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Performance of Mesophilic Biofilter for the Treatment of Ethyl Benzene Polluted Air: Effect of Process Parameters

Rajamohan Natarajan*, Jamila Al-Sinani and Saravanan Viswanathan[1]

Faculty of Engineering, Sohar University, Sohar, Oman

[1]Department of Chemical Engineering, Annamalai University, India

✉ natrajmohan@gmail.com

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Abstract: The removal of ethyl benzene in a laboratory scale biofilter was investigated using a mixed microbial culture immobilized on a tree bark and proflex media based biofilter. The effect of operating variables namely inlet VOC concentration and flow rate were studied in the range of 1.0-4.0 g/m^3 and 0.25-1.0 m^3/h. The maximum removal efficiency of ethyl benzene attained was 98 % with an initial concentration of 1.0 g/m^3 and Empty Bed Residence Time of 282 s. Increasing the inlet ethyl benzene concentration and the air flow rate reduced the removal performance of the biofilter. A maximum elimination capacity of 130.5 g/m^3/h was achieved under optimal conditions. Temperature variation of approximately 4.4°C was observed during the entire biofiltration period confirming the exothermic nature of bio-reaction. Maximum carbon dioxide production of 310.1 g/m^3/h was recorded when the maximum elimination capacity was achieved.

Key words: Biofilter, ethyl benzene, volatile organics.

Introduction

Volatile Organic Compounds (VOCs) are gaseous emissions characterized by a vapour pressure of 10 Pa or higher at 293 K that are released into the atmosphere due to the industrial processes (Volckaert et al., 2013). Most of the VOCs are likely to cause ill effects to humans such as neuro and bronchial disorders, nausea, cancer initiation and also cause substantial damage to the plants and animals exposed to them. In addition, some VOCs like freons are reported to take part in ozone layer depletion and urban smog formation. Different types of air pollution control technologies are developed due to the diversity and volume of the VOCs released from various industrial activities. They are either phase transfer or VOC concentration technologies (Delhomenie and Heitz, 2005). In comparison to the physico-chemical treatment methods, bioprocesses gained significant attention due to the fact that they did not utilize additional energy other than the microbial capacity to metabolize the organic pollutants. The most popular bioprocesses are biofilters, bio scrubbers and bio trickling filters (Rene et al., 2009; Lee et al., 2008). Biofilters are reactors in which a humid pollutant loaded air stream is passed through a porous bed on which immobilized microbial culture capable of degrading the organic pollutant is present. Biofilters are used in Europe in more than 600 chemical process industries for deodourizing and treatment of VOCs and are more suitable for the treatment of large volumes of air containing low concentrations of VOCs (Mudaliar et al., 2010).

The removal efficiency and biodegradation rate of the hazardous contaminants are dependent on the reactivity,

*Corresponding Author

solubility of the organic compound in the biofilm and its biodegradability (Garcia Pena et al., 2008; Rene et al., 2009). The effectiveness of the bioprocess largely depends on the acclimatization of the microbes to the pollutant, ability to form biofilm and decomposing the organic compound (Estrada et al., 2013). Ethyl benzene is one of the popular organic pollutants which need to be degraded in the exit air streams. Biofiltration of ethyl benzene has been tried using different biofilter media like compost, *Macadamia ternifolia* nutshells under different operating environments and removal efficiency in the range of 90% was achieved (Son and Striebig, 2001; Volckaert et al., 2013). In this research study, the performance of a laboratory scale biofilter employing mixed microbial culture immobilized on a proflex media-tree bark packing to degrade ethyl benzene was studied. The effect of process variables, namely, initial ethyl benzene concentration and flow rate of ethyl benzene on the removal efficiency was studied. In addition, carbon dioxide production rate was monitored and correlated to VOC removal rate. The removal performance was studied axially along the biofilter column.

Materials and Methods

Microbial Seed

A mixed microbial culture was collected from the activated sludge of the municipal sewage treatment plant and used to inoculate the biofilter. The concentrated sludge was cultured in an aerated batch reactor and diluted in 1 L of nutrient solution containing the following composition: K_2HPO_4 – 3.84 gL^{-1}, KH_2PO_4 – 1.94 gL^{-1} and NH_4Cl – 3.00 gL^{-1}, at pH 6.9 (Saravanan and Rajamohan, 2009).The packing media consisting of a mixture of date palm tree barks and proflex media was mixed with the sludge in the biofilter column and drained after 24 h and this procedure was repeated several times until visible biomass was noticed on the biofilter media.

