

Full Length Research Paper

Removal of xylene in gas - phase using compost - ceramic ball biofilter

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Biofiltration is fast emerging as a feasible option for treating odorous compounds and other volatile organic compounds (VOCs) from process waste - gas streams using microorganisms attached to porous support matrix. Compost, owing to its inherent physico-chemical and biological characteristics, has shown to be a promising filter material in biofiltration to treat both hydrophobic and hydrophilic gas - phase VOCs at low concentrations and high gas flow rates. This study aimed at evaluating the potential of a laboratory-scale biofilter, inoculated with mixed culture, to remove gas-phase xylene from a synthetic waste gas stream. The performance of the biofilter was studied by varying the flow rate from 0.024 to 0.072 m³/h, corresponding to empty bed residence times varying between 0.81 - 2.45 min and by changing the inlet loading rates (ILR) between 3.5 to 208 g/m³.h. Removal efficiencies higher than 68% were achieved for xylene loading rates lesser than 60 g/m³.h. However, due to the hydrophobic nature of the pollutant, xylene, that hinders mass transfer, and/or substrate inhibition to the microorganisms, a significant reduction in the removal efficiency was observed at high xylene concentrations. The results demonstrate the potential of compost biofilter to handle microorganism- tolerable xylene loads under steady-state conditions.

Key words: Biofiltration, compost, gas-phase xylene, mixed culture, performance capacity.

INTRODUCTION

Xylene (C₈H₁₀), a hydrophobic volatile organic compound, is among the 188 hazardous air pollutants listed under the USEPA - Clean Air Act 1990. Xylene and its isomers are widely used as solvents in chemical industries, viz., paint and varnish, printing and semiconductor manufacture industries. Due to improper practices and treatment, a substantial amount of vapors containing various xylenes are being emitted to the ambient atmosphere from process industries. It is reported to have significant effect on human health and natural environment (WHO, 1986). Repeated contact can produce dermatitis (dryness and cracking) due to degreasing action, besides being toxic to the liver, kidneys and the central nervous system (Forsyth and Faust, 2009). These severe ill-effects have led to increased attention from the regulatory authorities, who have emphasized on the continuous develop-

ment and modernization of the existing VOC control technologies. The Occupational Safety and Health Administration (OSHA) have set a maximum airborne concentration of 100 ppm xylene in workplace for an 8 h work day, 40 h work week.

Biodegradation, through immobilized bioreactor configurations such as a biofilter, appears to be a promising alternative for the complete mineralization of xylene to innocuous end products. The simplicity in the operation of biofilters has resulted in its emergence as a more practical treatment option for the treatment of hydrophobic VOCs such as xylene (Wu et al., 2006; Jeong et al., 2008; Saravanan and Rajamohan, 2009). A complex phenomenological step consisting of adsorption, absorption, diffusion and biodegradation takes place (Kennes and Thalasso, 1998). Furthermore, the removal and oxidation rates of these hazardous contaminants depend principally on the biodegradability, reactivity and largely on the solubility of the pollutant in the liquid layer of the biofilm. Jeong et al. (2008) studied the removal of *o*-xylene in batch and continuous systems using a xylene

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degrading bacterium, *Rhodococcus sp.* Strain BT062 and showed that a maximum specific degradation rate of $0.24 \times 10^{-7} \mu\text{mol/h/cell}$ could be achieved in batch systems. However, when the same strain was inoculated in the biofilter, under non-sterile conditions, a maximum elimination capacity (EC) of $240 \text{ g/m}^3 \cdot \text{h}$ was reported. A 2 month acclimatized culture, seeded with activated sludge from a secondary clarifier of a pharmaceutical industry, was used to inoculate a biofilter packed with commercial press mud. The removal efficiency profiles were reported to be decreasing function of an increased flow rate. The maximum RE was 98% at a gas flow rate of $0.03 \text{ m}^3/\text{h}$ and at an inlet xylene concentration of 0.2 g/m^3 (Saravanan and Rajamohan, 2009). These experimental studies have proved biofiltration as an efficient waste gas treatment process and a reliable technology for the control of hydrophobic VOCs like xylene. This paper present the performance of a compost based mixed culture biofilter treating xylene vapors at high concentrations. Besides, important literatures pertaining to gas - phase xylene treatment and the corresponding elimination capacities achieved in biological systems have been presented.

