BIOLOGICAL TREATMENT OF CONTAMINATED AIR STREAMS CREATED DURING THE CLEANUP OF CONTAMINATED SOILS AND AQUIFERS

PROJECT REPORT A-129

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TABLE OF CONTENTS

List of Figures
List of Tables
Acknowledgements
Statement of the Problem
Objectives of the Project
Literature Review
Methodology
Findings
Discussion
Summary
References

LIST OF TABLES

...

Table 1. Experimental Conditions for Compost Column.	17
Table 2. Experimental Conditions for DE Column.	18
Table 3. Compairson of Different Research Results.	50

TABLE OF CONTENTS

List of Figures
List of Tables
Acknowledgements
Statement of the Problem
Objectives of the Project
Literature Review
Methodology
Findings
Discussion
Summary
References

.

LIST OF FIGURES

Fig. 1. Schematic of Degradation of VOCs with a Compost Biofilter
Fig. 2. Schematic of Degradation of VOCs with a DE Biofilter
Fig. 3. Breakthrough Curve for Compost
Fig. 4. Average Gas Influent and Effluent Concentrations—Compost Column 21
Fig. 5. Record of Column Temperature
Fig. 6. Record of Column Humidity
Fig. 7. Applied Loading vs. Elimination Rate
Fig. 8. Applied Loading vs. Elimination Efficiency
Fig. 9. Residence Time vs. Elimination Efficiency
Fig. 10. Air Flow Rate vs. Pressure Drop
Fig. 11. Water Content of Compost Column
Fig. 12. Total Organic Carbon of Compost
Fig. 13. Protein Content of Compost
Fig. 14. Bacterial Population Counts of Compost
Fig. 15. Separation of Physical Adsorption and Biological Elimination
Fig. 16. Average Gas Influent and Effluent Concentration from the DE Column 37
Fig. 17. Record of Column Temperature
Fig. 18. Record of Column Humidity
Fig. 19. Applied Loading vs. Elimination Rate
Fig. 20. Applied Loading vs. Elimination Efficiency
Fig. 21. Residence Time vs. Elimination Efficiency

Fig. 22. Air	Flow Rate vs. Pressure Drop	.45
Fig. 23. Wa	ter Content of DE Column	.46
Fig. 24. Bio	mass on DE	47
Fig. 25. Khe	eldahl Nitrogen Content of DE	48
Fig. 26. Col	lumn Weight Gain During Experiment	49
Fig. 27. Ave	erage Consumption Rates of Nutrients	52

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STATEMENT OF THE PROBLEM

Groundwater is a valuable, limited resource that serves as a major source of water for domestic, industrial, and agricultural uses. A major concern exists over the contamination of groundwater by synthetic organic compounds. The Central Plain, including Oklahoma and Kansas, is dotted with several mid-continent refineries and crisscrossed with many pipelines. Benzene, toluene, ethylbenzene and xylene (BTEX) are considered the main contaminate compounds in groundwaters and soils due to the leakage of gasoline from underground storage tanks (USTs), ruptured refined product pipelines, and spills at refineries and pipeline terminals.

During the cleanup of contaminated groundwaters and soils with volatile organic compounds (VOCs), air stripping or soil venting are frequently used. With both techniques a contaminated waste gas stream is generated. In many instances this gas stream is allowed to escape untreated into the ambient air.

With the reauthorization of the Clean Air Act in 1990, a new focus has been placed on limiting the discharge of toxics into the air—especially from treatment processes. Obviously it is necessary to find a reasonable, practical, and economical method for the treatment of a contaminated air stream.

OBJECTIVES OF THE PROJECT

This project was undertaken to investigate the potential of using biological filters as air pollution cleanup technology. The objectives of this study are as follows:

- 1) to demonstrate the feasibility of eliminating selected BTEX compounds by biofiltration;
- 2) to provide elimination data in biofilters for selected BTEX compounds;
- 3) to compare elimination rates of selected BTEX compounds obtained from biofilters containing two different support media; and
- 4) to present some design criteria which can be used in a full scale case.

LITERATURE REVIEW

Background of Biofiltration

Biofiltration is considered to be an air pollution control (APC) technology. As early as 1923, Bach discussed in the literature the basic concept for the control of H₂S emissions from sewage treatment plants. Reports on the application of this concept dating back to the 1950s were published in the U.S. and in West Germany. Pomeroy received U.S. Patent No. 2,793,096 in 1957 for a soil bed concept and described a successful soil bed installation in California. In the U.S., the first systematic research on biofiltration removal of H₂S was conducted by Carlson and Leiser in the early 1960s (Leson and Winer, 1991).

During the 1960s and 1970s, biofilters were successfully used in the control of odors. Since the early 1980s, an increased interest in biofiltration as a technology to control VOCs and other air toxics emitted from industrial facilities has occurred (Leson and Winer, 1991).

The principle of biofiltration is that when a contaminated air stream containing biodegradable volatile organic compounds (VOCs) or inorganic air toxics is vented through a biologically active material, the contaminants will be adsorbed to the filter medium and then biologically destroyed. The byproducts in this process, if taken to completion, are carbon dioxide, water, and new cells.

Biofiltration has been considered as one of the most promising APC technologies since it can offer significant economic advantages over other technologies and it is also most effective when applied to dilute, easily biodegradable waste streams (Ramaratnam, et al., 1993). Environmental benefits with this technology include low energy requirements and the avoidance of cross media transfer of pollutants (Leson and Winer, 1991).

