Biofiltration of BTEX contaminated air streams using compost-activated carbon filter media

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Abstract

Three laboratory-scale biofilter columns were operated for 82 days to investigate the removal of benzene, toluene, ethylbenzene, and *o*-xylene (BTEX) from a waste gas stream. The columns contain a mixture of yard waste and sludge compost as the base biofilter material. Different amounts of granular activated carbon (GAC) are mixed with the compost in two of the three columns to evaluate the extent to which biofilter performance can be enhanced. The columns displayed preferential utilization of benzene followed by toluene, ethylbenzene, and *o*-xylene, respectively. Removal efficiencies of $\geq 90\%$ were achieved for inlet concentrations of ≥ 200 ppm of each of the BTEX compounds and a gas loading rate of 17.6 m^3/m^2 h in all columns over the period of study. During sudden increases in the gas flow rate or the BTEX influent concentration, biofilters containing GAC exhibited significantly higher removal efficiencies and more stable operation than the biofilter containing only compost. © 1998 Elsevier Science B.V. All rights reserved.

1. Introduction

The attention of various US air pollution control agencies has been increasingly focused on the control of volatile organic compounds (VOCs) and other air pollutants from stationary sources. Considerable quantities of VOCs are produced from industrial sources such as printing and coating facilities, foundries, electronics and paint manufacturing. In many instances, the off-gas streams are characterized by large flow rates and

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concentrations of less than 1000 ppm. This applies, in particular, where these pollutants are first released into the work place before being discharged to the atmosphere [1]. VOCs also appear as principal pollutants at contaminated and Superfund sites [2].

The literature identifies a number of processes that involve chemical or physical principles for the treatment of air streams with low VOC concentrations [3,4]. Chemical processes such as incineration, catalytic incineration, chlorination and combustion involve relatively complicated procedures, require the addition of chemicals or fuels, and involve considerable expense. Physical processes such as activated carbon adsorption and scrubbing are also costly and may involve regeneration or transportation of hazardous wastes. Most importantly, all the above mentioned processes generate secondary wastes that may require further treatment or disposal, thereby creating additional environmental problems.

Biological processes can also be used for the treatment of VOC polluted air streams. These processes involve the use of either bioscrubbers, trickling filters, or biofilters [5]. A biofilter consists of a filter bed usually comprised of natural material (soil, compost, or peat) that is kept wet to maintain a biologically-active layer surrounding the biofilter material particles, known as the 'biofilm.'

During the 1970s, major improvements in biofilteration technology were made in Europe where it has been successfully applied to control odors and both organic and inorganic air pollutants from a variety of industrial and public sector sources. It is currently a well established air pollution control technology in the Netherlands and Germany [6]. In the US, the first systematic research on biofiltration involved the treatment of odors resulting from hydrogen sulfide gases in sewage works [7]. Studies on the efficiency of removing sewage odors were conducted using a pilot-scale biofiltration unit. Much of the applicable knowledge about the use of biofilters in odor control, and in the treatment of volatile organic compounds (VOCs) in air streams, is owed to Bohn [8] and Bohn and Bohn [9]. He showed that biofiltration in soil and compost beds effectively and inexpensively disposes of air pollutants. Recent efforts include the work of Yang and Allen [10] on biofiltration control of hydrogen sulfide in waste gas stream. They evaluated the effects of several operating parameters and observed removal efficiencies of greater than 99.9% for H_2S inlet concentrations in the range 5 to 2650 ppm. Results from laboratory experiments using compost, granular-activated carbon inoculated with different amounts of active biomass, and a mixture of compost and diatomaceous earth biofilter material, were modelled and evaluated by Hodge and Devinny [11] and Devinny and Hodge [12] for the removal of ethanol vapors. They reported that granular activated carbon provided the best overall treatment, while the compost microbial community had a higher degradation rate constant.

Biofiltration offers a number of advantages for the treatment of low concentration polluted air streams. Besides its high removal efficiency, low capital and operating costs, safe operating conditions, and low energy consumption, it does not generate undesirable byproducts and converts many organic and inorganic compounds into harmless oxidation products (e.g. water and carbon dioxide).

The focus of this research is on a group of volatile organic compounds (VOCs) that is widely used in industry and that poses serious adverse effects on the quality of air. These are the aromatic hydrocarbons benzene, ethylbenzene, toluene, and xylene

(BTEX); the primary components of many petroleum products [13]. BTEX compounds are among the top 50 chemicals manufactured in the US [14]. They are employed in the production of other chemicals such as monomers, plasticizers in polymers, and are also widely used as solvents. Furthermore, gasolines are comprised of a variety of aliphatic and aromatic hydrocarbons, the aromatic portion consisting primarily of BTEX compounds. Benzene, ethylbenzene, and toluene are among the compounds on the Environmental Protection Agency's priority pollutants list [15].

The objective of this study is to examine the application of biofiltration for the treatment of gas streams polluted by BTEX vapors. The study evaluates the use of compost versus a mixture of compost and granular activated carbon as a biofilter media, and the influence of inlet concentration and residence time on the performance of the biofiltration system.