MATERIALS AND METHODS

Microbial consortia and medium composition

A mixed microbial culture obtained from a municipal sewage treatment plant was acclimatized with Xylene as the carbon source in a well defined mineral salt medium having the following composition: (gram per liter in distilled water): K_2HPO_4 - 0.8, KH_2PO_4 - 0.2, $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$ - 0.05, $\text{MgSO}_4 \cdot 7\text{K}_2\text{O}$ - 0.5, $(\text{NH}_4)_2\text{SO}_4$ - 1.0 and FeSO_4 - 0.01. The pH of the mineral salt media was adjusted to 6.85 and the cultures were grown under ambient conditions in a rotary shaker at 175 rpm. This culture, taken at its exponential growth phase, was then used to inoculate the biofilter.

Biofilter

The biofilter was constructed from acrylic tube (5 cm diameter and 70 cm height). The packing in the biofilter consisted of a mixture of sieved compost (3 - 6 mm) and ceramic - balls (4 - 6 mm), mixed together in a 60:40 proportion (v/v). A perforated plate was provided at the bottom to support the packed bed. The addition of ceramic beads offered several benefits to the biofilter: They are inert, stable and homogenous, that allows minimizing pressure drop and channeling problems. On the other hand, they also maintain the structural integrity of the biofilter. Gas sampling ports sealed with rubber septa were provided at equal intervals along the biofilter height.

Experimental

A schematic of the experimental setup is given in Figure 1. Liquid xylene placed in a trough, was vaporized and humidified at constant air flow rate, controlled through valves and was passed through the bed in an up - flow mode. The compost - based filter bed was inoculated with xylene acclimatized mixed culture, that was initially grown under batch conditions in 250 ml Erlenmeyer flasks, at 30°C and with initial xylene concentrations of 150 mg/l . These stock cul-

tures were repeatedly added from the top of the biofilter and drained after 2 h. This procedure was repeated everyday, for the first 4 days, before feeding gas - phase xylene to the biofilter. The bed moisture content was maintained, at about 55 - 60%, by periodically adding fresh mineral salt medium from the top of the bioreactor. The leachate thus generated, was removed with the help of a port at the bottom. Experiments were carried out by varying the flow rates of the xylene vapors and humidified air, independently to get different initial concentrations and empty bed residence times in the biofilter. Gas samples were collected from different ports and analyzed for residual xylene concentration.

Analytical methods

Xylene (mixture of isomers) concentration in gas samples were measured by gas chromatography (Model 5765, Nucon gas chromatograph) fitted with a stainless steel column and a flame ionization detector. Nitrogen was employed as the carrier gas at a flow rate of 20 ml/min . The temperatures at the GC injection, oven and detection ports were 150 , 120 and 250°C , respectively.

RESULTS AND DISCUSSION

Biofilter performance was evaluated in terms of the removal efficiency (RE, %) and the elimination capacity of the filter bed (EC, $\text{g/m}^3 \cdot \text{h}$), which can be represented by:

$$RE = \frac{C_{gi} - C_{go}}{C_{gi}} \times 100 \quad (1)$$

$$ILR = \frac{Q(C_{gi})}{V} \quad (2)$$

$$EC = \frac{Q(C_{gi} - C_{go})}{V} \quad (3)$$

Where;

C_{gi} = the inlet xylene concentration in the biofilter, g/m^3 ;
 C_{go} = the outlet xylene concentration in the biofilter, g/m^3 ;
 ILR = the inlet loading rate, $\text{g/m}^3 \cdot \text{h}$;
 V = the volume of the filter bed m^3 and
 Q = the gas flow rate, m^3/h .

