



# Biofiltration of gasoline vapor by compost media

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**“Capsule”:** *A compost medium was effective in biofiltering gasoline vapor.*

## Abstract

Gasoline vapor was treated using a compost biofilter operated in upflow mode over 4 months. The gas velocity was 6 m/h, yielding an empty bed retention time (EBRT) of 10 min. Benzene, toluene, ethylbenzene and xylene (BTEX) and total petroleum hydrocarbon (TPH) removal efficiencies remained fairly stable approximately 15 days after biofilter start-up. The average removal efficiencies of TPH and BTEX were 80 and 85%, respectively, during 4 months of stable operation. Biodegradation portions of the treated TPH and BTEX were 60 and 64%, respectively. When the influent concentration of TPH was less than 7800 mg TPH/m<sup>3</sup>, approximately 50% of TPH in the gas stream was removed in the lower half of the biofilter. When the influent concentration of BTEX was less than 720 mg BTEX/m<sup>3</sup>, over 75% of BTEX in the gas stream was removed in the lower half of the biofilter. Benzene removal efficiency was the lowest among BTEX. A pressure drop could not be detected over a 1-m bed height at a gas velocity of 6 m/h after approximately 4 months of operation. Results demonstrated that BTEX in gasoline vapor could be treated effectively using a compost biofilter. © 2002 Elsevier Science Ltd. All rights reserved.

**Keywords:** Biofiltration; Gasoline vapor; BTEX; Biodegradation; Compost

## 1. Introduction

From the 1930s through the 1960s, approximately 2 million underground storage tanks were installed at gasoline stations, petroleum distribution points, industries and residencies in the United States. It is estimated that between 10 and 30% of the remaining tanks have leakage problems (Galaska et al., 1990). Contamination of one million gallons of groundwater by one gallon of gasoline renders water unsuitable for consumption (Galaska et al., 1990), hence, treatment of contaminated sites is critical to preserving human and environmental health.

Soil vapor extraction (SVE) is one possible method for treatment of sites contaminated with gasoline. As of August 1995, about 45% of the diverse innovative treatment technologies for superfund remedial actions in the United States were SVE technology (US EPA, 1995). Gases containing hazardous volatile organic compounds (VOCs) extracted during the SVE process

must be treated prior to being discharged to the atmosphere. Traditional VOC treatment technologies such as carbon adsorption, liquid scrubbing, condensation, thermal incineration, and catalytic incineration have been used to remove VOCs from waste gases. Even though the desired VOC removal efficiencies are achieved by these technologies, the technologies may suffer from high operating costs and secondary waste stream problems (Bohn, 1996; Lu et al., 2000; Deshusses, 1997a). Biofiltration is one possible VOC treatment method that may be more cost-effective than other technologies and minimize generation of secondary contaminated waste streams (Gribbins and Loehr, 1998; Deviny et al., 1995).

Most biofiltration research has been conducted with only gasoline total petroleum hydrocarbon (TPH) or only benzene, toluene, ethylbenzene and xylene (BTEX) as research parameters. Little research has been done on removal of BTEX in the presence of other gasoline compounds. Furthermore, removal mechanisms such as biodegradation or physicochemical degradation have not been identified. The objectives of this research were to examine the biofiltration of gasoline TPH, biofiltration of BTEX in gasoline, influence of biological activity

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on biofilter performance, and spatial gradients in TPH and BTEX concentration in the biofilter.

## 2. Materials and methods

### 2.1. Experimental setup

Biofiltration units consisted of a gasoline solution chamber, a vacuum pump (GAST Vacuum Pumps, DOAP104AA, USA), a humidifier, and a biofilter reactor (Fig. 1). This system was installed in a 20 °C incubator to maintain isothermal conditions. The main reactor was composed of four transparent acrylic columns with an inner diameter of 5 cm and height of 25 cm (total column height was 100 cm). Five gas-sampling ports were located 0, 25, 50, 75, and 100 cm from the bottom of the reactor. A valve at the bottom allowed condensate to be drained from the reactor.

