



Biofiltration of Benzene Vapour using Agro Waste Packing Material in a Biofilter

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Abstract: The biofiltration technology has gained acceptance worldwide as an economical and reliable air pollution control technology for treating biodegradable volatile organic compounds (VOCs). In this work, the biofiltration of benzene has been studied. The casuarina seed is used as packing materials for the removal of benzene. The performance of the biofilter has been investigated over a period of 90 days. Experiments are carried out in three stages. For each stage, the biofiltration of pollutant is performed for different inlet concentrations of benzene (0.2 - 1.2 g/m³) and gas flow rates (0.03 to 0.09 m³/h). The biofilter performances are investigated by their removal rates, elimination capacity, and variation of bed height at the time of operations. For the inlet concentration of 0.2 ± 10% gm⁻³ and gas flow rate of 0.03 m³h⁻¹, the removal efficiency of benzene is 94%. The maximum EC is obtained at the inlet concentration of 1.2 g/m³ and gas flow rate of 0.09 m³/h for all tested conditions.

Keywords: Benzene, Casuarina seed, Removal Efficiency, Elimination Capacity

I. Introduction

Volatile organic compounds (VOCs) are considered to be a likely group of contaminants for atmospheric air pollution produced through ozone layer depletion and greenhouse effect. Benzene, along with toluene, ethyl benzene and xylene, forms a group of VOCs called as BTEX compounds and are reported to contribute 59 mass% of gasoline pollutants [1,2]. Benzene is a potential chemical while the others are mutagenic. Due to the harmful nature of benzene, Environmental Protection Agency has set a target to lower the benzene concentration to 0.62% by 2011 [4]. Long term exposure of benzene leads to leukemia, excessive bleeding, anemia and immune system disorders [5,6]. Major sources of VOCs occur through storage tank losses, process vessel vents, refining operations, automotive emissions, heat exchangers and leaks from piping network and equipment [7]. In addition, VOCs are released by several industries including chemical industries, foundries, printing and coating industries, electronics and paint manufacturing industries.

Various technologies available for VOCs control include absorption, incineration, ozonation, membrane separation, etc. However, these methods suffer from drawbacks like high operating costs, lower removal efficiency and secondary pollution problems [8]. In this contextual, biotechnological processes changed and gained popularity due to their simplicity and cost effectiveness. Different bioreactor configurations, namely biofilters, bio trickling filters, bio scrubbers and suspended growth bioreactors are available for VOC treatment [9]. Biofiltration is one of the biotechnological processes which involve the use of packing media over which pollutant decomposing microorganisms are immobilized as a biofilm. The pollutant laden air is passed through the biofilter column and transferred from the air stream to the biofilm due to the difference in concentrations [10]. A series of processes like absorption, diffusion and degradation are involved in this process [11] and involves physical, chemical and biological interactions [12]. In all instances, the microorganisms are reported to convert the organics to carbon dioxide and water vapor. The food source is provided by the organic compound which helps in multiplication or activity of microorganisms [13]. Various factors like particle size and surface area of packing media, pH, carbon source, temperature and solubility of target pollutant are reported to affect the efficiency of biofilters [14,15].

Biofilter operations have been reported under unsteady-state, transient, short term and long term shock loads, shut down and starvation conditions [16,17]. Different packing media like compost and pumice [18], press mud [19] and corn stalks [20] have been investigated in various biofiltration studies. Biofiltration of benzene in a compost based biofilter was investigated in the concentration range up to 1.7 g/m³ and removal efficiency of 90% was reported [22]. The maximum elimination capacities of benzene obtained, at an inlet load of 6.12 g m⁻³ h⁻¹, were 3.50 and 3.80 g m⁻³ h⁻¹ with raw and ground sugarcane bagasse based biofilter, respectively [24].

Studies on biofiltration of benzene at very high concentrations were few and the choice of date palm tree bark as a filter media was not investigated earlier. In this experimental work, the feasibility of a novel biofilter media for biofiltration of benzene under mesophilic conditions was studied. The biofilter performance was evaluated in terms of removal efficiency and elimination capacity. The influence of inlet loading and flow rate of the pollutant on the elimination capacity was investigated. In addition, axial concentration profile and biomass growth variations are monitored.

II. Materials and Methods

A. Inoculum and packing material

Casuarina plant seeds used in this study were obtained from Villupuram district, Tamilnadu, India. The seeds were washed with distilled water and dried in an oven at 70°C for 12 hours. The dried seeds were stored in airtight plastic container and ready to be used. Fresh activated sludge was used as the inoculum source for the biofilter. Activated sludge was obtained from a paper industry wastewater treatment plant, Sivakasi, India. Microorganisms in the activated sludge were acclimated to VOCs in order to accelerate the adaptation period. For acclimation, one liter of the activated sludge was enclosed in an aerated tank and diluted with 3 L of nutrient solution. The composition of nutrient solution per liter of distilled water was: K_2HPO_4 – 0.91 g; $Na_2HPO_4 \cdot 2H_2O$ – 2.39 g; $(NH_4)_2SO_4$ – 1.97 g; $FeSO_4 \cdot 2H_2O$ – 0.2 g; $MgSO_4 \cdot 7H_2O$ – 2.0 g; $MnSO_4 \cdot 7H_2O$ – 0.88 g; $Na_2MoO_4 \cdot 2H_2O$ – 1.0 mg; $CaCl_2$ – 3.0 mg; $ZnSO_4 \cdot 7H_2O$ – 0.04 g and $CoCl_2 \cdot 6H_2O$ – 0.04 mg.

