Co-treatment of benzene and toluene vapours in a biofilter: A factorial design approach

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ABSTRACT

Biofiltration has now become an indispensable treatment technique for the removal of low concentrations of Volatile Organic Compounds (VOCs) from process vent streams. This study involves performance evaluation of a laboratory scale compost based biofilter for the treatment of mixtures of benzene and toluene (BT) vapours. Experiments were conducted as per a statistical design of experiment, the 2^k full factorial design, with the initial concentrations of benzene and toluene and the gas flow rate as the independent variables and the elimination capacity (EC) and removal efficiency (RE) as response variables. The maximum EC attained was 31.7 $g/m^3.h$ for benzene and 85.9 $g/m^3.h$ for toluene, while the total maximum EC at an inlet loading rate (ILR) of 150.2 $g/m^3.h$ was 91.2 $g/m^3.h$. It was also observed that while there was mutual inhibition, benzene removal was severely inhibited by the presence of toluene than toluene removal by the presence of benzene. Statistical analysis in the form of analysis of variance (ANOVA) was carried out to determine the main and interaction effects of variables on the RE and EC values. This study establishes the potential application of biofilters to handle mixtures of VOCs effectively through a statistically authentic approach.

1 INTRODUCTION

The growth of industries and heavy vehicular traffic has contributed tremendously to the decline in ambient air quality and pollution of the environment. Among these, the Volatile Organic Compounds (VOCs) emitted from process industries pose a significant threat to human health and environment. Benzene and toluene (BT) are two commonly noticed VOCs that arise from the petrochemical industry, pharmaceutical and printing works. BT, even in low concentrations has been found to cause significant damage to the liver and kidney and paralyse the central nervous system (Martin *et al*., 1998; Murata *et al*., 1999). In practice, emissions from these process industries are often characterized by the presence of mixtures of VOCs rather than as just one pollutant. Biological techniques are attracting growing interest for control of VOC emissions. Biofiltration, a process that removes VOCs from air stream by passing it through a packed bed of biofilm grown on an inert support, has been scaled up to industrial scale. Biological treatment begins with the treatment of contaminants from the air phase to the water phase (Devinny *et al*., 1999). The efficiency of this process primarily depends on the kinetics of micro processes such as absorption, adsorption, diffusion and biodegradation. However, while treating mixtures interaction effects between pollutants can play an important role in both mass transfer and biodegradation steps of the biofiltration process (McNevin and Barford, 2000). This study primarily aims in evaluating the removal pattern of BT mixture at different concentrations and flow rates in a lab scale compost biofilter.

2 THE 2^k FULL FACTORIAL DESIGN

Experiments with biofilters would normally involve in studying the effects of two or more factors (parameters) on a response variable. Factorial design is widely used in experiments where main and interaction effects of factors are likely to have a significant effect on the final response. For example, in biofiltration of mixtures of toluene and xylene, Jorio *et al*. (1998) reported that the degradation of toluene was inhibited by the presence of xylene in mixture. Similarly in a binary mixture of ethanol and methanol, Arulneyam and Swaminathan (2004) reported that the presence of methanol inhibited the degradation of ethanol and vice versa. The 2 level factorial design central composite design was used in this study with three factors. The factors that were chosen were benzene concentration, toluene concentration and flow rate, while the response variables were removal efficiency (RE, %) and elimination capacity (EC, $g/m^3.h$).

3 MATERIALS AND METHODS

3.1 Chemicals

Laboratory grade chemicals of benzene (>99%) and toluene (>99%, sulphur free) were purchased from Ranbaxy Fine Chemicals Limited, India.

3.2 Microorganism and media

The mixed microbial consortium obtained from a sewage treatment plant was acclimatized with benzene as the carbon source in 250 ml flasks. The nutrient solution had the following composition (in g/l); K_2HPO_4 – 0.8, KH_2PO_4 – 0.2, $CaSO_4 \cdot 2H_2O$ – 0.05, $MgSO_4 \cdot 7K_2O$ – 0.5, $(NH_4)_2SO_{4-}$ – 1.0 and $FeSO_4$ – 0.01 in distilled water.

