

## Biodegradation of mixture of VOC's in a biofilter

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**Abstract:** Volatile organic compounds (VOC's) in air have become major concern in recent years. Biodegradation of a mixture of ethanol and methanol vapor was evaluated in a laboratory biofilter with a bed of compost and polystyrene particles using an acclimated mixed culture. The continuous performance of the biofilter was studied with different proportion of ethanol and methanol at different initial concentration and flow rates. The result showed significant removal for both ethanol and methanol, which were composition dependent. The presence of either compound in the mixture inhibited the biodegradation of the other.

**Keywords:** biofilter; methanol; ethanol; elimination capacity; upset loading; VOCs; biodegradation

### Introduction

Deteriorating atmospheric air quality has resulting in more stringent regulations are being enforced to control air pollutants. Volatile organic compounds (VOCs) are among the new class of air contaminants generated from a variety of industrial sources. Efforts to control the emissions of VOCs have often failed due to inefficiency of conventional treatment systems such as wet scrubbing, ozonation, catalytic and thermal oxidation, and adsorption. Biotreatment of contaminants in gaseous streams provides an effective and inexpensive alternative to conventional treatment systems (Ottengraf, 1983; Leson, 1991; Deshusses, 1993). The possibility of complete mineralisation and effectiveness at low concentrations are other added advantage of biodegradation (Jorio, 1998; Sorial, 1997). Among the biological waste gas treatment methods, biofiltration is an innovative and a versatile, low technology approach to air pollution control which has attracted growing interest during the last few years (Ottengraf, 1983; Allen, 1991). A number of biomass supported on suitable matrices such as compost, peat, humus earth and wood chips have been used. The volatile compounds are adsorbed on the biofilm and subsequently oxidized into end products like water, carbon dioxide and salts. Many experimental studies have established biofiltration as an efficient treatment process and reliable technology for the control of VOCs (Hodge, 1995; Shareefdeen, 1993; Tang, 1995; Corsi, 1995). The majority of the literature on biofiltration is for removal of single compounds. However, industrial/field emissions are generally mixtures of VOCs. In mixed culture systems with multiple substrates the biodegradation process may be completely different from pure substrate system due to complex interactions between the substrates and the microorganisms.

Ottengraf *et al.* (Ottengraf, 1983) investigating the treatment of composite gas mixtures containing toluene, butylacetate, ethylacetate and butanol have reported that all the organic components were simultaneously eliminated with maximum elimination capacity for each compound ranging from 20–40 g/(m<sup>3</sup>·h). Corsi and Seed (Corsi, 1995) treated a gas stream mixture containing 1:1:1 benzene, toluene and xylene in a compost media biofilter and reported an average removal efficiency of 95% for loading rates up to 2.8 kg COD m<sup>3</sup>/d. Kiared *et*

*al.* (Kiared, 1996) have reported high elimination capacities for both toluene and ethanol in laboratory scale biofilters treating a mixture of both.

Jorio *et al.* (Jorio, 1998) have extensively investigated the performance of the biofilter for the removal of high concentrations of toluene, xylene and mixture of toluene and xylene. They achieved maximum elimination capacity of 115 g/(m<sup>3</sup>·h) for the mixture of toluene and xylene. They have reported that the metabolism of toluene degradation was inhibited by the presence of xylene in the mixture. This paper reports the results of study of biodegradation of a mixture of ethanol and methanol in a laboratory biofilter.