Applications in the Control of VOCs using Biofiltration

The following classification of gases, according to their degradability, was presented by Bahn (1992): (1) Rapidly degradable VOCs, i.e. alcohols, aldehydes, ketones, etc.; (2) Slowly degradable VOCs, i.e. aliphatic and aromatic hydrocarbons (xylene, toluene, benzene, and styrene), phenols, and methylene chloride; and (3) Very slowly degradable VOCs, i.e. halogenated hydrocarbons (trichloroethylene, trichloroethane, carbon tetrachloride, and pentachlorophenol), polyaromatic hydrocarbons, etc. In most APC situations, compounds in categories (2) and (3) are of the most concern.

Kampbell and Wilson (1987) evaluated the removal of volatile aliphatic hydrocarbons (propane, isobutane, and n-butane) from a waste air stream in the laboratory using a bench scale unit and a

prototype field scale soil bioreactor. Laboratory investigations indicated first-order kinetics for the removal of total hydrocarbon vapors were followed and the potential to degrade light aliphatic hydrocarbons and trichloroethylene, a compound ordinarily resistant to aerobic biological treatment. The predicted behavior of the bioreactor based on laboratory studies agreed closely with the actual behavior of the field system. The prototype bioreactor reduced the hydrocarbon concentration in the air by at least 90% with a residence time of 15 minutes at a pressure drop of 85 cm of water. The bioreactor functioned well through a temperature range of 12 - 24 °C.

Wilson et al. (1988) performed a laboratory study using fixed film bioreactors to biologically remediate contaminated air streams. In their work, the authors introduced TCE and TCA vapors into bioreactors containing diatomaceous earth on which a biofilm had developed. Headspaces of the columns were plumbed to receive vapors of TCE and TCA, a mixture of butane and air, and a nutrient media. Because TCE and TCA can be degraded by a monooxygenase enzyme, butane (8 mg butane/L air) was also introduced into the biological reactors to serve as a primary substrate. Influent TCE concentration ranged from 90 μ g/L to 770 μ g/L while the TCA concentration ranged from 200 μ g/L to 990 μ g/L. The maximum removal efficiency observed in this work was 94% for TCE and 93% for TCA. The authors did note a drop in removal efficiency for both compounds as their influent concentration increased.

Galaska et al. (1989) designed an upward flow bio-airtower to degrade the contaminated air originating from a gasoline service station soil venting project. High surface area inert plastic media within the bio-airtower was used to support the biofilm growth. Water containing inorganic nutrients was recirculated over the packing material in order to keep the biofilm moist and provide a working medium for biological degradation. The unit was tested at flow rates of approximately 10.4 and 17.1 cubic feet per minute (CFM). The percent removal for both specific compounds such as benzene, toluene, xylene (BTX), and total petroleum hydrocarbons approximately doubled when the air flow rate through the bio-airtower was lowered. The authors concluded that changing the water recirculation rate and the influent concentration appeared to have very little effect on either the individual BTX or total petroleum hydrocarbons percent removal. The most important variable affecting petroleum hydrocarbon removal in this system appeared to be air flow rate. Specifically, BTX removal increased from approximately 45.2% to 74.5%, and total petroleum hydrocarbon removal increased from approximately 30.6% up to 60.8% after the flow rate was lowered from 17.1 to 10.4 CFM. The contact time between the airborne contaminants and the biomass was very limited in this type of system. Therefore it is unreasonable to assume that a minimal air residence time would provide effective treatment.

Bioreactors similar to those used by Wilson et al. (1988) were utilized in the work of Speitel and Malay (1990). However, these authors fed their system, packed with diatomaceous earth, a methane mixture instead of butane. Nutrients and influent gas flowed co-currently in the columns. The primary objectives of their work was to investigate the effect of influent concentration in the gas stream and gas flow rate on the degradation of TCE and 1,2-dichloroethane (DCA). The influent concentration for TCE ranged from 273 to 569 μ g/L at an average gas flow rate of 28.5 L/day while the surface loading rates varied only slightly from 0.160 - 0.335 gm TCE/m² hr. In the two experiments conducted using TCE the average removals were 28 and 57%. The authors also noted reduced TCE removal at influent gas concentrations greater than 500 μ g/L and speculated that elevated concentrations may have been toxic to the organisms. However, they stated that more work is needed to prove this point. The two experiments conducted with DCA shown an average removal of 18% when the average surface loading rate was 0.76 gm/m² \cdot hr and the influent concentration varied from 600 to 1000 μ g/L at an average gas flow rate of 47.4 L/day as opposed to 81% removal at an average surface loading rate of 0.45 gm/m² hr with influent concentrations ranging from 870 - 1100 µg/L at an average gas flow rate of 22.7 L/day. Overall, the authors concluded that biofiltration is a viable process for treating gas streams containing chlorinated solvents at low concentration levels.

Douglass et al. (1991) studied the applicability of biofiltration for the treatment contaminated streams typically found in air stripper off gas. The five packing media used in their study were native sand with chemical nutrient amendments, saw dust with chemical nutrient amendments, peat buffered with calcium carbonate to control pH, a mixture of 20% manure and 80% sawdust, and a mixture of 20% aerobically digested waste water sludge and 80% vermiculite (or sawdust). In this study, the influent concentrations of total gasoline, toluene, and benzene were 61.4, 3.6 and 2.8 μ g/L, respectively. Through their investigation, the authors reported that the maximum degradation rates for benzene and toluene were about 53 to 78 mg/minute m³ and 18 to 28 mg/minute m³ for o-xylene. Because the biological rate of degradation appeared to be the limiting factor for treatment efficiency, the chemical composition of the contaminants could be very important since rates of biodegradation varied by substrate. For the design and application of a mobile treatment system, peat buffered with calcium carbonate is the lightest, least expensive, and among the best biological support media.