3.3 Filter material

The packing material consisted of sieved compost (3-6 mm) and ceramic beads (4-6 mm) mixed in a 6:4 volume ratio. The filter material was inoculated with the acclimatized mixed consortia and loosely packed into the biofilter.

3.4 Experimental setup

Figure 1 illustrates the schematic of the experimental setup. The biofilter was made of poly acrylic tubes (5×70 cm) having 6 sampling ports sealed with a rubber septa at 10 cm along the biofilter height. The filter material supported on a perforated plate was packed to a height of 50 cms. Mixtures of BT were generated by passing humidified air through one of the VOC reservoirs and mixing with other vapour stream in a mixing chamber. The ranges of factors selected for factorial design are shown in Table 1. Samples were collected at regular time intervals using a gas tight syringe and analyzed for residual benzene and toluene concentrations.

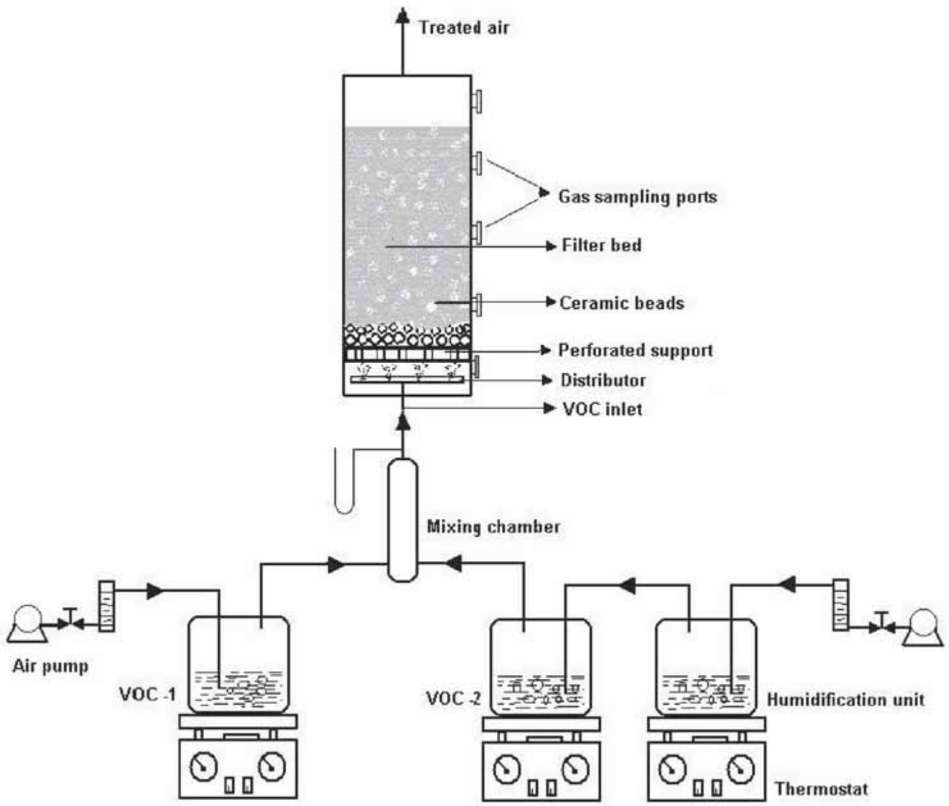

Figure 1. Schematic of the experimental setup.

Table 1.
Range of factors selected for factorial design.

Factors	Range and levels of factors		
	-1 (low)	0 (center point)	+1 (high)
Benzene, g/m^3	0.12	0.54	0.95
Toluene, g/m^3	0.14	0.81	1.48
Flow rate, m^3/h	0.024	0.036	0.072

3.5 Analytical methods

Benzene and toluene concentrations in the gas were measured using a Gas Chromatograph (Model 5765, Nucon gas chromatograph, Nucon Eng. India) with a poropak column (1/8" ID, liquid – 10% FFAP, solid – Ch-WIHP, 80/100 mesh) and flame ionization detector. Nitrogen was used as the carrier gas at a flow rate of 20 ml/min. The temperatures of the injection port, oven and detection port were 150, 120 and 250 °C respectively. The elution times were 1.1 min for benzene and 1.7 min for toluene respectively.

