Behavior of Trickle-Bed Air Biofilter for Toluene Removal: Effect of Non-Use Periods

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Published online 5 April 2005 in Wiley InterScience (www.interscience.wiley.com). DOI 10.1002/ep.10079

A laboratory-scale trickle bed air biofilter (TBAB) packed with synthetic, inorganic media for microbial attachment was used for toluene removal from an air stream. Three experimental strategies—including backwashing as the active biomass control and two non-use periods (starvation, which represents a period without toluene loading, and stagnant, which reflects no flow)—were considered in this study. The non-use periods can be considered as another means of biomass control for low VOC loadings (0.70 and 1.41 kg COD/m^3·day), whereas at high VOC loading (>3.52 kg COD/m^3·day) backwashing is deemed essential to remove the excess biomass accumulation in the biofilter. As the toluene loading rate was increased, a considerably longer reacclimation period was unavoidable for biofilter performance to reach the 99% removal for all experimental strategies (backwashing, starvation, and stagnant). Because the biofilter response was strongly dependent on the active biomass in the system, biofilter response after the non-use periods was significantly different from that after backwashing. © 2005 American Institute of Chemical Engineers Environ Prog, 24: 155–161, 2005

Keywords: biotrickling filter, trickle-bed air biofilter, toluene, non-use period

INTRODUCTION

Biofiltration is rapidly becoming a promising air pollution control technology for removing volatile organic compounds (VOCs) present in waste air stream, such as gaseous emissions from chemical process. In a biofiltration process, waste gases containing biodegradable VOCs or inorganic air toxics are degraded to environmentally benign end products by passage through a biologically active medium. In response to the growing concern over VOCs and because of its cost effectiveness, biofilters have been the subject of extensive research since the 1980s.

A trickle-bed air biofilter (TBAB), which allows consistent nutrient and pH control for optimizing the waste-using kinetics for micro-organisms, is conceptually an identical process to the biofilter, discussed in detail elsewhere [1–3]. The TBAB consists of a reactor containing microbes supported on the synthetic medium, with nutrients typically delivered at a minimal liquid flow rate. The main disadvantage of the TBAB is clogging arising from accumulation of excess biomass formation in the biofilter bed. Periodic *in situ* upflow washing (that is, backwashing) was found to be an effective strategy to control biomass growth [2].

In practice most off gas contaminated with VOCs have variable flow rates, unstable pollutant load, and contaminant compositions, which provided a challenge to industrial application of biofiltration [4, 5]. The biofilter is also exposed to non-use periods as a shutdown for equipment repair or during weekends and holidays. During such periods, the biofilter may be operated under the chemical starvation condition. It was reported that after non-use periods, the reacclimation period is an important factor in biofilter design and operation [6]. Generally, reacclimation to the previous removal performance increased after successive periods of non-use [6, 7]. During non-use periods, the microbial activity in biofilter decreased gradually [8], although significant loss in performance was not ob-

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Table 1. Experimental conditions and strategies for toluene biofiltration.

	Stage				
	I	II	III	IV	V
Experimental condition					
Inlet concentration, ppmv	50	50	100	250	500
Loading rate, kg COD/m^3 · day	1.14	0.70	1.41	3.52	7.03
EBRT, min	0.76	1.23	1.23	1.23	1.23
Experimental strategy (operational periods in days)					
Backwashing (1 h/week)	1–46	47–51	92–114	154–191	238–267
Non-use period					
Starvation (2 days/week)	—	52–71	115–133	192–210	—
Stagnant (2 days/week)	—	72–91	134–153	211–237	—

served after the restart of biofiltration after the short-term periods of non-use (such as 24 h) [7]. Moe and Qi [9] demonstrated that toluene removal in the fungal biofilter was adversely impacted for more than 2 days after weekend shutdown. However, the effects of non-use periods on biofilter performance as a function of contaminant loading rate have not yet been fully explored. To provide a constant high performance of the biofilter, the effect of non-use periods should be known to design the biofilter system effectively.

