



# Performance, influence of operational parameters and optimization of a corn cob based biofilter treating gas-phase mixture of MTBX

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## Abstract

The complete performance of a corn cob biofilter inoculated with microbial culture was optimized for the treatment of polluted air containing gas phase of MTBX. The biofilter was operated for the period of 149 days and this time period was divided into five phases (I-V) to differentiate between the outputs of biofilter. On the removal efficiency (RE) and elimination capacity (EC), the effects of various parameters like gaseous concentration and flow rate were investigated. The removal efficiency was measured with respect to inlet concentration of each component of MTBX and the elimination capacity was varied with change in loading rate of gaseous mixture. The maximum RE in MTBX was of *n*-butyle acetate (BA) with 99.98% with respect to inlet concentration of 0.118992 gm<sup>-3</sup> while the minimum RE was of *o*-xylene with 98.21% for the concentration of 0.189568 gm<sup>-3</sup>. In case of EC the maximum value was obtained as 86.27 g m<sup>-3</sup> h<sup>-1</sup> for *n*-butyle acetate at the highest value of inlet load of 114.69 g<sup>-3</sup> h<sup>-1</sup>. By performing an analysis of variance the results were statistically interpreted to elucidate the main and interaction effects.

**Keywords:** MTBX, Removal efficiency (RE), Elimination capacity (EC), Loading rate, Inlet concentration.

## Introduction

Characteristics of air pollution depend on the source of waste gas emissions, the type and concentration of existing contaminants in the gas stream<sup>1</sup>. Many air contaminants which are responsible for air contaminants can be produced from poultry buildings, livestock, on-site manure storage and treatment facilities. Generally these pollutants can be classified into four groups i.e. fixed gases or gases, odors, particulate matter (PM), and volatile organic compounds (VOC)<sup>2</sup>. In the petrochemical based VOCs, the composition of gaseous phase contain a mixture of solvents including ketones (e.g., methyl ethyl ketone, methyl isobutyl ketone, methyl propyl ketone), aromatic hydrocarbons (e.g., toluene and xylenes), and esters (e.g., *n*-butyl acetate). One of the most toxic ketone compounds is Methyl ethyl ketone (MEK) which is released into the atmosphere by various industries like chemical, petrochemical, food processing, pulp and paper mills, color printing, paint and coating, electronic industries, etc<sup>3</sup>.

In the many research studies, removal of individual volatile organic compounds (VOCs) from contaminated air streams has been found, and for these kinds of removal different removal technologies such as chemical or biological or a combination of both techniques have been proposed<sup>4,7</sup>. In comparison to biological techniques, the techniques based on chemical methods for treating VOCs have proven very expensive due to high maintenance, energy and operational costs<sup>8</sup>. Along with lower cost biological techniques are economically efficient as

well as friendly to environment for treating VOCs at low concentrations and high gaseous flow rates, they also produce offensive end-products<sup>9</sup>. Due to all above mention features biofilter is considered as most suitable, commonly used and also consider as successful reactor for the applications of various industrial sector<sup>10,11</sup>. From the efficiency point of view of the biomass which is used in a biofilter for treating the gaseous pollutants, according to a recent study it has been proved that degradation efficiency of bacterial biofilters are more than fungi based biofilters and the reason behind this is due to the presence of a highly varied microbial population used in bioreactor<sup>12</sup>.

In the previous study<sup>13</sup>, two different packing medium was used Press Mud and Corn stack for the removal of MEK by using biofiltration process. Experiments were performed for a period of 200 days and this experimental operation was divided into phases (I, II, III and IV) according to Empty Bed Residence Time (EBRT). The inlet concentration of pollutant was varied from 0.2 to 1.2 gm<sup>-3</sup>. The EBRT was varied from 2.81 to 0.7 min. The maximum RE is observed at the EBRT of 2.81 min, the RE is 97.3%<sup>13</sup>.

The objective of current research was to evaluate the steady state removal efficiency (RE) of gaseous phase of MEK, toluene, *n*-butyle acetate and *o*-xylene (MTBX) mixture with respect to different inlet concentrations and gaseous flow rates in a lab scale corn cob based biofilter. Inlet concentrations for MEK (0.073 - 0.59 g m<sup>-3</sup>), toluene (0.096 - 0.89 g m<sup>-3</sup>), *n*-butyl acetate (0.10 - 2.16 g m<sup>-3</sup>) and *o*-xylene (0.10 - 0.57 g m<sup>-3</sup>) were

varied from low to high level. The gas flow rates and EBRT in this experimental research were kept as 3-8 L min<sup>-1</sup> and 3.06 – 1.15 min respectively. The performance of the biofilter was tested by operating the biofilter for the periods of 1-149 days.

## Materials and methods

**Biofilter and operation:** For housing this biofiltration assembly a plywood chamber of size 1.2 m × 1.2 m × 2.2 m height was fabricated and all experiments were conducted in that temperature controlled chamber at 30 ± 2 °C. The double walled chamber was fabricated from 6 mm plywood sheets and glass wool was filled in between the walls to insulate the chamber. The chamber was equipped with the thermostatic heating device, operated and controlled by digital temperature controller (ESCORT, Japsin Pvt. Ltd, New Delhi). The temperature range could be controlled from room temperature to 70 °C with an accuracy ± 0.2 °C. Three individual sections were present in the bioreactor columns which were bolted together. These three sections were packed with packing material up to a height of 30 cm. To allow the sampling of gas for the redistribution of the contaminant stream between sections a 3 cm plenum was located between two sections. At the time of measuring bed depth, since there are not any conversions in these sections so only reactor height has been considered excluding plenum height. For the homogeneous distribution the packing material was supported on the acrylic sieve plate.

For starting the biofilter all packing material corn-cob was mixed with nutrient solution (BSM). Various nutrients (Table-1) were supplied on to the packing materials, these include KH<sub>2</sub>PO<sub>4</sub>, KNO<sub>3</sub>, (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub>, NH<sub>4</sub>Cl, NH<sub>4</sub>HCO<sub>3</sub>, CaCl<sub>2</sub>, MgSO<sub>4</sub>, MnSO<sub>4</sub>, FeSO<sub>4</sub>, Na<sub>2</sub>MoO<sub>4</sub>, and vitamins (B1, etc.) Initially the pH value this mixture was kept at 7.0 while temperature and moisture content were kept at 30°C and 5.1% respectively. After setting of operation parameters the bacteria *B. sphaericus* with MTBX and nutrient solution was continuously sprayed through the nutrients distribution system two times in a day for the time duration of 30 min on the top of the packing media. The distribution rate of nutrient solution was kept as 10 mL min<sup>-1</sup> to ensure suitable condition of moisture and nutrients for the activities of microorganism. When packing material was completely filled in the biofilter a gaseous stream of MTBX contaminated air was passed through the medium at constant loading rate from the bottom of the biofilter. To check the compatibility of result it was operated in five distinct phases (phase I-V) at different operating conditions. The concentrations of each components of MTBX were changed, and flow rate also vary from 3, 4, 5, 6 and 8 L min<sup>-1</sup> in Phase I, II, III, IV and V, respectively.