2. Materials and methods

2.1. Biofiltration system

A three column biofilter system was designed and constructed, the configuration of which is shown in Fig. 1. The three columns, identified as A, B and C, can be run simultaneously and controlled separately. The biofilter bed material is enclosed in transparent, rigid, plexiglass tubing, with an inner diameter of 5.1 cm and a height of 90 cm. Each of these columns was packed with the desired compost or compost-activated carbon mixture to a height of 75 cm. The packed biofilter material is supported by a stainless steel sieve plate. In this work, columns A, B, and C are distinguished according to the amount of activated carbon in each, with the composition and physical properties of the columns summarized in Table 1.

Sampling ports are fitted with rubber septa and located along each column for gas sampling and pressure measurements. The individual sampling ports are identified as inlet, upper, lower, and outlet ports. The humidified air stream was prepared by passing pressurized house air through a sparging bottle that contains water, defined as the humidification chamber. The flow rates of the house air and pollutant gas are controlled by valves located at the house air outlets and the inlet lines of the biofilter columns. The desired benzene, or other BTEX compound, inlet concentration and gas flow rate through the columns are controlled by adjusting the air valves at the outlets of the house air. An additional port was installed on the top of each column in order to introduce water to the filter material when required to maintain sufficient moisture content in the bed. All the gas and water lines are 1/4-in. diameter plastic pipes. The columns are sealed from the top and bottom by plastic covers provided with *o*-rings. These covers can be dismantled to replace the filter material and clean the filter columns before and after use. The system was operated at room temperature (22–25°C) throughout all experiments.

2.2. Packing material

The compost used in this study is a mixture of yard waste and sewage sludge marketed under the name 'earth blend leaf humus' from the composting facility of
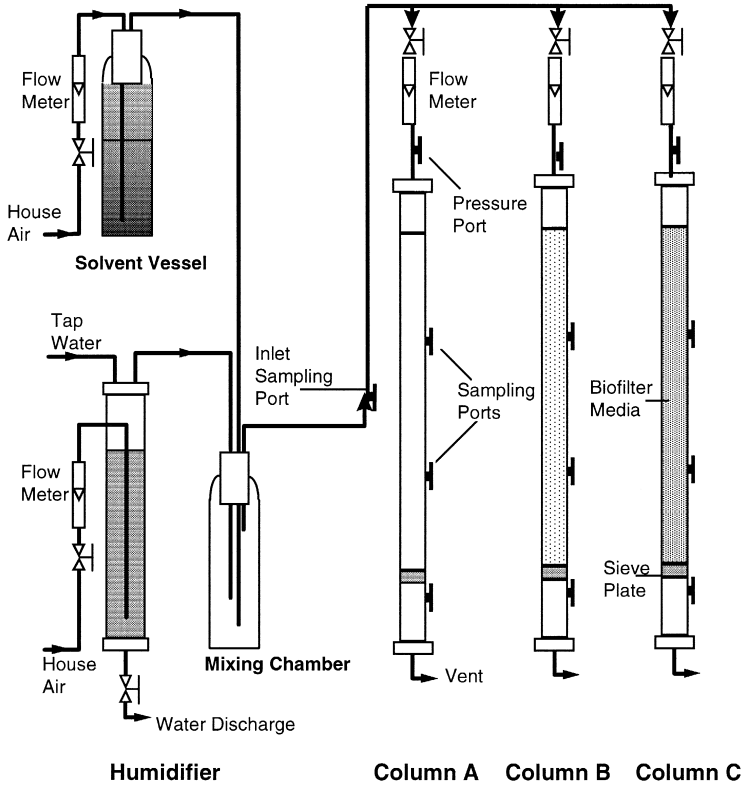
Fig. 1. Schematic of the three column biofilter system.

Table 1
Physical properties of the biofiltration columns

Physical property	Column		
	A	B	C
Wet compost weight, W_1 (g)	1109	1061	1058
Activated carbon weight, W_2 (g)	0.00	23.3	86.4
Dry compost weight, $W_3 = W_1 /(1 + 62.3\%)$ (g)	684	654	652
Percentage of activated carbon (dry weight basis) (%)	0	3.6	13.3
Diameter of column, D (cm)	5.1	5.1	5.1
Height of media in each column, H (cm)	75.0	75.0	75.0
Volume of media (cm^3)	1531	1531	1531
Volume of activated carbon, $V_c = W_2 / D_c$ (cm^3)	0	29.1	108.0
Volume of dry compost, $V_s = W_3 / G_s r_w$ (cm^3)	369	353	352
Bed void fraction (%)	76.5	76.3	72.5

Particle density of activated carbon $D_c = 0.8$ (g/cm^3).
Specific gravity of compost $G_s = 1.9$.

Recycling Systems (Independence, OH). The compost was stored in sealed plastic bags at room temperature before use to maintain its original moisture content. The compost was analyzed for its moisture content (62.3% on dry weight basis), specific gravity (1.9), and organic content (77.8%).

The activated carbon was type 'BPL 4×10' granular activated carbon obtained from Calgon Carbon (Pittsburgh, PA). This particular activated carbon is designed for use in vapor phase applications. It is made from selected grades of bituminous coal combined with suitable binder, and has been used for vapor phase adsorption of alcohols, chlorinated hydrocarbons, esters ketones, ethers hydrocarbons, and aromatics (Calgon Carbon 'Product Bulletin: BPL 4×10 Granular Activated Carbon').

2.3. Measurement methods

Periodic measurements of room temperature, gas concentration from sampling ports, pressure drop in the columns, and gas flow rate of each column in the biofilter system were carried out using the following devices.