### 2.2. Biofilter media

Media for the biofilter unit was compost produced at Nanji Composting Facility near the Nanjido landfill site in Korea. Physical and chemical characteristics of the media are provided in Table 1. The compost was sieved through a 2 mm sieve to remove large bulking agent and inert particles prior to biofiltration. The moisture content of biofilter media was adjusted to 60–80% of the water holding capacity and was approximately 50% on a dry weight basis. Each biofilter reactor was packed

with approximately 2.0 l of media with an estimated bulk density of 770 kg/m<sup>3</sup>.

### 2.3. Operational condition

Gasoline for the studies was obtained from a gas station near Konkuk University. Gasoline vapor vaporized by the vacuum pump (No. 2 in Fig. 1) was mixed with ambient air to control input vapor concentration. The input concentration was adjusted by the airflow control valve (No. 4 in Fig. 1). In fact, it was technically difficult to maintain uniform input concentration (Wright et al., 1997). The diluted vapor was passed through distilled water maintained at 30 °C. The relative humidity of the gas was maintained near 100% to prevent drying of the media. A constant flowrate of humidified, diluted gasoline vapor was injected into the biofilter reactor in an upflow mode. The gas flow rate was 0.2 l/min (6 m/h), yielding an empty bed retention time (EBRT) of 10 min. Biofiltration studies with and without HgCl<sub>2</sub> (biocide) addition were carried out for 1 and 4 months, respectively. Biocide was added at a concentration of 6000 mg/kg on a dry weight basis based on previous soil sterilization studies (Dooley et al., 1995; Frankenberger, 1992).

### 2.4. Gas sampling and analytical methods

Gas samples for analysis of VOCs were collected using charcoal tubes (SKC, Catalogue number 226–01). Vapor was passed through the tube at a constant flowrate using a handy sampler (Kimoto HS-6N, Japan).

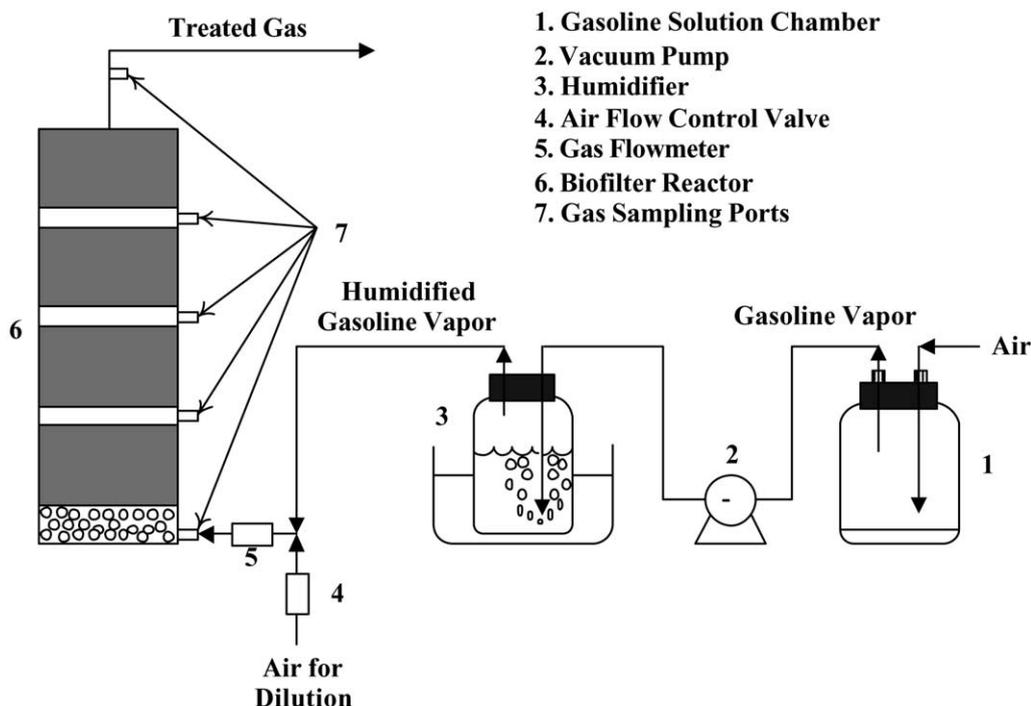


Fig. 1. Schematic of laboratory-scale biofiltration system.