**Analytical methods:** Concentration of MTBX compounds present in the gaseous mixture was analyzed by using a Netel India Limited (model- MICHRO 9100) gas chromatograph equipped with a capillary column type HP5 (30m × 0.249mm × 0.25 µm film thickness) and with a flame ionization detector.

The temperature of the injector, oven and detector were maintained at 210°C, 60°C and 230°C respectively. As fuel the hydrogen gas was used and as carrier gas nitrogen was used at the flow rate of 20mL min<sup>-1</sup>. A known amount of MTBX injected into a sealed bottle equipped with a Teflon septum was used to prepare the calibration curve. At experimental temperature range of 30°C the injected amount of MTBX gaseous mixture was allowed to evaporate in the air space within the bottle. After that the calibration process starts with the drawn of air samples from the bottle by a 1 mL gas tight syringe and after that analyzed by gas chromatograph.

**Table-1:** Concentration of Nutrients used in Biofilter.

Composition	Concentration of the Constituents (g L <sup>-1</sup> )	Essential nutrients	Concentration of the essential nutrients (g L <sup>-1</sup> )
Macro nutrients			
KH <sub>2</sub> PO <sub>4</sub>	0.91	P	0.207
K <sub>2</sub> HPO <sub>4</sub>	0.4	P	0.071
Na <sub>2</sub> HPO <sub>4</sub> .12 H <sub>2</sub> O	2.39	P	0.207
KNO <sub>3</sub>	2.96	N	0.41
(NH <sub>4</sub> ) <sub>2</sub> SO <sub>4</sub>	1.97	NH <sub>4</sub> <sup>+</sup>	0.534
MgSO <sub>4</sub> .7H <sub>2</sub> O	2.0	Mg <sup>++</sup>	0.364
FeSO <sub>4</sub> .7H <sub>2</sub> O	0.2	Fe	0.04
NaHCO <sub>3</sub>	0.5	Na <sup>+</sup>	0.136
Micro nutrients			
MnSO <sub>4</sub> .7H <sub>2</sub> O	0.88	Mn	0.175
ZnSO <sub>4</sub> .7H <sub>2</sub> O	0.04	Zn	0.009
CaCl <sub>2</sub> .2H <sub>2</sub> O	3.0	Ca <sup>++</sup>	0.816
Na <sub>2</sub> MoO <sub>4</sub> .2H <sub>2</sub> O	1.0	Mo	0.394
CoCl <sub>2</sub> .6H <sub>2</sub> O	0.04	Co	0.007

A gas tight syringe was used to analyze the air samples which were drawn from the various sampling ports. The concentration of CO<sub>2</sub> was determined with the help of GC equipped with a Porapak Q column (2 m length, 1/8' ID, 80/100 mesh) and thermal conductivity detector (TCD). Different samples of packing media were collected from each sampling point to determine the moisture content in the biofiltration unit. After collecting samples were heated at 105 °C in the oven. By finding the total weight different between both before and after

heating samples moisture contents in the packing media can be obtained.

A digital pH meter (model – NAINA NIG-333, India) was used to find pH values of nutrients solution and leachate from the biofilter. A spectrophotometer (Model Lambda 35, Parkin-Elmer, USA) was used to measure biomass concentration as well as optical density (OD) of the culture at 600 nm. The morphology of the packing materials and microorganisms before and after experiments can be understood by analyzing the samples using scanning electron microscope (SEM, LEO 435 VP, UK). The removal efficiency (RE) and elimination capacity (EC) of each component were calculated as follows:

$$RE = \left(1 - \frac{C_{go}}{C_{gi}}\right) \times 100$$
$$EC = \frac{(C_{gi} - C_{go})Q}{V}$$

Where  $C_{gi}$  is pollutant's inlet concentration used in the biofilter ( $\text{g m}^{-3}$ ),  $C_{go}$  is known as outlet concentration of pollutants obtained in the biofilter ( $\text{g m}^{-3}$ ), LR means inlet loading rate ( $\text{g m}^{-3} \text{h}^{-1}$ ) in biofilter, EC is elimination capacity ( $\text{gm}^{-3} \text{h}^{-1}$ ), V is the volume ( $\text{m}^3$ ) of biofilter bed and Q is the volumetric gas flow rate ( $\text{m}^3 \text{h}^{-1}$ ).

## Results and discussion

**Steady-state removal of gas-phase benzene and toluene mixture:** For evaluating the total performance of biofilter under variable loading rates, biofilter was operated on various range of MTBX concentration or on empty bed residence time (EBRT). The operation time duration was kept from 1-149 days. This conducted time period was further divided in five phases (I-V). The detail study of each phase for each element is described in the further sections. Various operational conditions were tested for several times and also at various times of intervals to ensure accuracy and compatibility of results. To evaluate the performance of the biofilter following parameters have also been studied in detail.

