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Optimum Operating Conditions for the Removal of Volatile Organic Compounds in a Compost-Packed Biofilter

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Abstract−Biofiltration was performed for 101 days in a compost-packed biofilter (I.D. 5.0 cm×height 62 cm) for the removal of nine volatile organic compounds (benzene, toluene, *m*-xylene, *o*-xylene, styrene, chloroform, trichloroethylene, isoprene, and dimethyl sulfide). Removal efficiency of the volatile organic compounds (VOCs) was dependent upon the column temperature, gas flow rate, and incoming concentrations of VOCs. At an empty bed residence time (EBRT) of 3 min and the incoming gas concentration of 66 g m^{-3} overall removal and efficiency increased up to 92.1 and 86.4% at 25 °C and 45 °C, respectively. Upon further increase of the incoming gas concentration to 83 g m^{-3}, the removal efficiency was 93.7% at 25 °C, but dropped to 73.1% at 45 °C. At incoming gas concentration of 92 g m^{-3} and EBRT of 1.5 min, the removal efficiency at 25 °C (91.6%) was comparable to 32 °C (95.5%). However, for 1 min of EBRT removal efficiency was better (86.6%) at 32 °C as compared to at 25 °C (73.6%). The maximum removal rates of VOCs were 3,561, 4,196, and 1,150 g m^{-3} h^{-1} at 25, 32, and 45 °C, respectively. At an EBRT of 1.5 min and 32 °C the removal efficiency of individual component was highest for toluene (98.9%) and *m*-xylene (97.6%), and lowest for TCE (86.1%) and chloroform (89.4%). Aromatic compounds (benzene, toluene, and xylene) were removed by 97.1-98.9%. After 101 days of operation profiles of pH and moisture content from the top to the bottom of the column were 7.2-6.3 and 53.8-67.2%, respectively, at 32 °C column, and 67% of the incoming VOCs was removed in the first quarter of the column. After 36 days of operation the cell concentration increased 108-fold from its initial value at 25 °C, and reached a maximum of 1.08×10^{8} cells·(g of dry compost)$^{-1}$.

Key words: Biofiltration, EBRT, VOCs, Removal Efficiency, Temperature

INTRODUCTION

Among volatile organic compounds (VOCs) emitted from industrial sites, benzene, chloroform, trichloroethylene, isoprene, toluene, styrene and *m*-xylene are known to be carcinogenic. They are also toxic to the liver and the kidneys, and paralyze the central nerve system even when exposed to low concentrations [Martin et al., 1998].

Removal of these VOCs by physical and chemical methods such as incineration, adsorption and ozonation is expensive and generates secondary pollutants [Markovska et al., 2001]. On the other hand, biological methods such as biofiltration are more economical, effective, and environmentally friendly processes that can degrade low level of VOCs and malodorous gases [Joseph et al., 1999]. In the early stage of development, biofiltration was mainly used to remove malodors from livestock and food processing wastes or gases from wastewater treatment plants. Recently, its application has been expanded to the removal of VOCs from petrochemical, chemical and solvent using processes [Corsi and Seed, 1995; Mattea and Ramsat, 1997; Sorial et al., 1997].

The basic concept of biofiltration is to remove the contaminants in the air by passing them through microbial layers established in a packed bed. Since microbes are grown on the packing material, a proper selection of the material is important to maintain a high performance of the biofilter. Commonly used packing materials are synthetic materials such as ceramics, activated carbon, and polystyrene beads as well as natural materials such as soil, peat, compost, and wood chips [Leson and Winer, 1991]. In the literature were reported degradation of VOCs using biofiltration [Leson and Winer, 1991; Mohseni and Allen, 2000; Ottengraf and Van, 1983] and the use of packing materials such as compost [Mattea and Ramsat, 1997], peat [Sorial et al., 1997] or a mixture of compost and perlite [Corsi and Seed, 1995].