B. Biofilter Set Up and Operation

The three biofilter column unit is made up of acrylic, with a height of 100 cm and an inside diameter of 5 cm. Five sampling points were provided at 0 cm (inlet), 25 cm (section-1), 50 cm (section-2), 75 cm (section-3) and 100 cm (exit), along the height of the biofilter. The nutrient media was added at periodic intervals through a nutrient distribution system at the top. Treated gas was collected at the reactor headspace and a 10 cm bottom space was utilized for leachate collection. Fig. 1 shows the biofilter unit with its components. The influent air stream was generated by injecting a compressed air stream into the storage vessel containing the respective volatile organic compound. In this study, the air is bubbled successively through two tanks filled with benzene and toluene respectively. The air stream loaded with benzene and toluene was adjusted for its concentration by mixing with the humidified pure air stream in the mixing chamber and fed into the biofilter reactor in an upflow direction. The operating parameters are varied and the experimental plan is given in Table 1. The samples were collected at periodic intervals for analysis for residual volatile organic compounds.

Figure 1 Experimental setup

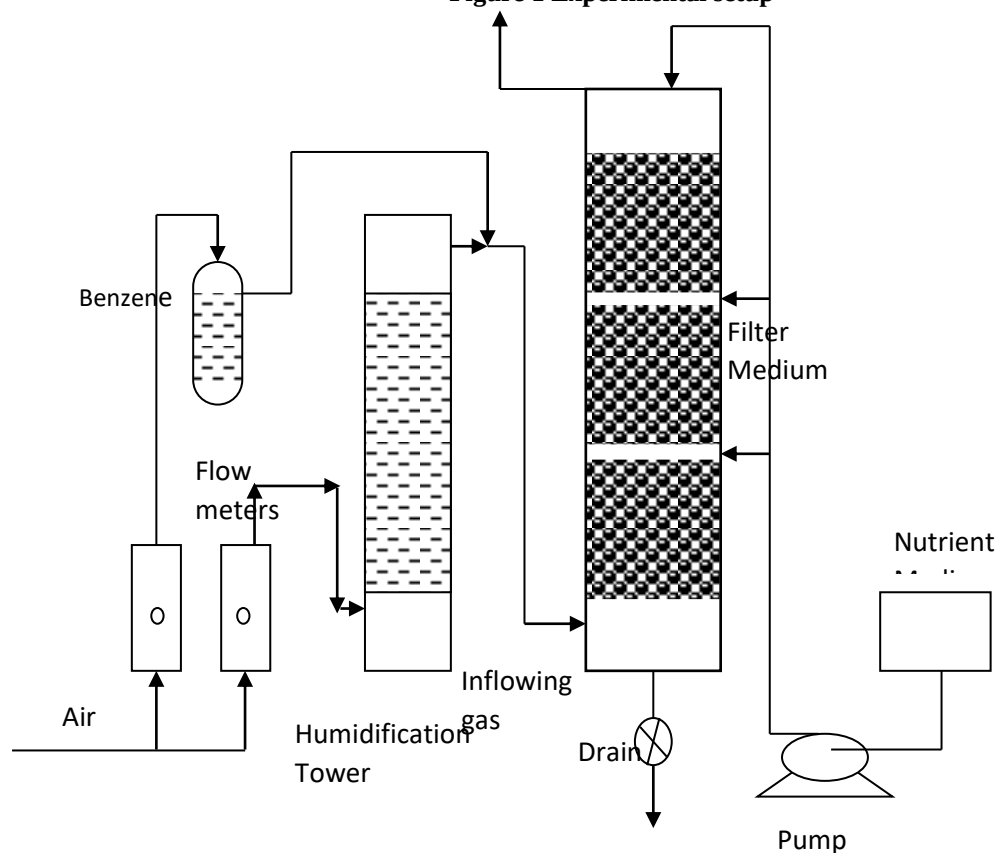


Table 1 Experimental Plan

Days	Gas Flow rate (m ³ /h)	Inlet Concentration (g/m ³)
0-10	0.03	0.2
11-20		0.2
21-30		0.4
31-40		0.6
41-50		0.8
51-60		1
61-70	0.06	0.2
71-80		0.4
81-90		0.6
91-100		0.8
101-110		1
111-120	0.09	0.2
121-130		0.4
131-140		0.6
141-150		0.8
151-160		1
161-170	0.12	0.2
171-180		0.4
181-190		0.6
191-200		0.8
201-210		1

C. Estimation of Concentration and Biofilter Parameters

The benzene and toluene concentrations in gas samples were measured by PID detector. The removal effect of the biofilter was measured in terms of the removal efficiency (% RE) and elimination capacity (EC), g/m³h. These parameters are defined as given below:

$$\text{Removal Efficiency} = \frac{C_{\text{in}} - C_{\text{out}}}{C_{\text{in}}} \times 100 \quad (1)$$

$$\text{Elimination capacity} = \frac{F(C_{\text{in}} - C_{\text{out}})}{V} \quad (2)$$

$$\text{Organic load rate} = \frac{F(C_{\text{in}})}{V} \quad (3)$$

Where C_{in} and C_{out} represent the inlet and exit concentrations of benzene or toluene or total (g/m³), F is the flow rate of the benzene or toluene (m³/h), V is the volume of the biofilter (m³).