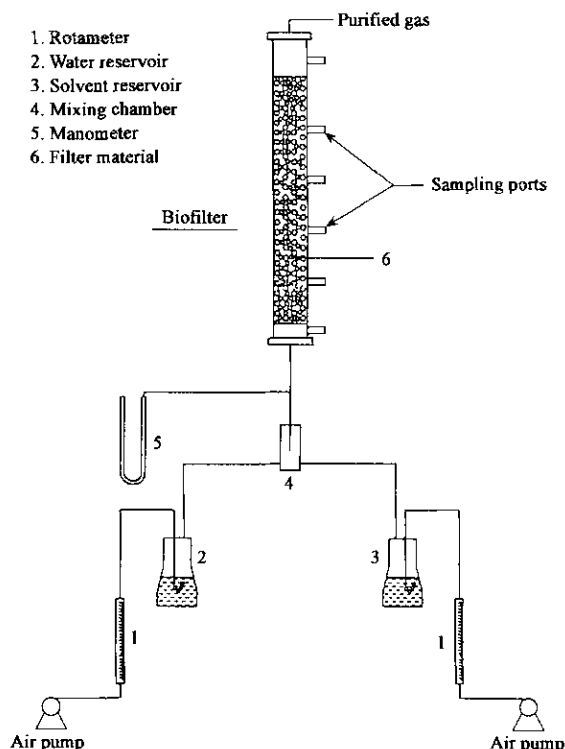


Fig. 1 Schematic diagram of the experimental set up

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## 1 Materials and methods

### 1.1 Filter materials

The packing material for the bed consisted of a mixture (60:40 v/v) of sieved compost (4 mm and 10 mm size granules, 1:1 ratio) and polystyrene inert particles (4 mm size). The inert material served to increase the bed porosity and to ensure a more homogenous gas distribution across the filter bed. The filter material was inoculated with methanol and ethanol acclimated mixed microorganisms.

### 1.2 Organisms and culture medium

A mixed culture of microorganisms obtained from an activated sludge plant treatment and acclimated to a mixture of ethanol and methanol as the carbon source was used as an inoculum. The nutrient solution consisted of (g/L):  $K_2HPO_4$ —0.8,  $KH_2PO_4$ —0.2,  $CaSO_4$ —0.05,  $MgSO_4 \cdot 7H_2O$ —0.5,  $(NH_4)_2SO_4$ —1.0 and  $FeSO_4 \cdot 7H_2O$ —0.01 in water.

### 1.3 Experimental set-up

Fig.1 illustrates the schematic diagram of the experimental set up. The biofilter made of transparent polyacrylic tube with an internal diameter of 5 cm. The tube was packed to a height of 50 cm. The filter material was supported by perforated plate. Sampling ports covered with rubber septa allowed collection of samples for analysis. A mixture of ethanol and methanol in humidified air was prepared from air streams of individual solvents in a glass-mixing chamber. The composition of the mixture was controlled by controlling the airflow rates using rotameter.

### 1.4 Biodegradation of mixtures

The biodegradation of mixtures of methanol and ethanol in the biofilters was studied at various proportions (20%—80%). The biomass was acclimated to a mixture concentration of  $2 \text{ g/m}^3$  (50:50) at a flow rate  $0.012 \text{ m}^3/\text{h}$ . After steady state removal was achieved, the biofilter was operated at different initial concentrations ( $2$ — $10 \text{ g/m}^3$ ), at different flow rates ( $0.012$ — $0.024 \text{ m}^3/\text{h}$ ). Experiments were run over a period of 70—80 hours. Effluent gas samples were collected at regular intervals and analyzed for residual ethanol and methanol concentrations.

### 1.5 Analytical methods

Ethanol and methanol were analyzed by gas chromatography (NUCON-5765, GC India) using a chromosorb-101 SS packed column (20 mm diameter  $\times$  3 m length) and a flame ionization detector. Nitrogen was used as the carrier gas. The temperatures of the column, injector and detector were maintained at  $150^\circ\text{C}$ ,  $160^\circ\text{C}$  and  $165^\circ\text{C}$  respectively.

## 2 Results and discussion

### 2.1 Start up and acclimation

Acclimation is an important operation for the successful performance of biofilter. Biofilter was first acclimated 50:50 mixture of ethanol and methanol continuously at a flow rate of  $0.012 \text{ m}^3/\text{h}$  and a total concentration of  $2.0 \text{ g/m}^3$ . When microorganisms were acclimated to the mixture, a steady state was achieved in about 30 d with a visible biofilm growth and high removals of ethanol and methanol.

### 2.2 Biodegradation of mixtures

The biodegradation pattern of mixtures methanol and ethanol at three different proportions, 20:80, 50:50 and 80:20 are shown in Figs. 2, 3, 4. It was observed that the removal efficiencies for both ethanol and methanol in the mixtures were much less than for the individual

substrate. Even a low proportion of methanol (20%) in the mixture inhibited the degradation of readily biodegradable ethanol.