2.3.1. Gas concentration

A Perkin Elmer gas chromatograph equipped with a mass spectrometer as a detection device (GC/MS auto system 910) was used for analysis of organic compounds. The GC unit is equipped with a 105 m $\times$ 0.53 mm I.D. with a 2.0 μm D_f of RTX-Volatiles capillary column and uses helium as a carrier gas. Gas samples of 1 ml each containing the target contaminants were obtained directly from the rubber septum sampling ports using 5-ml Hamilton gas-tight syringes and were injected immediately into the GC/MS unit for concentration determination. Each data point, for both inlet and outlet concentrations, represents the average of two sequential samples. Data precision, as given by relative percent difference (RPD) of the two measurements, ranged from 0.3–48.4% with an average RPD of 13.6% for inlet concentrations. For outlet measurements, the average RPD was 23.8%. This is biased, however, by expectedly high RPD values for the numerous measurements at less than 1 ppm (for instance, two consecutive measurements of 0.58 and 0.32 ppm produce a RPD of 58%). If the instances of measured outlet concentrations less than 1 ppm are not included, the average RPD reduces to 16%.

Unknowns were determined from a calibration curve that was developed using known concentration standards of the mixture of BTEX compounds. The standards were prepared by introducing known amounts of the compounds into a 2.8-l sealed glass bottle equipped with a rubber septum according to the methods described by Lodge [16]. Accuracy tests were performed regularly against a 75 ppm standard for all compounds. Percent recoveries were consistently between 95–105%, with recoveries in the range of 90–110% used as the quality assurance criterion.

2.3.2. Temperature

Temperature was measured using alcohol in a glass thermometer with a range from -20 to 110 and a scale division of 1°C.

2.3.3. Gas flow rate

Gas flow rates were measured using 'Key Instruments, Series FR 2000, Model 2A13 and 2A14' flow meters with units of l/min.

2.3.4. Pressure drop

Pressure drop was measured by a water manometer with a minimum division length reading of 1-mm water column (H_2O).

3. Results and discussion

3.1. Long term performance of laboratory biofilters

Biofiltration is an air pollution control technology that utilizes microorganisms present in the biofilter media to degrade the pollutants in a waste gas stream into water and carbon dioxide. The microbial population in compost is relatively inactive prior to contact with substrate, after which time the microorganisms grow in mass and number in the thin layer of water surrounding the support media of the biofilter forming a biofilm. The pollutants diffuse from the gas phase into the biofilm where the microorganisms are immobilized and in which oxidation of the pollutants into harmless byproducts such as carbon dioxide, water, and additional biomass occurs.

Activated carbon is widely used as an adsorbent, especially for organic compounds. Biological systems that employ Biological Activated Carbon (BAC) for the treatment of organic pollutants in water, wastewater, or air have been known to exhibit superior performance [17–19]. The enhanced performance may be manifest in higher removal efficiencies compared to conventional biological systems, shorter acclimation periods of the microorganisms in the system, and lower pollutant concentration in the effluent during step increases in the influent pollutant concentration (shock loading conditions). Simultaneous bioregeneration of the carbon is an additional economic advantage to these systems. Figs. 2–5 illustrate the impact of small additions of granular activated carbon (GAC) on the performance of compost biofilters used to treat a BTEX waste air stream.

