characteristics and removal yields of the odoriferous gas mixture in the two-stage biofilter are shown in Fig. 5. The removal yields of the ethanol and acetaldehyde were maintained at over 95% throughout

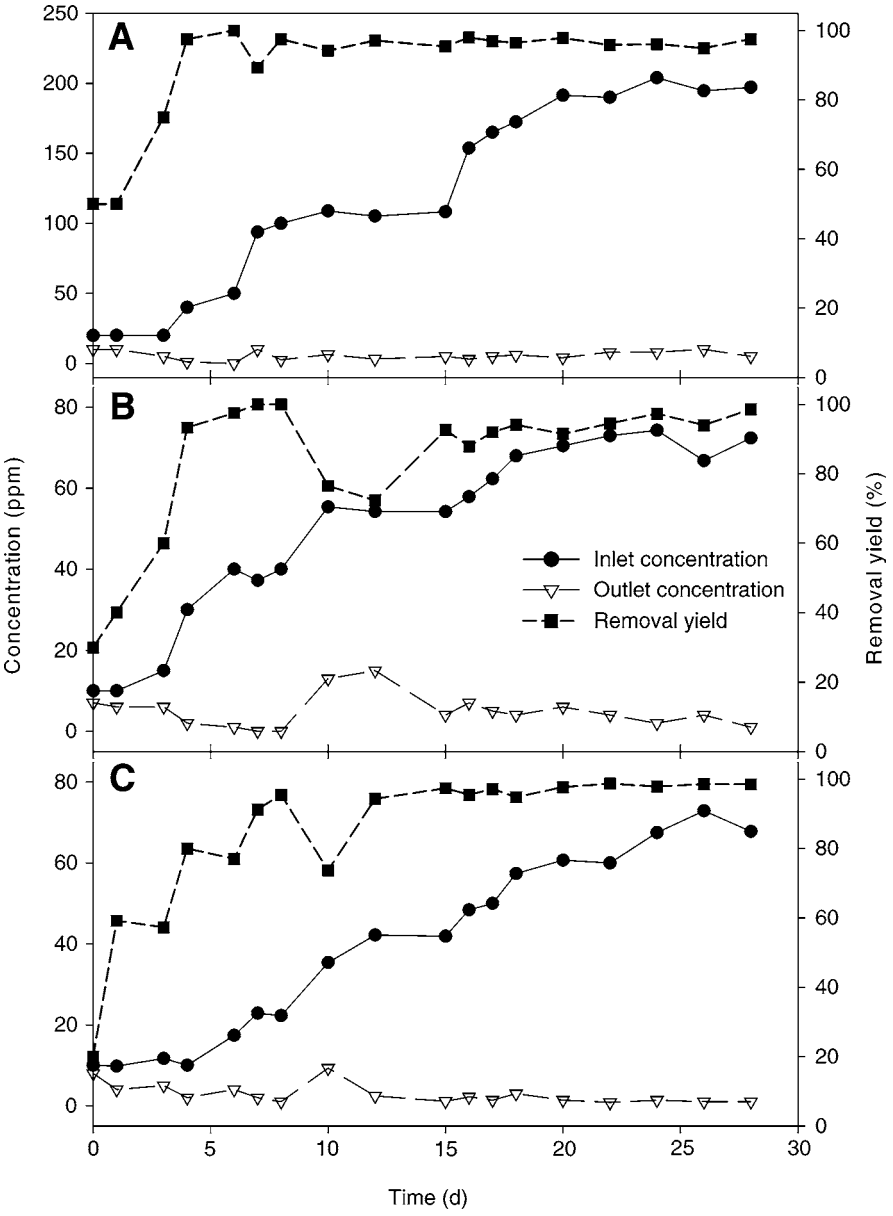

Fig. 5. Removal characteristics of ethanol, acetaldehyde and toluene in two-stage biofilter. **(A)** ethanol, **(B)** acetaldehyde, **(C)** toluene.

the 6 d of processing in reactor 1 (not shown), and after that period, the input concentration was increased in a stepwise manner using a 15 s residence time. When we had confirmed that the removal yields of the three odor gases were maintained at over 95% for 14 d, Reactor I and II were vertically connected, and then the mixed gas was supplied to Reactor II, after being passed through Reactor I. The input concentration was then increased in

a stepwise manner using a 30 s residence time. After the two biofilters had been connected, ethanol and acetaldehyde had removal yields over 97% (EC 240 $g/m^3/h$) at 100 ppm and 92–98% (*EC* 165 $g/m^3/h$) at 70 ppm, respectively, (Fig. 5A,B). Also, the toluene removal yield was maintained at between 95 and 98% (*EC* 239 $g/m^3/h$) at 50 ppm (Fig. 5C).

