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PERFORMANCE EVALUATION AND KINETIC STUDIES ON REMOVAL OF BENZENE IN UP-FLOW TREE BARK BASED BIOFILTER

Article Highlights

- The removal of benzene at different in loading rates were studied in a biofilter employing a novel biofilter media
- The performance was evaluated in terms of removal efficiency and elimination capacity
- The effect of flow rates of influent pollutant and biofilter bed height was studied
- Carbon dioxide gas production profile was recorded and related with elimination capacity
- Kinetic modeling was performed and the constants were estimated

Abstract

This study aims to evaluate the feasibility of Phoenix dactylifera tree barks as the novel filter medium in an upflow biofilter employing mixed culture to degrade benzene. The experiments were conducted at different benzene concentrations (1.5-6.0 g m^{-3}) and EBRT (1.2-4.7 min). The elimination capacity was found to vary linearly with inlet loading rate in the range of 0-306 g m^{-3} h^{-1}. Removal efficiency of 99% was achieved when the benzene concentration was 1.5 g m^{-3} and decreased with increase in benzene concentration. Lower flow rates resulted in higher benzene removal efficiency. The concentration profile was observed at different heights of filter media. Temperature increase during biofiltration experiments confirmed the exothermic nature of biofiltration. The carbon dioxide production rate was related to elimination capacity by the equation CPR = 1.76EC + 18. A Michaelis-Menten type model was applied and the kinetic constants, maximum elimination capacity, EC_{max}, and saturation constant, K_s were found to be 217.4 g $m^{-3}h^{-1}$ and 3.54 g m^{-3}, respectively.

Keywords: kinetics, benzene, pollution.

Volatile organic compounds (VOCs) are considered to be a potential group of contaminants for atmospheric air pollution caused 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 carcinogen while the others are mutagenic. These pollutants are listed in the European Pollutant Release and Transfer Register [3]. 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 demerits like high operating costs, lower removal efficiency and secondary pollution problems [8]. In this background, biotechnological processes evolved and gained popularity due to their simplicity and cost-

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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 stacks [20] have been investigated in various biofiltration studies. Biofiltration of benzene was reported using *Stenotrophomonas maltophilia* 3c in a polyurethane based biofilter [21]. Biofiltration of benzene in a compost based biofilter was investigated in the concentration range up to 1.7 g/m^3 and removal efficiency of 90% was reported [22]. BTEX degradation was evaluated as separate substrates and in mixtures, in liquid culture, and in packed biofilters with the filamentous fungus *Paecilomyces variotii* CBS115145 and a benzene elimination capacity of 10 g C m^{-3} h^{-1}, was reported [23]. 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. Kinetic modeling was performed and the constants were evaluated.

MATERIALS AND METHODS

Biofilter media

The biofilter media used in this study was date palm tree barks. Date palm tree, *Phoenix dactylifera,* belongs to the Palmae (Arecaceae) family and is a native to Middle East countries where it is found in large number and used as a staple food in Oman. The tree barks were cut into pieces and screened for sizes lesser than the diameter of the column by eight times in order to avoid preferential flow on the side of the column walls [25]. Physicochemical characterization studies were performed to verify the chemical resistance of the filter media. The chemical resistance of novel packing medium was tested by placing it in glass beakers containing pure benzene, tap water, acidic (pH 4.91) and basic (pH 9.23) solutions for 30 days. The filter media were then removed from the solution, rinsed repeatedly with deionized water, dried in an oven at 60 °C for 24 h, cooled in a desiccator, and then reweighed. A slight change in color was observed and the weight losses of 2.0 and 2.6% were recorded with the samples placed in acidic and basic solutions respectively. The choice of this filter media was justified by its extensive availability and environmentally friendly disposability due to the natural biodegradaing ability in the longer duration.

Inoculum

The biofilter media was sterilized with an autoclave four times at 120 °C for 60 min and mixed with 40 mL of an activated sludge (1.5% *w/V*) collected from a municipal wastewater treatment plant in a 5.0 L tank. After standing overnight, the biofilter was loaded with the filter media in different sections. The concentrated sludge was cultured in an aerated batch reactor and diluted in 1 L of nutrient solution containing the following composition: K_2HPO_4 – 3.84 g L^{-1}, KH_2PO_4 – 1.94 g $L^{-1}$and NH_4Cl – 3.00 g L^{-1}, at pH 6.9. The packing media was mixed with the sludge in the biofilter column and drained after 24 h and this procedure was repeated several times until visible biofilm formation was noticed on the biofilter media.

