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Research Article

Removal of MEK, Toluene and Xylene (MTX) from Air Using Modified Wood Charcoal Beads as Biofilter Media

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Abstract:

Biofiltration of air contaminated with mixture of methyl ethyl ketone (MEK), toluene and xylene was studied in a laboratory scale biofilter unit packed with a composite biofilter media (charcoal/PVA/ KNO₃ beads) in three phases for 56 days. Activated sludge from the aeration tank of a local sewage treatment plant after acclimation with MTX was used as inoculums. The maximum removal efficiency of 96.6% was achieved at the MTX loading of 236 g m⁻³ h⁻¹ with an EBRT of 28.4 s. The maximum elimination capacity of 770 g m⁻³ h⁻¹ was obtained at the MTX loading of 850 g m⁻³ h⁻¹. Results showed that the conditions were most favourable for toluene degradation followed by MEK and xylene. The Michaelis–Menten kinetic constants r_{max} and K_s values obtained for MEK, toluene and xylene were 0.088, 0.101, 0.103 g m⁻³ h⁻¹, and 1.043, 0.826, 0.696 g m⁻³, respectively. The experimental results were compared with predictions from the Ottengraf–van den Oever model for zero-order diffusion-controlled region. The critical inlet concentration, critical inlet load and biofilm thickness were estimated using the model predictions. The adsorptive properties of media were also examined.

Keywords: Bio-filter, Modified media, MTX Vapours, Michaelis–Menten kinetic model.

1.0 Introduction:

Bioremediation of gases containing VOCs using Biofilters is a relatively new application of bioprocess engineering in waste management. It applies microorganisms, attached on porous support media, for the breakdown of VOCs and has potential application to a number of industries. It has a very low operating cost and is very effective for treating large volumes of moist air streams with low concentrations of the biodegradable pollutants (Leson and Winer, 1991; Mohseni and Allen, 2000). The contaminated constituents from automobile industries and paint spray booth off gases vary depending on the type of paint utilized. However, typical composition includes a mixture of solvents including ketones (e.g., methylethylketone, methyl isobutyl ketone, methyl propyl ketone), aromatic hydrocarbons (e.g., toluene and xylenes), and esters (e.g., *n*-butyl acetate). Although the use of reformulated paints containing reduced quantities of VOCs can markedly decrease emissions from many facilities, there is also an increasing need for cost-effective and reliable air pollution control technologies that can remove VOCs from these emissions (Qi et al., 2005). So far, both organic and inert materials have been used as filter bed in industrial filtration.

However, the former have a relatively low durability (up to a few years), while the latter tend to be expensive and usually require a periodical nutrient supply. Consequently, the development of a material comprising the advantages of both organic and inorganic materials might lead to an improvement in the performance of biofilters (Hernandez et al., 2010). When they are chemically modified, the physico-chemical properties of the synthetic materials such as porosity, surface area, buffer capacity and water retention capacity can be improved (Hirai et al., 2001).

Being a heterogeneous biochemical process controlled either by mass transfer or biochemical reaction or both, the removal efficiency of biofilters varies with media characteristics (e.g. porosity, pore size, density, retention capacity), carbon and energy source, nutrients, pH and temperature (Upadhyay and Kumar, 2004). Thus, the packing material is considered the core of a biofilter because its nature influences both removal performance and operational costs (Liang et al., 2007). So far, both organic and inert materials have been used as filter bed in industrial filtration. However, the former have a relatively low durability (up to a few years), while the latter

tend to be expensive and usually require a periodical nutrient supply. Consequently, the development of a material comprising the advantages of both organic and inorganic materials might lead to an improvement in the performance of biofilters (Hernandez et al., 2010). Synthetic materials are of great importance because they simplify the modeling of biofilters. Also, when they are chemically modified, the physico-chemical properties of the synthetic materials such as porosity, surface area, buffer capacity and water retention capacity can be improved (Hirai et al., 2001). So far, only a few studies have investigated the biofiltration of paint solvents using various packing material like mixture of coal and matured compost, coal, polypropylene Pall Rings by various researchers (Rahuvanshi and Babu, 2009; Mathur and Majumder, 2008; Lu et al., 2001; Boswell, 2001; Kazenski and Kinney, 2000).

The objective of this investigation is to evaluate the performance of a biofilter packed with modified wood charcoal based biofilter media for removal of MTX under varying loading conditions and without supply of nutrient. The media was characterized and variations in pH, temperature, pressure drop and CO₂ production rate were monitored throughout the experiments. The experimental results were also validated with the results obtained using Ottengraf-van den Oever model.

2. Materials and Methods:

2.1 Preparation of PVA/Wood Charcoal/KNO₃ Composite beads:

Wood Charcoal purchased from the local market was selected as a base material for the preparation of composite beads. Poly vinyl alcohol (PVA), Boric Acid (H₃BO₃), Potassium Nitrate (KNO₃), Sodium monobasic phosphate (NaH₂PO₄·2H₂O) and Sodium dibasic phosphate (Na₂HPO₄·12H₂O) were used for the synthesis of composite beads. All chemicals used were analytical grade and laboratory grade reagent from Merck Specialities Pvt. Ltd., Mumbai, India. Poly vinyl alcohol (PVA) is a hydrophilic polymer capable of binding water by capturing OH group. Thus, PVA was used as a bonding agent and blended with natural packing material to make spherical beads. Boric acid was used to decrease the PVA hydrophilic property in the preparation of beads. The method of preparation of composite beads is similar to method adopted by Chan and Lin (2006).