Biofiltration Experiments

The biofilter column used in this study was made of acrylic with an inside diameter of 5 cm and column height of 100 cm. A 10 cm headspace for collection of the treated gas and nutrient feed addition and 10 cm bottom space for leachate collection were provided. The biofilter column was equipped with two sets of sampling ports located at 0 cm (inlet), 25 cm (section 1), 50 cm (section 2), 75 cm (section 3) and 100 cm (exit), for treated gas sampling and temperature measurements along the height of the biofilter. Figure 1 shows the biofilter set-up with its components. A novel biofilter media consisting of a mixture of date palm tree barks and proflex media present in the ratio of 3:1 was used as a packing material inside the column. Addition of proflex media was justified to prevent any chances of the occurrence of bed compaction with the natural packing media used in the study and also to provide more surface area per unit volume for the biofilm growth. The biofilter column is equipped with a carbon dioxide gas sensor connected at the exit. The compressed air is divided into two streams: namely minor and major stream. The minor air stream was passed into the storage tank containing ethyl benzene (99% pure) and through the humidifier. The minor air stream loaded with ethyl benzene was mixed with the major air stream in the mixing chamber in order to attain the desired inlet concentration and fed into the biofilter reactor in an upflow mode.

The air flow rates are regulated in the low (0 - 0.3 L min^{-1}) and high (0 -10.0 L min^{-1}) flow rate range using rotameters. All the gas flow rates are manipulated using brass control valves. The operating parameters are varied in the ranges: inlet ethyl benzene concentration 1.0 to 4.0 g/m^3 and flow rate 0.25-1.0 m^3/h and the samples were collected at periodic intervals for analysis for residual ethyl benzene. The nutrient solution, Basal salts medium, with the following composition: K_2HPO_4 – 0.91 g; $Na_2HPO_4.2H_2O$ – 2.39 g; $(NH_4)_2SO_4$ – 1.97 g; $FeSO_4.2H_2O$ – 0.2 g; $MgSO_4.7H_2O$ – 2.0 g; $MnSO_4.7H_2O$ – 0.88 g; $Na_2MoO_4.2H_2O$ – 1.0 mg; $CaCl_2$ – 3.0 mg; $ZnSO_4.7H_2O$ – 0.04 g and $CoCl_2.6H_2O$ -0.04 mg per litre of water was sprayed twice a day through the nutrient distribution system equipped with a peristaltic pump and spray nozzle. The nutrient solution along with the humidified air helps in maintaining the required relative humidity in the biofilter. The pressure drop in the biofilter was monitored using a pressure sensor.

Analytical Methods

Ethyl benzene concentration in gas samples was measured by gas chromatograph using a Perkin Elmer, USA equipped with a FID and a capillary column. The temperature conditions were 200°C for injector and 250°C for detector. The oven temperature was set at 60°C for the first 5 min and increased at a rate of 15°C per minute to reach 180 °C and held at 4 min. Helium was used as a carrier gas at a flow rate of 2 ml min^{-1}. The temperature of the filter bed was measured using temperature sensors connected to a data logger.

The head space gas was analysed for carbon dioxide concentration using gas analyzer (Extox, Germany).

Performance Evaluation

The performance of the biofilter was measured in terms of the removal efficiency (% RE), elimination capacity (EC), g $m^{-3}h^{-1}$, and carbon dioxide production rate (CPR), g $m^{-3}h^{-1}$. These parameters are defined as given below:

$$\% \, RE = \frac{EB_0 - EB_t}{EB_0} \times 100 \quad (1)$$

$$EC = \frac{Q(EB_0 - EB_t)}{V} \; (\mathrm{gm^{-3}\,h^{-1}}) \quad (2)$$

$$CPR = \frac{Q(C_{\mathrm{CO_2,out}} - C_{\mathrm{CO_2,in}})}{V} \; (\mathrm{gm^{-3}\,h^{-1}}) \quad (3)$$

The Empty Bed Residence Time (EBRT) is defined as

$$EBRT = \frac{V}{Q} \; (\mathrm{h}) \quad (4)$$

The Inlet Loading Rate (ILR) is defined as

$$ILR = \frac{Q(EB_0)}{V} \; (\mathrm{gm^{-3}\,h^{-1}}) \quad (5)$$

where EB_0 and EB_t represent the inlet and exit concentrations of ethyl benzene (g/m^3), Q is the flow rate of the ethyl benzene (m^3/h), V is the volume of the biofilter (m^3), and C_{CO_2out} and C_{CO_2in} represent exit and inlet concentrations of carbon dioxide (g/m^3).