III. Result and Discussion

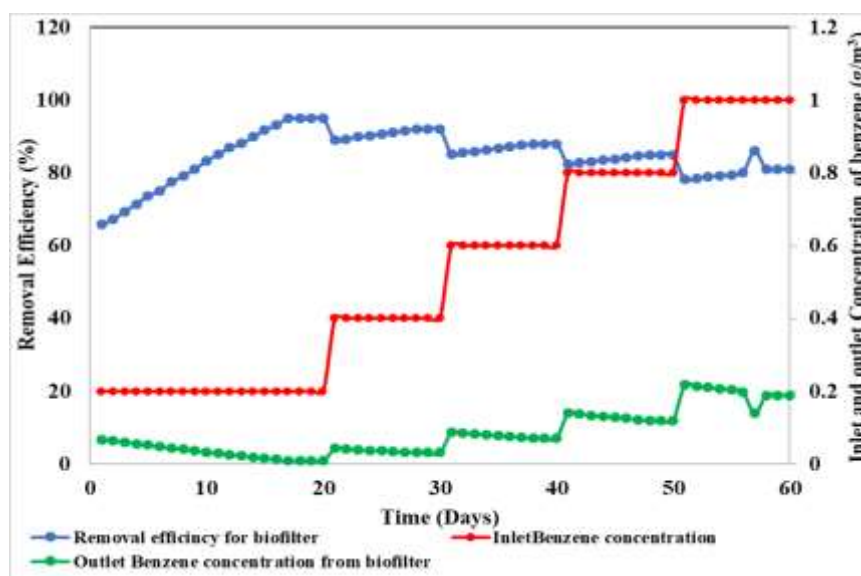
The biofiltration of gas stream containing pure benzene is carried out for 210 days at various operating conditions in an up flow mode biofilter. The experiment has been operated in four stages. Each stage is divided into five phases as shown in table 1. Various EBRT and concentration are maintained so that the corresponding loading rate could be maintained and regulated in the reactor to study the performance of the reactor. Table 1 shows the inlet concentration, inlet gas flow rate supplied to the biofilter.

A. Performance of biofilter at an EBRT of 2.81 min

During the first 60 days, the EBRT is maintained at 2.81 min as shown in figure 2. In the first phase of stage - I, benzene inlet concentration is maintained at $0.2 \pm 5\%$ g/m³, so that the average loading rate of 4.163 g/m³ h can be applied to the reactor. The corresponding gas flow rate is maintained at 0.03 m³/h. Continuous increase in removal efficiency is as shown in figure 2. It is clear from the figure 2 that a maximum removal efficiency of 95% removal is obtained after 8 days of operation. The results showed that the maximum removal

efficiency of pure benzene is 95%. In phase II, average loading rate has been increased by almost twofold from 4.163 to 8.143 g/m³ h. The flow rate and EBRT is kept at 0.03 m³/h and 2.81 min respectively. The average inlet benzene concentration is sustained at 0.4 ± 5 % g/m³. Due to this shock load there is a fall in the RE of benzene. A maximum RE of benzene is 88 %. In phase III, average loading rate is increased from 8.143 to 12.857 g/m³ h and the flow rate of main stream and EBRT is 0.03m³/h and 2.81 min respectively. The inlet concentration is kept constant at 0.8 g/m³. With the sudden increase in the loading rate to the reactor, the RE decreased from 88% to 81%. In phase IV, average loading rate is increased from 12.857 to 16.323 g/m³ h and the flow rate of main stream and EBRT is 0.03m³/h and 2.81 min respectively. The inlet concentration is kept constant at 0.8 g/m³. With the sudden increase in the loading rate to the reactor, the RE decreased from 81% to 78%. In phase V, average loading rate is increased from 16.323 to 20.408 g/m³ h and the flow rate of main stream and EBRT is 0.03m³/h and 2.81 min respectively. The inlet concentration is kept constant at 1.2 g/m³. With the sudden increase in the loading rate to the reactor, the RE decreased from 78% to 75%.

Fig 2. Overall performance of the biofilter for the removal of benzene for gas flow rate of 0.03g/m³



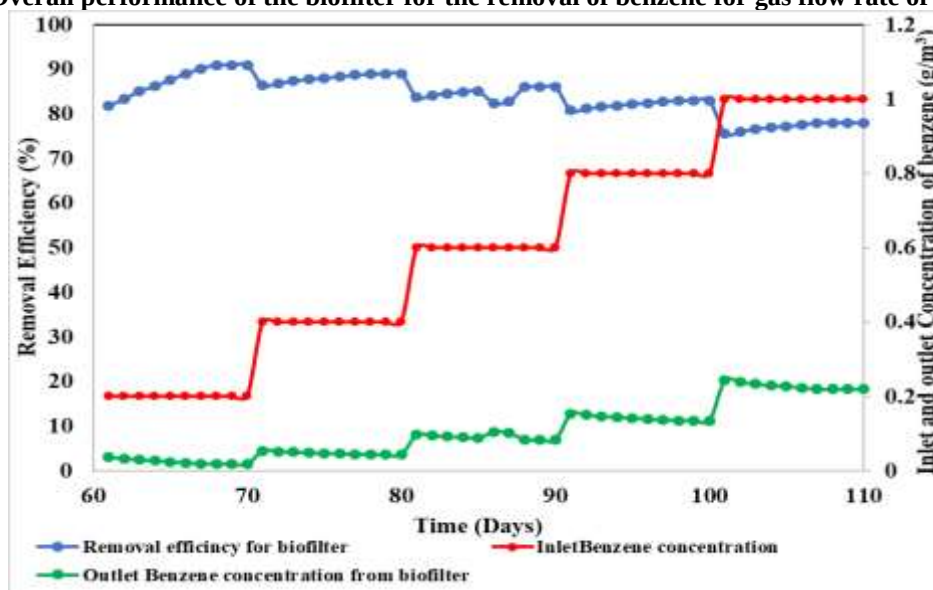
B. Performance of biofilter at an EBRT of 1.47 min