In order to obtain sufficient biomass concentration in the filter bed, the compost biofilter was acclimatized by passing low concentrations of gas - phase xylene ($0.1 - 0.3 \text{ g/m}^3$) at a gas flow rate of $0.024 \text{ m}^3/\text{h}$ for 36 days. This corresponds to an EBRT of 2.45 min of that pollutant within the biofilter. The degree of acclimatization primarily depends on the adaptive capability of the microorganisms present in the compost, substrate concentrations, nutrient concentration and its availability and other necessary environmental conditions. However, the efficient functioning of any biofilter unit strongly depends on the inlet pollutant load (Jorio et al., 1998). Initially, during the first 15 days, for low inlet loading rates during the acclimatiza-

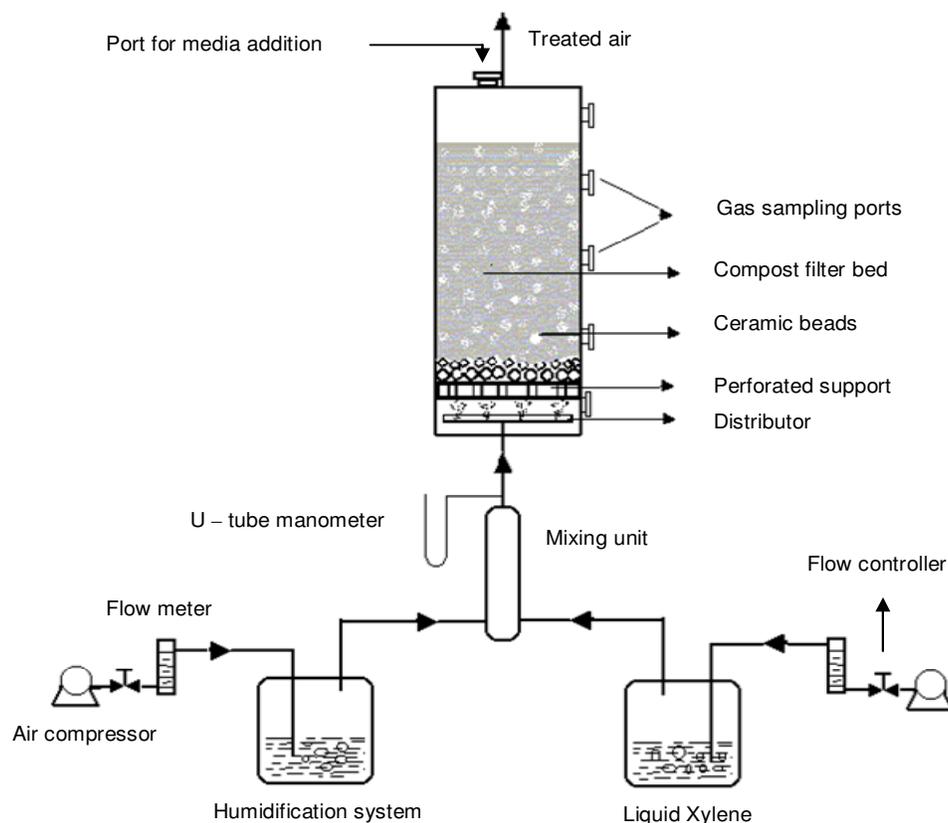


Figure 1. Schematic of the compost biofilter.

Table 1. Experimental scheme for continuous benzene degradation experiments.

Phases of biofilter operation	Empty bed residence time (min)	Range of ILR tested ($\text{g}/\text{m}^3\cdot\text{h}$)	Operating time (days)
Acclimatization	2.45	<7	36
Phase I	2.45	8 - 15.7	15
Phase II	1.63	12 - 54.9	39
Phase III	0.81	78 - 207.8	28

tion step ($< 7 \text{ g}/\text{m}^3\cdot\text{h}$), the RE was maintained at 70%. The removal efficiency increased slowly from the 16th day and reached a maximum of 91% on the 35th day. These profiles indicate that biomass growth occurred and that the mixed microbial consortium was able to utilize xylene as its sole carbon and energy source within the biofilter.