In order to determine the process characteristics and removal yields in the one-stage biofilter, mixed odor gas (ethanol, acetaldehyde, and toluene) was applied directly to Reactor II, using initial input concentrations of 20, 10, and 10 ppm, respectively. During the adaptation period to stabilize the biofilter, the retention time was set at 3 min. After 8 d, the input concentrations and retention time (30 s) began to be controlled at different levels in order to determine the characteristics of the removal of the mixed odor gases. Figure 6 shows the performance characteristics and the removal yields of mixed odor gas obtained using the one-stage biofilter. After the completion of the adaptation period, the removal yield of ethanol was maintained at over 97% (*EC* 481 $g/m^3/h$ at 100 ppm) (Fig. 6A). The 1-stage biofilter also removed approx 94% of the acetaldehyde (311 $g/m^3/h$ at 70 ppm) and approx 70–80% of the toluene (EC 388 $g/m^3/h$ at 50 ppm) (Fig. 6B,C).

When comparing Figs. 5 and 6, the removal profiles of ethanol and acetaldehyde exhibited similar patterns in the one- and two-stage biofilters. However, the toluene removal was generally poorer in the one-stage biofilter with the toluene removal yield determined to be approx 70–80% in the one-stage biofilter. However, the gas mixture was properly eliminated by the two-stage biofilter, with the toluene removal yield more than 95% using the two-stage biofiltration treatment.

Our results may be attributable to the fact that, in the continuous treatment of complex odors cause by hydrophilic and hydrophobic materials, the microorganisms which feed on hydrophilic materials exhibit overgrowth in one-stage biofilters, as reported previously (3). This can attenuate the efficacy inherent to the treatment of hydrophobic materials in such a system. Otherwise, we found that the hydrophilic and hydrophobic compounds could effectively be removed by a two-stage biofilter system.

Conclusions

The application of one- or two-stage biofilter systems was conducted in order to determine the efficiencies with which single or complex odoriferous compounds could be removed when reticulated polyurethane foam was used as a packing material for the biofilter. Firstly, we separately determined the removal characteristics and yields of the single odor gases, limonene, α-pinene, and iso-butyl alcohol, in a one-stage biofilter. More than 90% of the limonene and α-pinene were removed at a 50 ppm input concentration and a 30 s residence time, but removal yields dropped to approx 80% when the input concentration was increased to 100 ppm.