In this study, TBAB for toluene removal was evaluated for various toluene loading rates. The experimental plan was designed to investigate the effect of non-use periods on the biofilter performance.

EXPERIMENTAL METHODS

The experimental work was performed on a laboratory-scale trickle-bed air biofilter for controlling VOC as a single contaminant. The model VOC was toluene (C_7H_8). The biofilter was constructed of seven cylindrical glass sections with an internal diameter of 76 mm and a total length of 130 cm. Each section was equipped with a sampling port that extends to the center of the column. The reactor was packed with pelletized diatomaceous earth biological support media (Celite® 6 mm R-635 Bio-Catalyst Carrier; Celite Corp., Lompoc, CA) to a depth of about 60 cm. The biofilter was initially seeded with an aerobic microbial culture, which was obtained from the secondary clarifier of an activated sludge system at a municipal wastewater treatment plant (Millcreek wastewater treatment plant, Cincinnati, OH). The biofilter had been maintained at a constant operating temperature of 20 ° C in a constant temperature chamber and was operated in a cocurrent gas and liquid downward flow mode.

The air flow to the biofilter was initially set up at the rate of 3.6 L/min that was regulated by a mass flow controller. Liquid VOC was injected by a syringe pump into the air stream where it vaporized, and entered the biofilter through the topmost side port of the column. The biofilter was equipped with an independent system for feeding 1.5 L/day of a buffered nutrient solution. The nutrient formulation for the biofilter contained the same amount of nutrient–nitrogen and phosphorus ratio for a given VOC loading (a COD-to-nitrogen ratio of 50:1 and a nitrogen-to-phosphorous ratio of 4:1). As the sole source of nutrient-nitrogen, nitrate (NO_3–N) was used. The feed nutrient was sprayed as a fine mist onto the top of the medium bed through a spray nozzle.

Gas-phase samples for toluene analysis were taken with gas-tight syringes. These toluene concentrations were measured by using a gas chromatograph (GC) equipped with a flame ionization detector (FID). A GC equipped with a thermal conductivity detector (TCD) was used for determining the CO_2 concentration in the effluent gas phase. Detection limits for toluene and CO_2 were 0.5 and 300 ppmv, respectively. Liquid-phase samples were analyzed for nitrate, total carbon (TC), inorganic carbon (IC), and volatile suspended solid (VSS) concentration. Nitrate was determined by using a Shimadzu UV mini 1240 UV–vis spectrophotometer at a wavelength of 220 nm. TC and IC were determined by using a Shimadzu TOC 5000 analyzer. The VSS concentrations in the effluent and backwashing solution were determined according to Standard Methods 2540 G [10].

Experiments were designed for maintaining a consistent high biofilter performance. The biomass control strategy for this purpose involved *in situ* upflow washing with nutrient solution, that is, backwashing. The backwashing was conducted while the biofilter was off-line by using 18 L of the buffered nutrient solution to induce full medium fluidization at about 50% bed expansion for defined time periods. The backwashing duration and frequency were initially set at 1 h once per week.

Two different non-use periods (starvation and stagnant) were considered to simulate a shutdown or breakdown of VOC source. The first was a period without VOC loading (that is, starvation), which means pure air with nutrient flow through the biofilter. The second was a stagnant period that reflects no flow (VOC, nutrient, and air) passing through the biofilter. The duration and frequency for each non-use period were 2 days per week for a period of 3 weeks at each VOC loading rate (0.7, 1.41, and 3.52 kg COD/m^3·day). The conditions and strategies for this experiment are summarized in Table 1.

