

Original Article

Removal of styrene from waste gas stream using a biofilter

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ABSTRACT

Background: Styrene is produced in large quantities in the chemical industries and it has been listed among the 189 hazardous and toxic atmospheric contaminants under Clean Air Act Amendments, 1990, due to its adverse effects on human health. The biofiltration has been widely and efficiently applied during recent decades for the treatment of air streams contaminated by volatile organic compounds at low concentrations. Also this technology has been applied widely and efficiently in the removal of styrene from waste gas streams.

Methods: Biofiltration of waste gas stream polluted by styrene vapor was investigated in a three-stage bench scale reactor. Yard waste compost using shredded hard plastics as a bulking agent in a 75:25 v/v mix of plastics:compost was used to packing biofilter. The system inoculation was achieved by adding thickened activated sludge obtained from municipal wastewater treatment plant and the effects of loading rate, inlet concentration, and empty bed retention time variations on the performance and operation of biofilter were studied.

Results: Microbial acclimation to styrene was achieved with inlet concentration of 65 ± 11 ppm and bed contact time of 360 s after 57 days of operation. Under steady state conditions experimental results showed equal average removal efficiency of about 84% at loading rates of 60 and 80 $\text{g m}^{-3} \text{h}^{-1}$ with empty bed retention time of 60 s. Maximum elimination capacity was obtained up to 81 $\text{g m}^{-3} \text{h}^{-1}$ with organic loading rate of about 120 $\text{g m}^{-3} \text{h}^{-1}$. Reduction in performance was observed at inlet concentrations of upper than 650 ppm related to organic loading rates up to 160 $\text{g m}^{-3} \text{h}^{-1}$ and then removal efficiency was decreased sharply. Evaluation of the concentration profile along the bed height of column indicated that the most value of elimination capacity occurred in the first section of biofilter. Elimination capacity also showed higher performance when empty bed retention time was reduced to 30 s.

Conclusion: Microbial acclimation period for biofiltration of styrene inoculated with activated sludge was longer than inoculation with adapted or enriched bacteria. Addition of compost improved performance of the biofilter because of high water retention capacity. Also high pollutant load with significant concentration to the first sections has a clear inhibitive effect on biofilter performance along the bed height of biofilter.

Key Words: biofilter, styrene, Volatile Organic Compounds (VOCs), waste gas, compost.

Styrene is produced in large quantities in the chemical industries and its monomer constitutes the building block for the production of polystyrene, styrene copolymers, polyester resins, and rubber. It is also used in the production of fiberglass boats, storage tanks, pipes, shower units and car parts. Besides the known industrial releases from the production and processing units, styrene is also generated in smaller quantities from other sources such as natural microbial and fungal metabolism, cigarette smoke,

automobile exhaust, and the pyrolysis and cracking of petroleum and its derivatives¹⁻³. Statistically significant differences exist in styrene emissions between the plants, for example the maximum emission of 122 ppm (522 mg m^{-3}) has been measured in manufacturing fiberglass products¹. In general, the concentration of styrene in industrial waste gases is up to 1 g m^{-3} , with an average range of 150 - 400 mg m^{-3} ⁴. Styrene has been listed among the 189 hazardous and toxic atmospheric contaminants under Clean Air Act Amendments,

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1990, due to its adverse effects on human health, which threshold limit value (TLV) vs. Time Weighted Average (TWA) was set to 0.085 g m^{-3} (50 ppm). Acute exposure to styrene in humans results in mucous membrane and eye irritations as well as gastrointestinal disorders. Chronic exposure to styrene in humans results in a variety of discomforts such as headache, fatigue, weakness, depression, central nervous system dysfunction, hearing loss, and peripheral neuropathy⁵.

Consequently, in the industries involved in generating potential products, the development of viability and effective emissions control measures have been got a necessity. Among the various emerging air pollution control technologies, biotechnological processes are effective, reliable, simple, and cheap option, for treatment of volatile organic compounds (VOCs) emissions. Biological treatment of VOCs in biofilters offers an inexpensive alternative to conventional technologies such as catalytic and thermal oxidizers, wet scrubbing, ozonation, and activated carbon adsorption⁶.