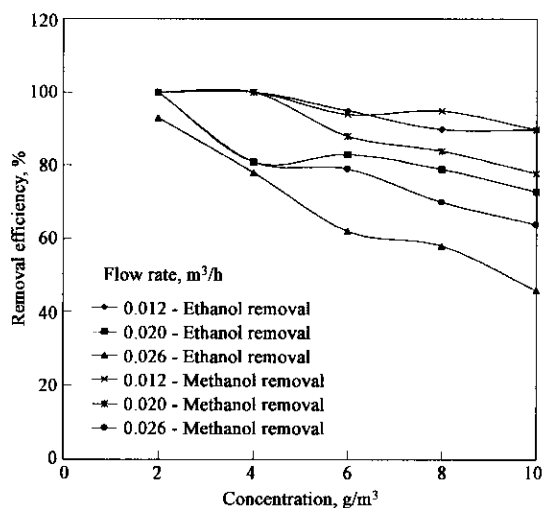


Fig.2 Removal efficiency of the filter bed at different gas flow rates and concentrations for mixture (methanol 20% + ethanol 80%)

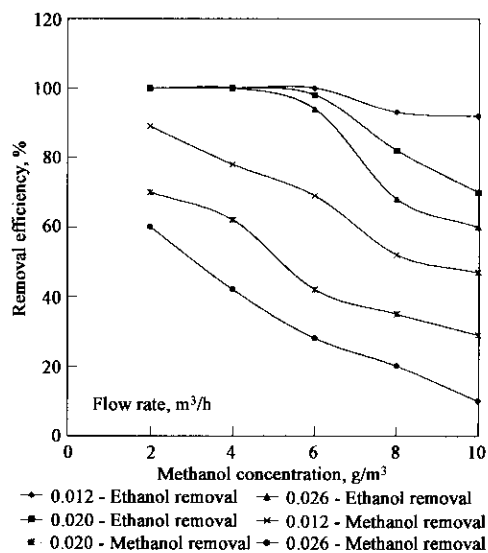


Fig.3 Removal efficiency of the filter bed at different gas flow rates and concentration for mixture (methanol 50% + ethanol 50%)

The presence of ethanol also decreased the removal of methanol. These inhibitory effects were more clearly seen with equal proportions of methanol and ethanol. In this case the ethanol removal was inhibited by about 30% while the methanol removal was reduced from 40% to 8%. At higher proportion of methanol (80% in the mixture) the effect of ethanol on methanol degradation was highly significant while the inhibitory effect of methanol on the ethanol removal is not very evident. It is 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 substrates and microbial species present. Though, some evidences of such metabolic effects are reported in mixtures in liquid phase cultures, there are very few reports in biofiltration of mixtures.

Arvin *et al.* (Arvin, 1989) observed stimulation of biodegradation of benzene in liquid phase by the presence of either toluene or xylene. Chang *et al.* (Chang, 1993) found mutual inhibition in the

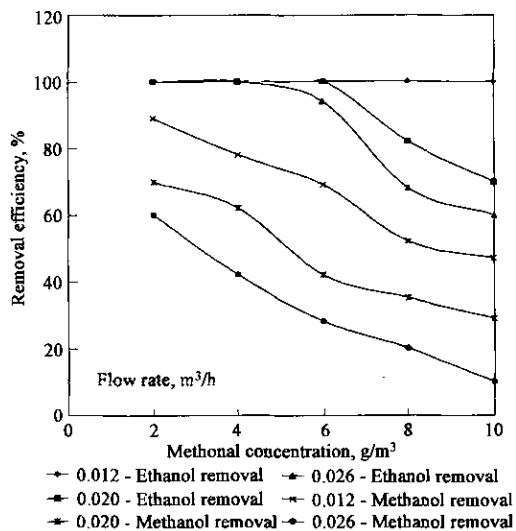


Fig. 4 Removal efficiency of the filter bed at different gas flow rates and concentration for mixture (methanol 80% + ethanol 20%)

biodegradation of a mixture of toluene and xylene in a suspension. In biofiltration, Deshusses and Hamer (Deshusses, 1993) observed inhibition of methyl isobutyl ketone (MIBK) biodegradation in the presence of methyl ethyl ketone (MEK). Jorio *et al.* (Jorio, 1998) have also reported that the removal efficiency for toluene decreased due to the presence of xylene while the presence of toluene had negligible effect on the xylene removal efficiency. Interestingly, so far no explanation other than a competitive inhibition has been offered for the cross interactions between pollutants in multi pollutant biodegradation.