Hodge et al. (1991) investigated the aerobic biodegradation of gas phase jet fuel and diesel by microorganisms supported on 4 different filter media. The filter media used in this work were: activated carbon (GAC), diatomaceous earth (DE), a soil mixture, and a mixture of activated carbon and diatomaceous earth (50% each by volume). For the GAC and DE columns, a microbial seed

culture and nutrients were completely mixed throughout the media. Individual components of the jet fuel and diesel were not determined and all the degradation rate data were reported as total hydrocarbon as methane equivalents. The authors found that the biodegradation rates ranged from 0.3 - 5.2 gm HC/m³·hr (for jet fuel) and 0.2 - 1.3 gm HC/m³·hr (for diesel) on the four different support matrices. The jet fuel was degraded faster than the diesel in all cases. The diatomaceous earth supported the lowest biodegradation rates. They also noted the importance of relative humidity for operational efficiency of the biofilter. The optimum water content levels were different for each filter medium. Overall, they concluded that this technology appears to be a viable method for treating hydrocarbon vapors.

Medina et al. (1992) conducted a bench scale experiment where granular activated carbon (GAC) was used as the microorganism support medium in a biofilter to treat soil vapor extraction off gases. In this study, gasoline vapors were introduced into the column in a downflow mode. Nutrients and microbial seed materials were vigorously mixed with the GAC medium. This material was used to fill the column. Excess solution was drained and returned to the top of the column. The excess was again drained and discarded. Following the seeding process, no further attempt was made to add nutrients or organisms. Initial adsorption provided a high percentage of removal (>85%). However, after the adsorption capacity was saturated, the treatment efficiency dropped dramatically (down to 7-20%). Later, biological activity increased to the point where effective treatment was possible (85-95% of removal efficiency). The main obstacle to effective treatment was the slow growth of biomass in the lower portions of the column. The average and maximum Elimination capacities of the biofilter were 64 and 119 gm/m³ hr, respectively. The residence time ranged from 5.9 - 7.0 minutes, averaging 6.4 minutes. Elimination rates were proportional to pollutant loading. Plate counts shown that both bacterial and fungal numbers increased as the column operated.

Vaughn et al. (1993) carried out research on the biodegradation of vapor phase p-xylene. Two bioreactors were packed with different media. One contained glass spheres and the other had diatomaceous earth (DE) pellet. The process parameters tested were gas and liquid flow rates and influent xylene concentrations. A sterile liquid medium, which served as a nutrient source, was distributed into the columns at a rate of 5 - 10 mL/minute. The concentration of xylene, introduced to the two columns, was 150 - 1500 ppm (v/v) at a gas flow rate of 600 - 6000 mL/minute. Their results indicated that total mass degradation rates were significantly higher in the DE pellet column (1.4 - 1.8 mg/minute) than in the glass sphere column (0.3 - 0.6 mg/minute). Protein levels per unit surface area were significantly higher in the DE pellet column.

Apel et al. (1993) demonstrated the utility of a fixed film biofilter for removal of methane, TCE, and xylene. Comparisons between the gas phase bioreactors, conventional shaken cultures, and sparged liquid bioreactors shown that the gas phase bioreactors offer advantages over the other two systems for the degradation of methane in air. Rates of methane removal with the gas phase bioreactors were 2.1 and 1.6 fold greater than those exhibited by the shaken cultures and sparged liquid bioreactors, respectively. The gas phase bioreactors were shown to have application for the removal of TCE vapors from air with a removal rate of approximately 2.6 mg TCE /day m^3 . Xylene vapors were also scrubbed from air using gas phase bioreactors. At a feed rate of 140 μ g of xylene per minute, approximately 46% of the xylene was mineralized to carbon dioxide in a single pass through a bench scale gas phase bioreactor.

Rho et al. (1993) worked on a demonstration project to study the feasibility of biofiltration. Some microbial strains identified as being responsible for the degradation of VOCs (ethanol, methanol, n-propanol, 2-butanone, propyl acetate, and propanol) were <u>Pseudomonas sp.</u>, <u>Bacillus sp.</u> and <u>Klebsiella sp.</u>. In this study, peat was used as a packing material and nutrients (nitrogen source only) were recirculated through the filter with a microbial suspension on a weekly basis. The results from these authors shown that over a 7-month period, the average inlet concentration was 1000 mg C/m³ gas and the organic load was 100 g C/m³ bed /h whereas the residence time was 18 seconds. The maximum Elimination capacity was 72 g C/m³ bed /h.

Cox et al. (1993) set up the experiments to enrich styrene-degrading fungi in biofilters under conditions representative for industrial off-gas treatment. Through each biofilter, a 43 L/hr styrene/ air mixture was passed with a relative humidity of 80 - 85% and a styrene concentration of initially 290 mg/m³. From the support materials tested, polyrethane and perlite proved to be most suitable for enrichment of styrene-degrading fungi. The biofilter with perlite completely degraded styrene when amounts ranging between 290 and 675 mg/m³ in the influent gas were present. An elimination capacity of at least 70 g styrene per m³ perlite per hour was calculated.

Gregg et al. (1993) used a bench-top bioscrubber testing unit to treat vapor phase toluene. In their work, activated carbon served as packing material. The inlet concentration of toluene was 10 -20 ppm with a air flow rate of 0.5 - 2.0 L/minute. Inorganic nutrients, required for biological growth, were fed downflow to the column at a 0.1 mL/hr rate. The authors reported that the bioscrubber achieved effective removal of low levels of toluene vapor. Column lengths ranging from 5 to 10 inches were required to confine the wavefront of the filter for the empty bed contact times ranging from 1 to 4 seconds.