The three columns (Fig. 1) were regulated to maintain identical loading conditions of gas flow rate and BTEX inlet concentration, and were operated continuously for 82 days. In Figs. 2–5, the inlet and outlet concentrations of the respective target compounds are plotted against the cumulative operation time. A major objective of this research was to investigate biofilter response to substantive perturbations in influent characteristics. Therefore, the experiment was operated in 3 different phases based on the influent gas flow rate. The first phase began on day 1 and lasted 57 days. The gas flow rate in this phase was 0.6 l/min, yielding an empty bed residence time (EBRT) for the contaminated air steam of 2.6 min in each column. At about the midway point of this initial phase, the influent concentration of all four BTEX compounds was increased four- or fivefold to observe biofilter performance for shock increases at the same gas flow rate. The second phase of the experiment was from day 57–71 using a decreased gas flow rate of 0.3 l/min (EBRT = 5.1 min). Influent concentrations were maintained at a relatively high level for most of this period, but reduced beginning at day 66. The last phase of the experiment commenced on day 71 by increasing the flow rate to 1.3 l/min, thereby reducing the EBRT to 1.2 min. After operating at low influent concentrations (< 50 ppm) from day 71–75, inlet values were increased approximately to 125, 300, 250, and 150 ppm for benzene, toluene, ethylbenzene, and *o*-xylene, respectively. The
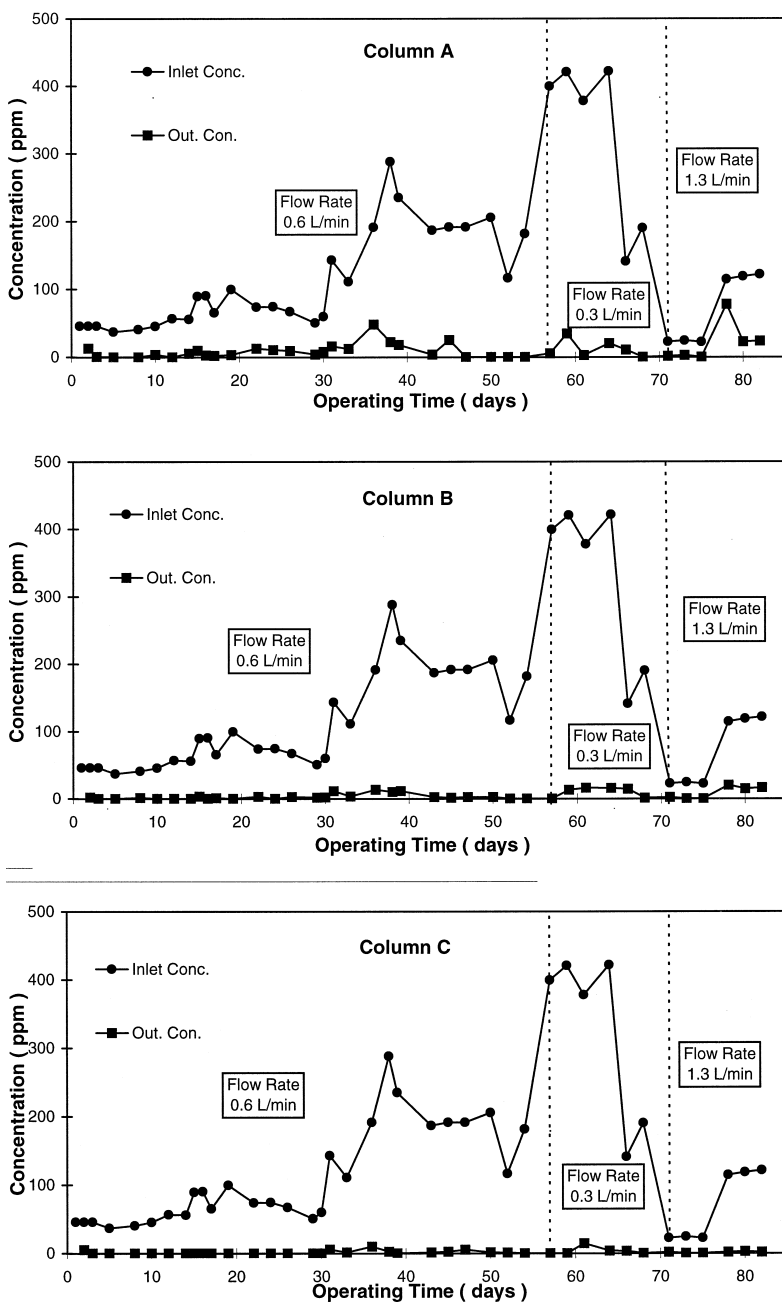
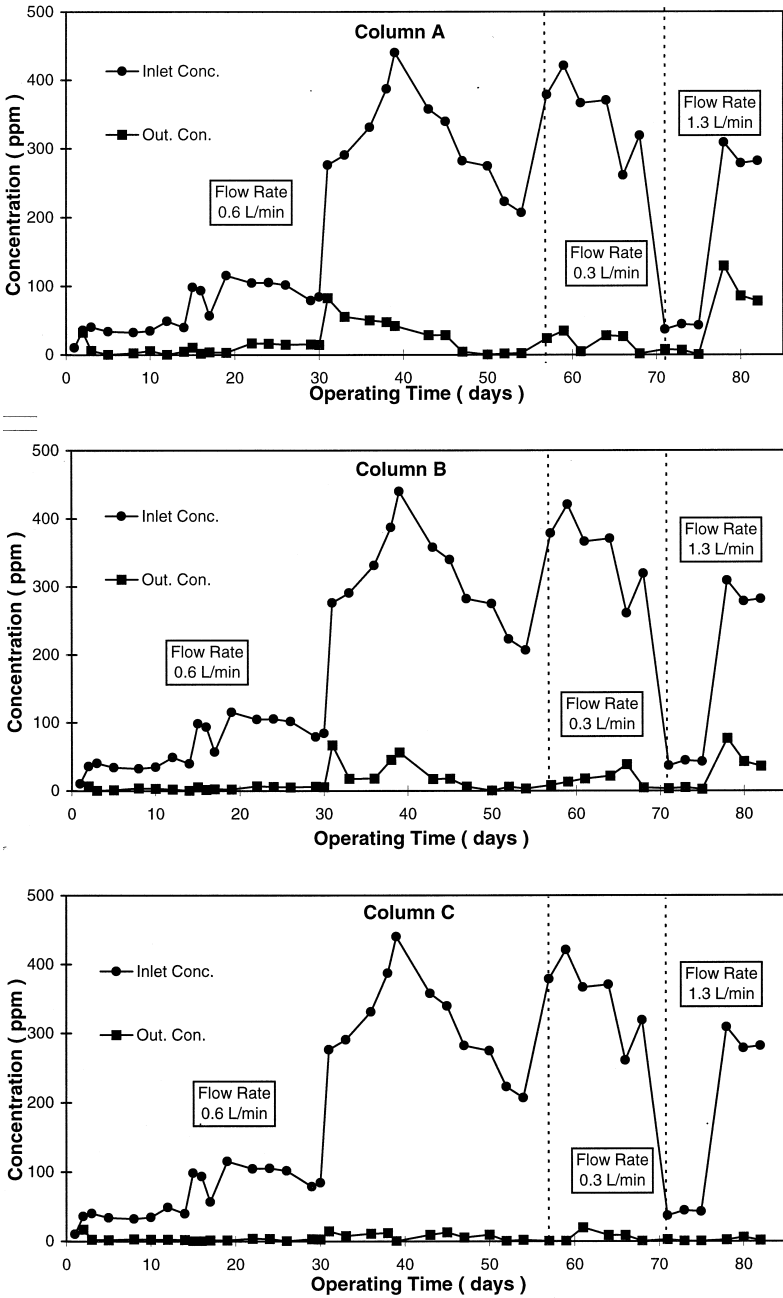Fig. 2. Time-variable benzene concentration in inlet and outlet gas streams of columns A, B and C.