EXPERIMENTAL RESULTS

Biofilter Performance

The biofilter performance with respect to toluene removal is shown in Figure 1. The first operation, stage (I), was recognized as a start-up period, which demon-

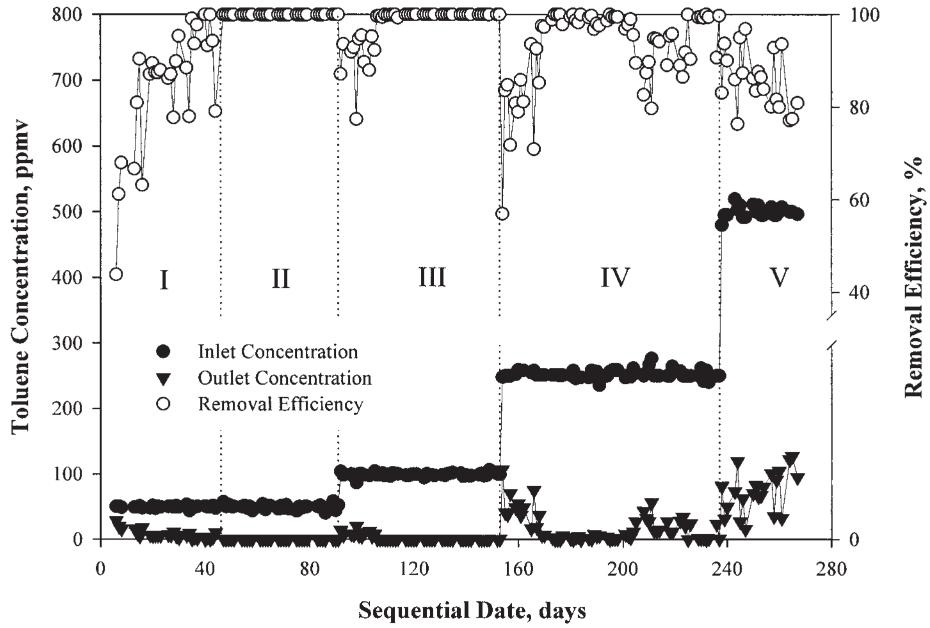

Figure 1. Biofilter performance with respect to toluene removal.

strates the acclimation period at start-up. Stage (II) was conducted under a VOC loading rate of 0.70 kg COD/m^3·day. The overall toluene removal efficiency reached over the 99% level for all three experimental strategies (backwashing, starvation, and stagnant). When the VOC loading rate was increased to 1.41 kg COD/m^3·day [stage (III)], a period of 10 days was required to acclimate the biofilter. After that, the biofilter was maintained at the 99% removal efficiency for all experimental strategies. During the non-use period strategies for VOC loading rates of 0.70 and 1.41 kg COD/m^3·day, the 99% removal efficiency was attained without backwashing. For a VOC loading rate of 3.52 kg COD/m^3·day [stage (IV)], a period of 15 days was required for acclimating the biofilter. After the acclimation period, the biofilter had been maintained at the 99% removal efficiency. During the non-use period strategies for VOC loading of 3.52 kg COD/m^3·day, the removal efficiency decreased occasionally <90% and demanded the backwashing strategy as the active biomass control. It is interesting to note that the behavior of the biofilter depended on the biomass control because its performance decreased substantially with buildup of back pressure arising from accumulation of excess biomass [1, 3, 11]. During stage (V), the overall toluene removal efficiency had not reached >99% under the backwashing strategy. Work by Smith *et al.* [12] demonstrated that oxygen limitation might occur in the biofilter operated over 500 ppmv of inlet toluene concentration, resulting in its failure. Figure 2 confirms that a corresponding inlet toluene concentration of 500 ppmv leads to substantial deterioration of the biofilter performance.

A generalized summary of biofilter performance, with respect to the experimental strategies, is presented in Figure 3. It is worth noting that the non-use period strategies provided biofilter performance similar to that with the backwashing strategy for toluene loading rates of 0.70 and 1.41 kg COD/m^3·day. Mao and Irvine [13] found that a biofilter loaded periodically can be operated without any contaminant breakthough. Cox and Deshusses [8] demonstrated that the biomass content in the reactor decreased during starvation, although this decrease was not critical for future reacclimation of the biofilter performance. Thus the non-use periods can be considered as another means of biomass control at a relatively low toluene loading rate.

Biofilter Response after Backwashing and Non-Use Periods

The biofilter response after restart-up after backwashing and non-use periods was explored by collecting effluent samples at prescheduled time intervals to determine the time-varying response of the biofilter (see Figure 4). The following observations can be deduced from Figure 4.