Table 1  
Physical and chemical characteristics of the compost biofilter media

| Parameters                                     | Value |
|--|-------|
| Bulk density (kg/m <sup>3</sup> ) <sup>a</sup> | 770   |
| Moisture content (%) <sup>a</sup>              | 20    |
| Water holding capacity (%)                     | 73    |
| pH   | 8.8   |
| Uniformity coefficient                         | 5.0   |
| Volatile solids (%) <sup>b</sup>               | 67    |
| Total organic carbon (%) <sup>b</sup>          | 41    |
| <i>Chemical composition (%)<sup>b</sup></i>    |       |
| C  | 35.7  |
| H  | 4.4   |
| O <sup>c</sup>                                 | 22.8  |
| N  | 3.0   |
| S  | 1.0   |
| Ash  | 33.1  |
| <i>Heavy metals (mg/kg)<sup>b</sup></i>        |       |
| Pb   | 41.3  |
| Zn   | 95.3  |
| Cu   | 76.6  |
| Ni   | 10.6  |
| Cd   | 1.0   |

<sup>a</sup> Wet weight basis.

<sup>b</sup> Dry weight basis.

<sup>c</sup> O = 100 – (C + H + N + S) – Ash.

Each tube was packed with coconut activated carbon and consisted of a front part of 100 mg and a rear part of 50 mg. According to the manufacturer's specifications, VOC breakthrough in the front part of the tube occurs when the VOC concentration in the rear part exceeds 25% of the front part. Gas sampling time to avoid breakthrough was 1–3 min. Breakthrough was not observed for any of the samples analyzed. The VOCs trapped in the tube were extracted by soaking with 1 ml methylene chloride.

Extracted compounds were analyzed using a gas chromatograph (Hewlett Packard model 5890) equipped with an integrator (Hewlett Packard model 3395) and flame ionization detector. A stock standard solution for gasoline TPH was prepared from commercial gasoline fuel. TPH concentration was calculated by total peak area of gas chromatogram (Sironi and Botta, 2001). A Modified GRO Mix Standard Solution (Gasoline Range Organics, Catalogue No. 4-8167, Supelco, Inc.) was used as the BTEX standard. Methylene chloride was used for preparation of standards to eliminate any effect of solvent type on the chromatographic output. Standard solutions and sample extracts were allowed to reach room temperature before analysis because of the temperature sensitive characteristics of methylene chloride.

Dehydrogenase activity was used as an indicator of microbial activity during biofiltration. Dehydrogenase activity was determined by reduction of triphenyltetrazolium chloride (Cole et al., 1994; Hwang et al., 2001) and was measured in units of  $\mu\text{g}$  triphenylforma-

zan (TPF)/g. Compost samples collected at different heights in the biofilter were analyzed throughout the experiments.

Pressure drop across the biofilter was measured using a U-manometer. It was measured at the completion of the 4-month biofiltration studies.

### 3. Results and discussion

The removal efficiency of gasoline TPH during 4 months of biofilter operation is shown in Fig. 2. During the first 7 days of operation, the influent TPH concentration was approximately 120 mg/m<sup>3</sup> to allow for microbial acclimation. After that, influent TPH concentration ranged from 50 to 7800 mg/m<sup>3</sup> during 4 months of operation. A removal efficiency of approximately 80% was observed for TPH during the operation period. The TPH removal efficiency in this study was similar to efficiencies reported by other researchers (Table 2).

A similar trend was observed for the removal of BTEX. The influent concentration of BTEX ranged from 30 to 1600 mg/m<sup>3</sup> except on day 75 when the concentration was 3000 mg/m<sup>3</sup>. This exceptionally high concentration may have been due to an analytical error or experimental system failure. The average removal efficiency of BTEX was 85% during 4 months of operation, which is consistent with BTEX and BTX removal efficiencies reported by other researchers (Table 2).