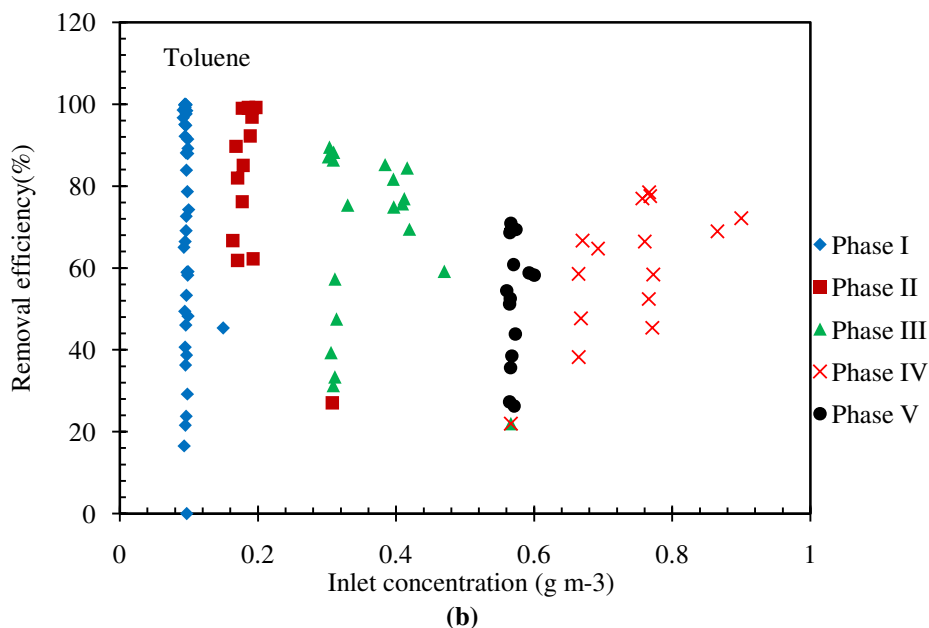
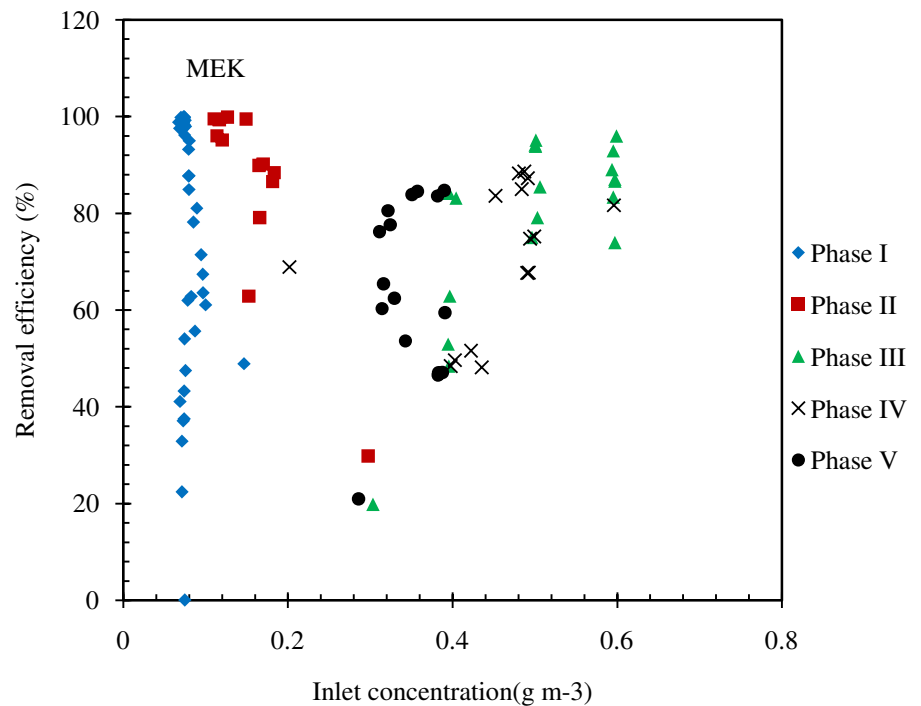
**Removal efficiencies:** Figure-1(a) shows the overall effect of inlet concentrations of MEK with respect to correspondent removal efficiency. From this figure it has been observed that the average loading rates and EBRT in all phases was increased during each phase change due to which a large variation in removal efficiencies have been found in each phase. With the variations of loading rates the observed maximum removal efficiency in each phase was 99.98%, 99.90%, 96%, 88.64% and 84.73% for phase I, II, II, IV and V respectively. The maximum removal efficiency of MEK was observed as 99.98% in phase I along with inlet concentration  $0.073233 \text{ gm}^{-3}$ . The relative inlet concentrations of these maximum efficiency was observed as  $0.126444 \text{ gm}^{-3}$ ,  $0.599059 \text{ gm}^{-3}$ ,  $0.487083 \text{ gm}^{-3}$  and  $0.389732 \text{ gm}^{-3}$  respectively. Figure-1(b) contains study of all phases of Toluene and shows that with the variations of loading rates the observed maximum removal efficiency in each phase

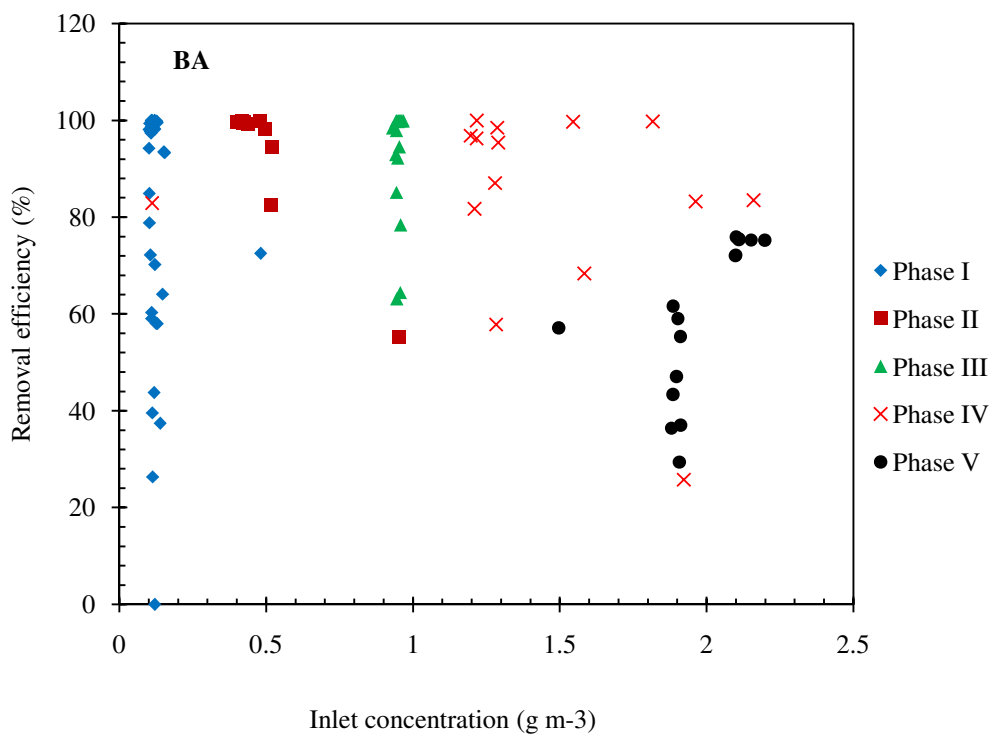
was 99.98%, 99.18%, 89.47%, 78.51% and 70.92% for phase I, II, II, IV and V respectively. The relative inlet concentrations of these maximum efficiency was observed as  $0.0948267 \text{ gm}^{-3}$ ,  $0.186182 \text{ gm}^{-3}$ ,  $0.303664 \text{ gm}^{-3}$ ,  $0.766282 \text{ gm}^{-3}$  and  $0.566282 \text{ gm}^{-3}$  respectively. The maximum removal of Toluene found in phase I with 99.98% and was continuously decreased during phase change. According to Figure-1(c) the overall effect of removal efficiency of n-butyl-acetate varies with respect to correspondent inlet concentrations. With the variations of loading rates the observed maximum removal efficiency in each phase was 99.98%, 99.88%, 99.94%, 99.73% and 75.87% for phase I, II, II, IV and V respectively. The relative inlet concentrations of these maximum efficiency was observed as  $0.118992 \text{ gm}^{-3}$ ,  $0.477967 \text{ gm}^{-3}$ ,  $0.962458 \text{ gm}^{-3}$ ,  $1.816612 \text{ gm}^{-3}$  and  $2.099732 \text{ gm}^{-3}$  respectively. The maximum removal efficiency of n-butyl-acetate was observed as 99.98% in phase I along with inlet concentration  $0.118992 \text{ gm}^{-3}$ . Study of all phases of o-xylene shows that with the variations of loading rates the observed maximum removal efficiency in each phase was 98.21%, 92.19%, 89.12%, 81.04% and 61.92% for phase I, II, II, IV and V respectively. The relative inlet concentrations of these maximum efficiency was observed as  $0.189568 \text{ gm}^{-3}$ ,  $0.148539 \text{ gm}^{-3}$ ,  $0.299948 \text{ gm}^{-3}$ ,  $0.367581 \text{ gm}^{-3}$  and  $0.432568 \text{ gm}^{-3}$  respectively. The maximum removal of o-xylene found in phase I with 98.21% and was continuously decreased during phase change.