Fig. 3. Time-variable toluene concentration in inlet and outlet gas streams of columns A, B and C.
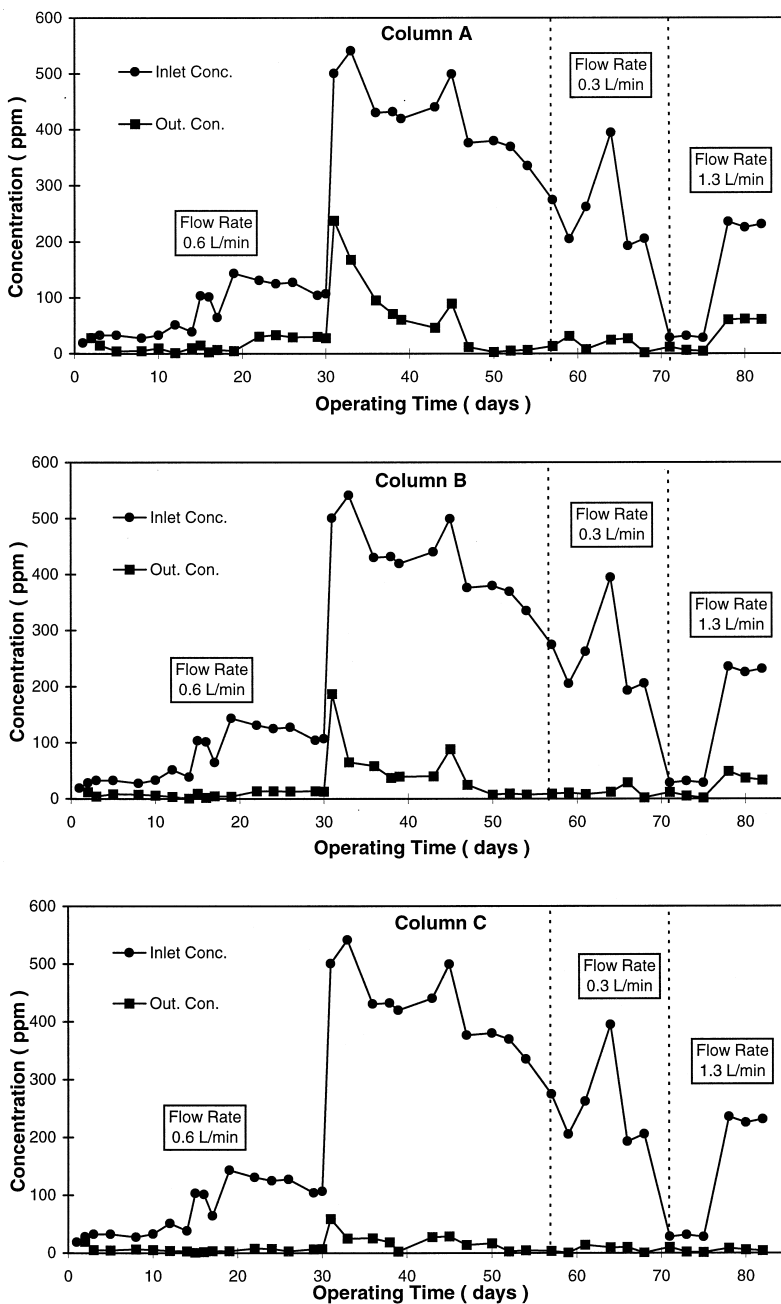
Fig. 4. Time-variable ethylbenzene concentration in inlet and outlet gas streams of columns A, B and C.
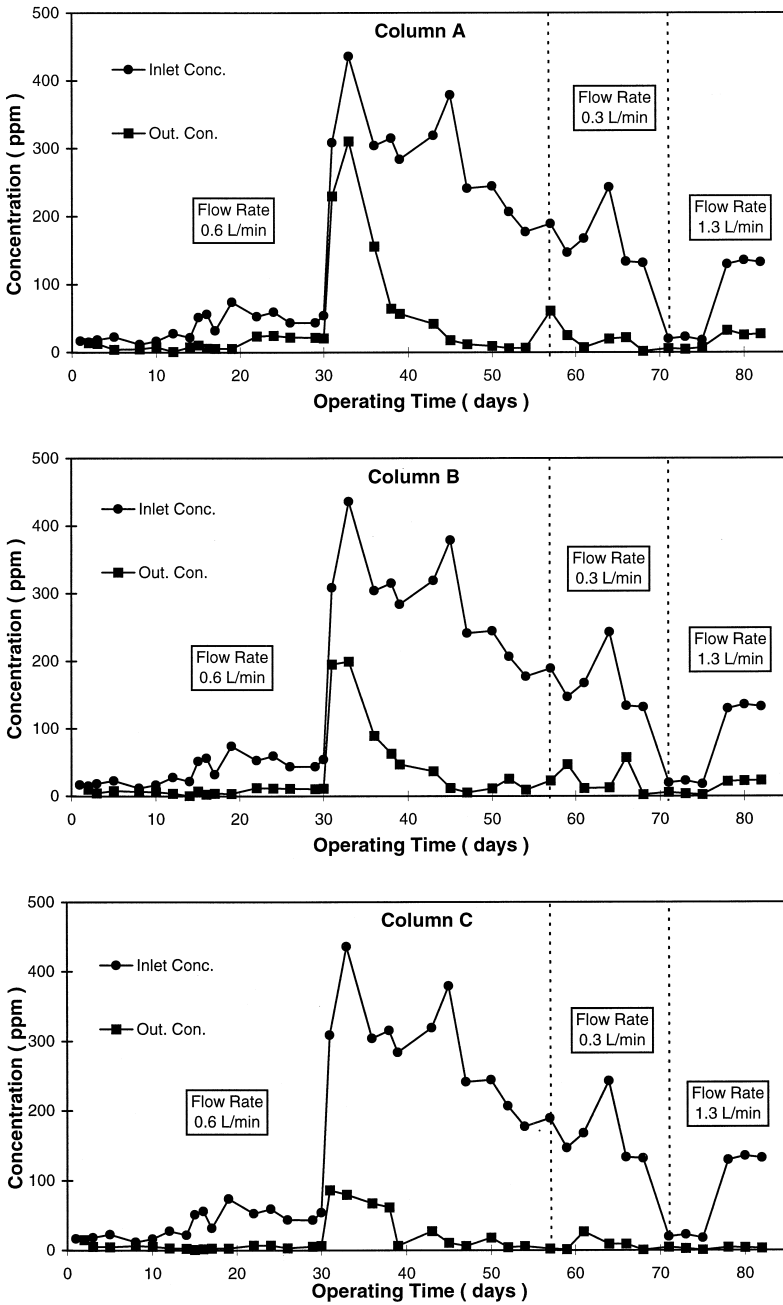
Fig. 5. Time-variable *o*-xylene concentration in inlet and outlet gas streams of columns A, B and C.