Zilli et al. (1993) evaluated biofiltration technology for removal of phenol from waste gases. With more than 1 year of continuous operation of a lab-scale unit at a gas flow rate of 40 L/minute, corresponding a residence time of 54 seconds, very low phenol concentrations were achieved in the outlet.

Hodge and Devinny (1994) conducted bench scale and continuous flow experiments to evaluate biofiltration in the treatment of waste gas streams contaminated with ethanol. The efficiency of three different packing materials—granular activated carbon (GAC), compost, and a mixture of compost and diatomaceous earth (compost/DE)—was compared for different operating conditions. A mathematical model was developed that describes basic transport and biological processes. For each column, the model predictions were in agreement with experimental data under the conditions of the two lower loading (79 - 156 g/m²·hr). However, for the high loading (256 - 270 g/m²·hr), possible channeling effects caused variability in terms of ethanol concentration, resulting in the completely different shapes of the concentration gradient profiles.

Sorial et al. (1994) investigated three microbial attachment media, which were channelized and pelletized media as well as compost mixture medium in biofilters for the treatment of air contaminated with volatile organic compounds (VOCs). They found that all the three media achieved more than 95% toluene removal efficiencies at an organic loading of 0.725 kg COD/m³/day.

Apel et al. (1994) developed a biofilter for the biodegradation of gasoline vapors. The overall goal of this effort was to provide information necessary for the design, construction, and operation of a commercial gasoline vapor biofilter. Experimental results indicated relative high amounts of gasoline vapor adsorption can occur during initial exposure of the biofilter bed medium to gasoline vapors. Biological removal occurred over a temperature range of 22-44 °C with removal being completely inhibited at 54 °C. At lower gasoline concentrations the vapor removal rates were considerably lower than those at higher gasoline concentrations, indicating that substrate availability (i.e. transport) was limiting in the system. In addition, total BTEX removal over the operating conditions employed was 50-55%. Removal of benzene was approximately 10-15% while removal of the other members of the BTEX group was typically higher than 80%.

Main Problems in the Control of VOCs Using Biofiltration

Although much work has been done on the control of VOCs using biofiltration, there are still some problems listed below, which have not been attacked or addressed sufficiently:

- For the BTEX compounds, most research has dealt with a single BTEX compound such as benzene, or toluene, or xylene instead of multiple compounds. In a practical sense, it is necessary to investigate the behavior of all the BTEX compounds.
- 2) Compared to other filter media, such as GAC and DE, research work regarding compost was found less often, especially concerning BTEX biodegradation. In fact, compost has several advantages over other filter media. For instance, it is very cheap and no nutrient addition is required.
- 3) There was little research concerning the comparison of a biofilter with a nutrient addition and a biofilter without a nutrient addition in the degradation of BTEX compounds. Furthermore, in this field, what are the key design criteria? What kind of model can be applied to a specific type of biofiltration technique for the BTEX compounds? These questions need to be answered in detail.
- 4) There were few research papers which provided cost estimations for the degradation of BTEX compounds for a specific type of biofiltration technique.

The above existing problems provide a guide for the objectives of this research project. These objectives are as follows:

- to demonstrate the feasibility of eliminating selected BTEX compounds by biofiltration;
- to provide Elimination rate data in biofilters for selected BTEX compounds;
- to compare Elimination rates of selected BTEX compounds obtained from biofilters containing two different support media; and
- to present some design criteria which can be used in a full scale case.

METHODOLOGY

Materials and Methods

Description of the schematic of the experimental apparatus

In this study, two columns constructed out of plexiglass pipes were employed as biofilter reactors. The columns, having an inside diameter of 3.1 cm and a length of 119 cm, were packed with either compost or diatomaceous earth (DE) which served as support media for the growth of

microorganisms. Seven sampling ports, having a 18.5-22.5 cm spacing between ports, were placed along the length of each column. Two separate reservoirs, containing three pure solvents of selected BTEX compounds(toluene, ethylbenzene, o-xylene) and distilled water, respectively, were used to create the contaminated air stream which was sent to each column. The air flow rate to each column was controlled by needle valves on the air inlet, outlet and waste lines. The influent concentration to each column was adjusted using the ratio of two air flow rates from the contaminant and distilled water reservoirs. Air flow rates were monitored by four air flow meters. The seed bacterial culture was mixed with compost or diatomaceous earth when the medium was packed into each column. Schematic diagrams of these two biofilters are shown in Figures 1 and 2.

Selection of porous filtration materials

There are several materials previous research has demonstrated to be suitable as filtration materials. These include:

- 1) granular activated carbon (GAC);
- 2) diatomaceous earth (DE);
- 3) compost; and
- 4) soil.

Since DE and compost are economical, they were selected for use in this study.

Selection of BTEX compounds

BTEX refers to benzene, ethylbenzene, toluene and xylene. The compounds benzene, ethylbenzene, toluene, and o-xylene are representative of gasoline in terms of their mass percent. However, benzene is the most volatile and it was difficult to keep a consistent influent concentration during preliminary experiments. Therefore, ethylbenzene, toluene and o-xylene were selected to be used in this study.





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Figure 2. Schematic of Degradation of VOCs with a DE Biofilter

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Incubation of the microbial culture

Six liters of activated sludge from a refinery wastewater treatment plant were placed into three bottles, aerated, supplied with an inorganic mineral solution, and injected with stock solutions of toluene, ethylbenzene and o-xylene daily. The flow rate of mineral solution added was about 200 mL/day and the amount of each organic stock solution ($200 \mu g/mL$) was $200 \mu g/day$. Before introducing the culture into the biofilters, this incubation system was operated for at least one month.