**Elimination capacities:** The capacity of a bioreactor to remove the pollutants can be measured in terms of its elimination capacity and has been plotted in Figure-2a,b,c, and d as a function of inlet load. Significant variations of the Elimination Capacity (EC) and loading rate were observed in various phases in the change of influent concentration. As the influent MTBX loading increases the elimination capacities of MTBX were also increased. In the phase I MEK was treated at low volumetric load between  $1.45$  to  $1.95 \text{ gm}^{-3} \text{ h}^{-1}$  at this load the elimination capacity of MEK was observed from  $0.78$  to  $1.19 \text{ gm}^{-3} \text{ h}^{-1}$  respectively. As the loading rate increases in second phase II, III, IV & V from  $4.78$ ,  $19.47$ , and  $23.37$  up to  $20.38 \text{ gm}^{-3} \text{ h}^{-1}$  respectively, the EC also increases from  $4.22$ ,  $14.40$ ,  $19.08$  and  $12.11 \text{ gm}^{-3} \text{ h}^{-1}$  with respect to their load. From the figure it can be visualize that volumetric loading rate is continuously increased from one phase to the next phase but elimination capacity show a slight decrease which can be recovered later. From the graph it can be observe that the maximum elimination capacity is obtained as  $19.08 \text{ gm}^{-3} \text{ h}^{-1}$  in phase IV at the inlet load of  $23.37 \text{ gm}^{-3} \text{ h}^{-1}$ . Graphical representation of elimination capacities with their respective inlet loading of each phase of Toluene is presented in Figure-2(b). During phase I Toluene was treated at low volumetric load between minimum  $1.82$  to maximum  $1.88 \text{ gm}^{-3} \text{ h}^{-1}$  at this load the elimination capacity of Toluene was observed between  $0.30$  to  $1.87 \text{ gm}^{-3} \text{ h}^{-1}$  respectively. As the loading rate increases in second phase II, III, IV & V from  $5.14$ ,  $13.56$ , and  $35.28$  up to  $29.54 \text{ gm}^{-3} \text{ h}^{-1}$  respectively, the EC also increases from  $5.09$ ,  $11.44$ ,  $25.46$  and  $20.95 \text{ gm}^{-3} \text{ h}^{-1}$  with respect to their load. From the graph it can

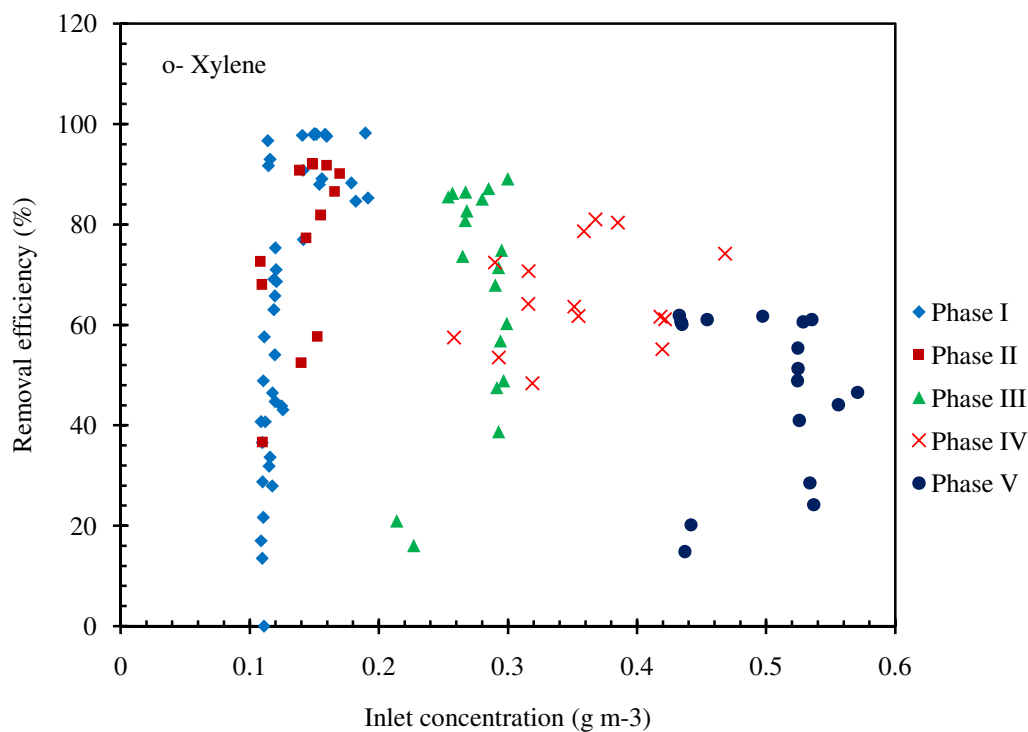
be observe that the maximum elimination capacity is obtained as  $25.46 \text{ g m}^{-3} \text{ h}^{-1}$  in phase IV at the inlet load of  $35.28 \text{ gm}^{-3} \text{ h}^{-1}$ . In case of *n*-butyl acetate initially the elimination was measured between between  $0.58$  to  $2.81 \text{ gm}^{-3} \text{ h}^{-1}$  at the loading of  $2.21$  to  $3.02 \text{ gm}^{-3} \text{ h}^{-1}$  respectively. With the increment of loading rate in phase II, III, IV & V from  $13.51$ ,  $31.53$  and  $71.23$  up to  $114.69 \text{ gm}^{-3} \text{ h}^{-1}$  respectively, the EC also increases from  $12.77$ ,  $31.50$ ,  $71.05$  and  $86.27 \text{ g m}^{-3} \text{ h}^{-1}$  with respect to their load. The maximum elimination capacity is obtained as  $86.27 \text{ gm}^{-3} \text{ h}^{-1}$  in phase V at the highest value of inlet load of  $114.69 \text{ gm}^{-3} \text{ h}^{-1}$ .

Again in phase I volumetric load of *o*-xylene was measured between  $2.14$  to  $3.75 \text{ gm}^{-3} \text{ h}^{-1}$  at this load the elimination capacity of *o*-xylene was observed between  $0.29$  to  $3.20 \text{ gm}^{-3} \text{ h}^{-1}$  respectively. With the increase in loading rate during second phase II, III, IV & V from  $4.42$ ,  $9.78$ , and  $18.35$  up to  $29.78 \text{ gm}^{-3} \text{ h}^{-1}$  respectively, the slightly increase in EC also measured from  $3.98$ ,  $8.71$ ,  $13.62$  and  $13.87 \text{ gm}^{-3} \text{ h}^{-1}$  with respect to their load. The maximum elimination capacity is obtained as  $17.06 \text{ g m}^{-3} \text{ h}^{-1}$  in phase V at the inlet load of  $27.93 \text{ gm}^{-3} \text{ h}^{-1}$ .



