International Journal on Emerging Technologies **8**(1): 97-102(2017)

ISSN No. (Print): 0975-8364
ISSN No. (Online): 2249-3255

A Study of Removal of Ethylbenzene and Xylene Using Biofilter

Ashish Kumar Yadav and S.K. Patidar

Department of Civil Engineering, NIT Kurukshetra, (Haryana), India

(Corresponding author: Ashish Kumar Yadav)
(Received 22 December, 2016 accepted 10 January, 2017)
(Published by Research Trend, Website: www.researchtrend.net)

ABSTRACT: In the present study, a laboratory scale corn-cob based biofilter inoculated with pseudomonas fluorescents (mtcc 9768) was used for investigating the degradation of ethylbenzene and m-xylene for a period of 60 days in two phases. for ethylbenzene maximum removal efficiency (re) was 72% at an inlet concentration of 4.26 gm^{-3} and for m-xylene re was 76% at an inlet concentration of 6.39 g m^{-3} at an ebrt of 59 seconds. maximum elimination capacity (ec) for ethylbenzene was 282.03 g m^{-3}h^{-1} at an inlet load of 368.52 g m^{-3}h^{-1} and for m-xylene maximum ec was 298.84 g m^{-3} h^{-1} at inlet load of 354.67 g m^{-3} h^{-1}.

Keywords: Biofiltration; BTEX; volatile organic compounds; removal efficiency; elimination capacity.

I. INTRODUCTION

Volatile organic Compounds (VOCs) emission contributed to the atmosphere causes adverse effects on the air quality. These air pollutants are generated from various chemical, petrochemical and allied industries. Benzene, toluene, ethylbenzene and xylene (BTEX) form a major group of aromatic VOCs because of their contribution in tropospheric chemistry and danger posed by them on human health. BTEX comprises 60% of non-methane VOCs and can be considered as one of the major pollutant because of its frequent use as industrial solvents and in production of agrochemicals, pharmaceuticals, explosive and many other everyday products. Among the aromatics hydrocarbons of BTEX, benzene is a human carcinogen which promotes myeloid leukemia, toluene exposure causes reproduction effects, ethylbenzene affects the blood, liver and kidneys and finally xylene exposure affect the central nervous system leading to cardiovascular and respiratory problems. The conventional technologies available for BTEX removal include adsorption, absorption, condensation, thermal incineration and membrane separation. However conventional technologies are not applicable everywhere due to high cost and generation of by-products. Unlike conventional technologies, gas phase biological treatment such as biofilter, membrane bioreactor, biotrickling filter and bioscrubber is an attractive option for BTEX control at a relatively low cost and minimum by-product generation. Biofiltration is one of the emerging biological treatment processes for removal of BTEX. In biofiltration process microbial attack occurs on the contaminants which are sorbed from the gas to the aqueous phase. During microbial attack microorganisms convert contaminants into CO_2, water vapour and organic biomass by the oxidation process. Various studies on BTEX biodegradation in biofilter over the last 10 years indicated that the substrate interaction between BTEX compounds as a rule differ with microbial culture and culture conditions. In the recent years, various agro-industrial residues including corn-cob have been used as a packing material in biofilter by the researchers. Many researcher have reviewed the principles of biological treatment for VOCs removal and its advantages over physical and chemical techniques, research is still going on to the biological treatment of VOCs for the use of effective new designs, packing media, microbial structure analysis and its modeling. Furthermore, there are many attempts being made to genetically modify the bacteria used in biological treatment to increase the degradation rate of a single pure micro-organism. In the present study, removal of ethylbenzene and m-xylene vapours was investigated using corn-cob as a filter material.

II. MATERIALS AND METHODS

A. Experimental Setup and Biofilter Operation

A schematic diagram of the experimental setup used for present study is shown in Figure 1. The biofilter consisted of four sections which were attached to each other with the help of screws. Each section has 20 cm height and 9.4 cm inner diameter. Acrylic sieve plates were placed at the joint of each section for supporting the packing material.