Experimental studies

The biofilter reactor set up consists of an acrylic column with an inside diameter of 5 cm and column height of 100 cm. The biofilter column was equipped with two sets of sampling ports located at 0 cm (inlet), 25 cm (section-1), 50 cm (section-2), 75 cm (section--3) and 100 cm (exit), for treated gas sampling and

temperature measurements along the height of the biofilter. The treated gas was collected at the reactor headspace and nutrient feed addition was performed at the top of the column while a 10 cm bottom space was utilized for leachate collection was provided. Figure 1 shows the biofilter setup with its components. The biofilter column is equipped with a carbon dioxide gas sensor (Extox, Germany) connected at the exit. The synthetic benzene polluted air stream was generated by injecting a low flow compressed air stream into the benzene tank. The air stream loaded with benzene was mixed with the humidified pure air stream in the mixing chamber in order to attain the desired inlet concentration and fed into the biofilter reactor in an up flow mode. The air flow rates are regulated in the low (0-0.3 L min^{-1}) and high (0-10.0 L min^{-1}) flow rate range using rotameter. All the gas flow rates are manipulated using brass control valves. The operating parameters are varied in the ranges: inlet benzene concentration 1.5-6.0 g m^{-3} and flow rate 0.25-1.0 m^3 h^{-1} and the samples were collected at periodic intervals for analysis of residual benzene. The nutrient solution, basal salts medium, with the following composition: 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 per litre of water was sprayed twice a day through the nutrient distribution system equipped with a peristaltic pump and spray nozzle. The nutrient solution along with the humidified air helps in maintaining the required relative humidity in the biofilter.

Analytical methods

Inlet and exit benzene concentrations in gas samples were measured by gas chromatograph (Perkin-Elmer, USA) equipped with a FID and a capillary column, operated in off-line mode. The temperature conditions were 160 °C for injector and 280 °C for detector. The oven temperature was set at 60 °C for the first 5 min and increased at a rate of 15 °C per min to reach 180 °C and held at 4 min. Helium was used as a carrier gas at a flow rate of 2 ml min^{-1}. The temperature of the filter bed was measured using temperature sensors connected to a data logger. The head space gas was analysed for carbon dioxide concentration using online gas analyzer (Extox, Germany).

Performance evaluation

The performance of the biofilter was measured in terms of the removal efficiency *(%RE*), elimination capacity (*EC*), g m^{-3} h^{-1}, and carbon dioxide production rate (*CPR*), g m^{-3} h^{-1}. These parameters are defined as given below:

$$\%RE = 100\frac{C_0 - C_t}{C_0} \tag{1}$$

$$EC = \frac{Q(C_0 - C_t)}{V} \text{ (g m}^{-3}\text{ h}^{-1}\text{)} \tag{2}$$

$$CPR = \frac{Q(C_{CO_2 out} - C_{CO_2,in})}{V} \text{ (g m}^{-3}\text{ h}^{-1}\text{)} \tag{3}$$

The *Empty Bed Residence Time* (*EBRT*) is defined as:

$$EBRT = \frac{V}{Q} \text{ (h)} \tag{4}$$

The inlet loading rate (*ILR*) is defined as:

$$ILR = \frac{QC_0}{V} \text{ (g m}^{-3}\text{ h}^{-1}\text{)} \tag{5}$$

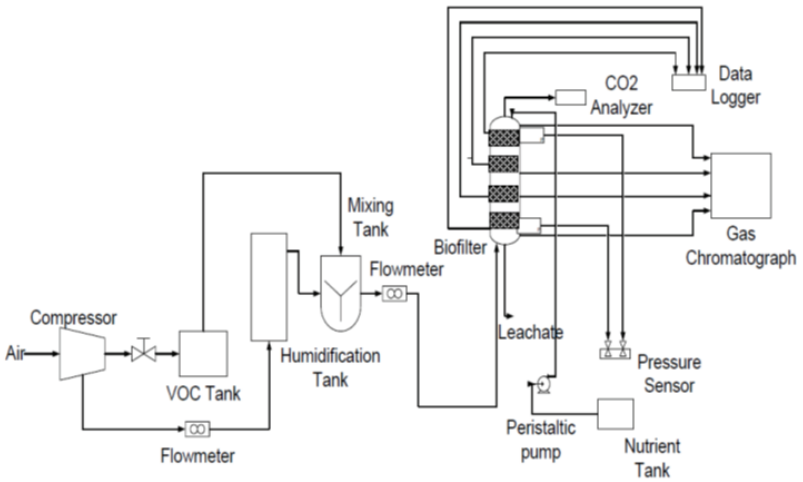

Figure 1. Biofiltration experimental setup.