The combined effect of gas - phase xylene concentration and gas flow rate was investigated in three phases (I, II and III) by subjecting the biofilter to different flow rates and inlet concentrations, as shown in Table 1, while the results are graphically explained in Figure 2. Steady state experiments were maintained for 3 to 7 day before switching over to the next operating condition that is changes in the concentration of gas – flow rate. On the

37th day, the concentration was increased to $0.55 \text{ g}/\text{m}^3$, while the same EBRT (2.45 min) was maintained. It was observed that the RE dropped by about 10%, due to a sudden increase of xylene concentration. In the later days of Phase - I, the concentrations were between $0.55\text{--}0.65 \text{ g}/\text{m}^3$, while RE was maintained close to 83%. During Phase - II, the EBRT was reduced to 1.63 min, while xylene concentrations were lesser than $1 \text{ g}/\text{m}^3$ (days 51 - 73). The ILR to the biofilter was maintained between $15\text{--}32 \text{ g}/\text{m}^3\cdot\text{h}$, with more than 75% RE. From thereon, during days 74 - 89, the inlet concentrations were increased from nearly 1 to $1.5 \text{ g}/\text{m}^3$. This increase in concentration (ILR: $44\text{--}55 \text{ g}/\text{m}^3\cdot\text{h}$) caused a 5% decrease in the removal profiles. At these flow rates, the compost biofilter

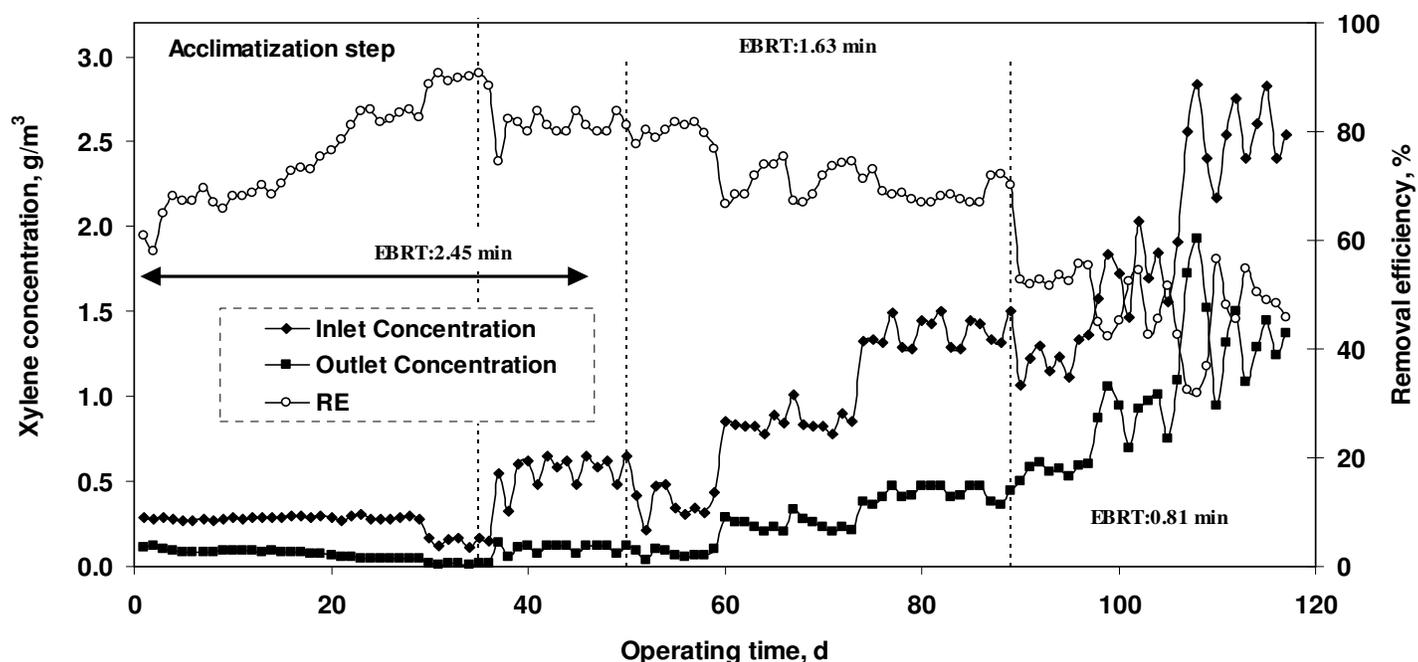


Figure 2. Different phases of biofilter operation - variation in removal efficiency with change in inlet xylene concentration at different empty bed residence time.