3.6 Software used

Statistical calculations and analysis (F – Fischer's variance ratio; P – probability value and T – test of significance) were done using the software MINITAB (version 12.2, PA, USA).

4 RESULTS AND DISCUSSIONS

4.1 Biofilter startup

Prior to carrying out experiments with mixtures of benzene and toluene (BT), the biofilter was operated for about 5 months under varied operating conditions with benzene vapours (*data not shown*). This biofilter was fed with near equal proportions of benzene and toluene for 18 days at a flow rate of 0.024 m^3/h and concentrations varying between 0.3-0.4 g/m^3. A slow gain in the removal of toluene could be noticed from the 6^{th} day of operation and this subsequently reached a steady state value of 74% on the 18^{th} day. This removal pattern indicates good acclimatization and high biological activity of the filter bed (*graph not shown*).

4.2 Removal of mixtures of benzene and toluene

Continuous experiments were carried out according to the 2^3 full factorial design (k=3), at different initial concentrations of benzene and toluene and at different flow rates after acclimatizing the biomass to a mixture of benzene and toluene. The low and high conditions for the variables are: benzene concentration – 0.12 and 0.95 g/m^3; toluene concentration – 0.14 and 1.48 g/m^3; flow rate – 0.024 and 0.072 m^3/h. Each experimental run was run for a period of 5 to 8 days to achieve steady state removal profiles and the average of these values were taken for calculating the RE and EC of the biofilter. The entire design of experiments along with their respective RE and EC is shown in Table 2.

Table 2.
Range of operating variables, removal efficiency and elimination capacity of biofilter for benzene and toluene removal.

Expt. No	Initial concentration, g/m^3		Flow rate, m^3/h	RE, %		EC, g/m^3.h	
	Benzene	**Toluene**		**Benzene**	**Toluene**	**Benzene**	**Toluene**
1	0.12	0.14	0.024	72.7	81.1	2.31	2.92
2	0.95	0.14	0.024	54.5	80.9	13.35	2.88
3	0.12	1.48	0.024	68.6	73.8	2.21	29.44
4	0.95	1.48	0.024	29.5	62.3	7.32	24.11
5	0.12	0.14	0.072	63.2	75.4	6.22	8.79
6	0.95	0.14	0.072	41.5	71.1	31.66	8.56
7	0.12	1.48	0.072	56.2	62.3	5.69	76.47
8	0.95	1.48	0.072	13.8	36.9	10.41	42.48
9	0.54	0.81	0.048	30.9	65.9	9.85	27.91
10	0.54	0.81	0.048	28.4	66.8	8.71	25.91

The results expressed in terms of RE for benzene and toluene is shown in Figs. 2 and 3 respectively. It was observed that the RE for both benzene and toluene were reduced in comparison to the individual study (Rene, et al., 2005). At low inlet loading rates (ILR, g/m^3.h) for both the substrates (total ILR $<$ 7 g/m^3.h), benzene removal was inhibited by around 19.2%, and toluene removal by 14.6%. At the same flow rate, when the benzene concentration was low and toluene concentration was high, the inhibition of toluene removal by benzene was less (4%) compared to that of benzene removal by toluene (23.4%). At high concentrations of both the substrate and at a total loading rate of 65 g/m^3.hr, benzene degradation was inhibited by 60.1%. At the highest ILR of 200 g/m^3.hr, the inhibition of benzene removal by toluene was 80.6%, while toluene removal inhibited by only 37%. Overall it is very obvious that the biofilter was much more effective in eliminating toluene than benzene.