1. For the backwashing strategy, the overall removal efficiency was initially below the 30% range but it increased over the 90% range within 600 min at all toluene loading rates. The biofilter performance immediately after backwashing was unexpectedly poor as a result of the loss of active biomass. It is interesting to discuss the carbon balance during each experimental strategy (see Table 2). It was found that there was a greater loss of biomass during the backwashing strategy compared to that during the non-use period strategies.
2. For the two different non-use strategies, the biofilter initially had relatively high removal efficiency compared to that of the backwashing strategy. This may be attributable to the active biomass in the biofilter. During the non-use strategies, less loss of biomass was encountered, as shown in Table 2.

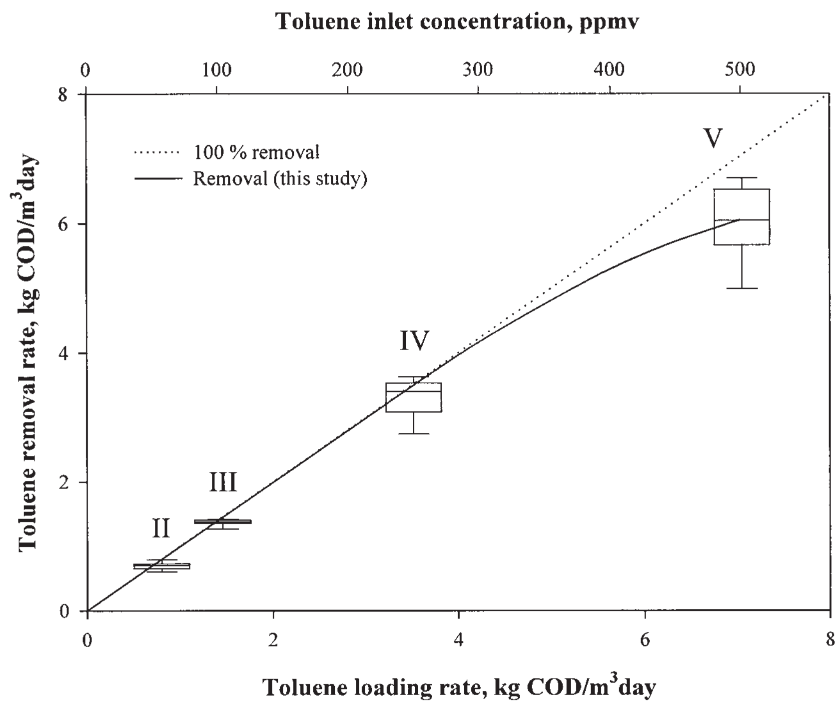

Figure 2. Toluene removal capacity with respect to toluene loading.

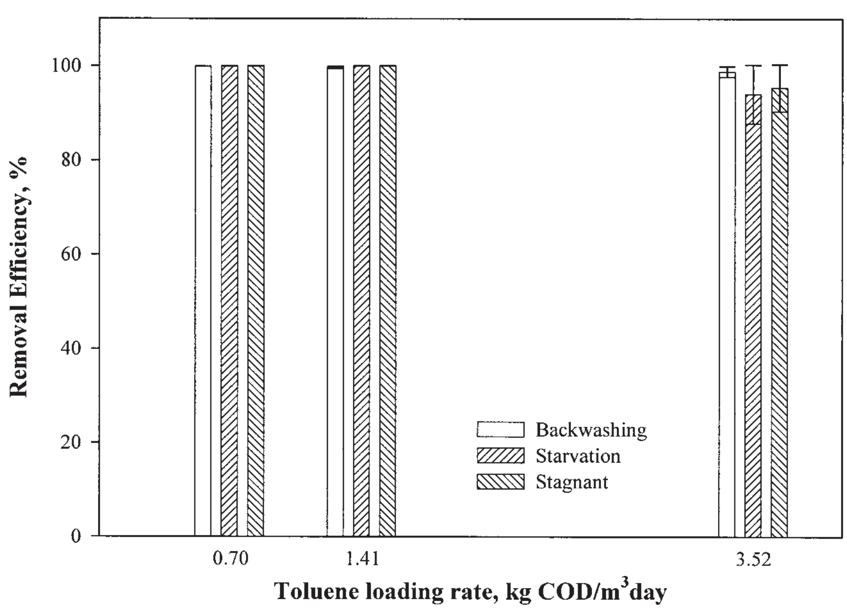

Figure 3. Biofilter performance with respect to experimental strategies.