operated at high efficiencies (68 - 90%) till an initial concentration of about 1.5 g/m^3 . The outlet concentration remained fairly uniform even when the inlet concentrations were fluctuating. At high gas flow rates in Phase - III, corresponding to EBRT of 0.81 min and at higher xylene concentrations, the removal efficiency decreased rapidly to 52%. When the concentration was increased beyond 2 g/m^3 , the removal efficiency declined gradually, reaching less than 50% for 2.8 g/m^3 of xylene inlet concentration. For high gas flow rates, low values of EBRTs, the contact time between the microorganism and gas - phase xylene was too short and the microorganism had insufficient time to perform the required degradation on the viable amount of xylene. In general, as observed throughout the experimental period, xylene concentrations greater than 1.5 g/m^3 , showed a significant reduction in removal efficiency, which may be due to substrate inhibition at high concentrations. This means that inlet concentrations greater than 1.5 g/m^3 , exceeds the biodegradation capacity of the microorganisms for achieving high removal efficiency at this gas flow rate. Moreover, the removal profiles of xylene at higher concentrations indicate that the rate limiting step for mass transfer appears to be diffusion through the aqueous phase. It is a well - known fact that elimination rate of VOCs in gas - phase depends highly on diffusion rate of the gas - phase pollutant (xylene) to the biofilm layer and the subsequent degradation rate within the biofilm. In other words, diffusion limitation occurs in the wet biofilm layer which is not fully active. It has been reported that aerobic degradation of xylene is usually initiated by progressive oxida-

tion of the alkyl side chain of the aromatic ring to produce carboxylic acids, or ring oxidation which produces substituted pyrocatechols. Carboxylic acids and pyrocatechols are then transformed to substrates of the citrate cycle through cleavage of the aromatic ring (Jindrova et al., 2002). *Pseudomonas* sp. represents the most common isolates of microorganisms found in domestic and Industrial wastewater treatment facilities. In a previous study, *Pseudomonas* sp., isolated from a muddy pond was able to grow utilizing *p* - xylene (strain P - X) and *m* - xylene (strain M - X) as the sole carbon and energy source. The methyl group of *p* - xylene was oxidized by P - X strain to *p* - toluate, which is converted by oxidization and decarboxylation to 3 - or 4 - methylpyrocatechol. *m* - Xylene was oxidized to *m* - toluate and 3 - methylpyrocatechol. Methyl pyrocatechol was then mineralized by meta-cleavage to hydroxymuconaldehydate and converted to 2 - pyridinecarboxylate in the presence of ammonia (Davis et al., 1968).

The elimination capacity, which reflects the capacity of the biofilter to remove the pollutant, is plotted as a function of the inlet xylene load in Figure 3. A near linear relation between the two variables was observed till an inlet load of $37 \text{ g/m}^3 \cdot \text{h}$, which corresponded to Phase I and phase II operations. However, for higher initial concentration and higher flow rate used in phase III, the elimination capacity of the filter bed increased initially and soon became constant at higher inlet loads. At inlet load beyond $100 \text{ g/m}^3 \cdot \text{h}$ and up to $208 \text{ g/m}^3 \cdot \text{h}$, the EC increased at a slower rate from $40 \text{ g/m}^3 \cdot \text{h}$ and reached a maximum EC of $101.3 \text{ g/m}^3 \cdot \text{h}$. This maximum EC corres-