Wood Charcoal 200 g (average diameter, 0.85 mm) was dried at 90°C before adding into an aqueous

solution (800 ml water mixed with 128 g KNO₃) in a 2000 ml bucket. The mixture was sealed and kept for approximately 24 h for charcoal to adsorb KNO₃ and water and reach equilibrium. PVA powder (200 g) in an aqueous solution (2000 ml water mixed with 128 g KNO₃) was heated to 90°C for dissolution. The wood charcoal/KNO₃ mixture was slowly added into the PVA/KNO₃ mixture at 90°C and then stirred for 1.5 h at 90°C and finally cooled to 40°C. The mixture was slowly dripped into a 6% boric acid aqueous solution (1500 ml) for 60 min leading to the formation of beads. The beads were subsequently transferred into the aqueous phosphate solution (150 g NaH₂PO₄·2H₂O and 335 g Na₂HPO₄·12H₂O in 450 ml water) and stirred for 30 min. Finally, the beads were dried in oven at 100°C for 24 h and then immersed in the 0.384M KNO₃ solution again because some amount of water soluble nitrogen may be dissolved out in boric acid solution. Physical properties of original wood charcoal and modified media were calculated and given in Table 1. CHN content of composite bead and wood charcoal were measured using CHN analyser (Perkin Elmer). The inoculum was prepared using a mixed bacterial culture obtained from the aeration unit of local wastewater plant. Methods of preparation of inoculum and its acclimation have been provided elsewhere (Singh et al., 2006).

2.2 Biofiltration setup and operation:

Biofilter setup (Fig. a) was the same as described earlier (Singh et al., 2010). Bed was filled with previously humidified modified biofilter media (PVA/Charcoal/KNO₃ composite beads) up to height of 50 cm. Vapour phase mixture of MTX was generated by bubbling the MTX mixture in a bubbler. Volumetric ratio of MTX in the bubbler was taken as 40:45:15 which is same as MTX mixture used paint solvent (Boswell, 2001). The filter material in the column was supported on a stainless steel sieve plate for avoiding excess compression of media at bottom. The biofilter was inoculated with mixed bacterial culture. No nutrient was supplied throughout the experiment however distilled water was added periodically to keep the bed sufficiently wet. The leachate was collected at the bottom of the column. Biofilter was operated for 56 days. Samples of inlet and outlet gas were collected from the sampling ports of the biofilter column in 15 ml gas samplers and analyzed using the Chemito -7610 gas chromatograph equipped with the flame-ionization detector (FID), Chromatopak column (1/8" dia., 2 m length and 80/100 mesh) and nitrogen as carrier gas. Hamilton gas (1 ml

capacity) and liquid (10 μl capacity) sampling syringes were used for collection and injection of samples. CO_2 was measured by bubbling outlet

gas into N/10 KOH solution for a fixed interval of time and titrating it by N/10 HCL solution using phenolphthalein as indicator.

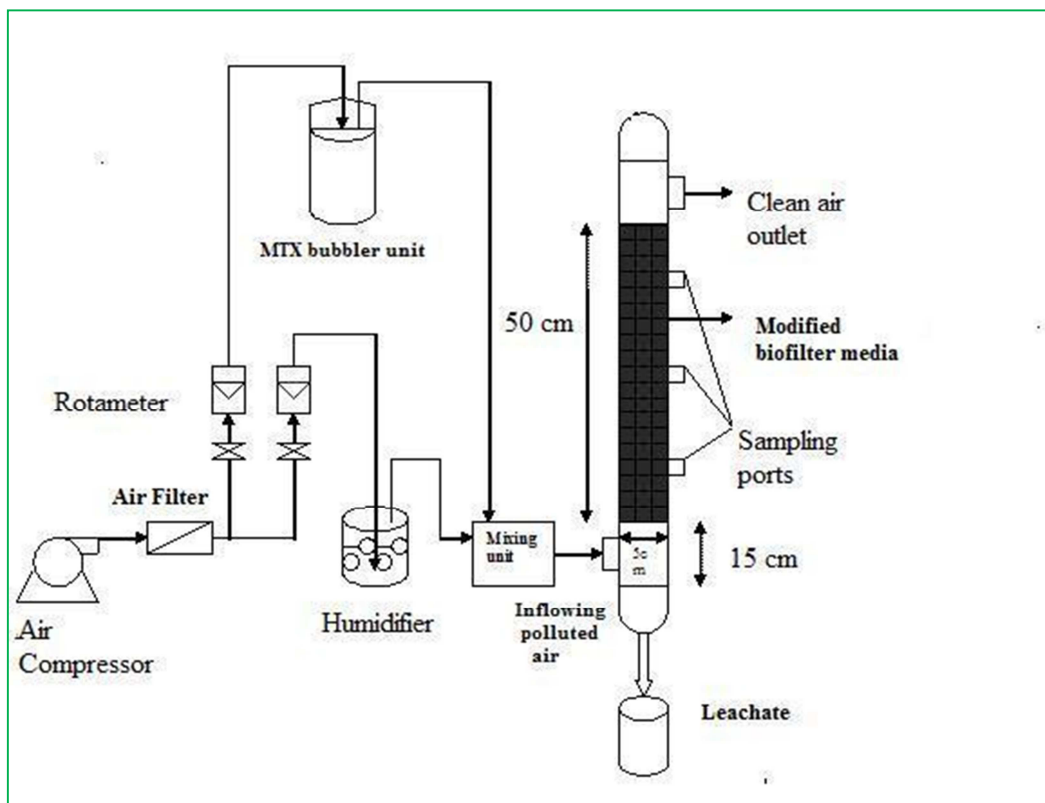


Fig a: Schematic set-up for the biofilter column.