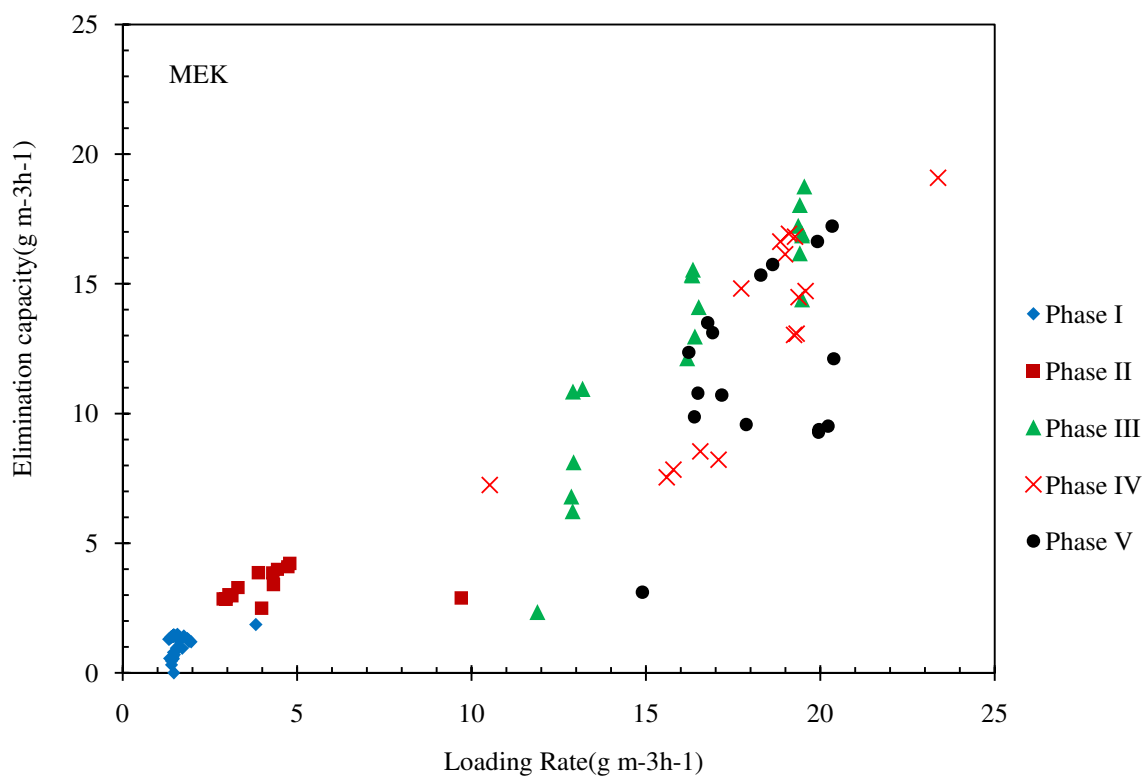


(c)

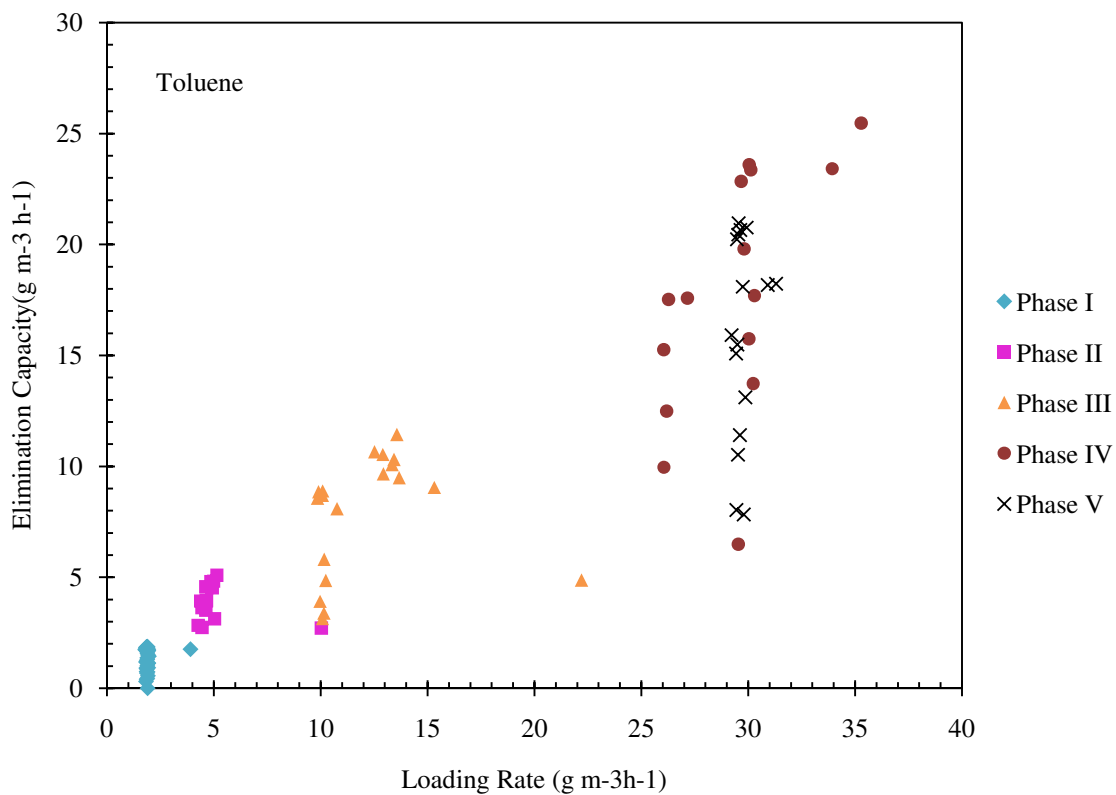


(d)