where C_0 and C_t represent the inlet and exit concentrations of benzene (g m^{-3}), Q is the flow rate of the benzene ($m^3 h^{-1}$), V is the volume of the biofilter (m^3), C_{CO_2out} and $C_{CO_2,in}$ represent exit and inlet concentrations of carbon dioxide (g m^{-3}).

RESULTS AND DISCUSSION

Effect of inlet benzene concentration

The effect of inlet benzene concentration was studied in the range of 1.5-6.0 g m^{-3} during a biofiltration period of 65 days. In order to acclimate the microbial culture to higher VOC concentrations, the concentration of input benzene was increased stepwise by maintaining an inlet concentration of 1.5 g m^{-3} for the first 16 days followed by 3.2 g m^{-3} during the second phase of 17-32 days of operation. The outlet concentrations of benzene were monitored once in 24 h and the removal efficiency was calculated. The variation in removal efficiency during the different phases is presented in Figure 2. The maximum removal efficiency obtained was 99% with the lowest benzene concentration and the efficiency dropped to 91.8, 85.1 and 76%, respectively, with the inlet concentrations of 3.2, 4.8 and 6.0 g m^{-3}, respectively. The experimental data confirmed the decrease in the removal efficiency and increase in exit VOC concentration when the inlet benzene concentration was increased. Excessive addition of input benzene beyond the threshold withstanding ability of microbes was reported to be a reason for this behaviour [26]. At higher inlet benzene concentrations,a transient state induced by high concentration shock to microorganism was reported to occur and it leads to kinetic limitation. In addition, drying of the bed axially, channeling of air stream due to biomass accumulation and product inhibition were reported to be the other factors [8].

Elimination capacity, defined as the amount of pollutant degraded per unit time, normalized to the packed bed volume is reported to be affected by the inlet loading of benzene. The influence of inlet benzene loading rate on the elimination capacity was studied in the range of 0-306 g m^{-3} h^{-1}. From Figure 3, a linear relationship between elimination capacity and loading rate was observed. A linear relation was reported in the biofiltration study on hydrogen sulfide using granular activated carbon [27]. The maximum elimination capacity achieved was 192.7 g m^{-3} h^{-1} as shown in the plot which is comparatively higher than the capacities of 3.8 and 44.9 g m^{-3} h^{-1} reported on other benzene biofiltration studies [6,23]. The rate of increase in elimination capacity was observed to be slower at higher loading rates of benzene. This phenomenon could be related to the attainment of maximum removal capacity of microorganisms. Biofiltration studies on hydrogen sulfide using pine bark filter media reported a similar relationship between elimination capacity and inlet loading rate [28]. The leachate was collected periodically from the tail end of the column and its volume was approximately 250 mL per day.

Effect of EBRT

The flow rate of the benzene laden air is identified as an important process variable as it decides the quantity of pollutant to be degraded per unit time. In this study, the influence of flow rate (or EBRT) was studied in the range of 1.2-4.7 min. Figure 4 presents a plot of EBRT versus removal efficiency attained at different times. At increased EBRT, the time of exposure of microorganism to the target pollutant was high and could possibly resulted in better removal efficiency [19,26]. It was observed that when the inlet benzene concentration was low in the range of 1.5-3.0 g m^{-3}, the removal efficiencies achieved at lowest flow