To assemble the biofilter column, o-rings were placed between each section. High vacuum silicon grease was used to make the biofilter air tight. The total packed bed volume was 4.65 L and total packed depth of bed was 70.5 cm. For measurement of temperature and collecting samples, sampling ports were provided at each section of biofilter. First of all, air from an air pump was passed through an activated carbon filter to eliminate any unwanted contaminants present in the air then air flow was separated into three streams such that two were having minor flow with the help of hand operated flow splitters. Two rotameters were used to record the air flow rate going to the ethylbenzene and m-xylene vessels so that concentration of these compounds in the inlet stream could be controlled. These compounds were added by bubbling air through liquid solutions filled in conical flasks containing ethylbenzene and m-xylene. The air flow rates through the conical flasks containing ethylbenzene and m-xylene were kept low. The major air flow stream was passed through humidifying chamber (5.5 cm diameter and 65 cm height) containing water and packed with PVC packing. After that major flow stream from humidifying chamber and the minor flow streams containing ethylbenzene and m-xylene were mixed in a mixing chamber. The humidified air, ethylbenzene and m-xylene mixture was then led to the reactor inlet for biodegradation. The above air mixture entered the top of the biofilter column as it was operated in down the flow mode.The biofilter was operated at various inlet concentrations of ethylbenzene and m-xylene. Samples were collected at a regular intervals of time from the inlet, outlet and as well as from the sampling ports using an airtight syringe and analyzed for residual ethylbenzene and m-xylene.

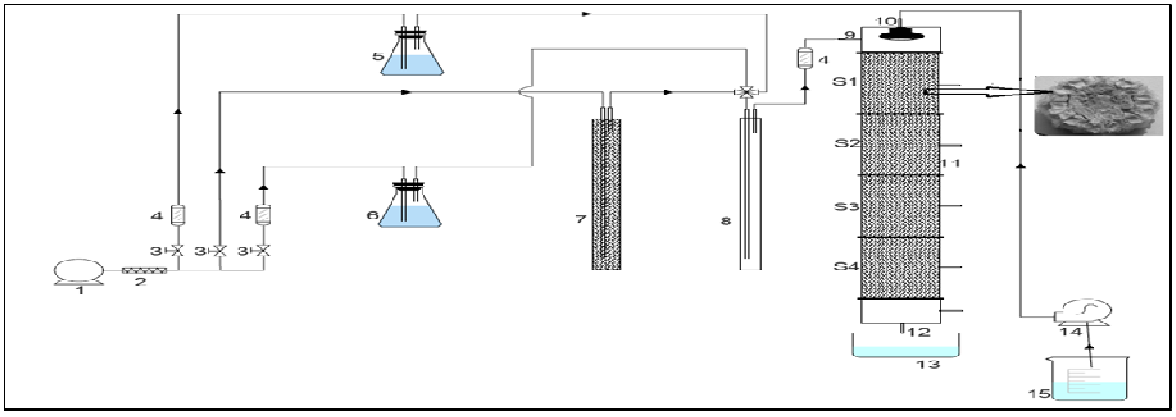

Fig. 1. Schematic Diagram of Biofilter Setup for treatment of Ethylbenzene and m-Xylene.

1. Air pump, 2. Carbon/ moisture trap, 3. Hand operated mass flow controller,4. Rotameter, 5. Ethylbenzene vessel, 6. m-Xylene vessel, 7. Humidifier, 8. Mixing chamber,
9. Spray nozzle, 10. Inlet gas,11. Biofilter, 12. Leachate collection port/ cleaned air, 13. Leachate collection vessel, 14. Peristaltic pump, 15. Nutrient solution container.

B. Packing Material

Corn-cob was used as a biofilter packing and inoculated with pure Pseudomonas fluorescens. The full size corn-cob were obtained from a local agricultural site, dried in an oven and cut into equal size cylindrical pieces (1.5 cm length and 2.0 cm diameter). They were washed twice with distilled water and finally dried in an oven at 105 °C for 5 hours. After drying, the pieces were sterilized at 15 psi for 20 min.

C. Micro-organism

Bacterial strain, namely MTCC 9768 Pseudomonas fluorescens (gram-negative) was used in this study. The isolated bacterial strain was purchased from MTCC, Chandigarh, India.