The effect of biocide addition on biofilter removal of gasoline TPH is illustrated in Fig. 3. The average elimination fractions (elimination capacity/loading rate) for the compost biofilter with and without biocide addition were 0.8 and 0.2 of the loaded TPH, respectively. The dehydrogenase activity of samples from the biofilter

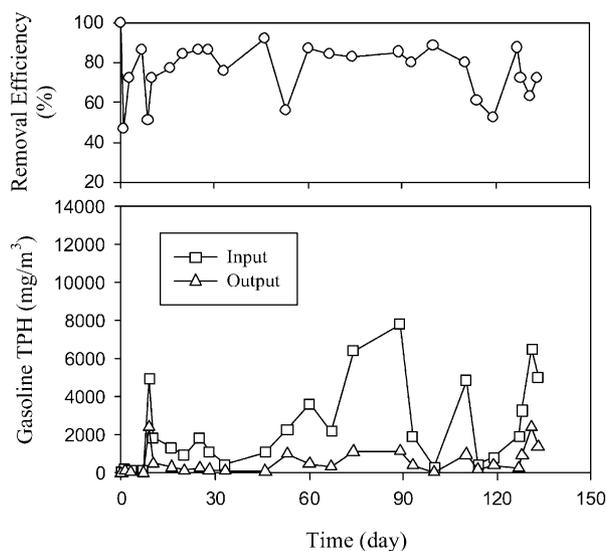


Fig. 2. Gasoline TPH influent and effluent concentrations and removal efficiency during a biofilter operation without biocide addition.

Table 2  
Maximum elimination capacity and removal efficiency of TPH and BTEX reported in the literature

| Contaminants | Biofilter medium | Critical loading rate <sup>a</sup><br>(g/m <sup>3</sup> /h) | Maximum elimination capacity (g/m <sup>3</sup> /h) | Average removal efficiency (%) | References                  |
|--------------|------------------|---|--|--------------------------------|-----------------------------|
| TPH          | GAC              | NA <sup>b</sup>   | 119  | 85–95                          | Medina et al., 1995         |
| TPH          | Compost          | 33  | 26   | > 98                           | Wright et al., 1997         |
| TPH          | Compost-based    | 55  | 34   | 82–98                          | Leson and Smith, 1997       |
| Benzene      | Compost-based    | 1   | 8  | NA                             | Johnson and Deshusses, 1997 |
| Toluene      | Compost-based    | 8   | 15   | NA                             | Johnson and Deshusses, 1997 |
| Toluene      | Compost          | 30–40   | 45–55  | NA                             | Seed and Corsi, 1994        |
| Toluene      | Compost          | NA  | NA   | > 98                           | Veir et al., 1996           |
| BTX          | Compost/perlite  | 35–40   | 50–60  | NA                             | Seed and Corsi, 1994        |
| BTX          | Compost          | NA  | NA   | 83–97                          | Ergas et al., 1995          |
| BTX          | Compost          | 162   | 108  | NA                             | Tahraoui and Rho, 1998      |
| BTEX         | Compost          | NA  | NA   | > 90                           | Wright, et al. 1997         |
| BTEX         | Compost          | NA  | NA   | > 80                           | Thompson et al., 1996       |

<sup>a</sup> Gas loading rate over which elimination capacity does not increase and remains constant.

<sup>b</sup> NA = Not Available.

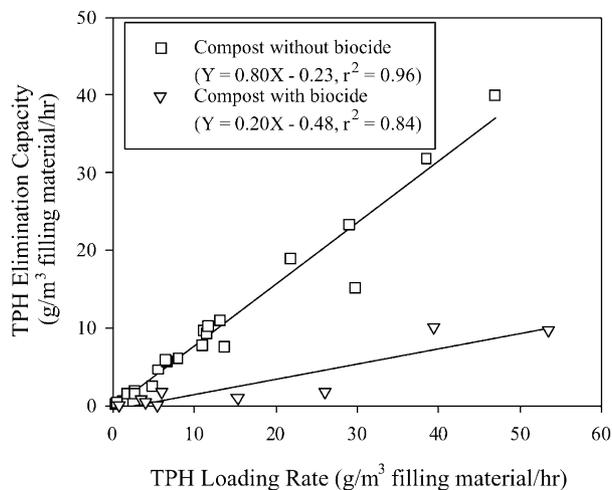


Fig. 3. Influence of biocide addition on the treatment of gasoline TPH by the compost biofilter.