The biofiltration has been widely and efficiently applied during recent decades for the treatment of air streams contaminated by VOCs at low concentrations¹. Jorio et al (2000) reported that the efficiency of biofilter is dependent on operational parameters such as bed media characteristic, contaminant inlet concentrations and, gas flow rate or detention time into the biofilter. He reported that at styrene inlet concentrations of upper than 2 g m^{-3} , elimination capacity (EC) decreases with increasing gas flow rate. However at low inlet concentrations ($< 1 \text{ g m}^{-3}$), the EC is increased as function of gas flow rate². Also Juneson et al (2001) reported that average EC of about $69 - 118 \text{ g m}^{-3} \text{ h}^{-1}$ has been observed in two months of operation period without any back pressure problems⁶. Arnold et al (1997) studied removal of styrene in a peat based biofilter and maximum elimination capacity and removal efficiency (RE) were $30 \text{ g m}^{-3} \text{ h}^{-1}$ and 98%, respectively⁷. At all, biofilters have been shown to be more effective for treating some poorly-water-soluble compounds than the other biological systems such as bioscrubbers or biotrickling filters because of that the high superficial area available for mass transfer and the highest removal efficiency of aromatic hydrocarbons air contaminants have been

achieved using biofilters⁸. Also, provided comparable performance can be obtained with similarly sized biofilters and biotrickling filters, while the simplicity and the easy operation of biofilters will favor their selection for the treatment of waste gases⁹.

Many studies have been performed on bacterial degradation of styrene mainly on *Pseudomonas* spp. and other reported styrene-degrading bacterial isolates for example, belong to such genera as *Alcaligenes*, *Corynebacterium*, *Mycobacterium*, *Rhodococcus* and *Xanthobacter*^{10, 12-17}. In the fungi, group only black yeast *Exophiala jeanselmei* and some white-rot fungi are reported to degrade styrene¹⁸⁻¹⁹. In the previous works natural residues like peat, perlite and especial preconditioned biomass pellets as the filter material and most of them have been inoculated with a specific activated consortium of microbial species^{1, 2, 7, 9, 20}. While compost-based media has been used extremely in recent years in comparison with the other filters, bed media is cheaper and has a large existing microbial population capability of degrading various pollutants^{21, 22}, but it has never been used as a packing material in the biofiltration of styrene.

Styrene-degrading bacteria enriched from activated sludge has been applied for inoculation of biofilters^{6,7,23}. Also activated sludge from petrochemical plant has been used as the inoculum of biotrickling filter and glucose has been added as a supplemental carbon source and enhancing the adhesion of microbial films to the media surface⁴. However, obviously process of enrichment to inoculate biofilter for faster achievement to the high performance has additional cost, especially in the full scales.

In this work, activated sludge from municipal wastewater treatment plant was used as styrene degrading consortiums to inoculate biofilter without any enrichment processes. In the present study biofiltration of styrene was studied through a biofilter packed with yard waste compost that mixed with activated sludge and shredded hard plastics as a bulking agent. The main objectives of this study were: (a) To estimate acclimation period of biofilter cultivated by activated sludge without any enrichment process and to demonstrate applicability and effectiveness of activated sludge microorganisms in degradation of styrene; (b) evaluation of

operational parameters effect such as gas retention time, styrene inlet concentration and organic loading rate on the performance of the system; and (c) application of experimental results in designing and operating of styrene treating biofilters in industries as well as petrochemical industries.

MATERIALS AND METHODS

Experimental set-up

A three-stages down flow bench-scale biofilter constructed from galvanized iron was used in this

study (figure 1). The column had an inner diameter of 8 cm and the effective height of bed media was 120 cm. Perforated steel plate plenums (pore diameter = 2 mm) placed between sections acted as a support for the packing material as well as for gas flow redistribution. A 7-cm space between the sections allowed for representative gas sampling. Provision of sampling ports at top, midpoint, and at the end of each section allowed bed media access. The synthetic waste air stream was prepared by sending compressed air through a granular activated carbon canister to adsorb residual oil and remove particles.