Results and Discussion

Effect of Initial Concentration of Ethyl Benzene

The biodegradation rate is related to the concentration and chemical configuration of VOC (Delhomenie and Heitz, 2005). The biofilter was operated with low inlet VOC concentration and low flow rates in order to acclimate and achieve substantial biofilm formation inside the biofilter. The biofilter was operated in an optimal plan to achieve stable and high removal efficiencies. In the first set of experiments, the biofiltration was carried out at ethyl benzene concentrations of 1.0, 2.0, 3.0 and 4.0 g/m^3 at fixed flow rates. The first phase of the experiment was carried over a period of 16 days with an inlet VOC concentration of 1.0 g/m^3 at different flow rates namely 0.25, 0.50, 0.75 and 1.0 m^3/h. The removal efficiency and exit ethyl benzene concentration were determined and plotted in Figure 2. The maximum removal efficiency attained with 1.0 g/m^3 of ethyl benzene concentration at the lowest flow rate of 0.25 m^3/h was 98% and the mixed microbial culture degraded the pollutant effectively. When the flow rate was increased at the same inlet concentration of 1.0 g/m^3, the removal efficiency decreased to a minimum of 79% corresponding to the flow rate of 1.0 m^3/h.

The second, third and fourth phases experiments were conducted at elevated ethyl benzene concentrations upto 4.0 g/m^3 in the similar pattern. The observations made during the different phases of the experiments showed that the removal efficiency decreased and exit VOC

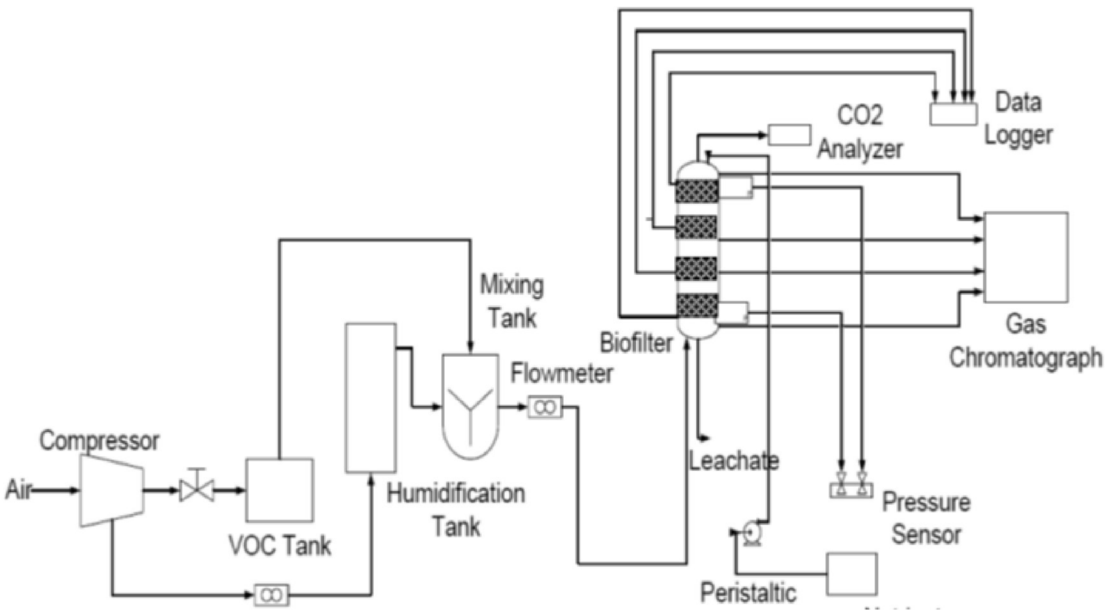

Figure 1: Experimental set up of biofilter reactor assembly.

concentration increased when the inlet ethyl benzene concentration increased. This phenomenon is due to the introduction of VOCs beyond a threshold limit at which the microbial growth and activity are inhibited (Delhomenie and Heitz, 2005). The results of this study are comparable with a similar research on ethyl benzene removal under mesophilic conditions in the concentration range of 0.5-3.0 g/m^3 (Volckaert et al., 2013). Studies on xylene removal using press mud as biofilter media employing mixed microbial culture also reported similar trends in effect of inlet concentration on removal efficiency (Saravanan and Rajamohan, 2009).