three phases (varying EBRT) and time variable inlet concentration data are noted in Figs. 2–5.

The concentration–time curves for all four compounds in column A (no GAC) indicate that biodegradation is the primary mechanism of pollutant removal. Although abrupt increases in influent concentration often resulted in effluent spikes of the target compounds, column A results suggest that the microbial population will eventually acclimate to the elevated substrate condition to again achieve removals of greater than 90% for all four compounds by biodegradation only. Furthermore, doubling the EBRT from 2.6 to 5.1 min (reduction in gas flow rate from 0.6 to 0.3 l/min) appears to mitigate the impact of sharp increases in inlet concentration, implying that gas-to-liquid mass transfer and bio-utilization kinetics are important considerations. These have been examined in greater detail in a companion paper [20]. In addition to the longer residence time, the improved utilization of spike inputs may also be due in part to a larger and better-acclimated biomass stimulated by the previous high inlet concentration period (32–56 h). Overall performance in column A favors removal of benzene followed by toluene, ethylbenzene, and o-xylene; although this ranking may be altered for selected time intervals based upon percent removal. The observed trend is consistent with that expected from compound structure–bioactivity relationships. Differences in percent removal between respective compounds are most pronounced during the initial phase of the experiment immediately following a shock increase in inlet concentrations. After 82 h of operation and a combined total loading of 8 g of BTEX compounds, there is still considerable biodegradation; however, the higher loading rate of the last segment produces elevated effluent levels with compound removal in the range 65–80%.

Data for columns B and C in Figs. 2–5 document improved biofilter performance for varying additions of GAC to the reactor media. BTEX removal in these columns followed several of the trends observed in column A (no GAC), namely: (1) overall removal is still best for benzene, followed by toluene, ethylbenzene, and o-xylene, and (2) distinction in compound removals are most observable as a result of the increase in inlet concentration during the initial phase (∼ 36 h). The results clearly indicate that columns B and C maintain better overall performance in terms of BTEX removal.

Voice et al. [19] suggested that the high adsorption capacity of activated carbon serves to concentrate not only organic compounds that can be utilized as microbial substrates, but also nutrients and oxygen at surface sites. It follows that a very high number of microorganisms tend to concentrate on the surfaces and even in the macropores of carbon particles. As a result, the mechanism by which pollutants are removed by BAC is believed to be adsorption and subsequent biodegradation by the dense microbial population within the matrix of carbon particles.