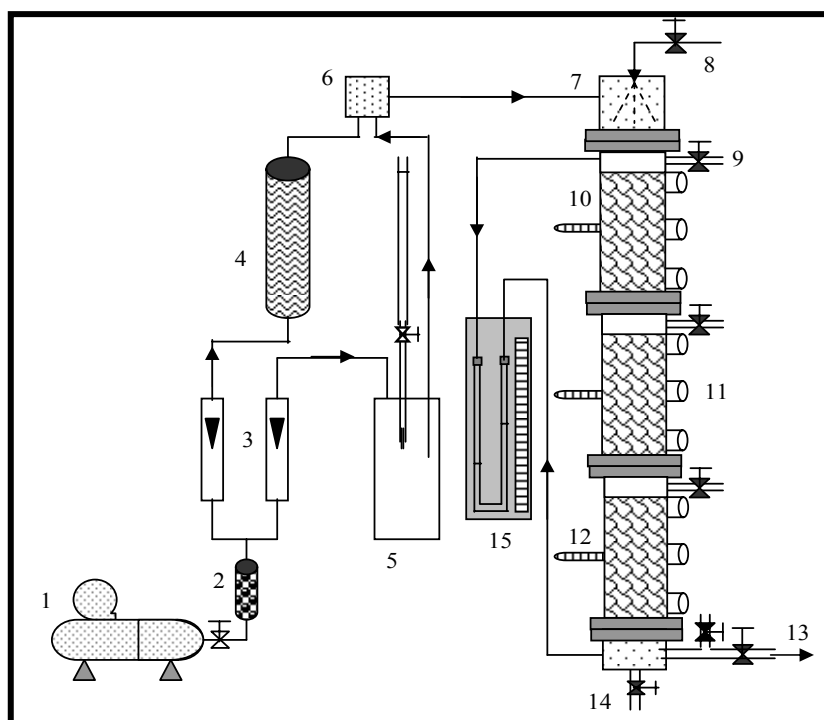


Figure 1. Schematics of biofilter system (1-compressor, 2-carbon filter, 3- rotameter, 4- humidifier, 5-pollutant vessel with styrene injector, 6-mixing chamber, 7-inlet, 8-nutrient, 9-gas sampling port, 10- biofilter bed, 11-bed sampling port, 12- thermometer, 13-outlet, 14-lechate, 15-manometer).

The air stream was then sparged through a 15-liter water container equipped with heated element for adjusting gas stream temperature and humidification. Pollutant vapor was prepared by introducing low flow air stream into a container receiving drop-wise styrene feed from a burette. After humidification, the main air stream was mixed with the stream containing pollutant vapor to generate feed air with the needed concentration. Changing water temperature in the

humidifier controlled variation of humidity in the influent gas stream and biofilter material. Temperature control of the bed material was achieved by using a heated tape wrapped around the exterior of reactor wall.

Yard waste compost that is used as nutrition for home plants was mixed with shredded high-density plastics (1.5 cm) as bulking agent to produce a 25:75 v/v ratio of compost-bulk agent. In preparing the

packing medium, thickened activated sludge obtained from municipal wastewater treatment plant (Tehran Water & Wastewater Co.), was added to this mixture to increase microbial density and develop compost particles attach on bulking agents. Nutrient and buffering solution was used according to the quantity of carbon inlet to the biofilter to keep the C: N: P ratio around 100:5:1 and adjusting pH in the medium, had the following composition (per liter of tap water): 0.694 g KH_2PO_4 , 0.854 g K_2HPO_4 , 1.234 g $(\text{NH}_4)_2\text{SO}_4$, 0.46 g $\text{MgSO}_4 \cdot \text{H}_2\text{O}$, 0.176 g $\text{CaCl}_2 \cdot 2\text{H}_2\text{O}$, 0.001 g $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$, and 5 ml trace element solution consist of 60 mg l^{-1} H_3BO_3 , 40 mg l^{-1} $\text{CoCl}_2 \cdot 6\text{H}_2\text{O}$, 20 mg l^{-1} $\text{ZnSO}_4 \cdot 7\text{H}_2\text{O}$, 6 mg l^{-1} $\text{MnCl}_2 \cdot 4\text{H}_2\text{O}$, 6 mg l^{-1} $\text{NaMoO}_4 \cdot 2\text{H}_2\text{O}$, 4 mg l^{-1} $\text{NiCl}_2 \cdot 6\text{H}_2\text{O}$, 2 mg l^{-1} $\text{CuCl}_2 \cdot 2\text{H}_2\text{O}$ with an overall pH around 6.9 ± 2^4 .