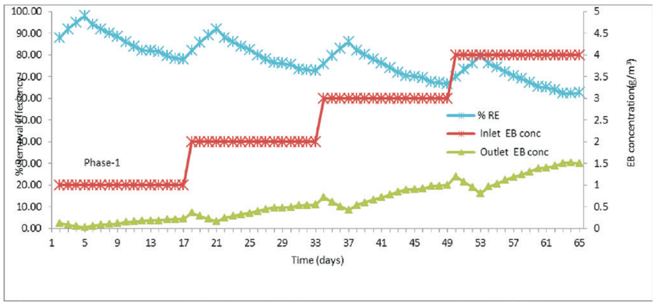

Figure 2: Biofiltration results for ethyl benzene removal at different inlet loads.

Elimination capacity is an indicative parameter of the capacity of the biofilter to degrade the pollutants. The effect of inlet loading rate on the elimination capacity was studied in the range of 0-204 g/m^{3}h. Figure 3 shows that when the inlet loading rate was increased, elimination capacity also increased. The pattern of increase was very linear in the initial phase and the slope of the line decreased at higher inlet loads. The maximum elimination capacity achieved was 130.5 g/m^{3}h as shown in the plot. At ethyl benzene inlet loads greater than 150 g/m^{3}h, the rate of increase of elimination capacity decreased and for a corresponding increase of 50 g/m^{3}h in inlet load, the elimination capacity increased by only approximately 30 g/m^{3}h. The variation patterns of elimination capacity was comparable with other studies on biofiltration of methyl tertiary butyl ether (Moussavi et al., 2009), benzene (Rene et al., 2009) and toluene (Saravanan et al., 2013).

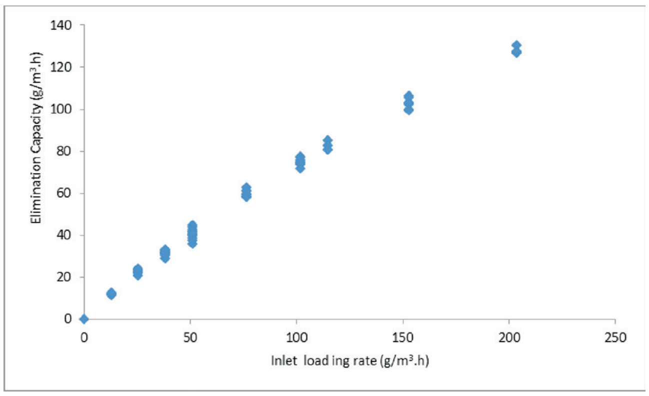

Figure 3: Effect of inlet loading rate on elimination capacity of ethyl benzene.

Effect of EBRT

The air flow rate and EBRT are vital parameters controlling the performance of biofilters. Depending on the flow rate and inlet VOC concentration, the biodegradation process is limited by either diffusion transfer from gas phase to biofilm or biochemical reaction or both simultaneously (Elmrini et al., 2004). The next phase of biofiltration studies was aimed at investigating the performance of biofilter at different residence times ranging from 70 to 282 s. The response of the biofilter was very effective at high EBRT of 282 s with efficiencies as high as 98% at 1.0 g/m^3 of ethyl benzene concentration and 74% at 4.0 g/m^3 of ethyl benzene concentration as shown in Figure 4. When the inlet concentration was increased to 2 g/m^3, removal efficiency achieved was 92% at the highest EBRT of 282 s and the efficiency decreased to 75.5% for a corresponding decrease in EBRT to 70 s. The impact of EBRT was more significant with higher inlet concentrations of VOC proven by the result of very low removal efficiency (64%) for 4 g/m^3 ethyl benzene concentration at a residence time of 70 s. This decrease in biofilter efficiency could be due to the non-availability of sufficient contact time between gas phase pollutant and the biofilm (Rene et al., 2012). In addition, substrate inhibition could also be a complimenting factor for reduced efficiencies at higher inlet loads. Research studies on removal of benzene, toluene, ethyl