Biofilter operation and its performance were evaluated in terms of percentage removal efficiency (RE) and pollutant elimination capacity (EC) as a function of pollutant loading at different EBRTs (Singh et al., 2006). Pressure drop across the bed height was measured at regular interval of time, with the help of U-tube manometer filled with water. Ambient and bed temperatures and relative humidity of inlet streams were measured by digital humidity – cum – temperature meter (Model No. DT-615). The pH of the leachate was also monitored regularly. The results of continuous bio-filtration runs were used to calculate various kinetic constants and operating parameters.

3. Results and discussion:

Without external supply of nutrient solution the biofilter demonstrated very good performance throughout the operation. Phyco-chemical

characterization results indicated improvement in some of the important properties (bed porosity, density etc.) of composite beads as compared to base material (wood charcoal) using which it was prepared (Table 1). No significant variation in the pressure drop across the bed was observed during whole operation. During whole operation inlet stream and bed temperature variations were found in the range of 24.5 - 33.2 and 25.9 - 34.3°C respectively. Bed temperature was always found more than the temperature of inlet stream which might be due to exothermic bioreaction in the biofilter. The pH of the leachate was found almost constant (6.93 - 7.02) and this may be due to the buffer capacity of phosphate solution in which the composite beads were immersed during the preparation. Relative humidity of inlet stream was found in the range of 80-95%.

Table 1: Initial characteristics of PVA/Charcoal/ KNO_3 composite beads and Wood Charcoal.

Sr. No.	Parameters	Composite bead	Wood charcoal cubes
1.	Particle size (mm)	6-10	10 - 15
2.	Dry weight g	0.4755	0.6532
3.	Moisture retention capacity %	82.8	64.4
4.	Bead porosity %	77	68
5.	Density g/cm^3	0.767	0.65
6.	C H N Content %		
	C	68.63	43.25
	H	3.65	1.635
	N	2.29	1.415

3.1 Batch Adsorption Tests:

Prior to the biofilter experiments, the batch isotherm tests were performed to determine the adsorption capacity of the modified media for MTX vapour as given in Singh et al. (2010). Equilibrium concentration in gas phase (C_e) and adsorbed solid phase concentration (q_e) of MTX on the composite beads were used to calculate Freundlich isotherm coefficients (K_F and N) using non-linear best fit method. The values of K_F and N were found to be 5.68 and 0.62. The value of K_F and N for composite beads is less than wood charcoal (Singh et al., 2010) which indicates reduction in the adsorption capacity of modified media as compared to wood charcoal.

3.2 Biofilter performance in terms of MTX removal

The continuous biodegradation of MTX vapour was carried out for a period of 56 days. This period was divided in three distinct phases as shown in Table 2. During the acclimation phase the biofilter was operated for 14 days with an initial flow rate of

$0.06 \text{ m}^3 \text{ h}^{-1}$ and initial inlet concentrations of 0.073, 0.26, and 0.047 g m^{-3} respectively for MTX (Fig. b). RE increased rapidly from very low value at start (>40%) reached at 78.7, 85.6, and 80.9% respectively for MTX on the 14th day.

On 14th day, flow rate was increased to $0.124 \text{ m}^3 \text{ h}^{-1}$ (Phase I). The inlet concentrations of MTX vapour in this phase were varied in the range of 0.51-1.81, 0.73 -2.6 g m^{-3} and 0.562 -2.34 g m^{-3} respectively and the removal efficiency was observed in the range of 93% to 97% for all components. The maximum overall RE of 96.55% was achieved at an average inlet concentration of 0.7 g m^{-3} and at loading rate of $235.65 \text{ g/m}^3 \text{ h}$. Further increase in inlet concentration and loading rates, reduce the removal efficiency to 74% may be due to high loading rates. In this phase initially the removal efficiency increased with increase in concentration and loading rates but further increase in loading rates resulted in decrease of the removal efficiency due to high concentrations.

Table 2: Operating conditions of each phase in the bio filter experiments for MTX.