Experimental schedule

Evaluation of biofilter performance and removal efficiency of system in treatment of styrene was performed for more than a 12-months period from April 2002 until May 2003 by varying airflow rate and styrene inlet concentrations. The biofilter set up was placed at Sharif University of Technology at the Water and Energy Research Center. In the start-up of biofilter to achieve short acclimation period the study was not operated in strong conditions. At first step system just was started with the mixture of bulk agent and thickened activated sludge but because of bed drying in the first section of biofilter, compost was mixed with the bed material in the 45th day of operation. After about 70 days of start-up and approach to steady-state condition the superficial gas velocity increased up to 168.75 m h^{-1} with empty bed

retention time (EBRT) of 120 s whereas styrene inlet concentration was kept constant up to 70 ppm for about 40 days. In the following days the efficiency of system was examined at the different inlet concentrations of about 250, 350, 500, and 650 ppm and finally reduced to 150, 60, and 30 ppm. Also system was operated in EBRTs of 60 and 30 s. In each of these experimental sets the operation of biofilter was continued for about one month to have a stable performance and collect more reliable data.

Analytical method

In order to determine the styrene concentration, gaseous samples were collected in 5-liter Tedlar bags by connecting the bag port onto the tube connected to the biofilter gas sampling ports. The Tedlar bags were flushed with activated carbon filtered air prior to sampling to clean previous samples contamination. Analysis was done within 3 h using a gas chromatograph (SRI 110 Inc.) equipped with a flame ionization detector and with a 30 m capillary column (GM 6210). Operating conditions were as follows: temperature schedule (injector 200 °C, oven 220 °C, detector 230 °C) and N_2 as carrier gas at 8 ml min^{-1} . Gas samples of 1000 μl containing styrene was withdrawn from Tedlar bags using 2000 μl gas-tight syringe (Series A-2, VICI, Inc.) and were injected immediately into the GC unit for concentration determination. Unknown samples were determined from a calibration curve that it is prepared from peaks observed for known concentration of styrene by introducing known volumes of styrene into the 5 L sealed Tedlar bag. For example to prepare a 5 L

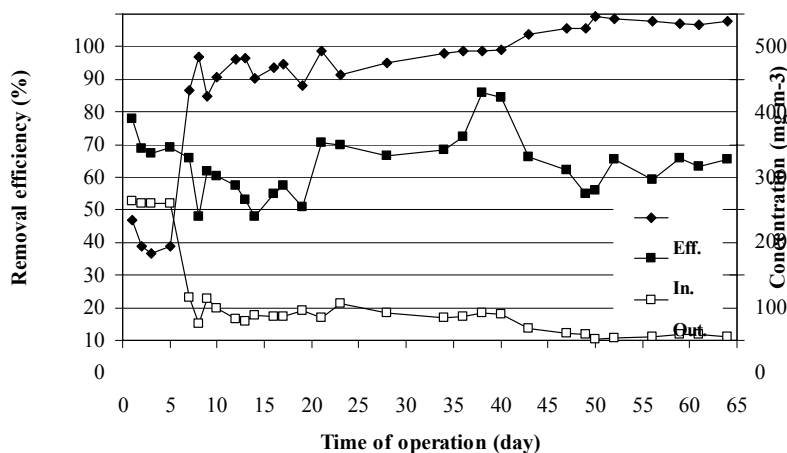


Figure 2. Performance of biofilter at the start-up period

Tedlar bag containing 100 ppm styrene at 25°C (room temperature), styrene should be spiked into the bags in the volume of 2.3 μl . To control the accuracy and preciseness of the examination, the reported inlet and outlet concentration represent an average of two sequential samples.

RESULTS

Gradual increase of removal efficiency of biofilter shows that microorganisms are acclimated to styrene. After about 2 months the steady state condition was obtained in biofilter column and no much fluctuation was occurred in the biofilter removal rate. Biofiltration of styrene inoculated with a microbial consortium has shown that at startup period with C_{in} of 500 mg m^{-3} and EBRT of 64 s the RE attained 40% at day 5 and by day 7 had increased to more than 97%². Also one of the

At the start up period system was operated with nearly constant empty bed retention time (EBRT) of 360 s and inlet concentration (C_{in}) of about 68 ± 11 ppm, which it is related to superficial gas flow rate of 56.25 m h^{-1} . Removal efficiency (RE) of the system reached up to 80% after about 6 days and finally it fixed up to 90% until the day of 44 (Figure 2).

biofiltration guys observed maximum styrene degradation rate of 80% after 12 days in a peat based biofilter which inoculated with enriched bacteria obtained from activated sludge of a petrochemical plant.

Analysis of styrene concentration profile along the biofilter column showed lower performance for the first section until about 44 days from the startup (Figure 3).