D. Acclimatization of Culture and Inoculum Development

Bacteria were grown in the nutrient broth solution and then they were separated with the help of centrifuge (at speed of 4500 rpm at 4 °C for 15 minutes). The separated bacterial pellets were then aseptically transferred to a flask containing 1.0 L of nutrient solution and a sterilized magnetic stir bar. The nutrient solution used had the following composition per liter of water: Glucose (2g), Na_2HPO_4 •$2H_2O$ (1.9 g) $KHPO_4$ (0.5g), KH_2PO_4 (1.3g), $NaNO_3$ (6g), $FeSO_4$•$7H_2O$ (0.00136g), $MgSO_4$•$7H_2O$ (0.5g), $MnSO_4$•$7H_2O$ (0.00101g), $CaCl_2$ (0.055g), $ZnSO_4$•$7H_2O$ (0.00058 g), $CuSO_4$•$5H_2O$ (0.0002g) and $CoCl_2$•$6H_2O$ (0.00024 g).

The flask was sealed with cotton for oxygen supply. Then the bacterial culture was stirred at low speed till an appreciable amount of growth was observed. For biofilter inoculation, 250 mL of the liquid culture was transferred into each of four 1.0 L beakers, and then 750 ml of autoclaved distilled water was added to make the total volume 1 L in each beaker. The corn-cob packing material for each section was submerged in the liquid culture in separate beakers and incubated for two days at 28 °C before transferring into biofilter column. Excess liquid was drained by gravity. All media and materials used for culturing were autoclaved at 15 psi for 30 minutes and aseptic techniques were used during bacterial culture transfers.

E. Analysis of Gas Samples

Gas-phase ethylbenzene and m-xylene concentrations were measured using a Thermo Scientific™ TRACE™ 1300 series gas chromatograph (GC), equipped with a capillary column (30 m × 0.25 mm × 0.25 μm, Thermo TG 5 MS) and a flame ionization detector (FID). Gas samples were collected from the biofilter sampling ports using glass gas-tight syringes (Thermo scientific, Bellefonte PA) of 500 μL capacity were introduced into the GC directly. The GC was operated at split ratio of 33.3:1 and carrier flow was 1 mL/min. The nitrogen carrier gas flow rate was 40 mL/min, air flow rate was 350 mL/min, and hydrogen flow rate was 35 mL/min. The injector and detector temperatures were set at 200 °C and 220°C, respectively and the oven temperature was 50°C for 1.0 min, followed by an increase at a rate of 10 °C per min to 150°C with a hold time for 1 min. Retention times for ethylbenzene and m-xylene were approximately 4.34 and 7.11 min, respectively. The calibration curve was prepared by injecting a known mixture of ethylbenzene and m-xylene into the GC from the tedlar bag.

F. Performance Evaluation of Biofilter

The performance of the biofilter was evaluated in terms of the removal efficiency (RE) and the elimination capacity (EC) of the filter bed, at various loading rate and were estimated using the following equations:

(1) Inlet loading rate, IL (g m^{-3} h^{-1}) = Q×C1/V

(2) Elimination capacity, EC (g m^{-3} h^{-1}) = Q (C1 – Co)/V

(3) Removal efficiency, RE (%) = (1-Co/C1) × 100

Where C1 and Co is the concentration of ethylbenzene and m-xylene at inlet and outlet, Q is volumetric gas flow rate (m^3 h^{-1}) and V is filter bed volume (m^3).

III. RESULT AND DISCUSSION

A. Biofilter Performance

The biofiltration of air stream containing ethylbenzene and m-xylene was carried out over 60 days at various operating conditions in a down flow mode corn-cob based biofilter. Reactor had been operated in two phases. The concentrations of ethylbenzene and m-xylene and flow rate were varied to maintain desired inlet loading rate during the study. Inlet concentration and removal efficiency for a period of 60 days are presented in Figure 2. Phase I lasted from days 1 to 30. Flow rate of main stream were maintained at 4 L min^{-1} and the average concentration of ethylbenzene and m-xylene were maintained at 3.25 g m^{-3} and 3.64 g m^{-3}, respectively, so that average loading rate of 188.62 g m^{-3} h^{-1} and 219.30 g m^{-3} h^{-1} for ethylbenzene and m-xylene can be applied for the biofilter. Corresponding EBRT(Empty Bed Residence Time) was 59 seconds. Gradual increase in removal efficiency from 46% to 65% for m-xylene and 43% to55% for ethylbenzene was observed after 30 days of operation. Elimination capacity at the end of phase I was 94.34 g m^{-3} $h^{-1}$for ethylbenzene and 148.03 g m^{-3} h^{-1} for m-xylene.