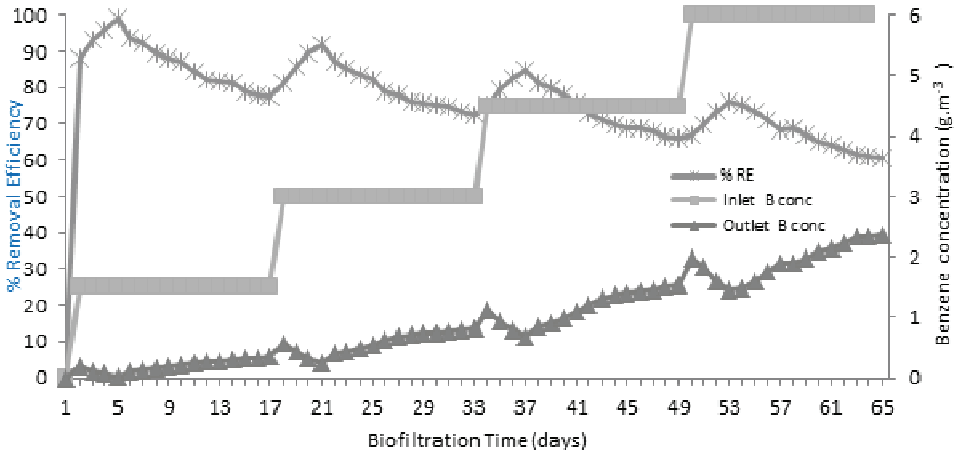

Figure 2. Biofiltration results for benzene removal at different inlet loading rates.

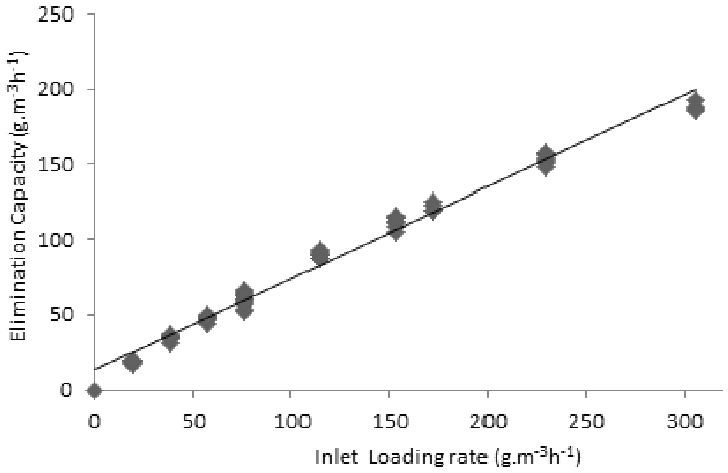

Figure 3. Effect of inlet loading rate on elimination capacity of benzene.

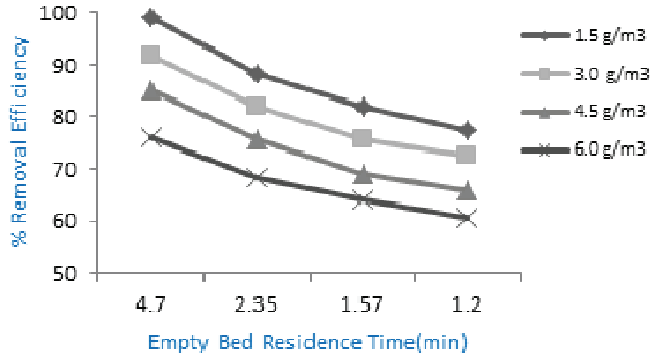

Figure 4. Effect of EBRT on benzene removal efficiency.

rate were higher by 20%. But, at higher benzene concentrations of 4.5 and 6.0 g m^{-3}, the effect is less pronounced with a reduced net difference in removal efficiency.

Effect of biofilter height

The biofilter reactor employed consisted of four different sections with different biomass distributions. In order to identify the relative contribution of each section in benzene degradation, the concentration profile was studied axially and plotted in Figure 5. The lowest section of the biofilter contributes to 40-45% of the total removal while the other sections degrade the remaining benzene. The reasons reported to be responsible for better removal at the section-1 were better biomass distribution and moisture content. During the course of biofiltration, the temperature of

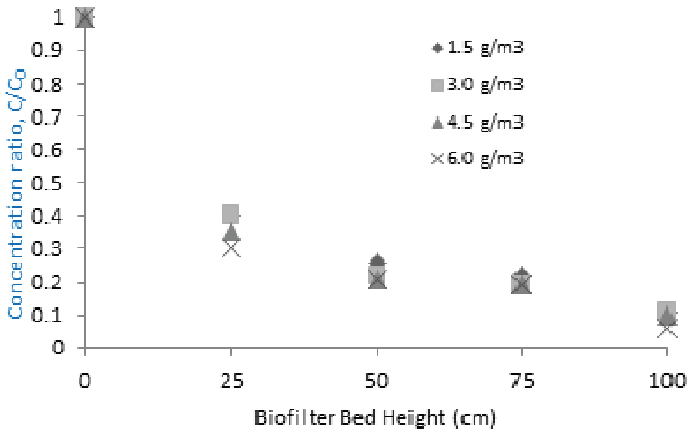

Figure 5. Concentration profile of benzene along the biofilter height.