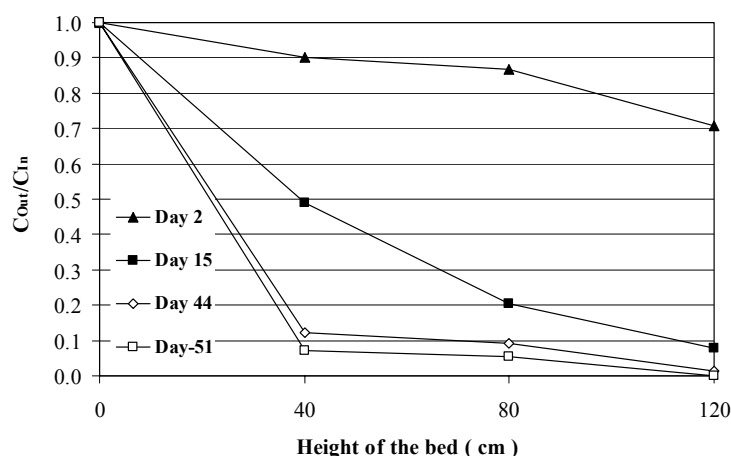


Figure 3. Styrene concentration profile along the bed height of biofilter at the start-up period

First section has not reached to its optimal performance comparison to second section. For example on day 41, there was 20% removal in the first section while more than 60% was removed in the next one. The difference in performance was believed to attribute to low moisture content since other conditions were as same as. Determination of water content confirmed the hypothesis as it was observed that a decrease in moisture down to less than 35% was encountered. This was so due to low water retention capability of plastic media used as bulking agent. Previous studies have confirmed that if the water content goes to less than 50%, removal efficiency can suffer by up to 50%²⁰. To improve

the biofilter performance, yard waste compost was added in a %25 v/v of the bed resulted gradually increase in RE up to 100% on days 45 and 51.

Also, performance of the system was studied as a function of EBRT in the range of 30 to 120 s and the results are shown in Figure 4. After the start-up period system was operated with EBRT of 120 s and styrene inlet concentration was held around 248.6 ± 16.4 ppm. RE of $87.4 \pm 4.4\%$ and EC of 26.9 ± 2.7 $\text{g m}^{-3} \text{h}^{-1}$ was obtained up to day 130. Then flow rate was increased to 6 l min^{-1} without any changes in C_{in} so organic loading rate (OLR) changed proportionally and reached to 60 ± 3.2 $\text{g m}^{-3} \text{h}^{-1}$. At the beginning of each step change in C_{in} ,

there was a larger than normal fluctuation in RE with gradual recovery and increasing trend with time. Step changes in C_{In} were from 250 to 320, 500 and 650 ppm at days 167, 197 and 225, respectively. For the first change removal efficiency was decreased but the time to recovery was very short and removal efficiency reached the original level prior to increase in the OLR. However for the third increase in C_{In} , removal efficiency was sharply decreased by about 14% and never recovered for

about 23 days. However for more evaluation, the biofilter was operated with EBRT of 60 s and inlet concentrations of about 146.3 ± 2.7 ppm as same as the styrene real average concentration in the off-gases of industries. In this condition the maximum RE was measured about $96.3 \pm 2.6\%$. Also system was operated at lower EBRT of 30 s and inlet concentrations of about 150, 65 and 30 ppm. Removal efficiency was not changed so much and it was fixed next to 100%.