Preparation of the mineral media

A mineral solution was used for the incubation of the microbial culture and recirculation through the DE column. The components of mineral solution were as follows:

 $KH_2PO_4 = 700 mg/L$ $NH_4(SO_4) = 500 mg/L$ $MgSO_4 \cdot 7H_2O = 600 mg/L$ $FeCl_3 \cdot 6H_2O = 1 mg/L$ $MnSO_4 = 1 mg/L$

The mineral solution was made up in tap water.

Start-up procedure

The procedure for starting the compost column was as follows:

- mix compost (530 gm) with the bacterial culture (20 mL);
- pack the mixture into the column;
- connect the compost column to a contaminated air stream;
- adjust air flow rates to the expected values; and
- take gas phase samples from the 7 sampling ports on the column after 30 minutes.

The procedure for initiating the DE column was as follows:

- mix DE (482 gm) with the bacterial culture (20 mL);
- pack the mixture into the column;
- recycle the mineral solution for 3 days;
- inject each organic stock solution (200 μg/L) with a 5-mL plastic syringe into the top of the column for 3 days;
- connect the DE column to a contaminated air stream;
- · adjust air flow rates to the expected values; and
- take gas phase samples from the 7 sampling ports on the column after 30 minutes.

Analytical techniques

Temperature and Humidity

Daily temperature and humidity measurements were performed on the influent and effluent of each bioflilter using a Digital Thermometer/Hygrometer (Model DTH1, Davis Instruments) that has effective ranges of -30-200 °F and 20-90 % RH.

BTEX concentrations

The concentrations of toluene, ethylbenzene and o-xylene from 7 sampling ports on each column were analyzed using a HP 5890 series II gas chromatograph equipped with a flame ionization detector. The GC was connected to a HP 3396 series II integrator. Each sample was injected into the GC with a gas tight syringe (Hamilton, 250 μ L) using an injection volume of 200 μ L. Gas standards for the three compounds were determined by equilibrating a series of known volumes of aqueous standards with the gas phase in sealed bottles. Henry's Law was used to calculate the resulting gas phase concentration. The calibration curve was run weekly.

Water content

At the beginning and end of each experimental run (or each loading rate), the water content of the medium (compost or DE) was measured. The detailed procedure for determining the water content of each medium was described in section 209F of *Standard Methods* (APHA et al., 1985).

Pressure drop

At the beginning and end of each experimental run (or each loading rate) on each column, the pressure drop across the column was determined. An Air Velocity Meter (Model 400, Dwyer Instruments, Inc.), which can function as a manometer, was employed to measure a pressure drop across each biofilter.

Protein content

At the beginning and end of each experimental run (or each loading rate) on the compost column, the protein content on the medium was determined. The protein content on the compost was determined by the Bio-Rad Protein Assay. The detailed procedure for determining the protein content was described in *User's Manual of the Bio-Rad Protein Assay* (Kit II, Bio-Rad Laboratories).

Volatile solids

At the beginning and end of each experimental run (or each loading rate) on the DE column, the volatile solids on the DE medium was determined. The detailed procedure for determining the volatile solids on DE was described in section 209F of *Standard Methods* (APHA et al., 1985).

Bacterial population counts

At the beginning and end of each experimental run (or each loading rate) on the compost column, the bacterial population counts on the compost was determined. The detailed procedure was described by Benson (1967).

Total organic carbon

At the beginning and end of each experimental run (or each loading rate) on the compost column, the total organic carbon of the compost was determined. The detailed procedure was described by Gaudette and Flight (1974).

Kjeldahl nitrogen

At the beginning and end of each experimental run (or each loading rate) on the DE column, the kjeldahl nitrogen on DE was determined. The detailed procedure was described in section 420B of *Standard Methods* (APHA et al., 1985).

Nitrogen and Phosphorus

At the beginning and end of each experimental run (or each loading rate) on the DE column, the inorganic nitrogen and phosphorus in the recirculation solution were determined. The detailed procedures were described in sections 420B.4d, 424C and 424D of *Standard Methods* (APHA et al., 1985).

Experimental design

Adsorption study

To separate the physical adsorption from biological elimination on the compost medium, a breakthrough test was performed wherein 700 cm³ of compost was placed into a glass column with a diameter of 5.2 cm and a height of 33.0 cm. The column and compost medium were sterilized in an autoclave for 30 minutes before they were used. The temperature and pressure were 248 °C and 15 psi, respectively, in the autoclave. The influent concentration of the contaminated air stream was controlled at about 1000 VOCs $\mu g/L$ with an air flow rate of 42 mL/minute, resulting in a residence

time of 10 minutes. The test was conducted until complete exhaustion of the column occurred; the point where the ratio of influent concentration to effluent concentration was 1.0.

Compost column study

The compost column was operated under conditions of two influent concentrations (low and high) in an upflow mode, where the contaminated air stream entered through the bottom of the column. For low influent concentration, the air flow rate was varied from 25 to 200 mL/minute, resulting in four residence times of 20, 10, 5 and 2.5 minutes, and for high inlet concentration, the range of the air flow rates was 25 - 100 mL/minute, corresponding to three residence times of 20, 10 and 5 minutes. No nutrients were added to the column. The experimental conditions are listed in Table 1.