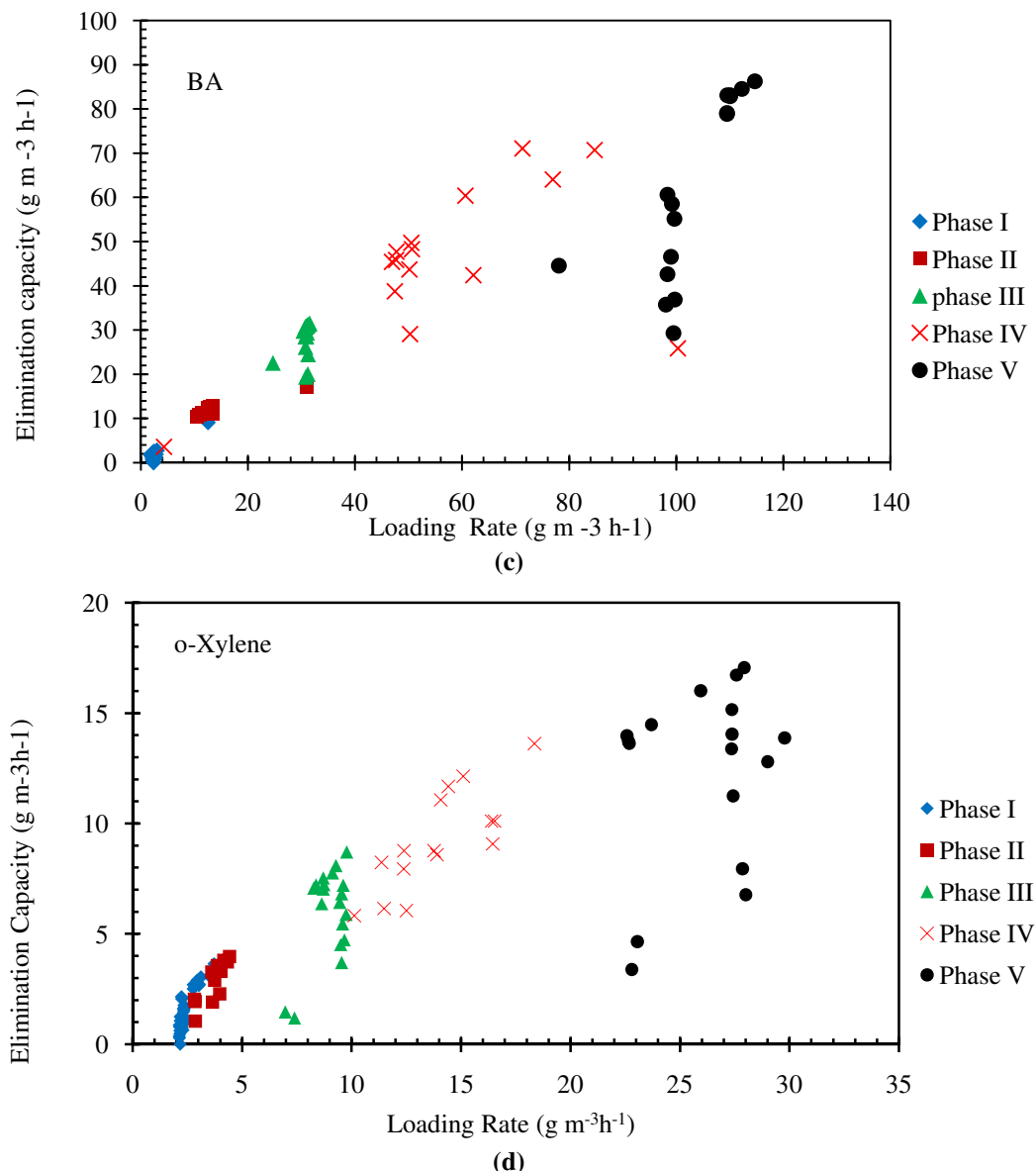
**Figure-1:** Removal efficiency of (a) MEK, (b) Toluene, (c) n-butyl acetate and (d) o-xylene in different phases with corresponds to respective inlet concentration.



(a)



(b)



**Figure-2:** Elimination capacity of (a) MEK, (b) Toluene, (c) n-butyl acetate (BA) and (d) *o*-xylene in different phases with corresponds to respective loading rates.

**Concentration profile of MTBX along with bed height:**

According to the results obtained during operation it has been observed that the removal efficiency was higher at the bottom of the reactor as compared to the obtained in the middle and on the top of the reactor. The reason behind this kind of performance of bioreactor may be the present of more carbon sources, moisture contents as well as nutrients present on the bottom of reactor due to which high degradation rate can be achieved. According to Figure-3(a) MTBX removal for the day 41 obtained. According to this figure in the first section (25 cm) the MTBX removal was 53%, 60%, 48% and 39% respectively while in the second section (45 cm) this removal was increased up to 79%, 80%, 75% and 89% respectively. Higher amount of removal was observed in the upper most part of biofilter.

MTBX removal profile in the phase II (65 day) was shown in Figure-3(b). For the first section of phase II the MTBX removal was 69%, 65%, 60% and 54% which was nearly same as previous phase. Removal profile of MTBX throughout the biofilter at the day 41, 65, and 102 was nearly stable but for the day 117 and 149 it was quite unstable. The curve presented in Figure-3 indicates that increased biofilter heights lead to greater contaminate removal (lower  $C_{go}/C_{gi}$ ). These observations show that in the greater biofilter heights, more biofilter media is available for the degradation of MTBX.

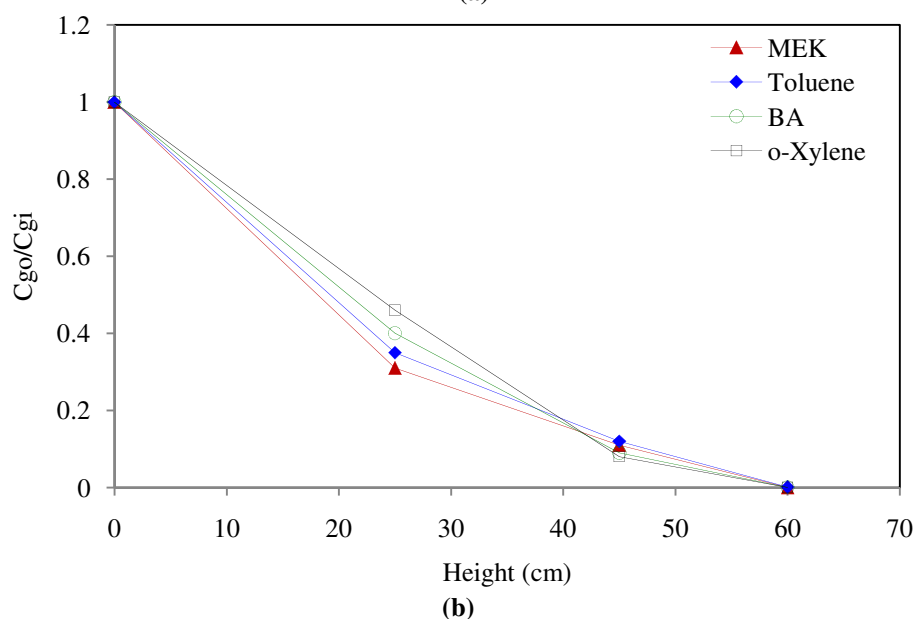
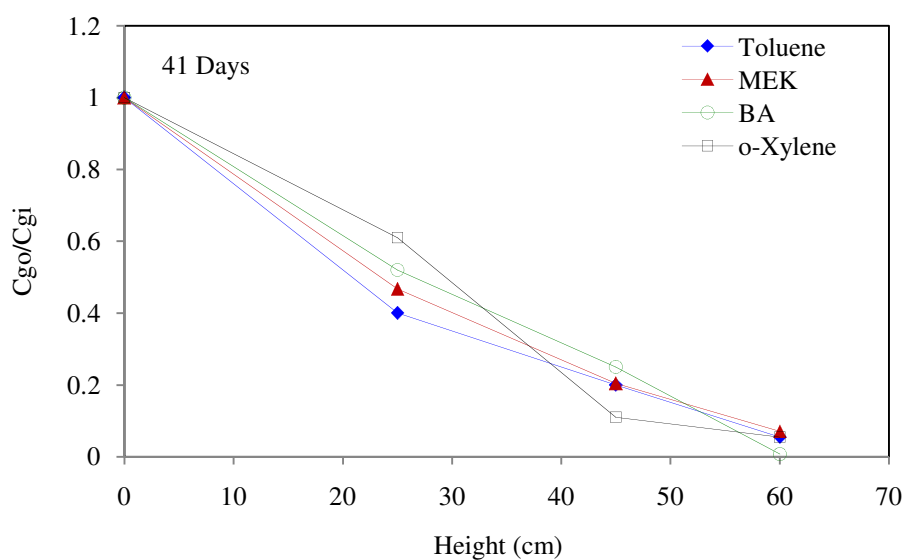
**Production of Carbon dioxide:** During biofiltration operation techniques, the organic pollutants which are present in gaseous stream are aerobically degraded into eco friendly components