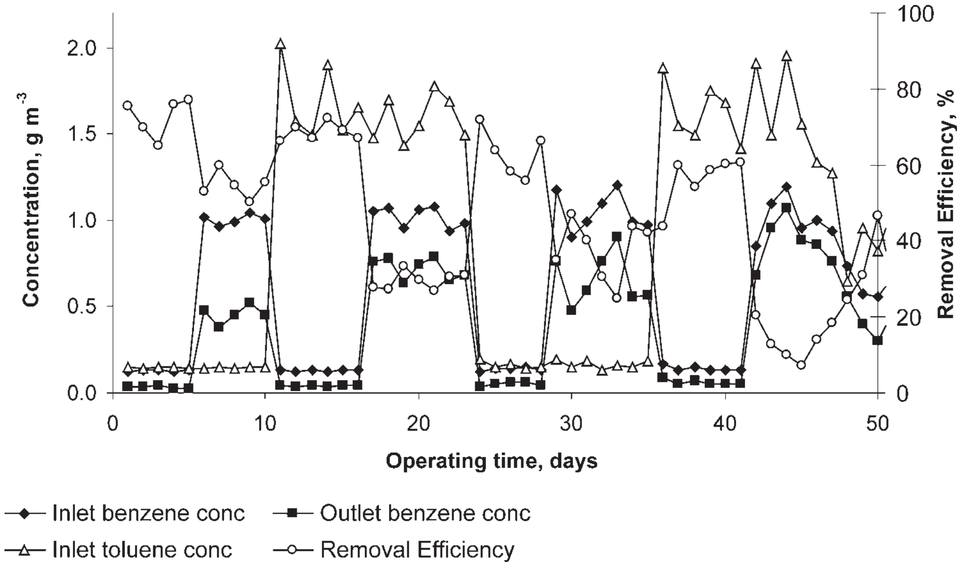

Figure 2. Removal profile of benzene under the influence of different toluene concentrations in biofilter.

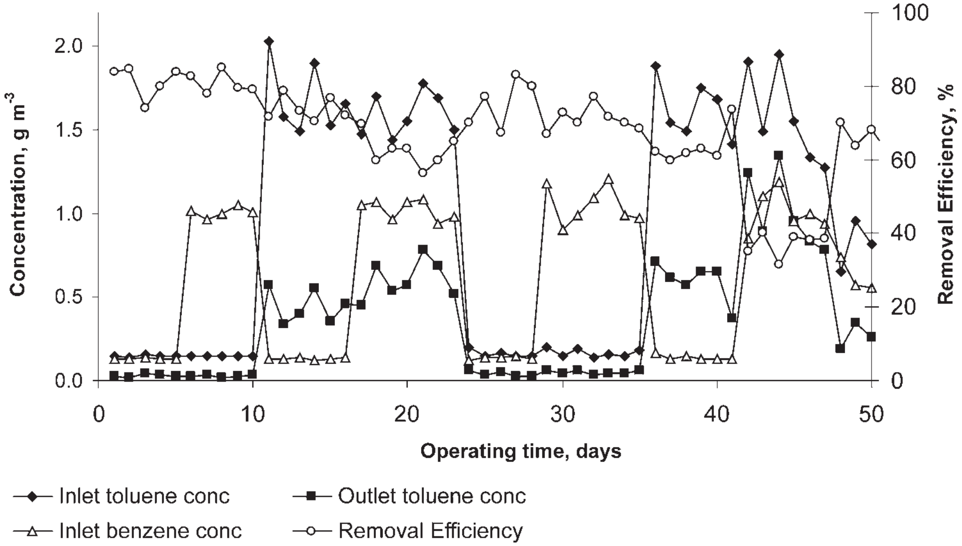

Figure 3. Removal profile of toluene under the influence of different benzene concentrations in biofilter.

This general feature of the biofilter for the treatment of BT mixtures is shown in Figure 4. The elimination capacities of the biofilter for both the pollutants were similar at lower ILRs. At high ILRs, the EC due to toluene removal was much higher than that of benzene. It may also be observed that at higher ILRs, the total EC of the biofilter decreased significantly due to inhibitory effects of the pollutants. The total maximum EC observed in this study is 91.2 g/m^3.h at a total inlet loading rate of 150.2 g/m^3.h. This EC value is nearly 25% higher than the EC values observed in the same biofilter when benzene was treated individually. It should be noticed that at this ILR, nearly 94% of the elimination was contributed due to toluene degradation at higher loading rates. Pressure drop and outlet temperatures were monitored during the operating period. Pressure drop values were nearly constant and varied between 3.5 – 7 cms of H_2O, while the outlet temperature varied between 27 – 35 ^oC.