the bed is expected to increase which could affect the moisture content at the top sections of the biofilter. Since the biofilter was operated in up-flow mode, the driving force varies from section to section. The local benzene concentration was high at the lowest section and lowest in the topmost section of the biofilter [6]. Higher microbial density and homogeneous biomass distribution in the lowest part of the bed could be the reasons for this behaviour. Moreover, the concentration gradients in the upper sections are comparatively lower, which resulted in lower removal efficiencies. Also, the filter bed could be dry in the upper sections due to the exothermic nature of the bio reaction. More efficient removal of toluene was reported in the first section of a compost based biofilter [24]. Studies on removal of xylene in the inlet loading range of 12-34 g m^{-3} h^{-1} reported most of the removal in the lower part of the biofilter column in a peat based biofilter [23].

Variation of bed temperature

The temperature variations during the biofiltration experiments were followed by recording the average bed temperature. Figure 6 shows the interactive variation patterns in temperature and elimination capacity. As expected, the biodegradation reaction of benzene was found to be exothermic which is proved by increase in temperatures at higher elimination capacities and microbial metabolic activity causes the increase of temperature. The temperature of the bed increased from 26.6 to 29.7 °C when there is a corresponding increase in elimination capacity from 16.8 to 192.7 g m^{-3} h^{-1}. The increase in temperature within the mesophilic range was reported to favour better removal of benzene by enhancing the activity of microorganisms [8].

Carbon dioxide production profile

The proposed biodegradation mechanism of benzene is as shown in Eq. (6), where the possible products are carbon dioxide, water and biomass:

$$a\mathrm{C_6H_6} + b\,\mathrm{O_2} \rightarrow c\,\mathrm{CO_2} + d\,\mathrm{H_2O} + \mathrm{e(biomass)} \quad (6)$$

The carbon dioxide production profile was recorded and plotted against elimination capacity in Figure 7. A linear relationship represented by an equation $CPR = 1.76EC + 18.6$ was found out with a high value of correlation coefficient, R^2 (> 0.99). When complete mineralization of benzene into water and carbon dioxide was assumed, the stoichiometric mole ratio between benzene and carbon dioxide is 1:6 and mass ratio could be 1:3.38. However, the actual conversion ratio was 1.76, which was lower than the theoretical value of 3.38. The difference in value was attributed to the possible consumption of benzene for the microbial growth inside the reactor during the biofiltration process. Also, the carbon dioxide could have accumulated in the liquid phase in the form of carbonates and bicarbonates [29]. Studies on removal of toluene in peat based biofilter [30] and xylene in an inert filter media biofilter [2] reported actual conversion ratios lesser than the stoichiometric ratios.

Biomass profile during biofiltration

Biodegradation efficiency depends on the establishment of uniform biofilm and growth intensity inside the biofilter column. The dry cell mass was measured in different sections during the benzene biofiltration experiments and plotted in Figure 8. In section 1 of the biofilter column, the dry cell mass concentration increased from 0.16 to 0.35 g g^{-1} of filter media and found to be the maximum biomass buildup compared