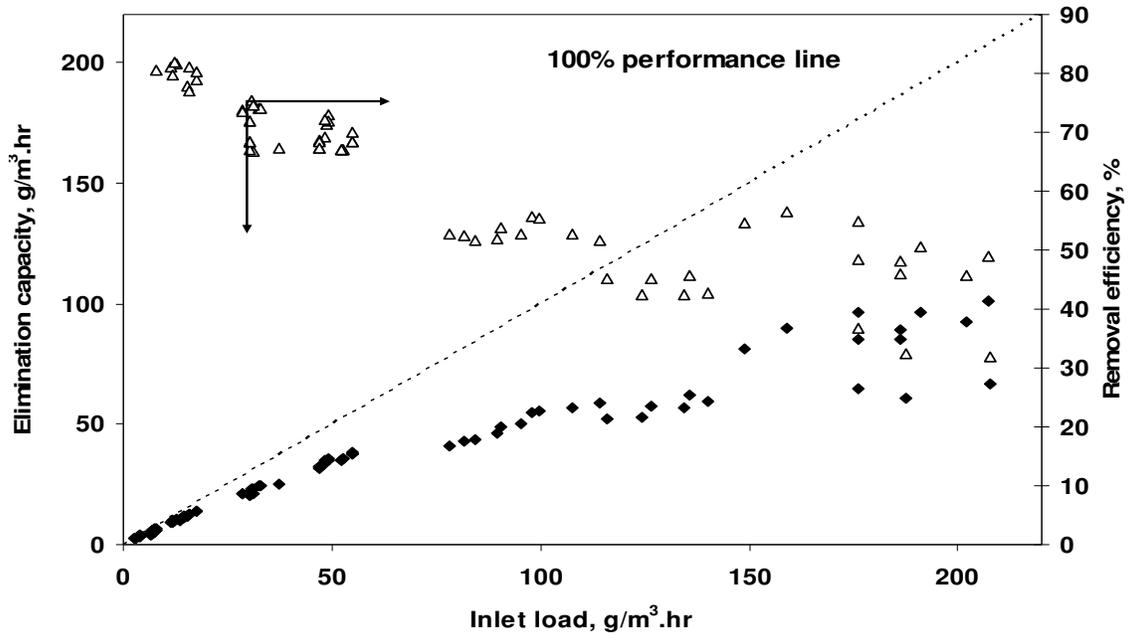


Figure 3. Influence of inlet xylene load on the elimination capacity and removal efficiency of the biofilter.

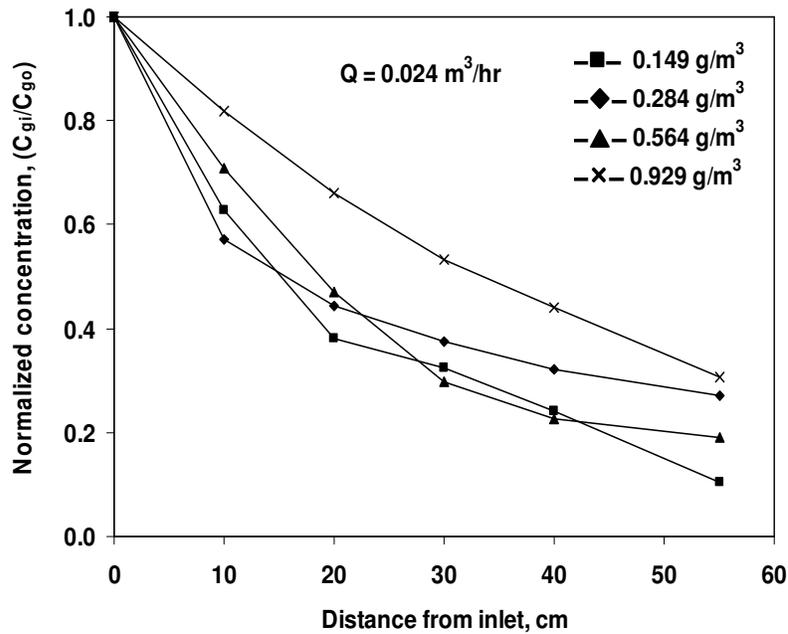


Figure 4. Normalized xylene concentration profiles as a function of the biofilter height .

ponds to an ILR of 208 g/m³.h with 49% RE. The biodegradation rate for xylene decreased as the inlet load to the biofilter increased, suggesting that inlet load range reached their inhibition levels for microbial growth. This trend also substantiates the fact that biomass concentrations may be the limiting factor during biodegradation of hydrophobic compounds like xylenes at higher concen-

trations. The results were compared with other biofiltration literature reports and it was found that the maximum EC observed herein this study is higher than most of the previously reported work on xylene removal (Table 2).