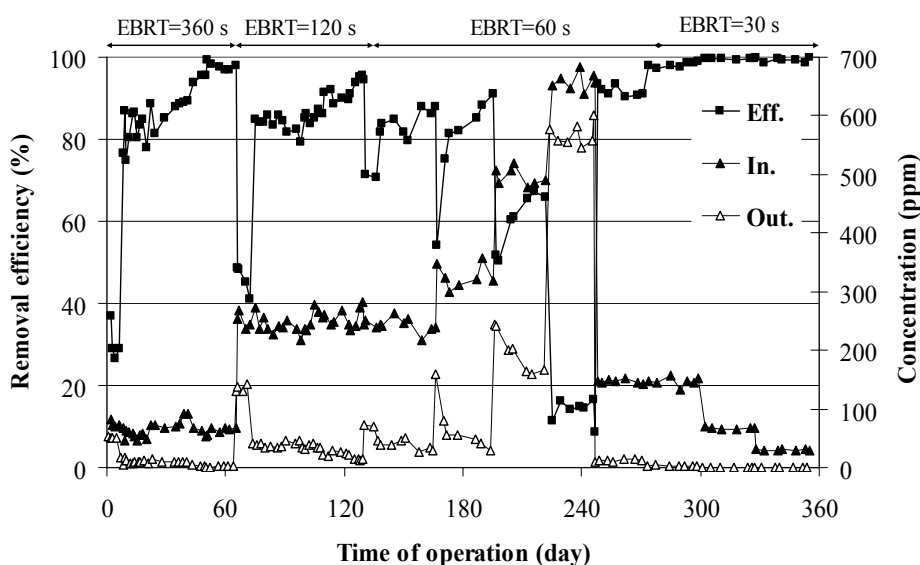


Figure 4. Overall performance of the biofilter at different EBRTs

Besides the nature of pollutants like water solubility and metabolic pathway for their microbial degradation, the input gas flow rate or empty bed residence time and pollutant inlet concentrations are among the most important process parameters influencing the biofiltration performance². Figure 5 shows the concentration profile along column of biofilter at EBRT of 60 and 30 s and different inlet concentrations. First section has showed better performances for concentrations below 150 ppm at the all empty bed retention times. In this work more than 50% of removal efficiency has been occurred in the first section of biofilter. Also it is probably due to the fact that higher concentration gradient results higher mass transfer rates into the biofilm⁶. At EBRT of 60 s and with inlet concentrations of 150 and 250 ppm related to organic loading rates of about 36.2 ± 0.7 and 60 ± 3.2 $\text{g m}^{-3} \text{h}^{-1}$ respectively, there were not any

differences in EC of biofilter. However, further increase in styrene inlet concentration showed an adverse impact on elimination capacity, because high pollutant load with significant concentration to the first sections has a clear inhibitive effect on biomass activity along the bed height of biofilter²⁸. It seems that under steady state conditions the degree of decompositions at inlet concentrations lower than 150 ppm system denoting first order kinetics. In the previous researches has been observed that overall kinetics of styrene removal is the zero-order kinetics with respect to styrene concentration^{20,28}.

As shown in Figure 6, maximum elimination capacity (EC) is about $81 \text{ g m}^{-3} \text{h}^{-1}$ for OLR close to $120 \text{ g m}^{-3} \text{h}^{-1}$. In the other works and at different conditions, maximum EC that have been reported were $50 \text{ g m}^{-3} \text{h}^{-1}$ with a novel biomass filter material, $63 \text{ g m}^{-3} \text{h}^{-1}$ using perlite-packed biofilter

inoculated with styrene degrading fungi, $63 \text{ g m}^{-3} \text{ h}^{-1}$ using compost packed biofilter enriched with different bacteria, and $66 \text{ g m}^{-3} \text{ h}^{-1}$ with a mixture of peat and glass beads as packing medium inoculated with the styrene oxidizing strain *Rhodococcus rhodochrous*^{2, 18, 20, 29, 28}, whereas lower elimination capacity (30 to $50 \text{ g m}^{-3} \text{ h}^{-1}$) were reported for peat biofilter using different inoculum's procedures^{7, 17}.

DISCUSSION

Acclimation period of the biofilter obtained after about 2 months. The results show that biofiltration with compost, soil, or activated sludge requires long acclimation periods ranging from several weeks to months to obtain good elimination capacities⁶. However inoculations of the biofilter media with specific and adapted microorganisms reduce the start-up time to achieve stable elimination capacities^{25, 26}. However achieved acclimation period in this study is compatible with others reported in the literature for styrene⁷.