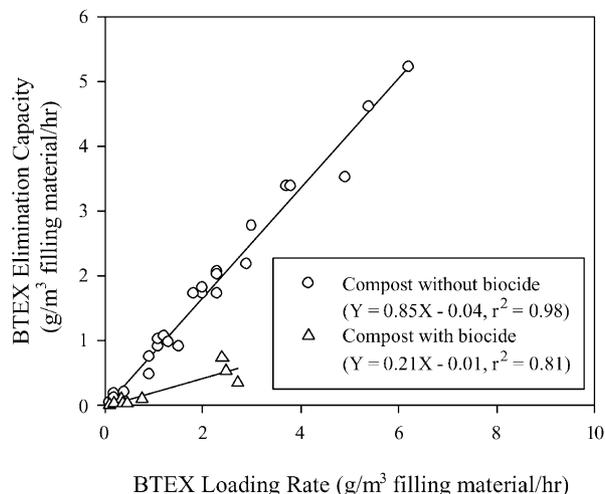


Fig. 4. Influence of biocide addition on the treatment of BTEX in gasoline by the compost biofilter.

with no biocide added varied between 2000 and 20,000  $\mu\text{g}$  TPF/g while samples from the biofilter that had biocide added never exceeded dehydrogenase activities of 250  $\mu\text{g}$  TPF/g (data not shown). These results suggest that the biocide addition significantly reduced biological activity during biofiltration and that 80% of the loaded TPH was removed by biological mechanisms.

The average elimination fractions of the biofilter for BTEX with and without biocide addition were 0.85 and 0.21, respectively (Fig. 4), hence the biodegradation portion of BTEX was 64%. Therefore, biological activity played a significant role in gasoline TPH and BTEX removal during biofiltration. Wright et al. (1997) reported that removal efficiencies of TPH and BTEX by biological reaction in a biofilter were 73 and 93%, respectively. These figures are higher than those obtained in this research and may be due to the use of cultured gasoline

degrading microbes and lower inlet concentrations for TPH and BTEX in the research conducted by Wright and co-workers. The maximum inlet concentrations for TPH and BTEX in their study were about 1.9 g TPH/m<sup>3</sup> and 0.37 g BTEX/m<sup>3</sup>, respectively.

The maximum elimination capacity of gasoline TPH observed in this research was 40 g TPH/m<sup>3</sup>/h at a loading rate of 50 g TPH/m<sup>3</sup>/h (Fig. 3). The maximum TPH elimination capacities reported for compost biofilters by other investigators were 26 and 34 g TPH/m<sup>3</sup>/h (Table 2). These differences might be due to the different types of compost or different loading rates used by other investigators.

The maximum elimination capacity of BTEX in the gasoline observed in this research was 5.3 g BTEX/m<sup>3</sup>/h at a loading rate of 6.2 g BTEX/m<sup>3</sup>/h. This was much lower than elimination capacities previously reported

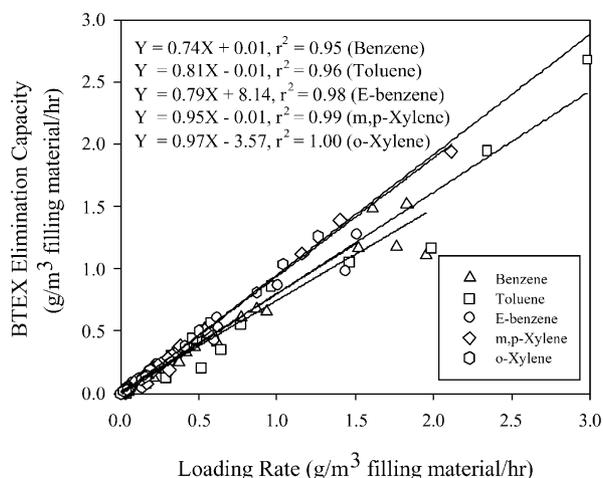


Fig. 5. Elimination capacity of benzene, toluene, ethylbenzene, and xylene in gasoline vapor during biofilter operation without biocide addition.

(Table 2). The difference may have been due to the presence of other compounds in the influent gas stream. Only BTX were used by Tahraoui and Rho (1998), while BTEX in the presence of gasoline were tested in this research. In general, the biofiltration of one organic compound is affected by the presence of other organic compounds (Deshusses et al., 1999). Hence, other compounds in the gasoline may have influenced the removal of BTEX in this study.