During the second 50 days, the EBRT is maintained at 1.47 min as shown in figure 3. In the second phase of stage - I, benzene inlet concentration is maintained at 0.2 ± 5 % g/m³, so that the average loading rate of 8.163 g/m³ h can be applied to the reactor. The corresponding gas flow rate is maintained at 0.06 m³/h. Continuous increase in removal efficiency is as shown in figure 3. It is clear from the figure 3 that a maximum removal efficiency of 92% removal is obtained after 8 days of operation. The results showed that the maximum removal efficiency of pure benzene is 92%. In phase II, average loading rate has been increased by almost twofold from 8.143 to 16.323 g/m³ h. The flow rate and EBRT is kept at 0.06 m³/h and 1.47 min respectively. The average inlet benzene concentration is sustained at 0.4 ± 5 % g/m³. Due to this shock load there is a fall in the RE of benzene. A maximum RE of benzene is 83 %. In phase III, average loading rate is increased from 16.857 to 24.848 g/m³ h and the flow rate of main stream and EBRT is 0.06m³/h and 1.47 min respectively. The inlet concentration is kept constant at 1.0 g/m³. With the sudden increase in the loading rate to the reactor, the RE decreased from 83% to 80%.

In phase IV, average loading rate is increased from 24.848 to 32.636 g/m³ h and the flow rate of main stream and EBRT is 0.03m³/h and 2.81 min respectively. The inlet concentration is kept constant at 0.8 g/m³. With the sudden increase in the loading rate to the reactor, the RE decreased from 80% to 76%. In phase V, average loading rate is increased from 32.636 to 48.818 g/m³ h and the flow rate of main stream and EBRT is 0.06m³/h and 2.81 min respectively. The inlet concentration is kept constant at 1.2 g/m³. With the sudden increase in the loading rate to the reactor, the RE decreased from 75% to 72%.

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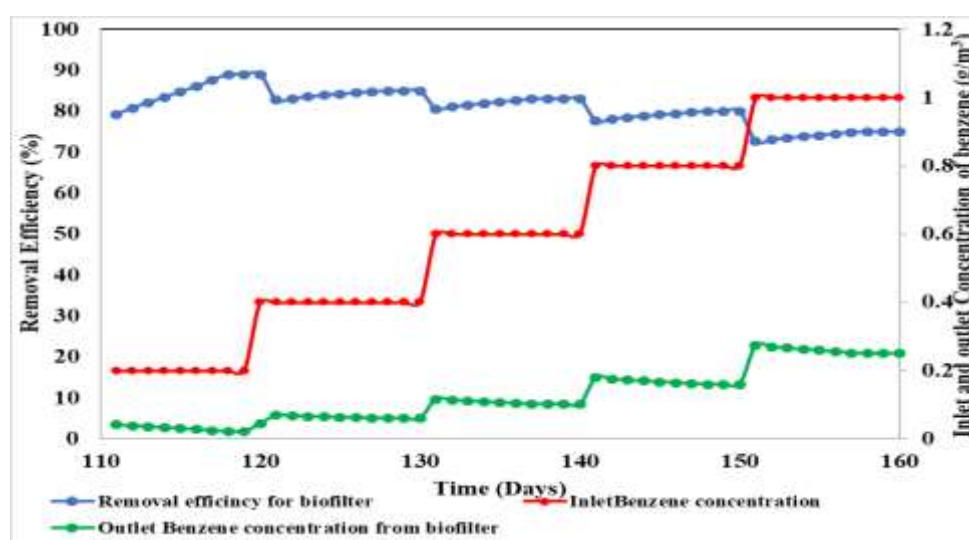
Fig 3. Overall performance of the biofilter for the removal of benzene for gas flow rate of 0.06g/m³



C. Performance of biofilter at an EBRT of 0.9 min

During the third 50 days, the EBRT is maintained at 0.9 min as shown in figure 4. In the third phase of stage - I, benzene inlet concentration is maintained at 0.2 ± 5 % g/m³, so that the average loading rate of 12.249 g/m³ h can be applied to the reactor. The corresponding gas flow rate is maintained at 0.09 m³/h. Continuous increase in removal efficiency is as shown in figure 4. It is clear from the figure 4 that a maximum removal efficiency of 89% removal is obtained after 8 days of operation. The results showed that the maximum removal efficiency of pure benzene is 89%. In phase II, average loading rate has been increased by almost twofold from 12.249 to 24.49 g/m³ h. The flow rate and EBRT is kept at 0.09 m³/h and 0.9 min respectively. The average inlet benzene concentration is sustained at 0.4 ± 5 % g/m³. Due to this shock load there is a fall in the RE of benzene. A maximum RE of benzene is 85 %. In phase III, average loading rate is increased from 24.848 to 36.736 g/m³ h and the flow rate of main stream and EBRT is 0.09m³/h and 0.9 min respectively. The inlet concentration is kept constant at 1.0 g/m³. With the sudden increase in the loading rate to the reactor, the RE decreased from 85% to 80%. In phase IV, average loading rate is increased from 36.736 to 48.78 g/m³ h and the flow rate of main stream and EBRT is 0.09m³/h and 0.9 min respectively. The inlet concentration is kept constant at 0.8 g/m³. With the sudden increase in the loading rate to the reactor, the RE decreased from 80% to 76%. In phase V, average loading rate is increased from 48.78 to 61.24 g/m³ h and the flow rate of main stream and EBRT is 0.09m³/h and 0.9 min respectively. The inlet concentration is kept constant at 1.2 g/m³. With the sudden increase in the loading rate to the reactor, the RE decreased from 76% to 72%.