Figs. 2–5 also illustrate that the impacts of shock loading due to increases in influent concentration or abrupt changes in gas flow rate are diminished in direct proportion to the amount of activated carbon in the biofilter. Percent removal of o-xylene immediately following the spike increase in influent at 32 h increases from 32% to 55% to 80% corresponding to 0%, 3.6% and 13.3% of activated carbon (dry weight basis) in the biofilter. A more than fourfold increase in gas flow rate (phase 3) had virtually no impact on the performance of column C as removals of all compounds were maintained at greater than 95%. The BAC, therefore, operates as a buffer, i.e. it adsorbs a large

portion of the increase, followed by gradual biodegradation of accumulated BTEX molecules. Furthermore, the dense microbial population continuously regenerates the activated carbon, thus maintaining higher removal efficiencies and the ability of columns B and C to respond to shock loads. In their work with BAC and compost/diatomaceous earth biofilters, Hodge and Devinny [11] noted that adsorption was the controlling mechanism of removal during startup, while biodegradation was dominant in the later stages of operation. In this work with compost/GAC biofilters, the same could be said for the initial spike increase in inlet concentration of BTEX compounds. However, it is also apparent that adsorption continues to play an important, even if not dominant, role in biofilter performance over time.

3.2. Column pressure drop

Resistance to gas flow is the major factor that determines the amount of energy needed by the blowers to force the contaminated gas through the filter bed material [20]. Special care should be taken in packing the filter bed material. It should not be packed too densely since this will increase the pressure drop across the filter material resulting in higher energy requirements. At the same time, the bed material should not be packed loosely since this will produce flow channeling and lower the efficiency of the biofiltration process as portions of the contaminated gas may escape with little or no treatment.

Packing material such as compost generally exhibits an aging pattern resulting in cluster formation, shrinkage of the bed, and formation of cracks in the material. These phenomena disturb the homogeneous flow distribution of the gas. Therefore, the pressure drop may also increase considerably over time [21].

During the course of the experiment, the pressure drop was continuously monitored using a water manometer, and its values were plotted as a function of time as presented in Fig. 6. The data indicate a near linear relationship between superficial gas flow rate and head loss which is to be expected for Reynolds numbers in the range 0.3–1.5 as given by the experimental conditions. In phase 1 (0.6 l/min gas flow rate), the pressure drop exhibited a small decrease for a short period followed by a gradual buildup over time. This behavior was similar in every phase given by a change in gas flow rate. Probable reasons for this behavior are the gradual compaction of the packing material during the extended operation time, and the continuous generation and accumulation of biomass with accompanying reduction in the bed void fraction as a result of the substrate oxidation reactions. Fig. 6 also illustrates that the three columns have almost identical pressure drop values at low gas flow rates, with some differentiation developing as the run proceeds. Not surprisingly, the extent of differentiation appears to increase with the gas flow rate. As illustrated most clearly in the 1.3 l/min flow region, the head loss is largest in column A, followed by B and C, which is inversely proportional to the amount of GAC in the bed. Several factors may contribute to this phenomenon, including the lower density of activated carbon versus compost particles and the amount and distribution of biomass in the respective columns. A post-run microbial enumeration was performed for a well mixed sample of media from each column for comparison with a count for the compost prior to the biofilter experiment. The average number of colony
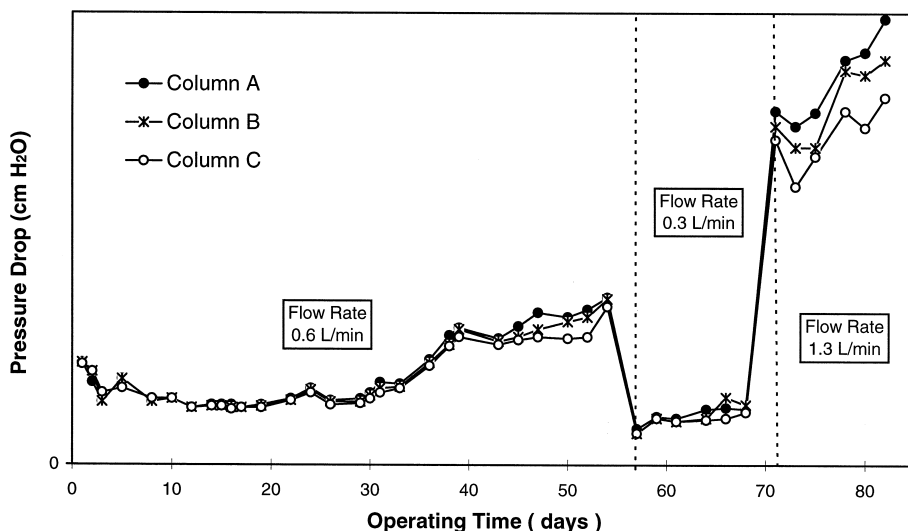
Fig. 6. Time-variable pressure drop in biofilters as a function of flow rate.

forming units per gram dry media (cfu/g dry) was 1.2×10^7 for pre-experimental compost. This value increased nearly 30-fold in post-run column A media to 3.1×10^8 cfu/g dry, and nearly 20-fold to $\sim 2.0 \times 10^8$ cfu/g dry in both B and C. These data suggest a greater production of biomass in column A resulting in a smaller bed void fraction at the end of the run, although no actual determinations of this parameter were made. It is also conceivable that the enhanced contribution of adsorption and biosorption provided by the presence of GAC promotes a more even distribution of biomass in the biofilter, thereby inhibiting build-up of biomass near the flow entrance that would increase head loss. The observed temporary decrease in pressure drop immediately following the sharp increase in gas flow rate from 0.3 to 1.3 l/min (at ~ 69–70 days) may be indicative of sloughing and redistribution of biomass in the bed; however, there was no evidence of biomass sloughing throughout the experiment.