The observation with mixtures in the present study may be explained in terms of two different groups of microorganisms namely ethanol utilizers and methanol utilizers, while the methanol utilizers have the ability to switch to ethanol utilization in the presence of ethanol, the ethanol utilizers seems to be inhibited by the presence of methanol. Thus, the lesser removal of methanol in the presence of ethanol appears to be due to preferential utilization of ethanol by the methanol degraders. The decrease in the ethanol removal in the presence of methanol seems to be related to the inhibitory effect of ethanol metabolism. However, more detailed microbiological studies are required to establish this differentiation in the biomass.

### 2.3 Elimination capacity

The rate of conversion in a continuously operating biofilter can be expressed by the elimination capacity (EC). Elimination capacity (EC) is a normalized measure of VOCs removal capacity of biofilter at a given mass loading. EC is defined as the amount of VOC removed in the biofilter per unit volume of biofilter per unit time ( $g/(m^3 \cdot h)$ ).

The elimination capacities for the individual compounds were estimated in the biofilter treating different proportions of mixtures. The results are presented in Figs. 5, 6, 7. It was observed that the elimination capacity for methanol reached the limiting values, particularly at higher proportions. However, the elimination capacity for ethanol did not reach the limiting value in the concentration range tested. Comparison of the elimination capacities for ethanol and methanol in the mixture with the elimination capacities obtained with individual compounds showed a distinct reduction in the elimination capacities of the mixture as explained in the previous section. The elimination capacity for ethanol decreased as the proportion of methanol in the

mixture is increased, which may be due to inhibitory effects of methanol on ethanol metabolism.

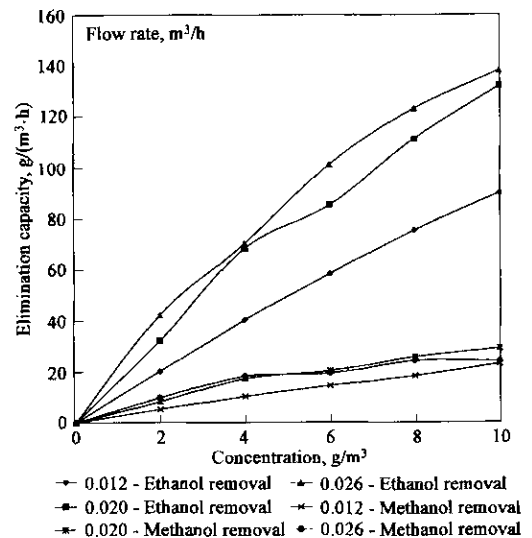


Fig. 5 Elimination capacity of the filter bed for mixture (methanol 20% + ethanol 80%)

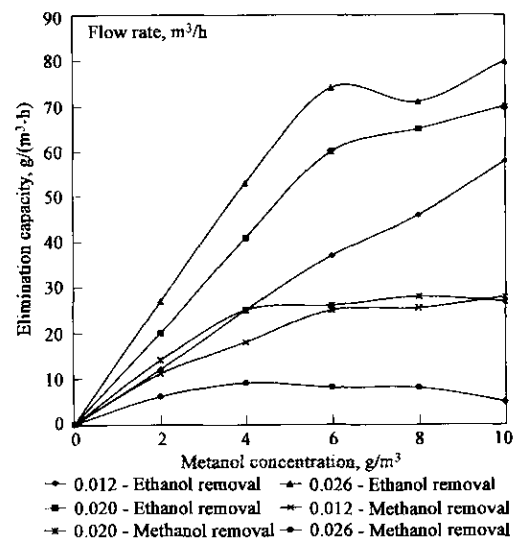


Fig. 6 Elimination capacity of the filter bed for mixture (methanol 50% + ethanol 50%)

Jorio *et al.* (Jorio, 1998) have also shown that the maximum elimination capacities for toluene in a mixture with xylene were much lower than its maximum elimination capacity as a single pollutant. On the other hand the maximum elimination capacity for xylene was similar both as a single pollutant and in the mixture. This behavior was attributed to them by the inhibition of toluene degradation with xylene. Since, elimination capacity is an important parameter in the stagewise design of biofilters, the performance of biofilters treating mixtures of VOCs has to be carefully analyzed to arrive at optimal design.