Fig 4. Overall performance of the biofilter for the removal of benzene for gas flow rate of 0.09g/m³



D. Performance of biofilter at an EBRT of 0.7 min

During the fourth 50 days, the EBRT is maintained at 0.7 min as shown in figure 5. In the fourth phase of stage - I, benzene inlet concentration is maintained at $0.2 \pm 5 \text{ g/m}^3$, so that the average loading rate of $16.323 \text{ g/m}^3 \text{ h}$ can be applied to the reactor. The corresponding gas flow rate is maintained at $0.12 \text{ m}^3/\text{h}$. Continuous increase in removal efficiency is as shown in figure 5. It is clear from the figure 5 that a maximum removal efficiency of 84% removal is obtained after 8 days of operation. The results showed that the maximum removal efficiency of pure benzene is 84%. In phase II, average loading rate has been increased by almost twofold from 16.323 to $32.656 \text{ g/m}^3 \text{ h}$. The flow rate and EBRT is kept at $0.12 \text{ m}^3/\text{h}$ and 0.7 min respectively. The average inlet benzene concentration is sustained at $0.4 \pm 5 \text{ g/m}^3$. Due to this shock load there is a fall in the RE of benzene. A maximum RE of benzene is 82 %. In phase III, average loading rate is increased from 32.656 to $48.979 \text{ g/m}^3 \text{ h}$ and the flow rate of main stream and EBRT is $0.12 \text{ m}^3/\text{h}$ and 0.7 min respectively. The inlet concentration is kept constant at 1.0 g/m^3 . With the sudden increase in the loading rate to the reactor, the RE decreased from 82% to 78%.

In phase IV, average loading rate is increased from 48.979 to $65.30 \text{ g/m}^3 \text{ h}$ and the flow rate of main stream and EBRT is $0.12 \text{ m}^3/\text{h}$ and 0.78 min respectively. The inlet concentration is kept constant at 0.8 g/m^3 . With the sudden increase in the loading rate to the reactor, the RE decreased from 78 % to 74%. In phase V, average loading rate is increased from 65.30 to $81.6 \text{ g/m}^3 \text{ h}$ and the flow rate of main stream and EBRT is $0.12 \text{ m}^3/\text{h}$ and 0.7 min respectively. The inlet concentration is kept constant at 1.2 g/m^3 . With the sudden increase in the loading rate to the reactor, the RE decreased from 74% to 70%.

E. Inlet load and Elimination Capacity effects

The Elimination Capacity (EC) allows a direct comparison between different biofilter systems. Pollutant elimination capacities for the different runs versus IL are presented in figure 6 to 9. A comparison of the EC of benzene as a function of inlet load for different EBRT of 2.8, 1.47, and 0.7 min is shown in Figures 6 to 9. A maximum EC of $49.33 \text{ g/m}^3 \text{ h}$ benzene is observed at a gas flow rate of $0.12 \text{ m}^3/\text{h}$ (EBRT = 0.7 min). When increasing the gas flow rate the elimination capacity also shows positive trend. From the figures 6 to 9, it is observed that the casuarina seed based biofilter tended to operate with slightly higher removal capacities. It suggests that, increase in inlet load enhances the transfer rate of benzene to the biofilm at constant gas flow rate. However, at higher inlet load, the elimination capacity curve deviated markedly from the 100% removal curve. Similar observation is obtained for the removal of toluene, Ethylacetate and xylene vapour using perlite as porous material [26].

Fig 5. Overall performance of the biofilter for the removal of benzene for gas flow rate of 0.12 g/m^3