Experimental Run No.	1	2	3	4	5	6	7
Influent Gas Conc. (µg/L)	1000	1000	1000	1000	2000	2000	2000
Air Flow Rate (mL/minute)	25	50	100	200	25	50	100
Applied Loading Rate (mg/cm ^{3.} day)	0.10	0.20	0.40	0.80	0.20	0.40	0.80
Residence Time (minutes)	20	10	5	2.5	20	10	5

Table 1. Experimental Conditions for Compost Column

DE column study

Most experimental conditions were the same as for the compost column; however, in this study, a recirculating nutrient solution at a flow rate of 5 mL/minute was added to the top of the column. The contaminated air stream was introduced into the top of the column. The two flow streams entering the column operated in a co-current mode. The experimental conditions are presented in Table 2.

Experimental Run No.	1	2	3	4	5	6	7	8
Influent Gas Conc. (µg/L)	1000	1000	1000	1000	2000	2000	2000	2000
Air Flow Rate (mL/minute)	25	50	100	200	25	50	100	200
Applied Loading Rate (mg/cm ^{3.} day)	0.13	0.26	0.52	1.04	0.26	0.52	1.04	2.08
Residence Time (minutes)	20	10	5	2.5	20	10	5	2.5

Table 2. Experimental Conditions for DE Column

FINDINGS

Adsorption study

Breakthrough curve for compost

The result of the breakthrough test is illustrated in Figure 3. It can be seen that the Ce/Ci reached 0.51 in an elapsed time of 3.3 hours, and after 9.6 hours, the value of Ce/Ci was 1.0. Using this breakthrough curve, a physical adsorption capacity of 0.33 mg VOCs/gm compost can be determined by the integration of the area in front of curve.

Compost column study

Average gas influent and effluent concentrations

During the entire period of the compost column study, the gas influent and effluent concentrations from the column were monitored on a daily basis. The average gas influent and effluent concentrations are shown in Figure 4. The low influent concentration was controlled at approximately 1000 μ g VOCs/L for Runs 1 to 4 and the high influent concentration was maintained at about 2000 μ g VOCs/L for Runs 5 to 7. Within each experimental run, it was difficult to control the gas influent concentration at a constant value through this period mainly due to the variability of the air source pressure.

Records of column temperature and humidity

The temperature and humidity of the column were also measured on a daily basis. Figure 5 and 6 are the records of the average temperature and humidity, respectively, of the column for the entire experimental run. For the 189-day period, the temperatures and humidities of both the influent and effluent air were fairly close, and the average temperature of the column was about 70 °F, which is appropriate for the growth of bacteria. The average humidity, however, was about 80%, which seems to be a little bit high in terms of the optimum value of about 50 - 60% (Ottengraf, 1986).

Effect of applied loading on Elimination rate

The effect of applied loading on the elimination rate is provided in Figure 7. As the applied loadings ranged from 0.1 to 0.77 mg VOCs/cm³ compost day with the low inlet concentration, the



Figure 3. Breakthrough Curve for Compost



Figure 4. Average Gas Influent and Effluent Concentrations from the Compost Column



Figure 5. Record of Column Temperature





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Figure 7. Applied Loading vs. Elimination Rate

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Elimination rates were $0.1 - 0.41 \text{ mg VOCs/cm}^3$ compost day. However, the Elimination rates were $0.11 - 0.36 \text{ mg VOCs/cm}^3$ compost day on the high inlet concentration when the applied loadings varied from 0.2 to 0.82 mg VOCs/cm³ compost day. Holding at a fixed applied loading, the higher the inlet concentration the lower the elimination rate. The reason could be the toxicity of BTEX compounds to the bacteria.

Effect of applied loading on elimination efficiency

Figure 8 shows that for applied loadings of $0.1 - 0.77 \text{ mg VOCs/cm}^3$ compost-day, the elimination efficiencies of the VOCs were 100 - 53.1% with low inlet concentrations, respectively. But, at the applied loadings ranging from 0.2 to 0.82 mg VOCs/cm³ compost-day, the efficiencies were 53.3 - 44.1\%, respectively, when fed the high inlet concentration. This revealed that the three VOCs could be more completely removed under the condition of a low inlet concentration.

Effect of residence time on elimination efficiency

In Figure 9, it is seen that with the residence times varying from 5, 10 to 20 minutes the resulting elimination efficiencies in terms of total VOCs were 74.2, 98.9 and 100% with a low inlet concentration, and 44.1, 50.4 and 53.3% with a high inlet concentration, respectively. The shorter the residence time, the lower the elimination efficiency.

Effect of air flow rate on pressure drop

The effect of an air flow rate on the pressure drop through the column is presented in Figure 10. Under the condition of a high inlet concentration, the smaller the air flow rate the lower the pressure drop. At air flow rates of 25, 50 and 100 mL/minute, the pressure drops were 0.05, 0.09 and 0.11 in. H₂O, respectively.

Water content of compost column

The values of the average water content of the compost column are shown in Figure 11. During the first 28 days, the difference in water content between the top and the bottom of the column, operated in an upflow mode, was small. However, from day 46 on, the difference gradually increased until day 116. After that the difference seemed to be a constant.

Total organic carbon of compost

Figure 12 presents the total organic carbon of the compost medium. During the first 28 days, the total organic carbon (TOC) values from both the top and bottom of the column were small, and there was little difference between them. After day 46, there was a significant increase on the TOC

for both the top and bottom. On day 147, the TOC difference between the top and the bottom reached its maximum.

Protein content of compost

The protein content of the compost medium is provided in Figure 13. For the bottom of the column, the protein content increased with elapsed time but for the top of the column, the protein content did not increase until day 63. After that, it decreased until day 172 and then remained a constant.