## 3 Conclusions

Biodegradation in biofilter containing compost as the main biomass support appears to be a cost effective treatment method for easily biodegradable volatile compounds like ethanol and methanol. The continuous studies on biodegradation of mixture of ethanol and methanol vapours in biofilters with acclimatized biomass have revealed the feasibility of treating these compounds under extended period of operation

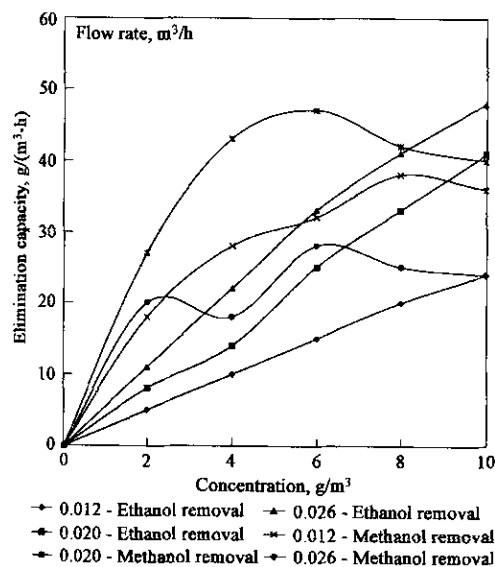


Fig.7 Elimination capacity of the filter bed for mixture (methanol 80% + ethanol 20%)

with preferential removal of ethanol over methanol. Both the removals of ethanol and methanol were comparatively less than those for the pure compounds. The effect on ethanol removal was due to the 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. Appreciable elimination rates of both ethanol and methanol were obtained at lower proportion of methanol to ethanol. This study established the potential application of biofilters for the treatment of VOCs.

## References :

Allen E R, Yang Y, 1991. Biofiltration control of hydrogen sulfide emissions[C]. Proceedings of the 84th annual meeting of the air and waste management association, Pittsburg, Pa.

- Arvin E, Jensen B K, Gundersen A T, 1989. Substrate interactions of benzene, toluene, and paraxylene during microbial degradation by pure cultures and mixed culture aquifer slurries[J]. *Appl Environ Microbiol*, 57: 3221—3225.
- Chang M L, Voice T C, Criddle C S, 1993. Kinetics of competitive inhibition and metabolism in the biodegradation of benzene, toluene, and *p*-xylene by two *Pseudomonas* isolates[J]. *Biotechnol Bioeng*, 41: 1057—1065.
- Corsi R C, Seed L P, 1995. Biofiltration of BTEX: media, substrate, and loading effects[J]. *Environ Prog*, 3: 151—158.
- Deshusses M A, Hamer G, 1993. The removal of volatile ketone mixture from air in biofilters[J]. *Bioprocess Engg*, 9: 141—146.
- Hodge D S, Devanny J S, 1995. Modelling removal of air contaminants by biofiltration[J]. *J Env Eng*, 121: 21—44.
- Jorio H, Kiared K, Brzizinski R *et al.*, 1998. Treatment of air polluted with high concentrations of toluene and xylene in pilot scale biofilter [J]. *J Chem Technol Biotechnol*, 73: 183—196.
- Kiared K, Bibeau L, Brzezinski R, 1996. Biological elimination of VOC's in biofilter[J]. *Environ Prog*, 15: 148—152.
- Leson G, Winer A M, 1991. Biofiltration: an innovative air pollution control technology for VOC emissions[J]. *J Air and Waste Management Association*, 41: 1045—1052.
- Ottengraf S P P, Vander oever A H C, 1983. Kinetics of organic compound removal from waste gases with a biological filter[J]. *Biotechnol Bioeng*, 25: 3089—3102.
- Shareefdeen Z, Baltzis B C, Oh Y S *et al.*, 1993. Biofiltration of methanol vapor [J]. *Biotechnol Bioeng*, 41: 512—524.
- Sorial G A, Smith F L, Suidan M T *et al.*, 1997. Evaluation of trickle bed air biofilter performance for toluene removal[J]. *J Air and Waste Management Association*, 45: 801—810.
- Swanson J, Loehr W R C, 1997. Biofiltration: fundamentals, design and operations principles, and applications[J]. *J Env Eng*, 123(6): 538—546.
- Tang H M, Hwang S J, Hwang S C, 1995. Dynamics of toluene degradation in biofilters[J]. *Haz Waste Haz Matls*, 12: 160—172.

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