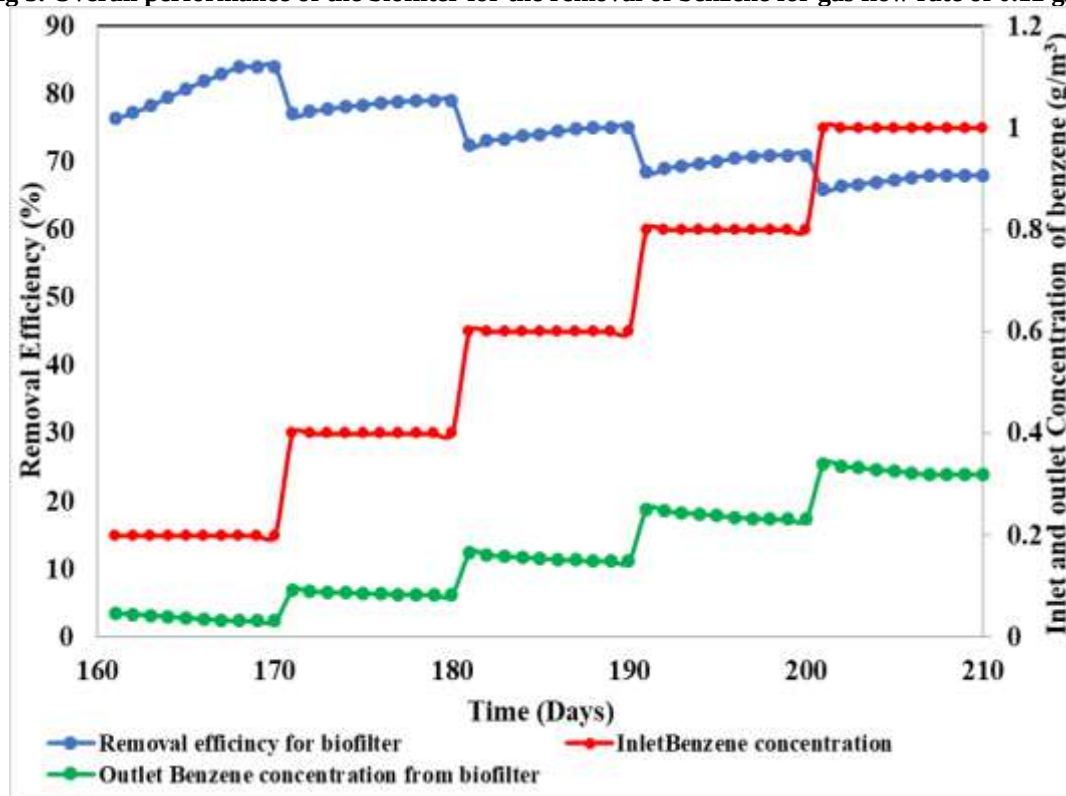


Fig 6. Elimination capacity and inlet load curve for removal of benzene from air at the gas flow rate of 0.03 g/m^3

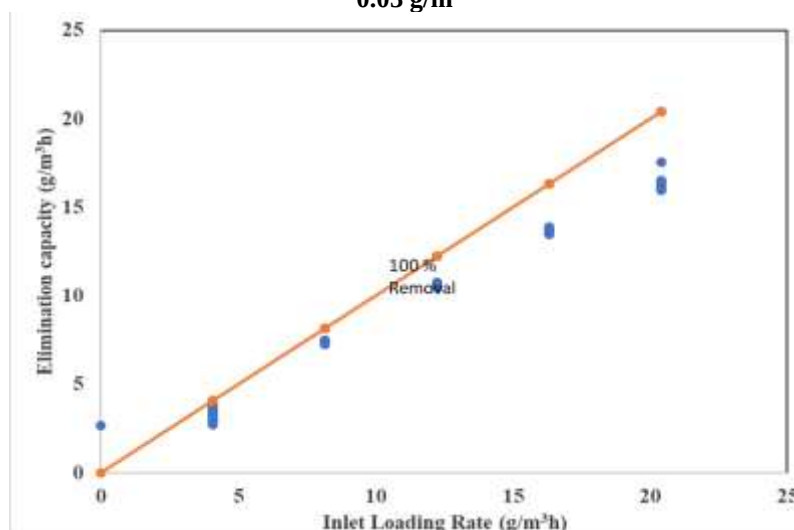


Fig 7. Elimination capacity and inlet load curve for removal of benzene from air at the gas flow rate of 0.06 g/m^3

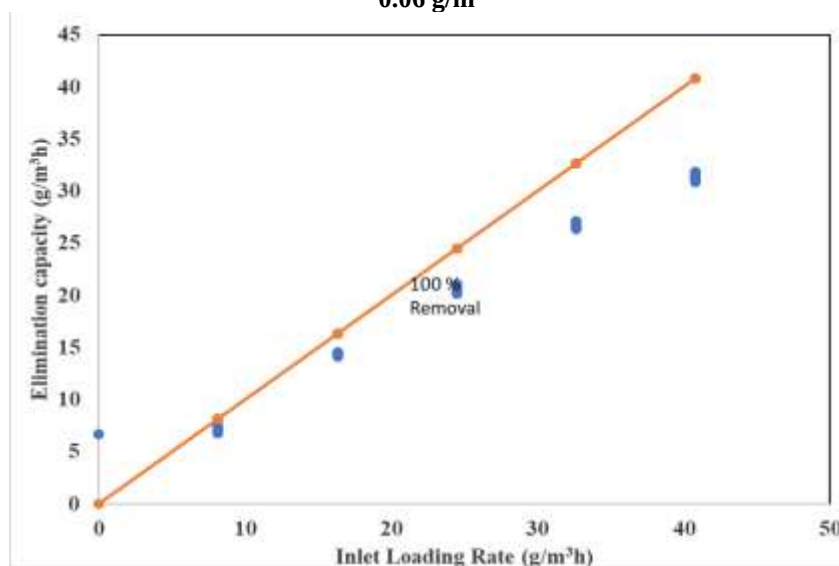


Fig 8. Elimination capacity and inlet load curve for removal of benzene from air at the gas flow rate of 0.09 g/m^3