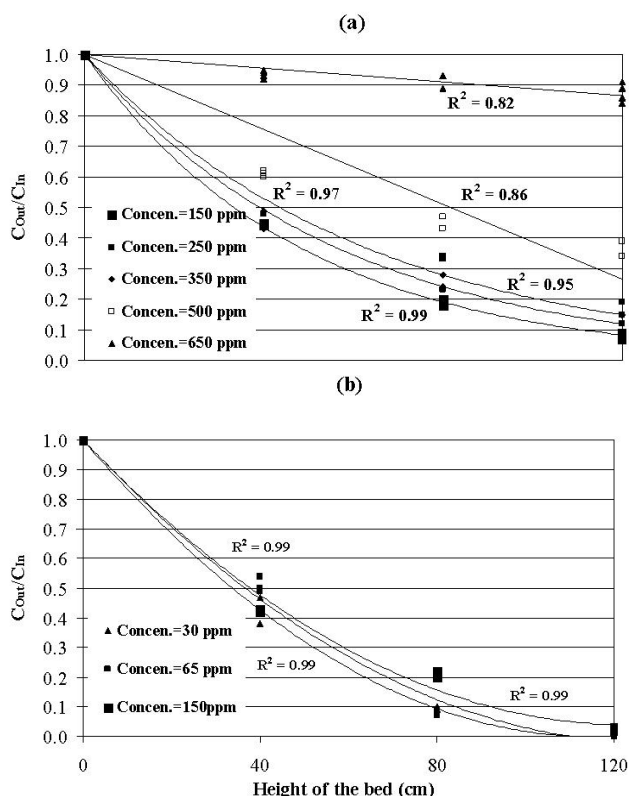


Figure 5. Concentration profile of styrene at steady state condition along the bed height of biofilter at different inlet concentrations; (a) EBRT= 60 s and (b) EBRT=30 s.

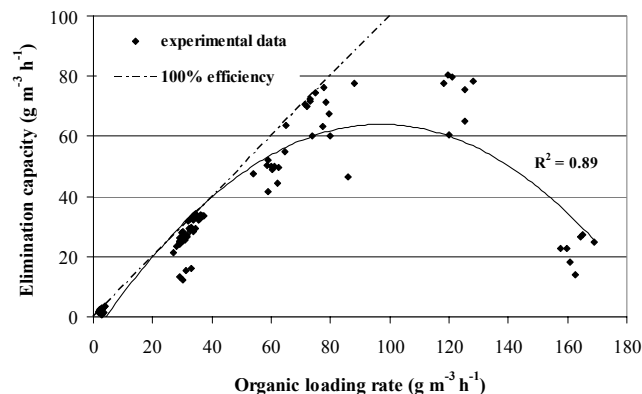


Figure 6. Overall performance of biofilter in the removal of styrene at different organic loading rates.

In the other hand activated sludge microorganisms capable for degrading styrene in the waste gas streams. Addition of compost to the filter bed material improved performance of the biofilter because of high water retention capacity. Inlet concentration of substrate is one of the most important parameter influencing performances of biofilter since at higher concentrations the pollutant transfer rates is increased. However excessive concentration of low biodegradable compounds may have adverse effects on microorganisms and suppress their metabolism functions. In this work styrene was removed completely at loading rates less than $80 \text{ g m}^{-3} \text{ h}^{-1}$ and removal efficiency was close to 100% trend line but at higher loading rates, styrene demonstrated inhibitory effect on biofilter performance and elimination capacity was decreased very drastically. Also Maximum styrene elimination capacity was about $78 \text{ g m}^{-3} \text{ h}^{-1}$ for organic loading rates close to $120 \text{ g m}^{-3} \text{ h}^{-1}$ at empty bed retention time of 60 s.

The processing of the results shows that at steady state condition, most of the overall performance of biofilter has been occurred at first section of biofilter. Also the results of other works have shown that average ratio of styrene removal in the upper half of biotrickling filter to overall removal is equal to 0.76 and indicating that most of biological reactions occur in the earlier sections of the biofilter²⁷. This was due to the fact that more carbon sources, moisture contents, and nutrients were present in the earlier sections of the biofilter, which caused a

higher metabolic reaction, and they led to a faster biodegradation rate of styrene waste gas.

CONCLUSIONS

- Microbial consortiums of activated sludge are capable of degrading styrene as a substrate in waste gas streams. Acclimation period for biofiltration of styrene inoculated with activated sludge was longer than inoculation with adapted or enriched bacteria, but it seems that in this way performance of the system for mixed cultures is more than single strains. So application of activated sludge for start up of biofilters in full scale would be practical and economical instead of using pure or enriched consortium.
- Lower empty bed retention times of about 30 s shows acceptable performance of biofilter in the removal of styrene. Then the results show the significant applicability of biofilters for high flow rates of waste gas streams containing low concentrations of styrene. For example at the inlet

concentrations of lower than 150 ppm system denoting zero order kinetics, also at this inlet concentration, empty bed retention time did not show significant effect on biofilter performance.

- At the inlet concentrations of lower than 150 ppm equal to styrene real average concentration in the off-gases of related industries, styrene outlet concentration (C_{Out}) was not detectable in the samples. It shows the ability of biofilters in approach to the occupational health standards in styrene handling and processing plants.

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