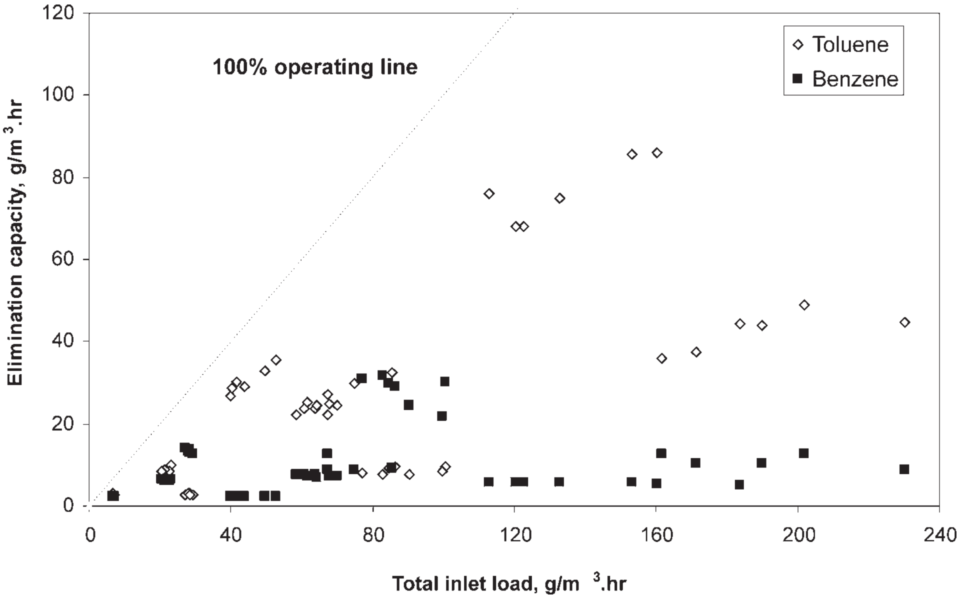

Figure 4. Effect of total inlet load on the elimination capacity of biofilter treating mixtures of benzene and toluene.

It is also well recognized that in the case of biodegradation of two or more pollutants the metabolic activity may involve the mechanism of induction, inhibition or co metabolism depending on the substrate and microbial species present. In biofiltration studies, Veir *et al*. (1996) studied the interaction of DCM and toluene in a compost biofilter that was earlier used to treat DCM. The presence of toluene caused an immediate decline in the DCM removal, but however the RE recovered considerably in the following weeks of operation. On the other hand toluene removal showed no inhibition during the recovery phase of DCM removal. Arulneyam and Swaminathan (2004) in their study on biofiltration of methanol and ethanol vapours observed that the removal efficiencies in mixtures were comparatively less than those for individual compounds. They further conclude that the effect on ethanol removal was due to inhibitory effect of methanol on ethanol utilizing organisms, while the effect on methanol removal was due to preferential utilization of ethanol by methanol utilizing microorganisms.

The main effect plot illustrating the effect of co-substrate on the removal of the primary substrate for benzene and toluene are shown in Figures 5 and 6. It was observed that the removal of the primary substrate decreased with increasing concentrations of both the primary and co-substrates. However, the effect of co-substrate was little less than that of the primary substrate. This indicates mutual inhibition between benzene and toluene on each others removal. Table 3 show the analysis of variance (ANOVA) for RE of benzene and toluene as binary mixtures under the conditions used in this biofiltration study. The main effects were significant for all the profiles as observed from the low P values (< 0.05). Among the main effects on toluene removal, toluene concentration (T = -39.25, P = 0.016) appears to play a major role than benzene concentration (-22.26, 0.029) and flow rate (-28.17, 0.023). The conclusion that could possibly be drawn from this study is that presence of toluene inhibits the degradation of benzene strongly at higher loading rates, while the presence of benzene has little effect on toluene degradation.