In phase II, the average loading rate was maintained at rate of 291.96 g m^{-3} h^{-1} for ethylbenzene and 304.10 g $m^{-3}h^{-1}$ whereas the average concentration of ethylbenzene and m-xylene were maintained at 5g m^{-3} and 5.2 g m^{-3}, respectively. In this phase, main stream flow rate and EBRT was kept constant at 4 L min^{-1} and 59 seconds. With increase in loading rate to the biofilter, removal efficiency of ethylbenzene and m-xylene increased upto 72% and 76%, respectively and after that decreased till the end of phase II due to the temperature drop of the biofilter bed causing reduction in microorganism population. The removal efficiencies observed were less than the removal efficiencies reported by some researchers (Cho *et al*., 2009; Rahul *et al.,* 2013; Mathur *et al.,* 2007; Tahraoui and Rho, 1998; Oh and Choi, 2002). The reason for low removal efficiency could be relatively higher inlet loading rate applied and temperature drop in the present study.The elimination capacity in this phase increased upto 282.03 g m^{-3} h^{-1} and 298.84 g m^{-3} h^{-1} for ethylbenzene and m-xylene respectively. The maximum elimination capacity observed in present study was more than the earlier studies (Oh and Bartha, 1997; Oh and Choi, 2002; Shim et al., 2002).These performances are mainly credited to appropriate physical and biological properties of the biofilter media, adequate control of operating parameters such as the moisture content of the bed, temperature of the biofilter bed, the nutrient solution addition to the biofilter media and the affinity of the microorganism population grown in the biofilter to the target pollutants.

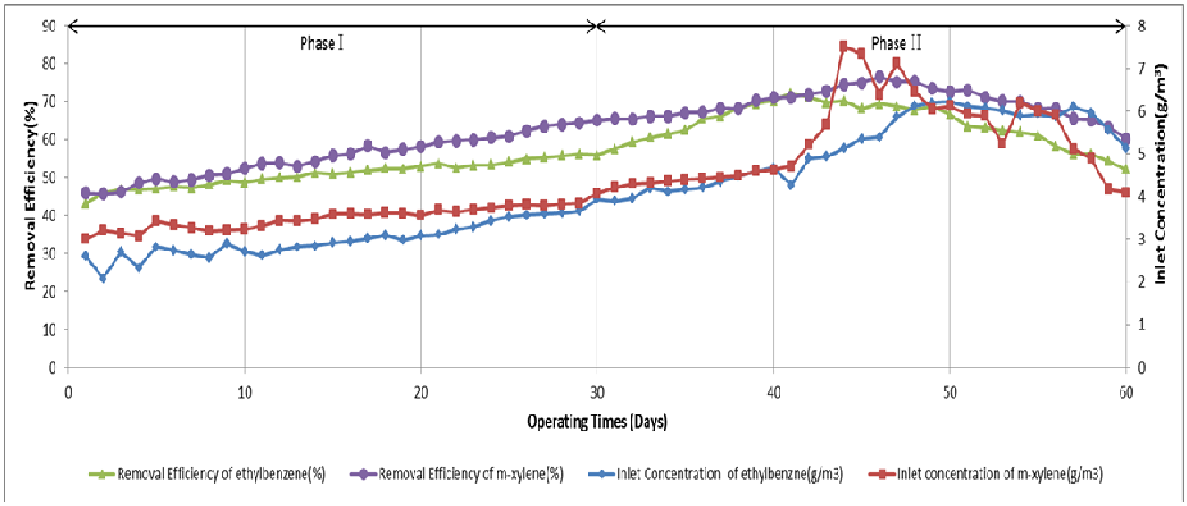

Fig. 2. Overall Performance of Corn-cob Based Biofilter in the Removal of Ethylbenzene and m-Xylene with Time.