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Figure 8. Applied Loading vs. Elimination Efficiency

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Figure 9. Residence Time vs. Elimination Efficiency

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Figure 10. Air Flow Rate vs. Pressure Drop



Figure 11. Water Content of Compost Column



Figure 12. Total Organic Carbon of Compost



Figure 13. Protein Content of Compost

Bacterial population counts of compost

The results of the bacterial population counts of the compost medium are plotted in Figure 14. During the first 63-day period, the SPCs (Standard Population Counts) from the column top were higher than those from the bottom. However, from day 116 through day 189, the SPCs from the column top were less than those from the column bottom.

Separation of physical adsorption and biological degradation

Figure 15 shows the separation of the physical adsorption and biological elimination capacities of the compost. During the acclimation period (Run No. 0), the total elimination capacity was 6.9 mg VOCs/gm compost, while the physical adsorption capacity was only 0.33 mg VOCs/gm compost (based on the integration of the area in front of curve). Therefore, the biological elimination capacity could be considered the most significant portion (6.57 mg VOCs/gm compost) in this period. Throughout Runs 1-6, it was assumed that if contaminants were physically adsorbed to the compost medium, they would finally be degraded by microorganism. Thus, the maximum physical adsorption capacity in each of these periods was 0.33 mg VOCs/gm compost, and the total elimination capacity can still be attributed to the biological elimination capacity.

DE column study

Average gas influent and effluent concentrations

As with the compost column, during the whole period of the DE column study, the gas influent and effluent concentrations from the column were monitored daily. The average influent and effluent concentrations are shown in Figure 16. In this portion of the study, the low influent concentration was controlled at approximately 1200 μ g VOCs/L for Runs 1 to 4 and the high influent concentration was maintained at about 2300 μ g VOCs/L for Runs 5 to 8. Within each experimental run, it was difficult to control the gas influent concentration at a constant value through the experimental period mainly due to the variability of the air source pressure.

Records of column temperature and humidity

The temperature and humidity of the column were also measured on a daily basis. Figures 17 and 18 are the records of the average temperature and humidity of the column for each run. For the entire 97-day period, the temperatures and humidities of both the air influent and effluent were fairly close except those on day 62 (for temperature) and day 18 (for humidity), and the average temperature of the column fell within a range of 50-70 °F, which is appropriate for the growth of bacteria. The average humidity of the column was about 80%.

Effect of applied loading on elimination rate

The effect of applied organic loading on the elimination rate is provided in Figure 19. As the applied loadings ranged from 0.16 to 1.13 mg VOCs/cm³ DE·day with a low inlet concentration, the elimination rates were 0.16 - 1.12 mg VOCs/cm³ DE·day. The elimination rates were 0.30 - 2.39 mg VOCs/cm³ DE·day with a high inlet concentration when the applied loadings varied from 0.30 to 2.43 mg VOCs/cm³ DE·day. At any fixed applied loading, there was no significant difference in the elimination rates between the low inlet concentration and a high inlet concentration.



Figure 14. Bacterial Population Counts of Compost

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Figure 15. Separation of Physical Adsorption and Biological Elimination



Figure 16. Average Gas Influent and Effluent Concentrations from the DE Column



Figure 17. Record of Column Temperature

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Figure 18. Record of Column Humidity

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Figure 19. Applied Loading vs. Elimination Rate



Effect of applied loading on elimination efficiency

Figure 20 shows that for the applied organic loadings of $0.16 - 1.13 \text{ mg VOCs/cm}^3 \text{ DE} \cdot \text{day}$, the elimination efficiencies of the VOCs varied from 100 to 99.7% at the low inlet concentration, respectively. As the applied loadings increased to $0.30 - 2.43 \text{ mg VOCs/cm}^3 \text{ DE} \cdot \text{day}$, the elimination efficiencies ranged from 100 to 98%, respectively.

Effect of residence time on elimination efficiency

In Figure 21, it can be seen that with the residence times varying from 20, 10, 5 to 2.5 minutes, the resulting elimination efficiencies were 100, 100, 100, and 99.7% at the low inlet concentration and 100,100, 98, and 98%, respectively, with the high inlet concentration. Residence time less than 10 minutes had the lowest elimination efficiencies.

Effect of air flow rate on pressure drop

The effect of air flow rate on the pressure drop across the column is presented in Figure 22. While holding the air flow rate at 25, 50 and 100 mL/minute, the pressure drops were 0.02, 0.03 and 0.30 in. H₂O at the low inlet concentration, and 0.80, 4.5 and 3.0 in. H₂O at the high inlet concentration, respectively. Most of these values indicated that the larger the air flow rate, the higher the pressure drop. On the other hand, a pressure drop across the column might also be increased over the elapsed time due to accumulation of the biomass on the medium.

Water content of DE column

The values of the average water content of the column are shown in Figure 23. During the entire 94-day experimentation, the differences of water content between the top and the bottom of the column gradually increased. The values from the top samples were higher that those from the bottom samples.

Biomass on DE

The values of the biomass on the DE medium are provided in Figure 24. Throughout the period of 96 days, there was only small increase in biomass in the bottom portion of the column but a trend of increasing biomass in the top portion of the column was observed.

Kjeldahl nitrogen content of DE

The result of the kjeldahl nitrogen content of the DE medium is plotted in Figure 25. There was a significant increase of the kjeldahl nitrogen content in the top portion of the column during the first 18 days, and after that, the kjeldahl nitrogen content gradually decreased with elapsed time. Like the

biomass content, there was a little increase in kjeldahl nitrogen in the bottom portion of the column during the whole 96 day period.