Phase	Operating days	Flow Rate $\text{m}^3 \text{h}^{-1}$	Range of average pollutant concentrations, g m^{-3}			Total inlet loading range(MTX), $\text{g m}^{-3} \text{h}^{-1}$	EBRT sec
			MEK	Toluene	Xylene		
Acclimation	0-14	0.061	0.073-0.26	0.26-0.49	0.047-0.21	23-58.1	57.9
Phase I	15-34	0.124	0.51-1.81	0.73-2.60	0.56-2.34	222.1-848.97	28.4
Phase II	35-56	0.191	0.18-1.24	0.37-2.04	0.266-1.71	164.9-895.65	18.5

The phase III lasted, for 23 days from 35-56 days. During this last phase gas flow rate was increased to $0.18 \text{ m}^3 \text{h}^{-1}$ with corresponding EBRT of 18.5 sec. The inlet concentration of MTX vapour was varied between 0.18 to 1.24 g m^{-3} , 0.37 to 2.04 g m^{-3} , and 0.26 to 1.71 g m^{-3} . With sudden decrease in concentration, the removal efficiency increased gradually and reached 95.67%. with further increase in concentration along with loading rates the removal efficiency decreased and reached a value of 74.36% at a high total loading rate of $895.65 \text{ g/m}^3\text{h}$. Removal efficiency was increased to 95.67% as concentration increased. At an average inlet concentration of 1.66 g m^{-3} removal efficiency decreased to 71%. The column behaviour throughout the three phases is observed to be same. With increase in inlet concentrations and loading rates removal efficiency increased but further increase in loading rates and inlet concentrations resulted in the decrease of removal efficiencies of the mixture. A maximum removal efficiency of 96.35% for MEK, 97.87% for Toulene and 95.2% for Xylene are achieved during entire biofilter operation. Biofilter responded very quickly for the change in flow rates and inlet concentration. This result shows an active bio film present in the biofilter.

In the present study, the RE is obtained as 96.55% achieved during phase II (after 18 days) by the use of Composite Beads bed made of wood charcoal and acclimated mixed culture for an total inlet MTX load of $222-297 \text{ g m}^{-3} \text{h}^{-1}$ while Mathur and Majumder (2008) showed a removal efficiency of around 99% after 40 days for the removal of MTBX using coal as the packing material for a lower inlet load. While Qi et al. (2006) using two biofilters one

with continuous loading and other with intermittent loading carry out similar phase-wise studies for the removal of paint solvent mixture. As compared with the systems reported by previous studies, the time taken to achieve maximum removal efficiency in the present study is less for higher inlet loads and the column worked successfully under high loading rates. An increase in the inlet pollutant concentration enhances the transfer rate of the pollutant to the bio film. However higher concentrations of some recalcitrant pollutants may produce inhibition effects on the metabolic activity of the microbial population. On the other hand, the elimination capacity reflects the capacity of a biofilter to remove the pollutants. The maximum elimination capacity of a biofilter depends on the microbial population and their activities of the VOC-oxidizing bacteria exist in the biofilter media. Fig. c shows the effect of different inlet loading rates on removal efficiency and elimination capacity. As the inlet concentration of MTX vapour increases the value of corresponding inlet loading and elimination capacity increases but an opposite trend was observed for the removal efficiency. The total inlet load of MTX during the experiment was in the range of 23 to $895.65 \text{ g m}^{-3} \text{h}^{-1}$.

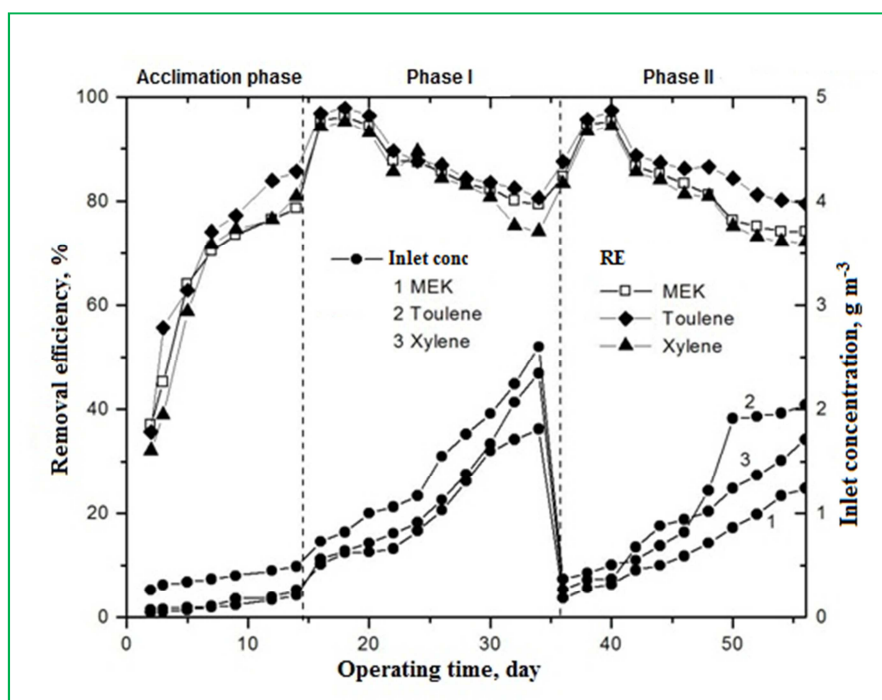


Fig b: Variation of removal efficiency with change in inlet concentration of MTX and gas flow rates.