3. At a toluene loading rate of 3.52 kg COD/m^3·day, higher removal efficiency had been observed right after the restart-up after the non-use periods compared to that after backwashing. It is speculated that, initially, toluene adsorption on the biofilm might have occurred in the active biomass within the biofilter.

These observations suggested that the preservation of biomass may reduce the period of reacclimation of the biofilter performance after non-use periods. However, high accumulated biomass can deteriorate biofilter performance. It is shown in Table 2 that the ratio of unaccounted carbon to carbon equivalent of VOC removed increased with loading rates. Although backwashing is helpful in removing the accumulated biomass in the biofilter, the remaining biomass in the biofilter will contribute to the unaccounted carbon in the carbon balance.

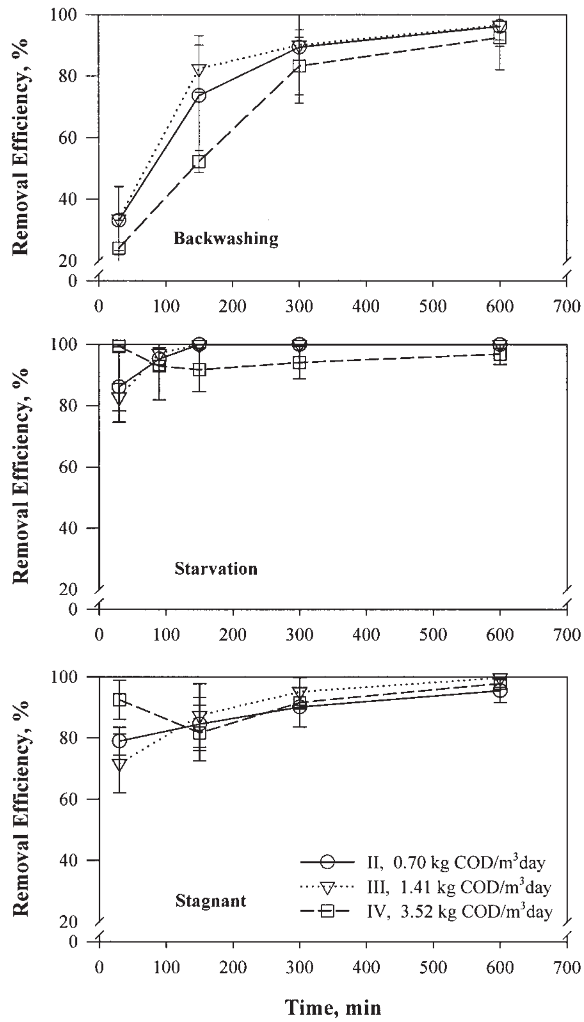

Figure 4. Biofilter response after the restart-up after backwashing, starvation, and stagnant periods.

Table 2. Carbon balance at different toluene loading rates and experimental strategies.

Loading rate, (kg COD/$m^3 \cdot$ day)	Experimental strategy	Ratio of carbon equivalent of CO_2 produced in gas to carbon equivalent of VOC removed (%)	Ratio of IC produced in liquid to carbon equivalent of VOC removed (%)	Ratio of TOC produced in liquid to carbon equivalent of VOC removed (%)	Ratio of carbon equivalent of lost biomass to carbon equivalent of VOC removed (%)	Ratio of unaccounted carbon to carbon equivalent of VOC removed (%)
0.70	Backwashing	79.9	0.4	1.5	14.3	4.0
	Non-use periods	83.0	0.7	1.3	0.8	14.2
1.41	Backwashing	75.0	0.6	0.8	12.3	11.3
	Non-use periods	88.2	0.8	0.5	0.2	10.1
3.52	Backwashing	69.2	0.2	0.0	15.4	15.2
	Non-use periods	80.9	0.5	0.2	0.2	18.2
7.03	Backwashing	63.2	0.3	0.1	15.5	20.9

Note: IC, inorganic carbon; TOC, total organic carbon (TOC = total carbon − IC); loss biomass = VSS in the effluent and backwashing solution during backwashing.