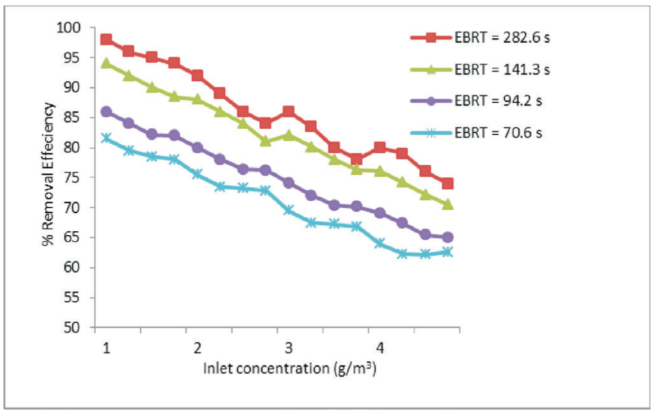

Figure 4: Effect of EBRT on removal efficiency.

benzene and xylene in a fungal biofilter (Rene et al., 2012) and thermophilic bioreactor (Mohammed et al., 2007) reported similar fluctuations.

Effect of Biofilter Height

The VOC concentration profile was plotted as dimensionless ratio (C/C_0) against the height of the biofilter in order to understand the hydrodynamics of ethyl benzene removal and shown in Figure 5. The results indicated that the ethyl benzene removal patterns were identical in the specific sections of the biofilter axially and did not vary appreciably when the inlet loading and flow rates were changed. In the first section of the biofilter, nearly 50-60% of the inlet ethyl benzene was biodegraded and the subsequent sections accounted for the remaining 40-50% removal. The average removal percentages in sections 2 and 3 of the biofilter were approximately 20% and the last section of the biofilter towards the exit end contributed to 10-12% of the removal efficiency. Higher microbial density and homogeneous biomass distribution in the lowest part of the bed could be the reasons for this behaviour. Moreover, the concentration gradients in the upper sections are comparatively lower which resulted in lower removal efficiencies. Also, the filter bed could be dry in the upper sections due to the exothermic nature of the bio-reaction. Similar axial variation of concentration profile in a biofilter column has been reported (Rene et al., 2009).

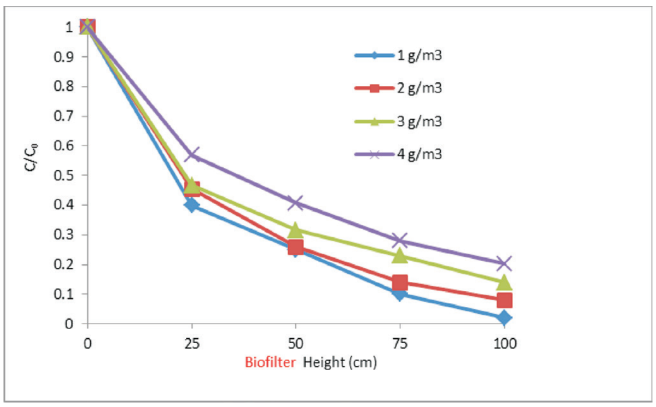

Figure 5: Concentration profile of ethyl benzene along the biofilter height.

Temperature Profile along the Biofilter

The temperature variation in the biofilter bed was observed during the study and plotted against time along with changes in elimination capacity in Figure 6. The results showed that the elimination capacity changes were accompanied by corresponding temperature changes with increase in temperature observed at higher elimination capacities. This phenomenon confirmed the exothermic nature of the biodegradation reaction of ethyl benzene. Metabolic activity of the biofilm releases heat during the biofilter operation (Devinny and Ramesh, 2005). The maximum temperature difference during the entire experimental phase was observed to be approximately 4.4°C. Also, the temperature variation along the height of the bed influences the microbial activity inside the biofilter column.