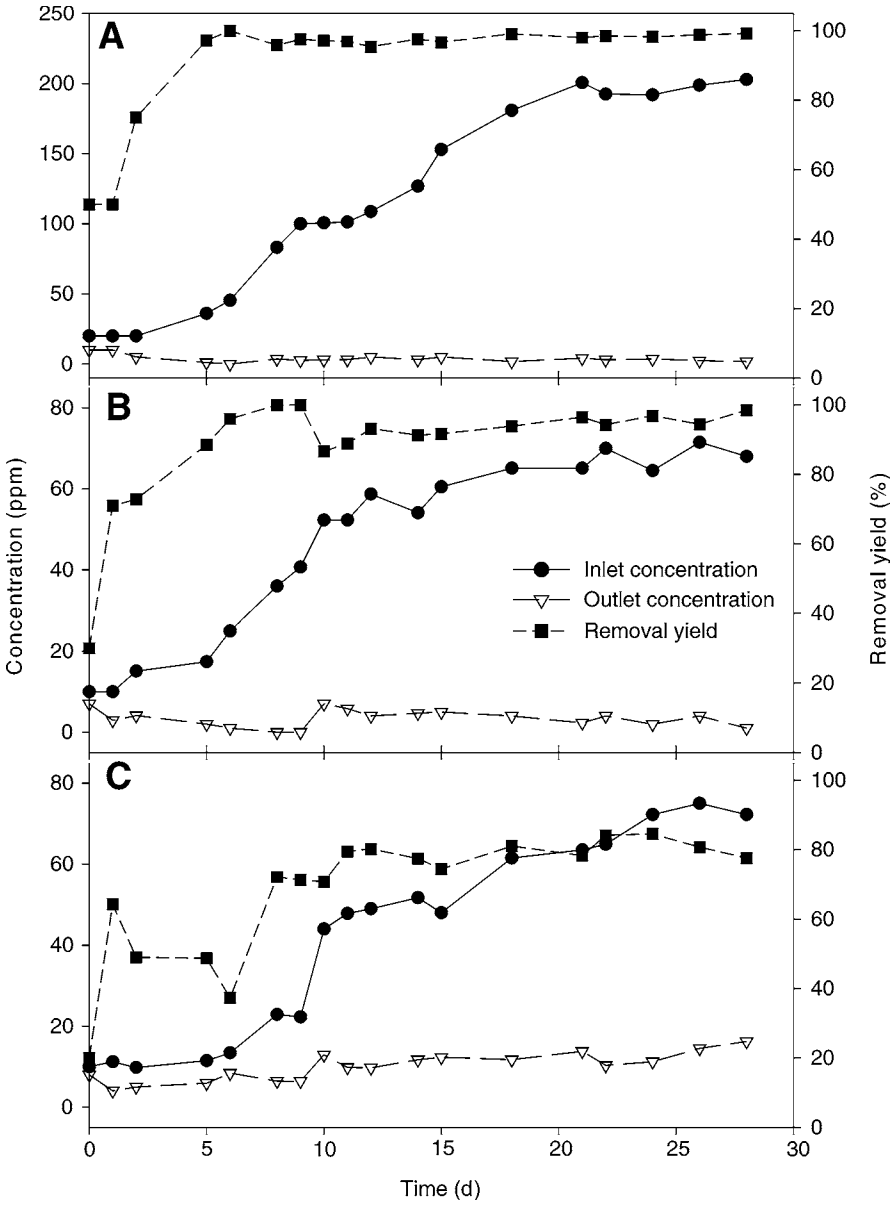

Fig. 6. Removal characteristics of ethanol, acetaldehyde and toluene in one-stage biofilter. **(A)** ethanol, **(B)** acetaldehyde, **(C)** toluene.

An effective iso-butyl alcohol removal yield above 90% was maintained at a 100 ppm input concentration. Secondly, we attempted to eliminate complex odoriferous gases with a two-stage filtration scheme using a residence time of 45 s and an ethanol inlet concentration of 200 ppm, an acetaldehyde inlet concentration of 70 ppm, and a toluene inlet concentration of 70 ppm, in both one- and two-stage biofiltration systems. Toluene removal yields were

determined to be significantly lower than those of other gases in the one-stage biofilter, but not in the two-stage system. Mixture experiments showed that complex gases could be properly eliminated using a two-stage biofilter.

Acknowledgment

This study was financially supported by the Program for the Training of Graduate Students in Regional Innovation, which was conducted by the Ministry of Commerce, Industry, and Energy, of the Korean Government.

References

1 Ruokojärvi, A., Ruuskanen, J., and Martikainen, P. J. (1995), *J. Air Waste Manage. Assoc.* **51(1),** 11–16.
2 Auria, R., Aycaguer, A. C., and Devinny, J. S. (1998), *J. Air Waste Manage. Assoc.* **48,** 65–70.
3 Mohseni, M. and Allen, D. G. (1999), *J. Air Waste Manage. Assoc.* **49(12),** 1434–1441.
4 McNevin, D. and Barford, J. (2000), *Biochem. Eng. J.* **5(3),** 231–242.
5 Shinabe, K., Oketani, S., Ochi, T., et al. (2000), *Biochem. Eng. J.* **5(3),** 209–217.
6 Park, D. H., Cha, J. M., et al. (2002), *Biochem. Eng. J.* **11,** 167–173.
7 Morales, M., Perez, F., Auria, R., and Revah, S. (1994), in *Advances in Bioprocess Engineering* (Galindo, E. and Ramirez, O. T., eds.), Kluwer Academic, Dordrecht, The Netherlands, pp. 405–411.
8 Shareefdeen, Z., Baltzis, B., Oh, Y. -S., and Bartha, R. (1993), *Biotech. Bioeng.* **41,** 512–524.
9 Deshusses, M. A., Hamer, G., and Dunn, I. J. (1995), *Environ. Sci. Technol.* **29,** 1059–1068.
10 Williams, T. O. and Miller, F. C. (1992), *Biocycle Magazine* **33(10),** 72–77.
11 Lee, G. Y. (2004), PhD Thesis, Chonnam National University, Gwangju, Korea.