Kinetic Analysis

Kinetic analyses were based on pseudo-first-order removal rates as a function of depth in the biofilter for each experimental strategy. To avoid misinterpretation of the data arising from the possibility of degradation occurring above the media bed along the reactor free-board walls at top of the biofilter and in the bottom disengagement chamber used for separation between water and air, data obtained from sampling ports within the media were used for plotting the natural logarithmic scale of the ratio of residual concentration to inlet concentration as a function of the cumulative empty bed residence time. The obtained results are shown in Figure 5. It is seen that the first-order reaction rate constants were reduced with an increase in the toluene inlet concentration for all experimental strategies. Assuming that aerobic heterotrophic organisms mainly accounted for the toluene consumption as the substrate in this study, a higher concentration of toluene may result in a substrate inhibition effect on the growth of microorganisms, as demonstrated by Schroder *et al.* [14]. At low toluene inlet concentrations corresponding to loadings of 0.70 and 1.41 kg COD/m^3·day, the non-

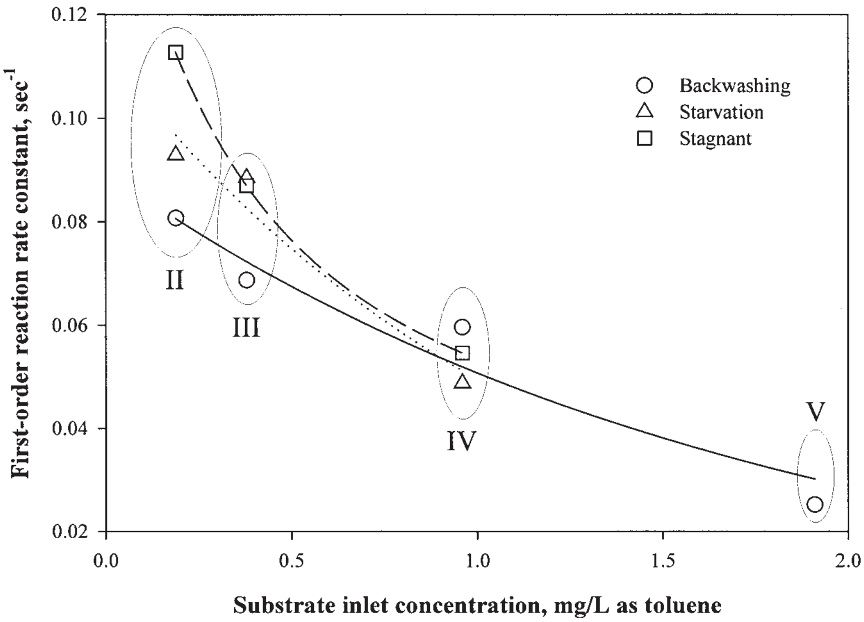

Figure 5. First-order reaction rate constants at different toluene inlet concentrations.

use period strategies provided higher first-order rate reaction constants than those of the backwashing strategy, whereas at high inlet concentration corresponding to a loading of 3.52 kg COD/m^3·day, the backwashing strategy had slightly high first-order rate constants. This experimental result indicates that the active biomass may play an important role in this effect, as discussed in section 3.2.