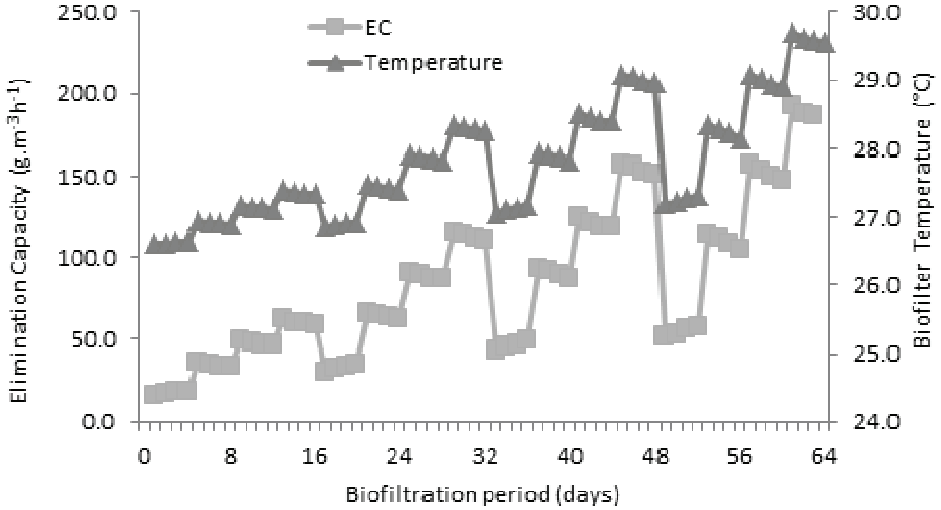

Figure 6. Temperature variation profile during the biofiltration experiments.

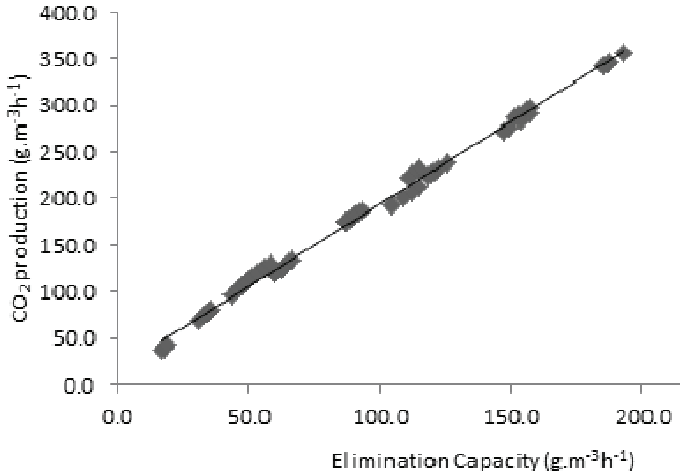

Figure 7. Elimination capacity versus carbon dioxide production.

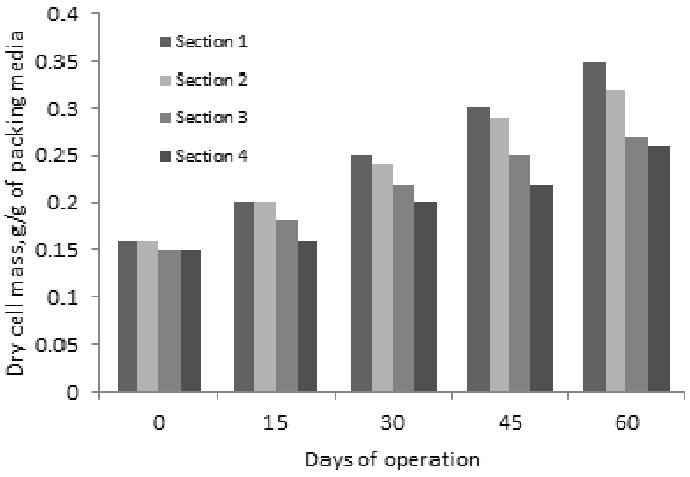

Figure 8. Biomass profile during benzene biofiltration.

to other sections of the biofilter. This observation was supplemented by enhanced removal percentages at section-1. These results show the attainment of a stable microbial density which is higher than the initial biomass in the filter media. The cell mass build up was found to be less at the top section due to the lower availability of benzene.

Kinetic modeling

The removal rate of benzene in the immobilized cell biofilter was fitted to a modified Michaelis- Menten model, shown below in a linearized form as Eq. (7) [31]:

$$\frac{1}{EC} = \frac{K_s}{EC_{max} C_{ln}} + \frac{K_s}{EC_{max}} \tag{7}$$

where EC_{max} (g m^{-3} h^{-1}) is the maximum EC, C_{ln} (g m^{-3}) is the logarithmic mean of inlet and outlet benzene concentrations and K_s (g m^{-3}) is the saturation constant. The slope and intercept of the linear fit between $1/EC$ and $1/C_{ln}$ plot shown in Figure 9 gives the values of the kinetic parameters, EC_{max} and K_s. The value of R^2 was found to be 0.61. The kinetic constants will depend on the microorganisms attached to the filter media. The values of EC_{max} and K_s were found to be 217.4 g m^{-3} h^{-1} and 3.54 g m^{-3}, respectively. The value of EC_{max} observed in this study was found to be in the range of values reported for ethyl benzene removal using *Macadamia ternifolia* nutshells as filter media [32]. The physical meaning of K_s corresponds to the benzene concentration that must be treated to achieve $EC_{max}/2$. A material having a small K_s value was reported to have a greater affinity to benzene and vice-versa. The value of K_s observed in this study was comparatively less than the values of 70 g m^{-3} reported with biofiltration of hydrogen sulfide in a novel media biofilter [28] and 47 g m^{-3} reported with biofiltration of hydrogen sulfide and ammonia in an alginate beads based biofilter [33].