Column weight gain during experiment

The column weight gain, using the DE medium, is shown in Figure 26. After each experimental run, the column was weighed using a balance. The column weight gain was assumed to be caused by a biomass gain on the DE medium. A large number of weight gain was found individually during Runs 1, 2 and 4; however, the relatively small number of the weight gain from Runs 3, 5 and 6, respectively was recorded.



Figure 20. Aplied Loading vs. Elimination Efficiency



Figure 21. Residence Time vs. Elimination Efficiency

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Figure 22. Air Flow Rate vs. Pressure Drop









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Figure 25. Kjeldahl Nitrogen Content of DE

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Average consumption rates of nutrients (nitrogen and phosphorus) in the recirculation solution

Figure 27 presents the average consumption rates of nutrients (nitrogen and phosphorus) in the recirculation solution. During the entire 96 days of the experiment, there were a higher average consumption rate of total inorganic nitrogen than that of total inorganic phosphorus.

DISCUSSION

A comparison of the results of this study with those obtained by other investigators is presented in Table 3. The literature data cited in Table 3 did not use exactly the same contaminants as used in this study. In two cases, Medina (1992) and Apel (1994), the investigators used more complex contaminants (actual gasoline vapors).

Researchers' Name	Veenstra (1995)	Medina (1992)	Vaughn (1993)	Apel (1994)
Contaminants	Toluene Ethylbenzene o-Xylene	Gasoline Vapors	p-Xylene	Gasoline Vapors (in terms of: BTEX or TEX)
Packing Material(s)	Compost (Com.) DE	GAC	Glass Spheres(GS) DE	Compost
Applied Loading Rate (mg/cm ^{3.} day)	0.10-0.80 (Com.) 0.16-2.43 (DE)	0.07-3.11	0.55-1.29 (GS) 2.37-3.09 (DE)	0.67
Residence Time (minutes)	5-20 (Com.) 2.5-20 (DE)	5.9-7.0	5.0-10.0 (GS) 1.7-2.5 (DE)	3.4
Elimination Rate (mg/cm ^{3.} day)	0.10-0.36 (Com.) 0.16-2.39 (DE)	0.01-2.86	0.02-0.29 (GS) 0.19-1.63 (DE)	0.34-0.54
Elimination Efficiency (%)	44.1-100 (Com.) 99.7-100 (DE)	85-95	43-75 (GS) 73-88 (DE)	50-55 (BTEX) >80 (TEX)

Table 3. Comparison of Different Research Results

The analysis of the data in Table 3 shows only large trends. More specific matching of individual experimental points, such as similar residence times under similar loading conditions, is not

possible since the authors of the work cited in Table 3 do not give enough information in their papers to specifically determine under which conditions the elimination rate and elimination efficiency reported occur.

A review of the data presented in Table 3 points out some significant characteristics of the two filter media used in this work:

• The DE biofilter has the higher elimination efficiency (at least 99.7%) for total VOCs so that it can be used where a high degree of elimination is required.



Figure 27. Average Consumption Rates of Nutrients



- The DE biofilter has the larger elimination rate (up to 2.39 mg VOCs/cm³·day) at the lowest residence time (down to 2.5 minutes) therefore volume of both the biofilter, packing medium, is small, resulting in a low cost requirement.
- The DE biofilter has the highest applied loading (up to 2.43 mg VOCs/cm³·day) couple with a high elimination efficiency (99.7%), which should allow the biofilter to be used in cases of a high influent gas concentration of total VOCs.
- The DE biofilter is better than the compost biofilter in terms of allowable applied loading, elimination rate and elimination efficiency.
- A compost biofilter can still be used in some cases where the elimination efficiency requirement is low or there is no space limit on the biofilter.

SUMMARY

The major findings of this project, which investigated two biofilters in a lab-scale study, are as follows:

- The elimination of selected BTEX compounds by biofiltration from a contaminated air stream is a feasible and effective method. For the selected BTEX compounds, this technology can provided between 44.1 - 100% elimination efficiency in terms of total VOCs at an applied loading of 0.82 - 0.1 mg VOCs/cm³ compost day, and 99.7 - 100% at an applied loading of 0.82 - 0.1 mg VOCs/cm³ DE day, respectively.
- 2) The elimination rates of the selected BTEX compounds varied with an applied loading, residence time (or air flow rate), and influent gas concentration as well as a type of filter medium. The ranges of the elimination rates were 0.1 0.41 mg VOCs/m³/day for the compost biofilter and 0.16 2.39 mg VOCs/m³/day for the DE biofilter. This result shown that the DE medium is better than the compost medium in terms of elimination rates.
- 3) The main design criteria were gas influent concentration, residence time (or air flow rate) for a specific filter medium. In the compost column, the elimination efficiency varied from 44.1 -

100% with the residence times ranging from 5 -20 minutes, while the elimination efficiency was 98 - 100% with the residence times of 2.5 -20 minutes for the DE column.

- 4) In the compost biofilter, before the compost was saturated by contaminants, the total elimination capacity was attributed to both the physical adsorption and biological elimination. But the biological elimination capacity was about 95% in terms of mg VOCs/gm compost. Once the medium was saturated with selected BTEX compounds, since all the contaminants, attached to the compost medium, would finally be eliminated by microorganism, the maximum physical adsorption capacity was 0.33 mg VOCs/gm compost, and the total elimination capacity in this situation could still be attributed to the biological elimination.
- 5) It was difficult to maintain a constant gas influent to the columns using the apparatus that was used during this work, mainly due to the variability of the air source throughout the whole experimental period. But if a cylinder type of air source is available, it may improve the consistence of the influent concentration in the contaminated air stream.

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