such as water and carbon dioxide and these components, obtained after filtration process are used as the essential source of carbon for the growth microbial community. Hence, the concentration profile of carbon dioxide in the gaseous phase at the inlet and the outlet of the biofilter provide valuable information related to the biofilter performance<sup>13</sup>.

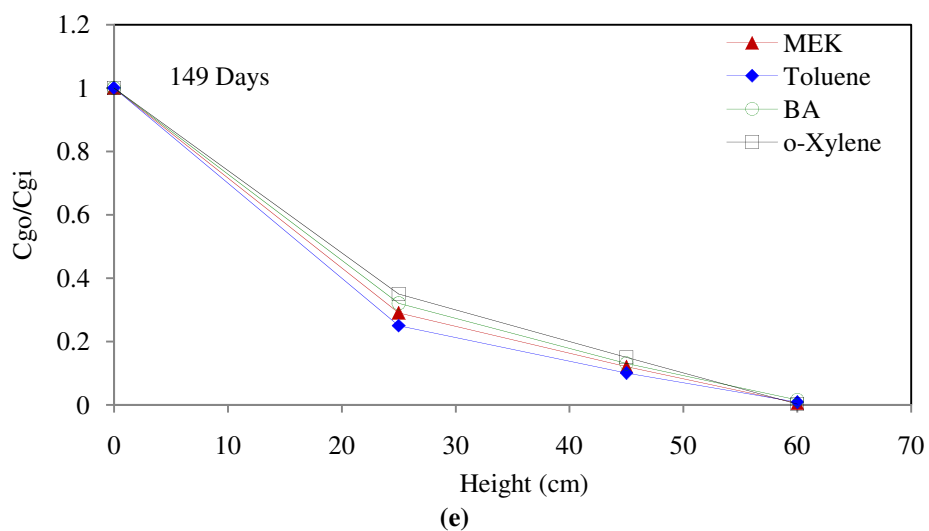
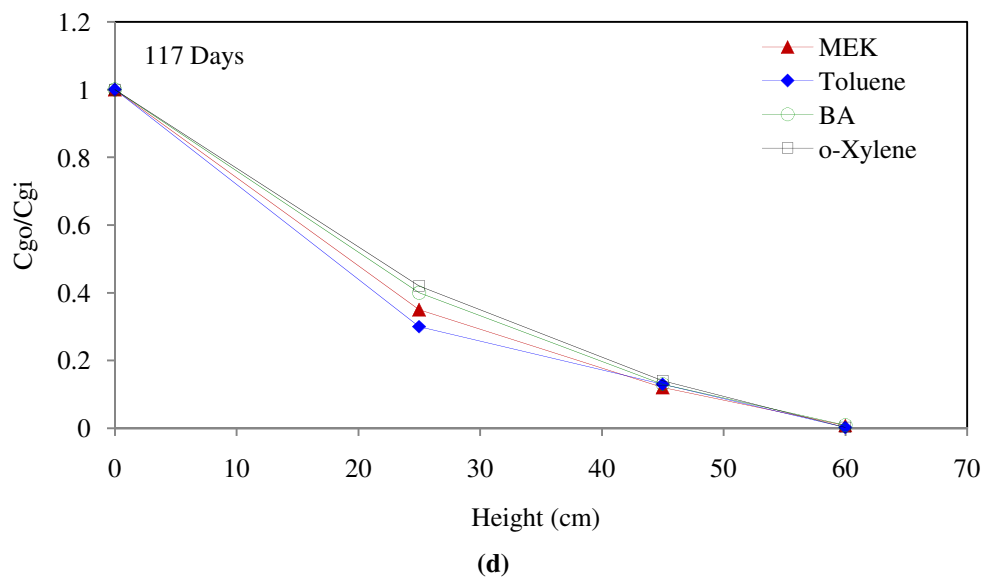
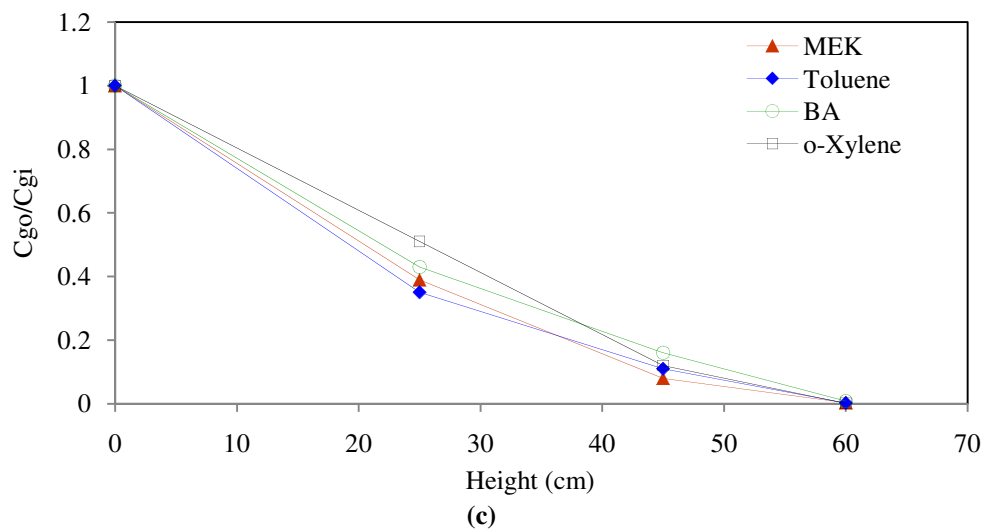
The production of CO<sub>2</sub> is the most important parameter to evaluate the degradability of pollutants. Figure-4(a) shows the production rate of carbon dioxide (PCO<sub>2</sub>) with respect to elimination capacity (EC). The amount of carbon utilized by microorganisms is defined as following expression<sup>14</sup>.