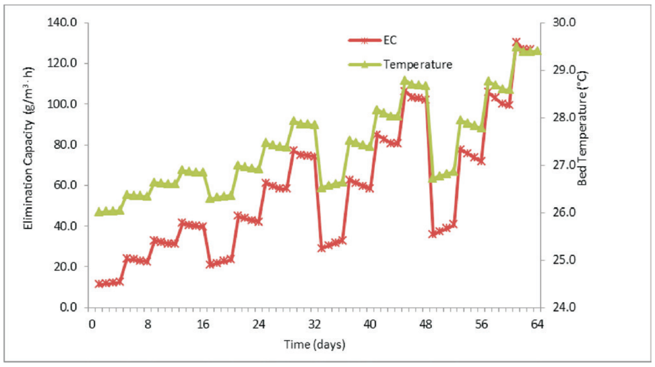

Figure 6: Temperature variation profile during the biofiltration experiments.

Carbon Dioxide Production and Bioreaction Stoichiometry

The possible end products of the aerobic bio-degradation processes are carbon dioxide and water vapour. Based on the fundamental principles of chemical kinetics, by-product concentration profiles always give meaningful conclusions to the mechanism of reaction. In the biofiltration process, carbon dioxide concentrations were measured at the exit of the reactor and correlated with elimination capacity. Figure 7 shows the rate of carbon dioxide production observed during various stages of biofiltration experiments. A linear relationship was observed between the elimination capacity and the carbon dioxide production rate represented by the equation: CO_2 production rate ($gm^{-3}h$) = 2.49 EC + 9.64 with a regression coefficient (R^2) value of 0.986. During the different phases of the study, the outlet carbon dioxide concentration is always greater than the inlet concentration and the better production rates were observed at lower gas flow rates or higher residence times. This observation was consistent with the general requirement for optimal biofilter performance. When the ethyl benzene concentration was 1 g/m^3, the carbon dioxide production rate was higher at a higher EBRT of 282 s compared to 70.6 s. For higher inlet ethyl benzene concentration, reduction in production rate of carbon dioxide was observed which was in agreement with reduced elimination capacity. Assuming complete conversion of ethyl benzene into water and carbon dioxide according to the following stoichiometry shown in equation (6),

$$C_8H_{10} + 10.5\ O_2 \rightarrow 8\ CO_2 + 5\ H_2O \qquad (6)$$

the stoichiometric mole ratio observed between ethyl benzene and carbon dioxide is 1:8 and mass ratio could be 1:3.3. However, the actual conversion ratio was 2.49 which was lesser than the theoretical value of 3.3. The difference in value was attributed to the possible consumption of ethyl benzene for the microbial growth inside the reactor during the biofiltration process. Also, it was reported that some of the carbon dioxide could have accumulated in the liquid phase in the form of HCO_3, H_2CO_3, and CO_3^{2-} (Rene et al., 2012).

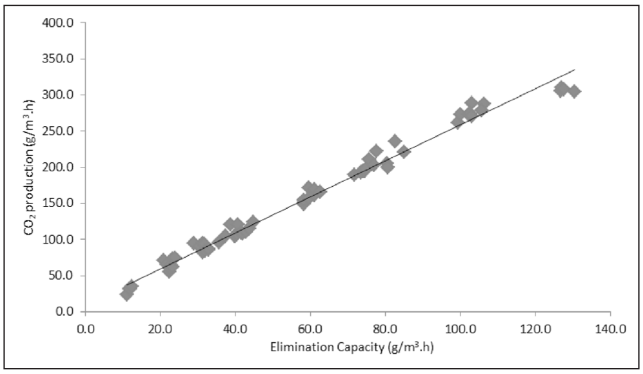

Figure 7: Elimination capacity versus carbon dioxide production.

Conclusion

This experimental work presents the performance analysis of a mesophilic biofilter operated continuously at different inlet concentrations and flow rates. The findings revealed the acclimation of mixed microbial culture from municipal sewage treatment plant to the ethyl benzene environment and withstood an inlet concentration upto 4 g/m^3. The biofilter removal efficiency decreased marginally with increase in inlet ethyl benzene concentration and a low gas flow rate of 0.25 m^3/h corresponding to a high EBRT of 282 s resulted in better removal in all the cases. The elimination capacity increased linearly in the inlet loading range of 0-150 g/m^3 h. The axial concentration profile of the ethyl benzene was studied along the height of the bed and the lower part of the bed was found to contribute more to the overall removal efficiency. High degree of VOC conversion was proved by recording the carbon dioxide production profiles in the biofilter. The actual conversion ratio was observed to be 2.49. Temperature increases were found to happen during the aerobic biodegradation and did not affect the process efficiency significantly.

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