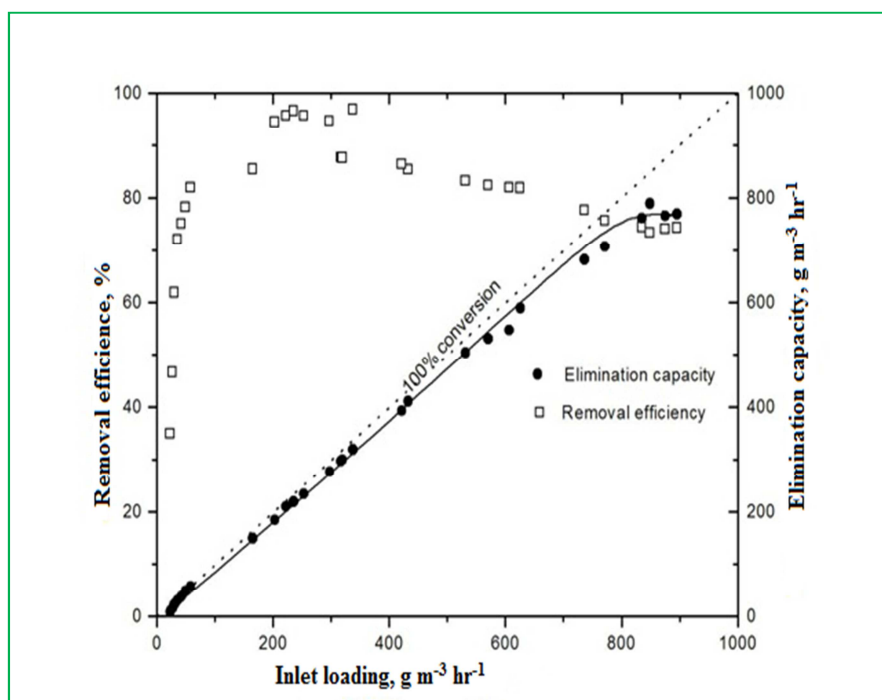


Fig c: Influence of MTX loading rate on elimination capacity and removal efficiency of biofilter

During the acclimation phase, the removal efficiency of MTX increased from 35% to 82% as total inlet load varied from 23 to 58.1 g m⁻³ h⁻¹. In phase I, total inlet load was increased from 222.10 to 848.97 g m⁻³ h⁻¹ and removal efficiency varied in the range of 95.63% to 74%. The maximum removal efficiency of 96.55% was achieved at a

total inlet load of 235.65 g m⁻³ h⁻¹. In phase II, the total inlet load decreased to 164.90 g m⁻³ h⁻¹ due to decrease in inlet concentration and varied up to 895.65 g m⁻³ h⁻¹. An average removal efficiency of 95.67% was achieved up to the total inlet load of 252.67 g m⁻³ h⁻¹. A nearly linear relation between the inlet load and the elimination capacity was

observed till a total loading rate of $625.81 \text{ g m}^{-3} \text{ h}^{-1}$, shows that up to this loading the overall process may be controlled by mass transfer mechanism. In phase II, the maximum elimination capacity of $769.23 \text{ g m}^{-3} \text{ h}^{-1}$ was observed at total inlet load of $895.65 \text{ g m}^{-3} \text{ h}^{-1}$, if the loading rate was increased beyond this value the process may reach in the reaction controlling zone.

In the acclimation period, the maximum total elimination capacity of $56.93 \text{ g m}^{-3} \text{ h}^{-1}$ was achieved at total inlet loading rate of $58.10 \text{ g m}^{-3} \text{ h}^{-1}$ and in phase I, the maximum elimination capacity of $779.98 \text{ g m}^{-3} \text{ h}^{-1}$ was achieved at total inlet load of $848.97 \text{ g m}^{-3} \text{ h}^{-1}$. Further increase in loading rate resulted decrease in removal efficiency and constant elimination capacity.

3.3 Production of Carbon Dioxide:

Carbon dioxide Production (PCO_2) is an important factor indicating the degree of VOCs mineralized completely, because VOCs are finally biodegraded to water and carbon dioxide and utilized as carbon source to format biomass for microbial growth Delhomenie et al., (2002). The PCO_2 was measured in this work for evaluating the degree MTX mineralization. Assuming that a complete oxidation and "mineralization" of the mixture of MTX took place, *i.e.* ignoring the biomass generated, the stoichiometric ratio of total carbon dioxide produced/VOCs(MTX) consumed (*i.e.*, PCO_2/EC) was 3.03. Fig. d presented the evolution of the practical production of carbon dioxide

(PCO_2) versus elimination capacity (EC) during the whole experimental period. It could be seen in Fig. 4 that a linear correlation was found between elimination capacity and production of carbon dioxide. Such a linear behaviour was also observed in reported work by Mathur *et al.*, (2007). The linear relation obtained from the data points was formulated as following equation:

$$\text{PCO}_2 = 2.355 \text{ EC} + 8.5 \quad (1)$$

The PCO_2 value was $8.5 \text{ gm}^{-3} \text{ h}^{-1}$, corresponding to no carbon source introduced into the biofilter (*i.e.*, the value of EC was zero), which was likely due to endogenous respiration of microorganism populations that do not utilize VOCs (MTX) as carbon source and the desorption of CO_2 generated by VOCs (MTX) biodegradation from the packing media. The discrepancy between the theoretical ratio (3.03) and the practical ratio (2.335) provided by Eq. (1), indicated that conversion to carbon dioxide was not the only fate of the carbon source (MTX) in a biofilter. The fact also suggested that approximate 77% of VOCs (MTX) were completely mineralized and the remaining 23% might be accumulated in various forms, such as biomass, extracellular polymers, degradation intermediates, and/or other forms, including HCO_3^- , H_2CO_3 , CO_3^{2-} . On the other hand, the difference between the theoretical ratio (3.03) and the practical ratio (2.335) indicated that removed VOCs (MTX) were mainly eliminated by biodegradation.