$$\% \text{ carbon recovery} = \frac{\text{Mole of } C_2 \text{ produced}}{\text{Mole of C in pollutant consumed}} \times 100 (\%)$$

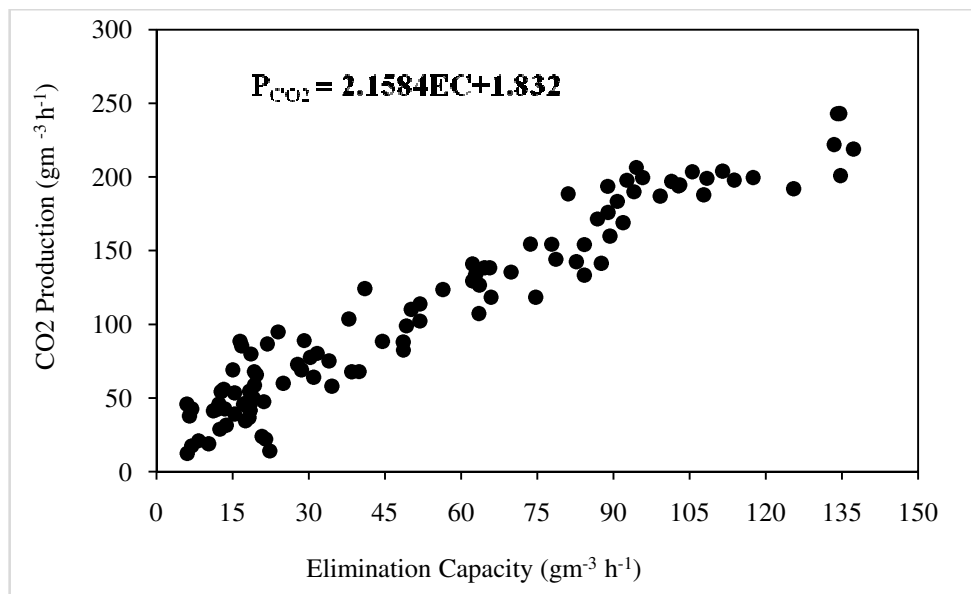
The total release of CO<sub>2</sub> directly correlate with the oxidized amount of MTBX in the biofilter. A linear expression calculated according to the least square method, provide the equation,  $P_{CO_2} = 2.1584EC + 1.832$  for up flow mode operation. During biofilter operation period, the presence of CO<sub>2</sub> concentration indicates the total production of CO<sub>2</sub> after the biodegradation of organic pollutants. Figure-4(a) represents the correlation between elimination capacity of biofilter and total CO<sub>2</sub> production. According to above figure, the changes in the elimination capacity directly indicate the changes in production of CO<sub>2</sub>. Both loading rates and elimination capacity were higher with the higher MEK, Toluene, n-butyle acetate and o-xylene (MTBX) concentrations and lead to higher amount of CO<sub>2</sub> production. However, Figure-4(b) indicates CO<sub>2</sub> evaluation and elimination capacity of biofilter continuously increased with the increase in loading rates.



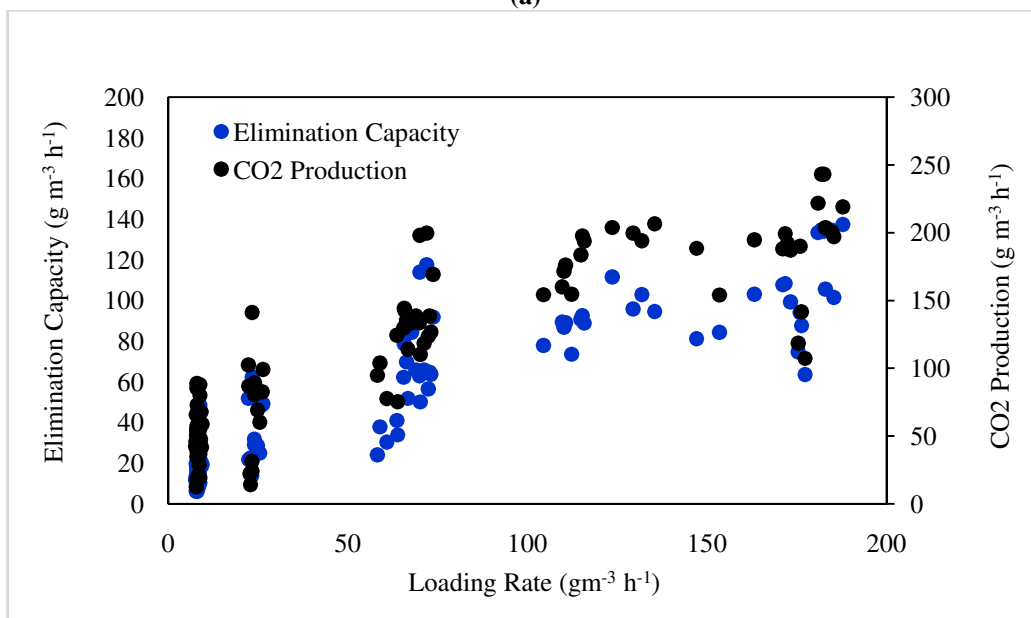




**Figure-3:** MTBX removal profiles along the biofilter bed height during days (a) 41 (phase I) (b) 65 (phase II) (c) 102 (phase III) (d) 117 (phase IV) and (e) 149 (phase V).



(a)



(b)

**Figure-4:** Carbon dioxide production in the experiment versus (a) elimination capacity (b) loading rate.

## Conclusion

In the present work, experiments have been performed for the removal of MTBX by using corn-cob as packing materials of the biofilter. After acclimatization of biofilter biofilms are grown on the packing materials by immobilizing microbial community. All the characteristics of biofilter related to steady state operation for the removal of MTBX have been studied. The complete performance of a compost biofilter inoculated with microbial culture was optimized for the treatment of polluted air containing gas phase of MTBX. The biofilter was operated for the period of 149 days and this time period was divided into five phases (I-V) to differentiate between the

outputs of biofilter. On the removal efficiency (RE) and elimination capacity (EC), the effects of various parameters like gaseous concentration and flow rate were investigated. The removal efficiency was measured with respect to inlet concentration of each component of MTBX and the elimination capacity was varied with change in loading rate of gaseous mixture. The maximum RE in MTBX was of BA with 99.98% with respect to inlet concentration of  $0.118992 \text{ gm}^{-3}$  while the minimum RE was of *o*-xylene with 98.21% for the concentration of  $.0.189568 \text{ gm}^{-3}$ . In case of EC the maximum value was obtained as  $86.27 \text{ g m}^{-3} \text{ h}^{-1}$  for n-butyle acetate at the highest value of inlet load of  $114.69 \text{ gm}^{-3} \text{ h}^{-1}$ . The results were

statistically interpreted by performing an analysis of variance to elucidate the main and interaction effects.

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