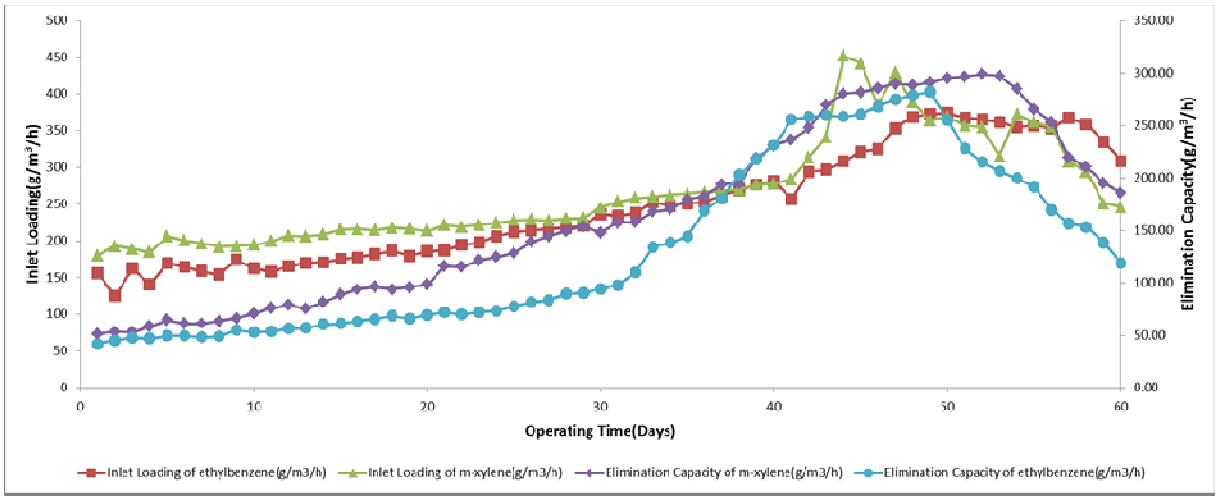

Fig. 3. Elimination Capacity of the Biofilter over a Period of 60 Days.

B. Microbial Population

Microorganism population count on corn-cob media and in the leachate was done on weekly basis by plate counting method. Based on results it was observed that as the time elapsed there was an increase in microorganism population on corn-cob media and in leachate. The increase in microbial concentration on corn-cob media was high as compared to increase in microbial concentration in leachate. Initially the microbial concentration was 73×10^8 CFU per gram of packing material, which increased to 110×10^8 CFU per gram on the 22nd day of operation. After that the microbial concentration declined to 85×10^8 CFU per gram on 46th day and after that it continued to decline to 56×10^8 CFU per gram on 56th day due to the toxicity produced by higher inlet concentration, temperature drop and pH reduction in the biofilter. In leachate maximum microbial concentration was recorded as 71×10^7CFU per ml of leachate on 22nd day of biofilter operation. Microbial concentration declined to 60×10^7 CFU per gram on 42nd day and after that it continued to decline to 45×10^7 CFU per gram on 56th day due to the toxicity produced by higher inlet concentration and pH reduction in the biofilter.