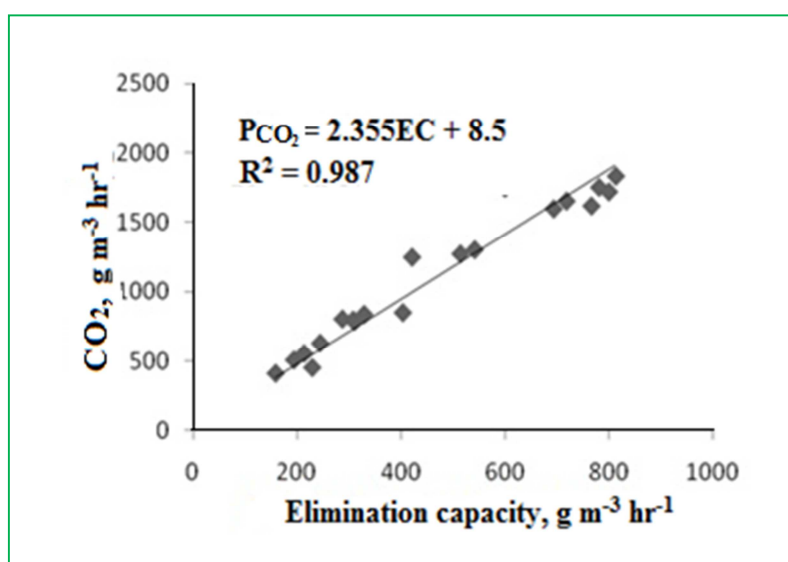


Fig d: Production of carbon dioxide vs. Elimination capacity

3.4 Kinetic Analysis:

Under the condition of no oxygen limitation in the biofilter and overall biodegradation rate being controlled only by the bio-reaction, the EC should commonly follow the Michaelis-Menten model (Hirai et al., 1990; Ramirez et al., 2008). The linear form of the Michaelis-Menten model gives which is commonly known as Lineweaver–Burk equation.

$$\frac{1}{EC} = \frac{K_s}{EC_{max} C_{in}} + \frac{1}{EC_{max}} \quad (2)$$

Here EC_{max} is the maximum EC in the biofilter without any inhibition or substrate limitation, K_s is the saturation constant corresponding to C_{in} at which $EC = (EC_{max}/2)$, and C_{in} is the logarithmic average of the inlet and outlet concentrations of VOCs in the gas phase. The kinetic constants were calculated using equation (2). The value of EC_{max} was found to be 0.088, 0.101, 0.103 $g\ m^{-3}\ s^{-1}$ for methyl ethyl ketone (MEK), toluene and xylene respectively. Similarly, the value of K_s was 1.043, 0.826, 0.696 $g\ m^{-3}$ respectively (Fig. e). In all calculations correlation coefficient (R^2) was more

than 0.95. Similar values of EC_{max} and K_s have been reported by Mathur et al. (2007), Krailas et al. (2004) and Raghuvanshi et al. (2009). It is clear that value of K_s obtained for MTX is neither following the condition $K_s \gg C_0$ nor $K_s \ll C_0$ (for Phase I and II) and so zero-order kinetic with diffusion limitation model proposed by Ottengraf-van den Oever (1983) will be most appropriate to describe the biochemical reaction process.

3.6 Modelling with Ottengraf–van den Oever model:

The Ottengraf’s zero order diffusion limitation model based on the steady - state plug flow behavior of the contaminants through a bed of solid filter particles coated with a biologically active biofilm is the widely used model. According to this model two distinguishable zones- diffusion and reaction limited zones, occur and EC is correlated with the inlet loading rate (IL), depending on the process limitation as given below

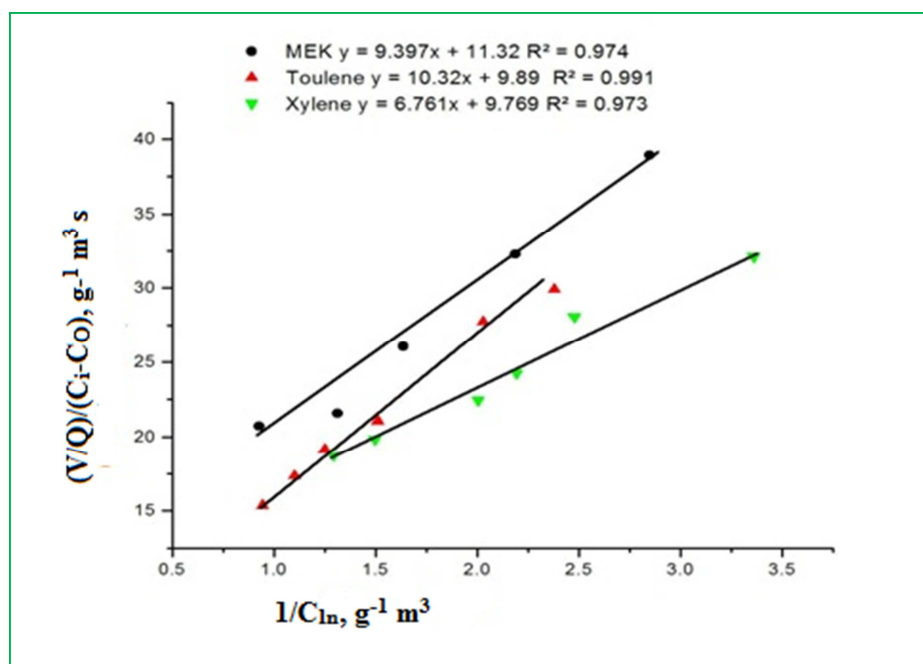


Fig e: Determination of Michaelis–Menten kinetic constants of MEK, Toluene, and Xylene