Linear relationships between biofilter loading rate and elimination capacity for benzene, toluene, ethylbenzene, and xylene are presented in Fig. 5. The slope of *o*-xylene was 0.97, which was the highest of the compounds tested, while the slope for benzene was the lowest (0.74). Mass transfer of contaminants from the air to the pore water of the biofilter media is a fundamental step for biofiltration (Devanny et al., 1999). Values of Henry's law constants for BTEX range from  $5.4 \times 10^{-3}$  to  $8.6 \times 10^{-3}$   $\text{m}^3 \text{ atm/mol}$  (Table 3), implying that these VOCs have relatively similar volatilities. However, the water solubility of benzene is over four times greater than the solubility of the other compounds. Considering only physical properties of these compounds, benzene is

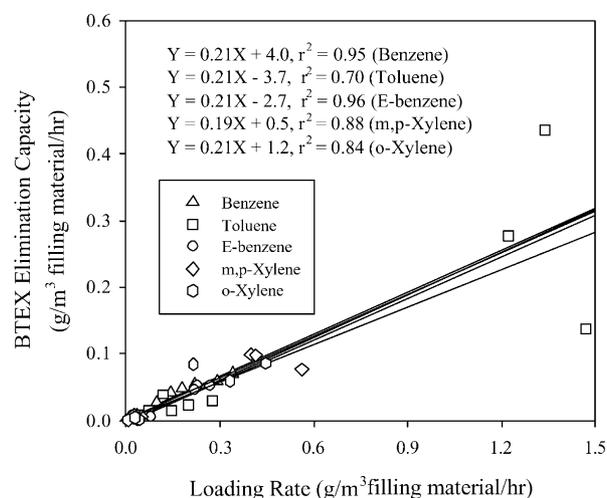


Fig. 6. Elimination capacity of benzene, toluene, ethylbenzene, and xylene in gasoline vapor during biofilter operation with biocide addition.

the most easily sorbable compound to the filter media. When the compost biofilter was treated with biocide, however, there was no significant difference in the removal of the individual BTEX compounds (Fig. 6) suggesting there was little difference in physical and chemical removal mechanisms among BTEX.

Webster et al. (1995) reported that the removal efficiency of benzene was lower than toluene during biofiltration using yard waste compost. Ortiz et al. (1998) also reported that benzene removal efficiency was the lowest among BTX contained in gasoline vapor. The threshold limit value for benzene, classified as a suspected human carcinogenic compound, is  $32 \text{ mg/m}^3$ , which is much lower than the other compounds (Table 3). Hence, the low removal efficiency of benzene by biofiltration may have been due to its high toxicity.

Fig. 7 shows the variations in gasoline TPH and BTEX concentrations with depth. As the experimental data indicate, approximately half of the influent TPH were removed in the lower half of the biofilter (Fig. 7a). When the influent concentration was less than  $720 \text{ mg BTEX/m}^3$ , 80–100% of the influent BTEX were removed in the lower half of the biofilter (Fig. 7b). This

Table 3  
Physical properties<sup>a</sup> and health risks of BTEX<sup>b</sup>

| Item         | Vapor pressure at 20 °C (atm)      | Water solubility at 20 °C (mol/m <sup>3</sup> ) | $K_H$ at 20 °C (m <sup>3</sup> atm/mol) | TLV–TWA <sup>c</sup> (mg/m <sup>3</sup> ) | TLV–STEL <sup>d</sup> (mg/m <sup>3</sup> ) |
|--------------|------------------------------------|---|---|---|--|
| Benzene      | $1.25 \times 10^{-2}$              | 22.79   | $5.4 \times 10^{-3}$                    | 32  | –  |
| Toluene      | $3.75 \times 10^{-2}$              | 5.60  | $6.6 \times 10^{-3}$                    | 188                                       | –  |
| Ethylbenzene | $1.25 \times 10^{-2}$              | 1.4   | $8.0 \times 10^{-3}$                    | 434                                       | 543  |
| Xylene       | $1.09\text{--}1.15 \times 10^{-1}$ | 1.50–2.07                                       | $5.5\text{--}7.1 \times 10^{-3}$        | 434                                       | 651  |

<sup>a</sup> Smallwood (1993).