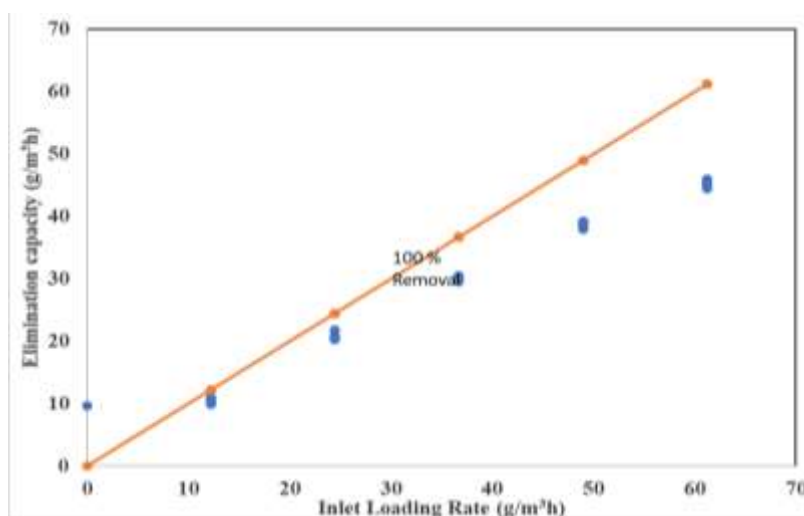
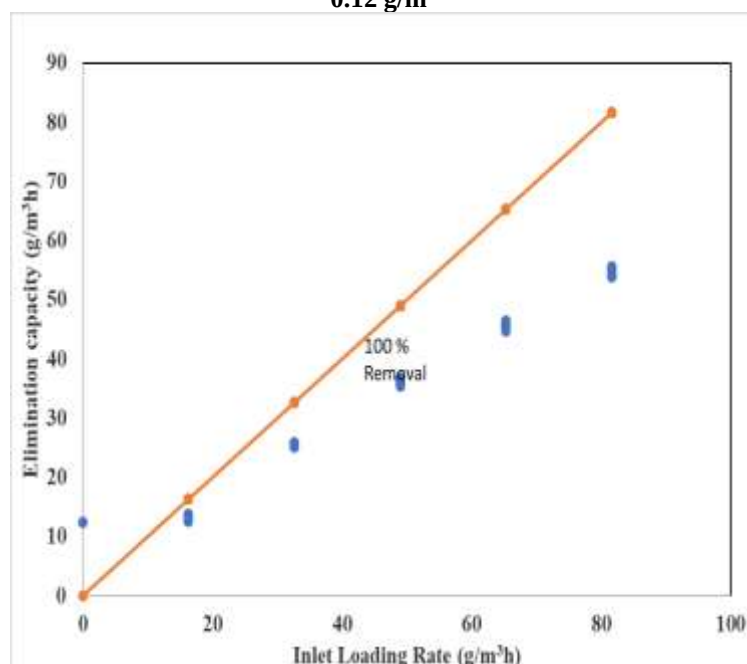


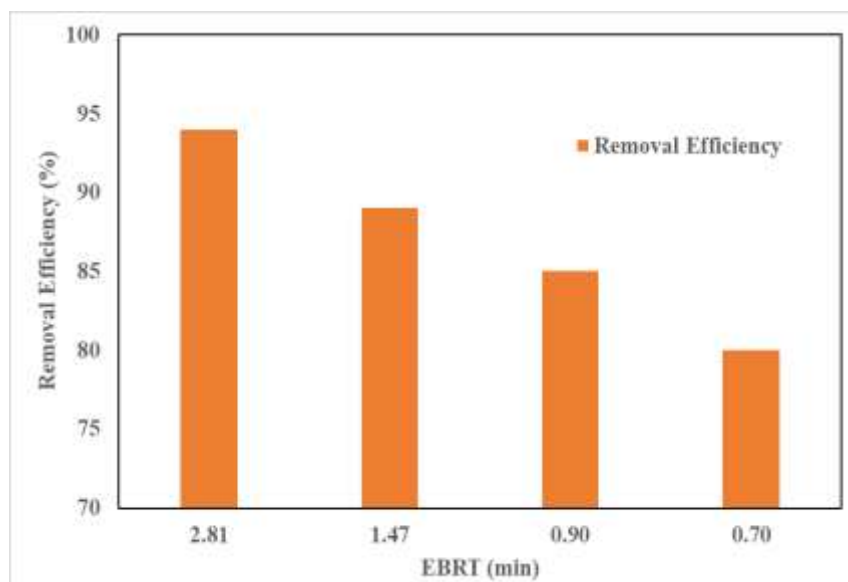
Fig 9. Elimination capacity and inlet load curve for removal of benzene from air at the gas flow rate of 0.12 g/m^3



F. Effect of Empty bed residence time on biofiltration operation

The influence of residence time on the removal efficiency, at different inlet load has been studied. All the results are shown in Figure 10. The maximum benzene elimination is observed at an EBRT of 2.81 min. A decrease in the EBRT from 2.81 to 0.7 min caused a decrease in the removal efficiency from 92% to 50%. But for residence times less than 1.47 min, a sharp linear decrease is observed, achieving a removal efficiency of 50% at 0.7 min. These results suggest that a minimum EBRT of about 1 min may be needed to reach adequate removal efficiencies, and an EBRT of 1.47 min could be considered safe to ensure the absence of limitations associated with contact time.

Fig 10. Effect of EBRT on removal efficiency for removal of benzene

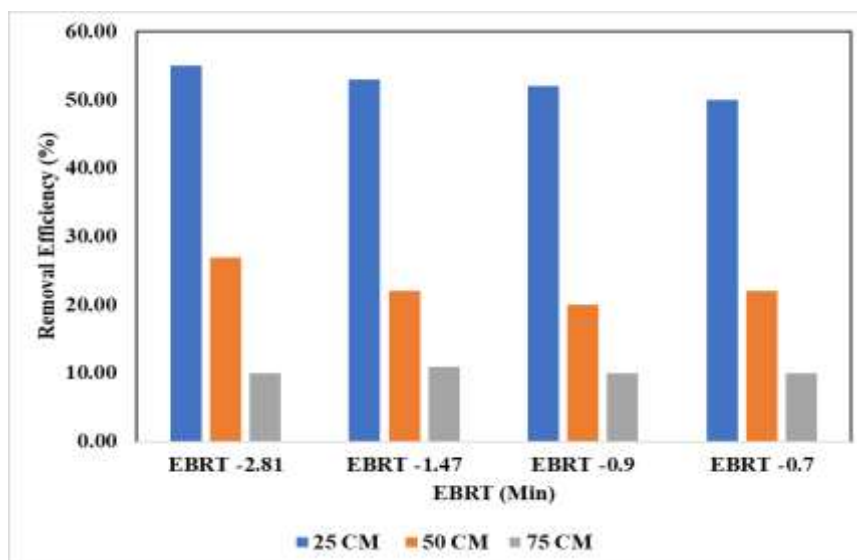


G. Effect of bed height on removal efficiency

The biofiltration column is subdivided into three identical sections. Three sampling ports are located across the bed height. For better understanding the performance of biofilter, samples are collected at the three sampling ports to measure the removal efficiency. In order to analyze the composition of the gas phase, samples are withdrawn from the inlet and outlet of the biofilter and the exit of the first two sections as well. As a result, the contribution of the three different sections of the bioreactor to its overall performance can be compared to each