3.3. Moisture content of the biofilter material

The moisture or water content of the biofilter media is an important operational parameter since it directly influences the efficiency of the biofilter and the pressure drop across the filter material. Typically, in biofilters, the pollutants in the gas stream diffuse into the biolayer surrounding the filter bed media particles. This biolayer consists of an aqueous film that contains microorganisms where the pollutants are oxidized. The optimum moisture content in biofilters is between 40 and 60% (dry basis). Values out of this range can result in compaction, breakthroughs of incompletely treated raw gas, and the formation of anaerobic zones that emit odorous compounds [1].

In order to maintain optimum moisture content of the material, the polluted air stream should be humidified before it is introduced to the biofilter. However, if the biofilter is
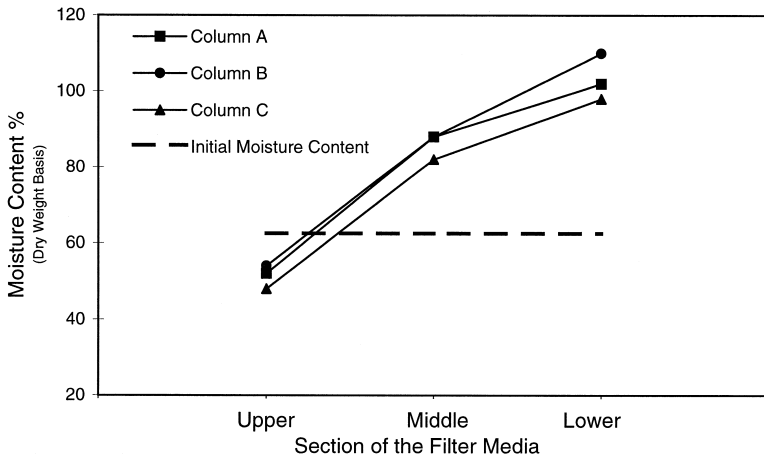
Fig. 7. Moisture content profile in biofilters media before and after the completion of the experiment.

loaded with high gas flow rates (i.e. more than 0.6 l/min in this experiment), humidifying the air stream was not sufficient to maintain the optimum moisture content of the media. In such cases, frequently watering the biofilter by spraying water on top of the filter material was necessary to maintain optimum moisture content. When the gas flow rate was low (less than 0.6 l/min), humidifying the air stream was adequate. At the end of the experiment, samples from all three biofilter materials were examined for water content. All the columns exhibited a decrease from the initial water content in the part of the biofilter closest to the inlet of the gas stream, while the deeper reaches of the bed had higher moisture content than initial levels. Stratification of the biofilter media water content values before and after the experiment is illustrated in Fig. 7. To some extent this may be due to the experimental design, namely the concurrent introduction of water and gas in downflow mode. The variable densities of the air and water would thus contribute to the observed moisture distribution. In addition to this physical factor, biomass distribution and activity can result in a temperature gradient in the biofilter. The occurrence of temperature gradients is inherent in microbial activity; where this activity is high, the temperature will be slightly higher than elsewhere as a result of the liberated energy of oxidation [21]. In this instance where the substrate is introduced at the top of the column, the upper zone is assumed to be more biologically active resulting in elevated temperature and evaporation, while less active zones in the deeper portion of the bed take up excess moisture by condensation.

4. Conclusions

As an air pollution control technology, the use of microbial biofilters for the treatment of air streams contaminated with BTEX compound vapors is reliable, highly efficient and easy to operate and maintain. Biofiltration has a number of advantages in the treatment of low concentration polluted air streams. Besides its high removal

efficiency and low capital and operating costs, it does not generate undesirable byproducts and converts many organic and inorganic compounds into harmless oxidation products (e.g. water and carbon dioxide).

Although the backyard waste compost used in this study is a suitable biofilter media for BTEX removal from waste gas streams under appropriate operating conditions, its unique properties were significantly enhanced after it was mixed with relatively small amounts of granular activated carbon. Two of the major advantages of adding activated carbon are the enhanced buffering capacity of the biofilters to different shock loads that may occur as a result of increases in the influent pollutant concentration or in the polluted gas flow rates, and improved overall removal efficiencies under the same operating conditions. Some operational considerations suggested by this work are as follows.

(i) Over a lengthy period of operation, there appears to be an inverse correlation between the amount of activated carbon and the pressure drop across the biofilter. Higher percentage use of GAC in the bed reduces head-loss buildup.

(ii) There is evidence of ongoing biological regeneration of activated carbon by the microorganisms in the biofilter media.

(iii) When the biofilter system is operated under moderate to low gas loading rates (less than 17.6 m^3/m^2 h), humidifying the polluted air stream by purging it in a water container is sufficient for maintaining the moisture content of the biofilter media within the optimum limits (40–60%). At higher gas loading rates, the biofilter media shows a decrease in the water content in the vicinity of the column inlet for the gas stream. In addition to humidifying the influent air, maintaining the media in a wet condition by spraying water on top was necessary. This situation may also be remedied by employing counter-current operation with introduction of the polluted gas stream at the bottom of the column.

Although several advantages have been noted regarding the use of GAC for enhancing the performance of less expensive compost biofilters, further investigations are required to develop a methodology to estimate the optimum quantity of GAC needed for specific applications to achieve the desired balance between biofilter operation and costs.

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