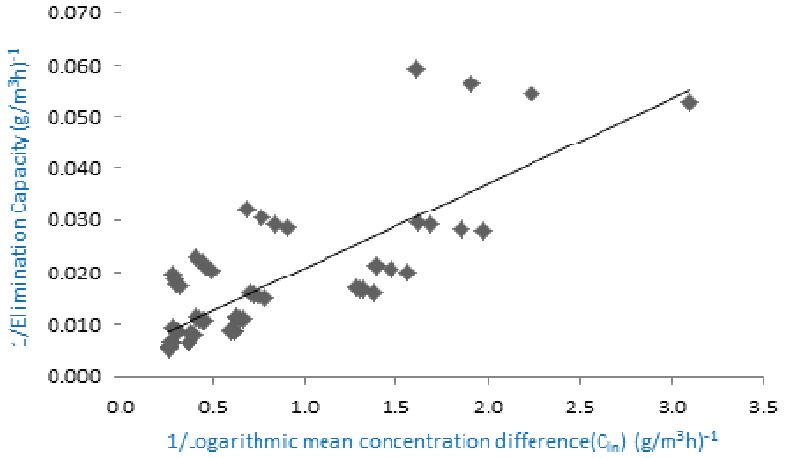

Figure 9. Kinetic model plot for biofiltration kinetics of benzene.

CONCLUSION

The performance of a biofilter to treat benzene at different inlet loading rates was evaluated using a novel tree bark filter medium. The influence of inlet benzene concentration on the removal efficiency was studied and the biofilter was found to handle high concentrations of benzene effectively. Higher retention times or lower flow rates favoured better removal of benzene in the biofilter. The axial concentration profile of benzene in the biofilter column proved a larger percentage of benzene removal in the lowest part of the column reactor. Carbon dioxide production rate was compared with the elimination capacity of benzene and a linear relationship was proposed. Temperature variations were recorded and found to vary with elimination capacity changes. The biokinetic constants were calculated using a modified Michaelis--Menten model. The results suggest that the date palm tree bark based biofilter is a suitable choice for treating benzene in the concentration range of 1.5-6.0 g m^{-3}.

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NAUČNI RAD

PROCENA PERFORMANSI I KINETIKA UKLANJANJA BENZENA U BIOFILTERU NA BAZI KORE DRVETA SA STRUJANJEM NAVIŠE

U radu je pogodnost primene kore drveta Phoenix dactylifera kao novog filter medija u biofilteru sa strujenjem naviše i mešanom kulturom za uklanjanje benzena. Eksperimenti su rađeni sa različitim koncentracijama benzena (1,5-6,0 g m^{-3}) i vremenom zadržavanja u filter bez biomaterijala (EBRT = 1,2- 4,7 min). Nađeno je da eliminacioni kapacitet linearno varira sa brzinom ulaznih količina benzena u opsegu od 0-306 g.m^{-3} h^{-1}. Pri koncentraciji benzena od 1,5 g m^{-3} postiže se efikasnost uklanjanja od 99%. Ona opada sa porastom koncentracije benzena. Pri manjim protocima efikasnost uklanjanja benzena je veća. Analiziran je koncentracioni profil za različite visine filter medija. Pošto temperatura raste za vreme biofiltracije eksperiment potvrđuje egzotermnu prirodu biofiltracije. Brzina stvaranja ugljen dioksida u odnosu na kapacitet eliminacije data je jednačinom CPR = 1.76EC + + 18.6. Primenom Michaelis-Menten modela nađene su kinetičke konstante, kao što su: maksimalni eliminacioni kapacitet 217,4 g m^{-3} h^{-1} i konstanta zasićenja 3,54 g m^{-3}.

Ključne reči: kinetika, benzen, zagađenje.