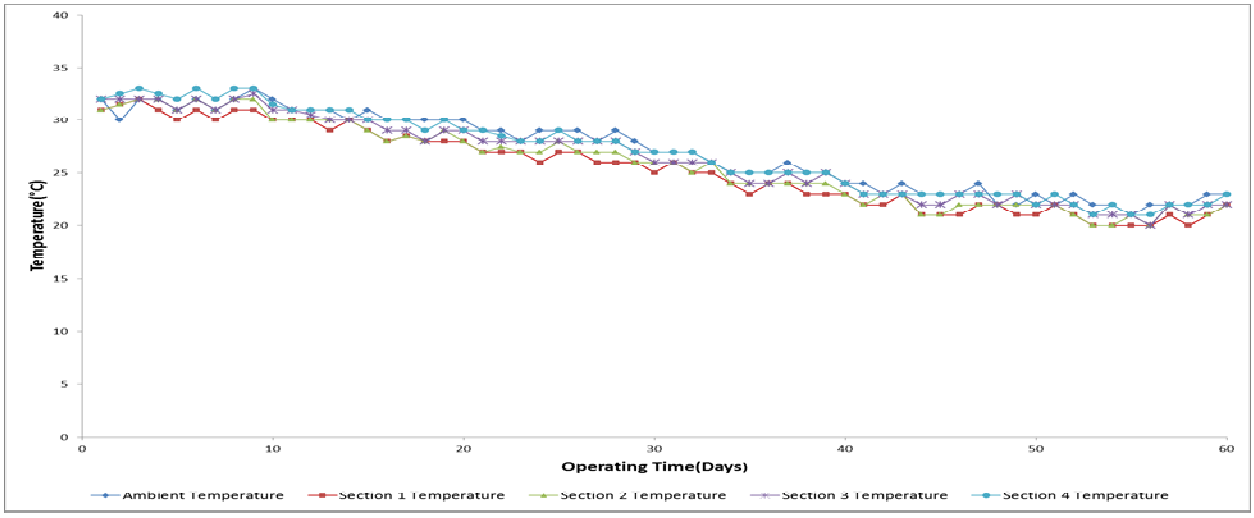

Fig.4. Temperature Variations at Various Levels of Biofilter.

IV. CONCLUSION

In present study, biodegradation of high concentration ethylbenzene and m-xylene laden air stream was successfully achieved with a bacterial culture Pseudomonas fluorescens (MTCC 9768) immobilized in the corn-cob based biofilter. The maximum removal of ethylbenzene and m-xylene were 72% and 76% at an average inlet concentration of 5 g m^{-3} and 5.2 g m^{-3}, respectively and the maximum elimination capacity of the biofilter were 282.03 g $m^{-3}h^{-1}$ and 298.84 g $m^{-3}h^{-1}$for ethylbenzene and m-xylene at an inlet load of 368.52 g $m^{-3}h^{-1}$ and 354.67 g $m^{-3}h^{-1}$, respectively with an EBRT of 59 seconds in both phases.

REFERENCES

[1]. Cho, E., Galera, M.M., Lorenzana, A. and Chung, W.J., (2009). Ethylbenzene, o-xylene, and BTEX removal by Sphingomonas sp. D3K1 in rock wool-compost biofilters. *Environmental Engineering Science*, **26**(1), pp.45-52.

[2]. Choi, S.C. and Oh, Y.S., (2002). Simultaneous removal of benzene, toluene and xylenes mixture by a constructed microbial consortium during biofiltration. *Biotechnology letters*, **24**(15), pp.1269-1275.

[3]. Delhoménie, M.C. and Heitz, M., (2005). Biofiltration of air: a review. *Critical reviews in biotechnology*, **25**(1-2), pp.53-72.

[4]. Jeong, E., Hirai, M. and Shoda, M., (2006). Removal of p-xylene with Pseudomonas sp. NBM21 in biofilter. *Journal of bioscience and bioengineering*, **102**(4), pp.281-287.

[5]. Jeong, E., Hirai, M. and Shoda, M., (2008). Removal of o-xylene using biofilter inoculated with Rhodococcus sp. BTO62. *Journal of hazardous materials*, **152**(1), pp.140-147.

[6]. Jorio, H., Kiared, K., Brzezinski, R., Leroux, A., Viel, G. and Heitz, M., (1998). Treatment of air polluted with high concentrations of toluene and xylene in a pilot-scale biofilter. *Journal of Chemical Technology and Biotechnology*, **73**(3), pp.183-196.

[7]. Lu, C., Lin, M.R. and Chu, C., (1999). Temperature effects of trickle-bed biofilter for treating BTEX vapors. *Journal of Environmental Engineering*, **125**(8), pp.775-779.

[8]. Lu, C., Lin, M.R. and Chu, C., (2002). Effects of pH, moisture, and flow pattern on trickle-bed air biofilter performance for BTEX removal. *Advances in Environmental Research*, **6**(2), pp.99-106.