other. Figure 11 shows the RE of the three sections of the biofilter for various inlet loads of benzene. Here, the RE of an individual section is defined as the ratio of the difference between the inlet and exit concentration of benzene in this section to the concentration of benzene at the entrance of the biofiltration column. Thus, the summation of the three individual section efficiencies is equivalent to the overall removal efficiency of the biofilter. For instance, at an EBRT of 2.81 min, the RE in the first section is 60% compared to 25% and 15% in the second and third sections all the cases of biofilter..

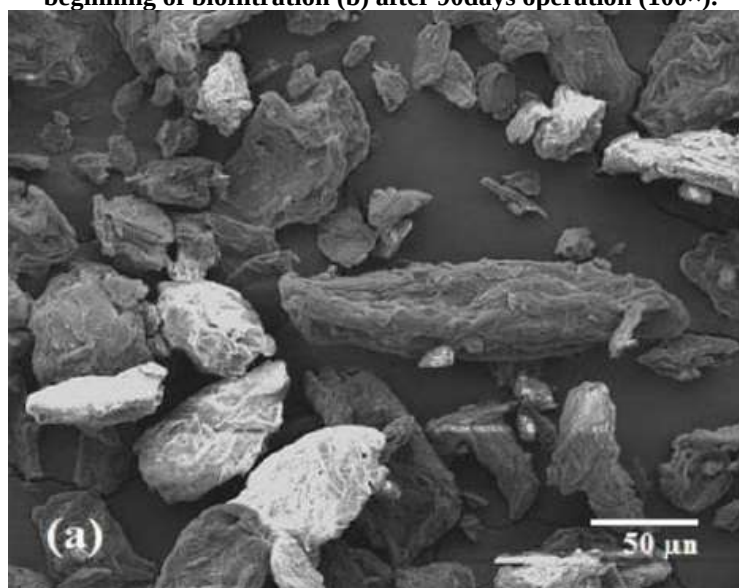
Fig 11. Effect of bed height on removal of benzene for different EBRT.

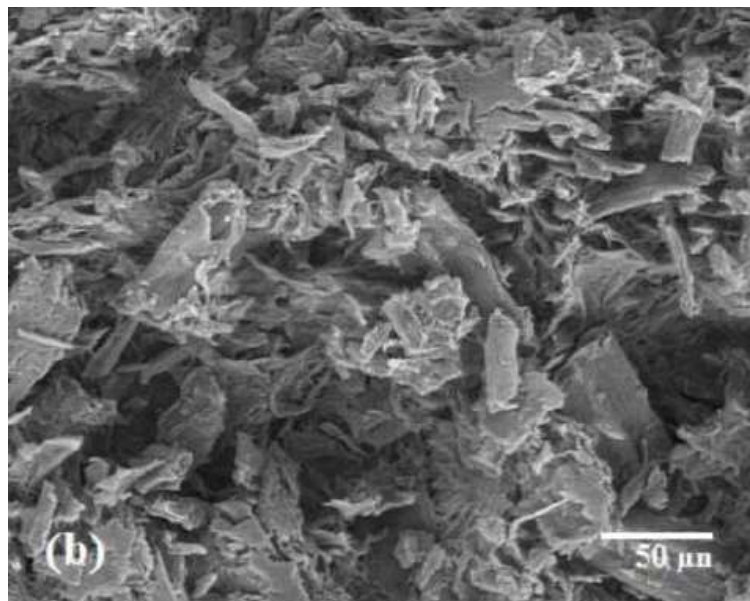


H. Microscopic observations

The scanning electron micrograph (SEM) provides information about the microbial community on the biofilter media. The biomass of individual particle can be mapped. From such precision, important factor such as filter media coverage, thickness and activity can be determined. The SEM of microbial growth on various types of media before and after experiment has already been shown by some researchers [24]. A comparison of the scanning electron microscopic images of, cassuriane seeds before and after experiment is shown in Figures 12(a) and 12 (b). A biofilm on the surface of the packing material was observed clearly after 210th days of operation. Even growths of microbial community on the surface of the pore of the packing materials are clearly visible. Initially, the degree of acclimatization depends upon the adaptive capacity of the microorganism in the packing material, substrate concentration and its availability and other necessary environmental conditions. Several groups of microorganisms are involved in the degradation of air pollutants in biofilters including bacteria, actinomycetes and fungi [25,26].

Fig 12 Biofilm morphology of microorganisms on the surface of casuarina seed by SEM: (a) at the beginning of biofiltration (b) after 90days operation (100×).





VI. Conclusion

A biofilter consisting of a casuarina seed as packing material be successfully started in 7 weeks for benzene removal. The RE decreased when increased benzene loading rate but did not decline significantly when reduced gas EBRTs. High ECs at high organic loading rates could be obtained under low pollutant concentrations. Excessive biomass accumulation was singularly occurred on the biofilter. The biofilter is suitable for treating waste gases with low benzene concentrations even at high flow rates over long periods of operation, reduces the footprint of gas biofilters, and overcomes the deterioration in removal observed with traditional biofilters.

V. References

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