The dynamics of xylene removal was understood by plotting the normalized xylene concentration profiles as a function of the biofilter height (Figure 4). The results indi-

Table 2. Literature comparing elimination capacities achieved in biofilters for xylene removal.

Packing material	Inoculum source	EC _{max} [†]	Reference
Press mud from sugar industry waste and berl saddles	Activated sludge from pharmaceutical industry	67	Saravanan and Rajamohan (2009)
Biosol® - made of foamed waste glass mixed with corrugated cardboard	<i>Rhodococcus</i> sp. – BT062 ^a	45* 240**	Jeong et al. (2008)
Mature pig compost, forest soil and packing material made of poly ethylene	Activated sludge	80	Wu et al. (2006)
Food waste compost and pig manure compost	Activated sludge from petroleum wastewater treatment plant	21	Hwang et al. (2007)
Poly urethane foam (PUF)	Mixed culture	80	
Conditioned spherical peat	Activated consortium ^b	61	Elmrini et al. (2004)
Foam cubes	Bacteria and fungi ^c	62	Li and Liu (2006)
Biosol® - made of foamed waste glass mixed with corrugated cardboard	<i>Pseudomonas</i> sp. NBM 21	150* 160**	Jeong et al. (2006)
Peat	Mixed microbial consortium	67	Jorio et al. (2000)
Peat	<i>Pseudomonas pseudoalcaligenes</i> strain BTX 02 ^d	8.1	Oh and Choi (2000)
Compost and ceramic beads	Mixed culture	101.3	This study

[†], units (g/m³.h); ^a, Isolated from activated sludge of a wastewater treatment plant;

^b, Specific aerobic and facultative anaerobic species were present;

^c, *Bacillus subtilis*, *Paenibacillus* sp. and *Aureobacterium* sp. were present in the first zone, while in the second zone, Eukaryotic organisms, *Aspergillus candidus* and *Penicillium frequentans* were present. First zone refers to a bacterial - suspended growth reactor and second zone refers to fungal - attached growth reactor;

^d, Isolated from wastewater of an industrial complex;

* under sterile conditions;

** under non - sterile conditions.

cate that the removal is more efficient in the lower part of the biofilter than in the upper part of the filter at constant flow rates (0.024 m³/h). At low concentrations (0.139 g/m³), more than 60% of xylene was removed in the first 20 cm of the bed height, while the rest of the 30 cm removed only an additional 30%. At high concentrations (0.929 g/m³), nearly 35% xylene was removed in the first 20 cm, followed by an additional 32% removal in the remaining parts. Similar observation has been reported in the literature. The first one - third of the whole length of the packed column, a cork - biofilter, removed as much as 79% of the incoming BTEX vapors and relationship between concentration versus column height, suggested a 1st order gradient (Kwon and Cho, 2009). The high removal in the first two sections of the filter bed may be due to higher concentrations of microbial population and higher moisture content (Rene et al., 2009) in sections near the inlet of the filter bed. Zilli et al. (2005) has shown that higher biomass concentration near the inlet of the biofilter was due to the higher benzene loading rates in the inlet zone that stimulated the pollutant bio - oxidation and cell development. The results from their study showed a maximum removal capacity of 20.1 g/m³.h at

benzene - loading rate of 24.8 g/m³.h.