<sup>b</sup> ACGIH (1993).

<sup>c</sup> Threshold limit value–time weighted average.

<sup>d</sup> Threshold limit value–short time exposure limit.

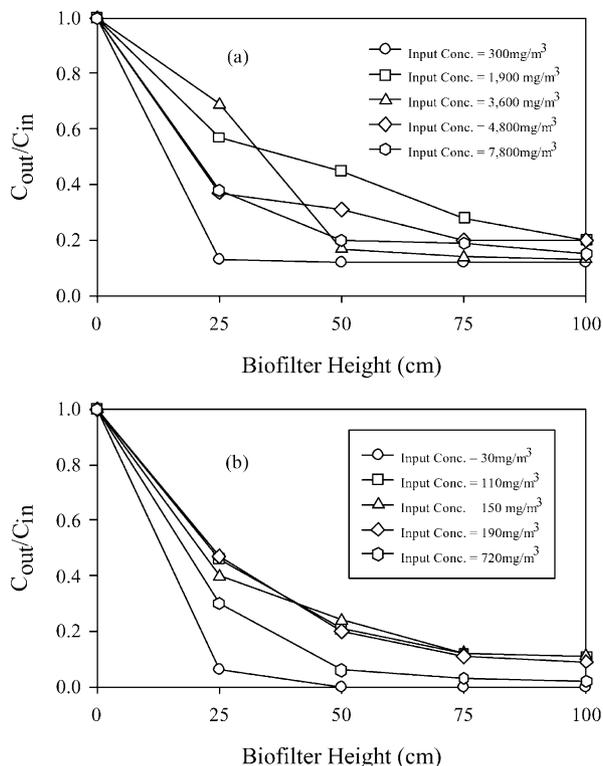


Fig. 7. Concentration profiles for (a) gasoline TPH, and (b) BTEX measured after 60 days of biofilter operation without biocide addition.

phenomenon may be attributed to a relatively higher microbial population in the lower part of the biofilter media.  $\text{CO}_2$  production and microbial protein levels were reported to be greater in the bottom portion of a biofilter than the upper portion (Morales et al., 1998; Vaughn et al., 1993), hence higher microbial activity levels may explain the greater removal of TPH and BTEX in the lower half of the biofilters used in these studies.

Pressure drop was not detected at a gas velocity of 6 m/hr after approximately four months of biofilter operation. Pressure drop was 60 mmH<sub>2</sub>O when velocity was increased to 15 m/h. An increase in pressure drop might cause a decrease in removal efficiency. Tang et al. (1997) stated that an increase in pressure drop was primarily due to bed compaction caused by the accumulation of the metabolic products and biomass, and compression of the filter material. Wright et al. (1997) reported that pressure drop of a compost biofilter was less than 10 mm H<sub>2</sub>O over a 1.8 m bed height after 250 days of operation. Pressure drop across a compost-based biofilter was also reported to be less than 50 mm H<sub>2</sub>O over a 1.5 m bed height (Deshusses, 1997b). Biofiltration of VOCs showed high removal efficiencies in these pressure drop ranges. Therefore, a pressure drop of 60 mm H<sub>2</sub>O over 1 m observed in this research should not cause significant operational problems.

#### 4. Conclusions

At the gas velocity of 6 m/h (EBRT = 10 min) and influent TPH and BTEX concentrations of less than 7800 and 1600 m<sup>3</sup>/h, respectively, the compost biofilter had an overall removal efficiency of 80% for TPH in gasoline vapor and 85% for BTEX in gasoline for an operating period of 4 months. Maximum elimination capacities observed in this research for TPH and BTEX were 40 g TPH/m<sup>3</sup>/h and 5.3 g BTEX/m<sup>3</sup>/h, respectively. Biodegradation portions of TPH and BTEX were 60 and 64%, respectively. Benzene removal efficiency was the lowest among BTEX. Half of the input TPH were removed in the lower half of the biofilter. When the influent BTEX concentration was less than 720 mg BTEX/m<sup>3</sup>, approximately 80% of BTEX was removed in the lower half of the filter. Negligible pressure drops were measured during this research. In summary, gasoline vapor including BTEX could be treated effectively by the compost biofilter for approximately 4 months without any operational problems.

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