1) Zero-order reaction with diffusion limited zone: EC can be calculated by the following equation:

$$EC = IL \left(1 - \left(1 - K_1 \sqrt{\frac{EBRT}{IL}} \right)^2 \right) \quad (3)$$

where,

$$K_1 = \sqrt{\frac{k_0 D_e a}{2m\delta}} \quad (4)$$

where k_0 is the zero-order kinetic constant, D_e is the effective diffusion coefficient ($m^2\ h^{-1}$), m is the Henry’s coefficient, a is the interfacial area per unit volume (m^{-1}), δ is the biofilm thickness (μm).

The gas phase VOC concentration at the outlet of the biofilter column is obtained by the following equation;

$$\frac{C_{go}}{C_{gi}} = \left[1 - EBRT \frac{K_1}{\sqrt{C_{gi}}} \right]^2 \quad (5)$$

2) Zero-order with reaction limitation kinetics is given by Eq 6.

$$EC = K_0 \quad (6)$$

where, K_0 corresponds to the maximum elimination capacity in that phase.

The critical inlet concentration ($C_{critical}$) is defined as the limiting inlet concentration at which the biodegradation mechanism changes from the diffusion-controlled to reaction rate controlled and is given by Eq (6):

$$C_{critical} = \frac{1}{4} \left(\frac{K_0}{K_1} + K_1 EBRT \right)^2 \quad (7)$$

The critical inlet load is calculated by using Eq. (8)

$$I_{critical} = C_{critical} \times \frac{Q}{V} \quad (8)$$

The experimental data obtained during steady state operation for all the phases except the acclimation phase have been used to test the model. The values of K_1 for MEK, T and X are obtained by linear regression of $1 - (C_0/C_i)^{0.5}$ vs. $1/(C_i)^{0.5}$ and given in Table 3. Using the value of K_1 critical inlet concentration ($C_{critical}$) was calculated for Phase I and II and found to be more than the maximum inlet concentration of MEK, T and X applied in that particular phase (Table 2) which confirms the presence of diffusion controlling mechanism in these phases. Raghuvanshi and Babu (2009) and Alvarez-Hornos et al. (2008) also used this model to analyse their data on biofiltration of MEK and ethylbenzene respectively and observed a similar trend. Using the value of $C_{critical}$ critical inlet loading rate ($I_{critical}$) was also calculated for Phase I and II (Table 3).

Table 3: Model parameters for various operating conditions.

Phase	Parameters	MEK	Toluene	Xylene
Phase I	$K_0(g\ m^{-3}\ h^{-1})$	221.23	315.5	269.54
	$K_1(g^{0.5}\ m^{-1.5})$	82.46	97.46	81.54
	$IL(g\ m^{-3}\ h^{-1})$	63.76-232.26	89.33-329.94	68.90-297.17
	$C_i(g\ m^{-3})$	0.51-1.81	0.73-2.60	0.562-2.34
	$IL_{critical}(g\ m^{-3}\ h^{-1})$	291.12	438.56	393.1
	$C_{critical}(g\ m^{-3})$	2.77	3.97	3.89
Phase II	$K_0(g\ m^{-3}\ h^{-1})$	195.49	352.67	265.23
	$K_1(g^{0.5}\ m^{-1.5})$	103.49	118.12	107.2
	$IL(g\ m^{-3}\ h^{-1})$	48.49-218.63	68.65-325.67	47.76-276.63
	$C_i(g\ m^{-3})$	0.18-1.4	0.37-2.04	0.266-1.71
	$IL_{critical}(g\ m^{-3}\ h^{-1})$	284.108	440.67	345.03
	$C_{critical}(g\ m^{-3})$	1.46	2.82	2.287

4.0 Conclusions:

The results obtained during the experimental period illustrate the effective removal of paint solvent mixture (MTX) in vapor phase using microorganisms supported on PVA/Charcoal/ KNO_3 Composite beads in a laboratory scale biofilter without periodical supply of nutrients. Characterization results shows that composite beads possess most of the favorable biofilter media properties such as; high porosity, low bulk density,

high water retention capacity and pH buffer capacity. The modified biofilter media shows good mechanical stability and negligible pressure drop throughout the experimental period. The pH of the media was maintained in a range of 6.91 to 6.93 throughout the whole experimental period. During the whole experimental period the removal efficiency of MTX was obtained in the range of 74-97%. The results revealed that when influent MTX loading were less than $600\ g\ m^{-3}\ h^{-1}$, good removal

efficiencies could be achieved. Maximum removal efficiency of toluene (97.87%) followed by methyl ethyl ketone (96.54%), Xylene (95.20%) was achieved. Maximum elimination capacities of MTX during the experiment were 220.23, 325.77, 269.23 g m⁻³ h⁻¹, respectively.