[9]. Mallakin, A. and Ward, O.P., (1996). Degradation of BTEX compounds in liquid media and in peat biofilters. *Journal of Industrial Microbiology*, **16**(5), pp.309-318.

[10]. Marek, J., Paca, J., Halecký, M., Koutský, B., Sobotka, M. and Keshavarz, T., (2001). Effect of pH and loading manner on the start-up period of peat biofilter degrading xylene and toluene mixture. *Folia microbiologica*, **46**(3), pp.205-209.

[11]. Mathur, A.K. and Balomajumder, C., (2013). Biological treatment and modeling aspect of BTEX abatement process in a biofilter. *Bioresource technology*, **142,** pp.9-17.

[12]. Mathur, A.K. and Balomajumder, C., (2013). Performance evaluation and model analysis of BTEX contaminated air in corn-cob biofilter system. *Bioresource technology*, **133**, pp.166-174.

[13]. Mathur, A.K. and Majumder, C.B., (2008). Biofiltration of Pyridine by Shewanellaputrefaciens in a Corn□Cob Packed Biotrickling Filter. *CLEAN–Soil, Air, Water*, **36**(2), pp.180-186.

[14]. Mathur, A.K., Majumder, C.B. and Chatterjee, S., (2007). Combined removal of BTEX in air stream by using mixture of sugar cane bagasse, compost and GAC as biofilter media. *Journal of Hazardous Materials*, **148**(1), pp.64-74.

[15]. Mathur, A.K., Sundaramurthy, J. and Balomajumder, C., (2006). Kinetics of the removal of mono-chlorobenzene vapour from waste gases using a trickle bed air biofilter. *Journal of Hazardous Materials*, **137**(3), pp.1560-1568.

[16]. Oh, Y.S. and Bartha, R., (1997). Construction of a bacterial consortium for the biofiltration of benzene, toluene and xylene emissions. *World Journal of Microbiology and Biotechnology*, **13**(6), pp.627-632.

[17]. Otenio, M.H., Silva, M.T.L.D., Marques, M.L.O., Roseiro, J.C. and Bidoia, E.D., (2005). Benzene, toluene and xylene biodegradation by Pseudomonas putida CCMI 852. *Brazilian Journal of Microbiology*, **36**(3), pp.258-261.

[18]. Sene, L., Converti, A., Felipe, M.G.A. and Zilli, M., (2002). Sugarcane bagasse as alternative packing material for biofiltration of benzene polluted gaseous streams: a preliminary study. *Bioresource technology*, **83**(2), pp.153-157.

[19]. Shim, H., Shin, E. and Yang, S.T., (2002). A continuous fibrous-bed bioreactor for BTEX biodegradation by a co-culture of Pseudomonas putida and Pseudomonas fluorescens. *Advances in Environmental Research*, **7**(1), pp.203-216.

[20]. Tahraoui, K. and Rho, D., (1998). Biodegradation of BTX vapors in a compost medium biofilter. *Compost Science & Utilization*, **6**(2), pp.13-21.

[21]. Thakur, P.K. and Balomajumder, C., (2012). Biodegradation of O-Xylene by Azotobacter chroococcum. *International Journal of Advanced Biotechnology and Research.*, **1**(3), pp.502-508.

[22]. Van Agteren, M.H., Keuning, S. and Oosterhaven, J., (2013). *Handbook on biodegradation and biological treatment of hazardous organic compounds* (Vol. 2). Springer Science & Business Media.

[23]. Wu, G., Dupuy, A., Leroux, A., Brzezinski, R. and Heitz, M., (1999). Peat-based toluene biofiltration: a new approach to the control of nutrients and pH.*Environmental technology*, **20**(4), pp.367-376.

[24]. Yadav, J.S. and Reddy, C.A., (1993). Degradation of benzene, toluene, ethylbenzene, and xylenes (BTEX) by the lignin-degrading basidiomycete Phanerochaetechrysosporium. *Applied and environmental microbiology*, **59**(3), pp.756-762.

[25]. Young-Sook, O., (2000). Selection of suitable packing material for biofiltration of toluene, m-and p-xylene vapors. *The Journal of Microbiology*.