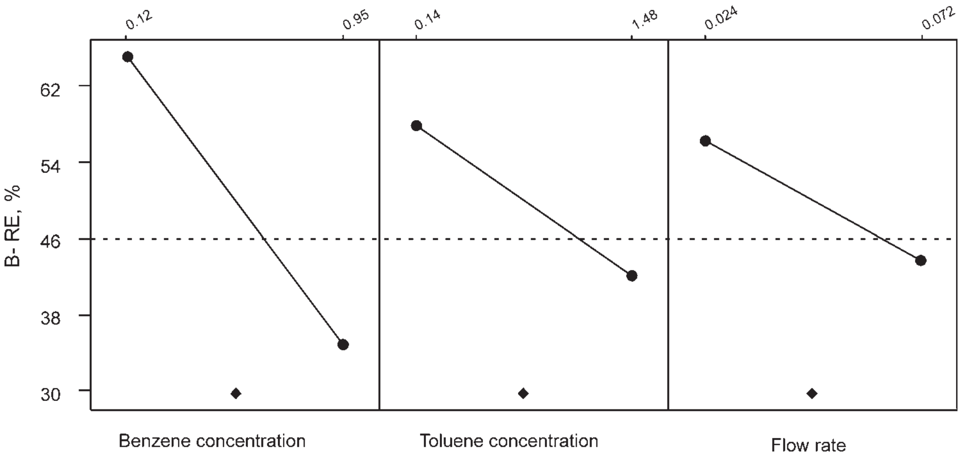

Figure 5. The main effects plot of variables for benzene removal in biofilter treating mixtures of benzene and toluene.

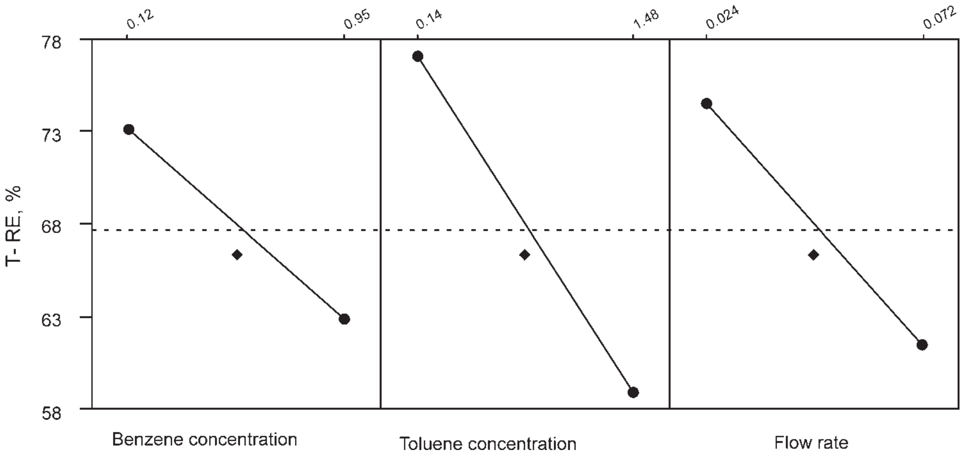

Figure 6. The main effects plot of variables for toluene removal in biofilter treating mixtures of benzene and toluene.

Table 3.
ANOVA for benzene and toluene removal from BT mixtures in biofilter.

Source	Benzene removal		Toluene removal	
	F	P	F	P
Main effects	284.92	0.044	943.15	0.024
2-Way Interactions	24.11	0.148	177.18	0.055
3-Way Interactions	0.00	0.975	28.91	0.117

5 CONCLUSIONS

Biofiltration of benzene and toluene vapours was investigated in a laboratory scale biofilter at varying operating conditions. Experiments were carried out according to the runs specified by the 2^k full factorial design. It was observed that at total ILRs less than 7 $g/m^3.h$, a maximum removal of 72.7% and 81.1% were achieved for benzene and toluene respectively. Moreover, an increase in the concentration of both benzene and toluene from low to high levels showed a significant decrease in the removal of both the VOCs. Furthermore, the results showed that the EC of the biofilter for both the pollutants were similar at low ILRs. At high ILRs, EC for toluene was much higher than that of benzene. The results from statistical analysis reveal that the main effects of the variables were significant with low P values (<0.05) than the interaction effects for the removal of both the compounds.

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