CONCLUSIONS

This study demonstrated the effect of non-use periods on the behavior of the biofilter under different toluene inlet loadings. Specific conclusions that can be drawn from this study include the following:

1. High performance of the biofilter had been observed for all experimental strategies (backwashing, starvation, and stagnant) for toluene loading up to 3.52 kg COD/m^3·day, corresponding to an inlet concentration of 250 ppmv.
2. The non-use periods can be considered as another means of biomass control for low toluene loadings (0.70 and 1.41 kg COD/m^3·day), but at high toluene loading (3.52 kg COD/m^3·day) the coordinated active biomass control such as the backwashing is needed for attaining high long-term removal efficiency. The biofilter response after restart-up after non-use periods was strongly dependent on the accumulated biomass in the biofilter.
3. As the toluene loading rate was increased, much longer reacclimation period was essential for biofilter performance to reach the 99% removal; moreover, the first-order reaction rate constants decreased for all experimental strategies (backwashing, starvation, and stagnant).

ACKNOWLEDGMENTS

The authors are pleased to acknowledge the financial support of the National Science Foundation under award number BES 0229135.

LITERATURE CITED

1. Smith, F.L., Sorial, G.A., Suidan, M.T., Breen, A.W., Biswas, P., & Brenner, R.C. (1996). Development of two biomass control strategies for extended, stable operation of highly efficient biofilters with high toluene loadings, Environmental Science & Technology, 30, 1744–1751.
2. Smith, F.L., Sorial, G.A., Suidan, M.T., Pandit, A., Biswas, P., & Brenner, R.C. (1998). Evaluation of trickle bed air biofilter performance as a function of inlet VOC concentration and loading, and biomass control, Journal of the Air & Waste Management Association, 48, 627–636.
3. Sorial, G.A., Smith, F.L., Suidan, M.T., Biswas, P., & Brenner, R.C. (1995). Evaluation of trickle-bed biofilter media for toluene removal, Journal of the Air & Waste Management Association, 45, 801–810.
4. Fitch, M.W., England, E., & Zhang, B. (2002). 1-Butanol removal from a contaminated airstream under continuous and diurnal loading conditions, Journal of the Air & Waste Management Association, 52, 1288–1297.
5. Popov, V.O., Bezborodov, A.M., Cavanagh, M., & Cross, P. (2004). Evaluation of industrial biotrickling filter at the flexographic printing facility, Environmental Progress, 23, 39–44.
6. Martin, F.J., & Loehr, R.C. (1996). Effect of periods of non-use on biofilter performance, Journal of the Air & Waste Management Association, 46, 539–546.
7. Dirk-Faitakis, C., & Allen, D.G. (2003). Biofiltration of cyclic air emissions of alpha-pinene at low and

high frequencies, Journal of the Air & Waste Management Association, 53, 1373–1383.

8. Cox, H.H.J., & Deshusses, M.A. (2002). Effect of starvation on the performance and re-acclimation of biotrickling filters for air pollution control, Environmental Science & Technology, 36, 3069–3073.
9. Moe, W.M., & Qi, B. (2004). Performance of a fungal biofilter treating gas-phase solvent mixtures during intermittent loading, Water Research, 38, 2259–2268.
10. American Public Health Association (APHA). (1995). Standard methods for the examination of water and wastewater (18th edition), Washington, DC: APHA.
11. Jorio, H., Bibeau, L., & Heitz, M. (2000). Biofiltration of air contaminated by styrene: Effect of nitrogen supply, gas flow rate, and inlet concentration, Environmental Science & Technology, 34, 1764–1771.
12. Smith, F.L., Sorial, G.A., Suidan, M.T., Biswas, P., & Brenner, R.C. (2002). Development and demonstration of an explicit lumped-parameter biofilter model and design equation incorporating Monod kinetics, Journal of the Air & Waste Management Association, 52, 208–219.
13. Irvine, R.L., & Moe, W.M. (2001). Period biofilter operation for enhanced performance during unsteady-state loading conditions. Water Science and Technology, 43, 231–239.
14. Schroder, M., Muller, C., Posten, C., Deckwer, W.D., & Hecht, V. (1997). Inhibition kinetics of phenol degradation from unstable steady-state data. Biotechnology and Bioengineering, 54, 567–576.