Conclusion

A compost biofilter, inoculated with activated sludge microbes acclimatized with xylene, was successfully tested for its maximum removal performance. The performance of the biofilter was assessed by studying the effects of flow rate and gas - phase xylene concentration on the removal efficiency and elimination capacities of the filter bed. The biofilter attained a maximum elimination capacity of 101.3 g/m³.h at an inlet load of 207.8 g/m³.h. The removal efficiencies were greater than 75% when the gas flow rate was 0.024 m³/h and inlet concentration was less than 0.5 g/m³. The results show that higher values of gas residence times and lower values of concentrations were favorable conditions for xylene degradation using mixed culture in compost biofilter.

The compost biofilter could be optimized further for its performance under high pollutant loads and transient operating conditions such as fluctuating - shock loads

and shut - down and re - start operations, which are frequently encountered in industrial situations.

REFERENCES

- Davis RS, Hossler FE, Stone RW (1968) Metabolism of p - and m - xylene by species of *Pseudomonas*. *Can. J. Microbiol.* 14: 1005 - 1009.
- Elmrini H, Bredin N, Shareefdeen Z, Heitz M (2004). Biofiltration of xylene emissions: bioreactor response to variations in the pollutant inlet concentration and gas flow rate. *Chem. Eng. J.* 100: 149 - 158.
- Forsyth CS, Faust RE (2009). Center for Integrated Risk Assessment (<http://cira.ornl.gov/>), Accessed - May 2009.
- Hwang JW, Jang SJ, Lee EY, Choi CY, Park S (2007). Evaluation of composts as biofilter packing material for treatment of gaseous p - xylene. *Biochem. Eng. J.* 35: 142 - 149.
- Jeong E, Hirai M, Shoda M (2006). Removal of p - xylene with *Pseudomonas* sp. NBM21 in biofilter. *J. Biosci. Bioeng.* 102: 281 - 287.
- Jeong E, Hirai M, Shoda M (2008). Removal of o - xylene using biofilter inoculated with *Rhodococcus* sp. BTO62. *J. Haz. Mat.* 152: 140 - 147.
- Jindrova E, Chocova M, Demnerova K, Brenner V (2002). Bacterial aerobic degradation of benzene, toluene, ethylbenzene and xylene. *Folia Microbiol.* 47: 83 - 93.
- Jorio H, Bibeau L, Viel G, Heitz M (2000). Effects of gas flow rate and inlet concentration on xylene vapors biofiltration performance. *Chem. Eng. Sci.* 76: 209 - 221.
- Jorio H, Kiared K, Brzezinski R, Leroux A, Viel G, Heitz M (1998). Treatment of air polluted with high concentration of toluene and xylene in a pilot - scale biofilter. *J. Chem. Technol. Biotechnol.* 73: 183 - 196.
- Kennes C, Thalasso F (1998). Waste gas biotreatment technology. *J. Chem. Technol. Biotechnol.* 72: 303 - 319.
- Kwon SH, Cho D (2009). A comparative, kinetic study on cork and activated carbon biofilters for VOC degradation. *J. Ind. Eng. Chem.* 15: 129 - 135.
- Li L, Liu JX (2006). Removal of xylene from off - gas using a bioreactor containing bacteria and fungi. *Int. Biodeter. Biodegrad.* 58: 60 - 64.
- Oh YS, Choi SC (2000). Selection of suitable packing material for biofiltration of toluene, m - and p - xylene vapors. *J. Microbiol.* 38: 31 - 35.
- Rene ER, Veiga MC, Kennes C (2009). Performance of a biofilter for the removal of high concentrations of styrene under steady and non-steady state conditions. *J. Hazard. Mat.* 168: 282 - 290.
- Saravanan V, Rajamohan N (2009). Treatment of xylene polluted air using press mud-based biofilter. *J. Haz. Mat.* 162: 981 - 988.
- WHO (1986). *Occupational Diseases*, Macmillan Publishers, Singapore.
- Wu D, Quan X, Zhao Y, Chen S (2006). Removal of p - xylene from an air stream in a hybrid biofilter. *J. Hazard. Mater.* B136: 288 - 295.
- Zilli M, Guarino C, Daffonchio D, Borin S, Converti A (2005). Laboratory - scale experiments with a powdered compost biofilter treating benzene - polluted air. *Proc. Biochem.* 40: 2035 - 2043.