References:

- 1) Alvarez-Hornos, F.J., Gabaldon, C., Martinez-Soria, V., Martin, M., Marzal, P., Penyaraja, J.M., 2008. Biofiltration of ethylbenzene vapours: influence of the packing material. *Bioresource Technol.* 99(2), 269–276.
- 2) Boswell, J.E.A., 2001. Biofiltration of VOCs from Paint Manufacturing. *Air & Waste Manage. Assoc.*, Orlando, Florida.
- 3) Chan, W.C., Lin, Z.Y., 2006. A process to prepare a synthetic filter material containing nutrients for biofiltration. *Bioresource Technol.* 97 (15), 1927–1933.
- 4) Delhomenie, M.C., Bibeau, L., Bredin, N., Roy, S., Broussau, S., Brzezinski, R., Kugelmass, J.L., Heitz, M., 2002. Biofiltration of air contaminated with toluene on a compost-based bed. *Adv. Environ. Res.* 6 (3), 239-254.
- 5) Hirai, M., Ohtake, M., Shoda, M., 1990. Removal kinetics of hydrogen sulfide, methanethiol and dimethyl sulfide by peat biofilter. *J. Ferment. Bioeng.* 70(5), 334-339.
- 6) Hirai, M., Kamamoto, M., Yani, M., Shoda, M., 2001. Comparison of the biological H₂S removal characteristics among four inorganic packing materials. *J. Biosci. Bioeng.* 91(4), 396–402.
- 7) Hernandez, J., Prado, J.O., Almarcha, M., Lafuente, J., Gabriel, D., 2010. Development and application of a hybrid inert/organic packing material for the biofiltration of composting off-gases mimics. *J. Hazard. Mater.* 178 (1-3), 665–672.
- 8) Kazenski, S.L., Kinney, K.A., 2000. Biofiltration of Paint Spray Booth Emissions: Packing Media Considerations and VOC Interactions, *Air & Waste Manage. Assoc.*, Salt Lake City, Utah.
- 9) Krailas, S., Tongta, S., Meeyoo, V., 2004. Macrokinetic determination of isopropanol removal using a downward flow biofilter. *Environ. Hazard. Manag.* 26(1), 55–64.
- 10) Leson, G., Winer, A.M., 1991. Biofiltration: an innovative air pollution control technology for VOC emissions. *J. Air Waste Manage. Assoc.* 41 (8), 1045–1054.
- 11) Lu, C., Lin, M., Wey, I., 2001. Removal of EATX from waste gases by a trickle bed air biofilter. *J. Environ. Eng.* 127 (10), 946–951.
- 12) Liang, J., Koe, L., Chiaw, C.K.L., X. Ning., 2007. Application of biological activated carbon as low pH Biofilter medium for gas mixture treatment. *Biotechnol. Bioeng.* 96 (6), 1092–1100.
- 13) Mohseni, M., & Allen, D. G., 2000. Biofiltration of mixtures of hydrophilic and hydrophobic volatile organic compounds. *Chem. Eng. Sci.* 55 (9), 1545-1558.
- 14) Mathur, A.K., Majumder, C.B., Chatterjee, S., 2007. Combined removal of BTEX in air stream by using mixture of sugar cane bagasse, compost and GAC as biofilter media. *J. Hazard. Mater.* 148(1-2), 64-74.
- 15) Mathur, A.K., Majumder, C.B., 2008. Biofiltration and kinetic aspects of a biotrickling filter for the removal of paint solvent mixture laden air stream. *J. Hazard. Mater.* 152 (3), 1027-1036.
- 16) Ottengraf, S.P., and van der Oever, A.H.C., 1983. Kinetics of organic compound removal from waste gases with a biological filter. *Biotechnol. Bioeng.* 25(12), 3089–3102.
- 17) Qi, Bing., Moe, W. M., and Kinney, K. A., 2005. Treatment of paint spray booth off-gases in a fungal biofilter. *J. Environ. Eng.* 131 (2), 180–189.
- 18) Ramirez, A.A., Benard, S., Giroir-Fendler, A., Peter Jones, J., Heitz, J., 2008. Kinetics of microbial growth and biodegradation of methanol and toluene in biofilters and an analysis of the energetic indicators. *J. Biotechnol.* 138(3-4), 88-95.
- 19) Raghuvanshi, S., Babu, B.V., 2009. Experimental studies and kinetic modeling for removal of methyl ethyl ketone using biofiltration. *Bioresource Technol.* 100 (17), 3855–3861.
- 20) Singh, K., Singh, R.S., Rai, B.N., Upadhyay, S.N., 2010. Biofiltration of toluene using wood charcoal as the biofilter media. *Bioresource Technol.* 101 (11), 3947–3951.
- 21) Singh, R.S., Agnihotri, S.S., Upadhyay, S.N., 2006. Removal of toluene vapours using agro-waste as biofilter media. *Bioresource Technol.* 97 (18), 2296–2301.
- 22) Singh R.S., Rai B.N., Upadhyay S.N., 2006. Performance evaluation of an agro waste based biofilter treating toluene vapour, *Environmental Technology*, 27(4), 349-357.
- 23) Upadhyay, S.N., Kumar, V., 2004. Biological control of air pollutant. In: Pandey, A. (Ed.), *Concise Encyclopedia of Biores. Technol.*, first ed. The Haworth Reference Press, New York.