Biofilter performance is affected by environmental factors such as moisture content, pH, and temperature. Operating parameters such as gas residence time, target compounds, and inlet concentrations of VOCs are important as well. However, comprehensive studies considering both environmental factors and operating conditions have not been well reported in the literature. Also the number of target compounds has been limited in the previous reports.

In this work we studied the effects of column temperature, gas residence time, and inlet concentrations on the removal of nine VOCs (benzene, toluene, *m*-xylene, *o*-xylene, styrene, chloroform, trichloroethylene, isoprene, and dimethyl sulfide) in a compost-packed biofilter.

MATERIALS AND METHODS

1. Inoculum and Cell Count

A sludge (200 g) was taken from the Jangrim river at Pusan, Korea, and mixed with 600 ml of sterilized distilled water by vor-

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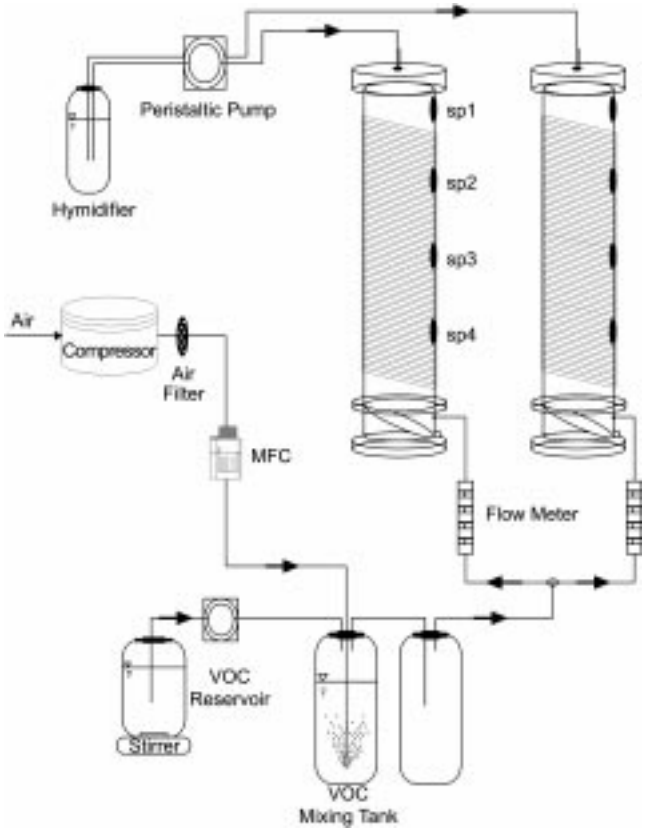

Fig. 1. Schematic diagram of the laboratory scale biofilter system (not to scale).
MFC: mass flow controller; sp: sampling port; VOC: volatile organic compound

texing for 3 min. After 30 min of settling, 40 ml (v/v) of the supernatant was taken by a sterilized syringe and inoculated into the compost of the column. The number of bacteria was determined by the Acridine Orange Direct Count (AODC) method. A formalin-treated (5% v/v) specimen was filtered through 0.2 μm polycarbonate membrane (Nuclepore Co., U.S.A). The membrane was examined under a fluorescence microscope (Olympus, BH-2, Japan) after staining by Acridin Orange, and the number of microorganisms was determined by counting 20 to 30 fields in a slide [Hobbie et al., 1977].

2. Biofilter Design and Experimental Set Up

The biofilter system consisted of two biofilter columns, a humidifier, a VOC reservoir, and VOC mixing tanks (Fig. 1). The biofilter column (1,020 cm^3 empty volume) was made of cylindrical glass column (I.D. 5.0 cm×L 62 cm), and packed with compost (Kellogg, Co., U.S.A.) up to 52 cm of height. A sieve plate (0.1 mm×0.1 mm) was placed at the bottom of the column to hold the compost. Four sampling ports sealed with rubber septa were located at 13, 26, 39 and 52 cm from the bottom of the column (sp1, sp2, sp3 and sp4 in Fig. 1, respectively). Membrane-filtered (0.2 μm pore size) air was sparged through VOC solution of the VOC mixing tank. The gas flow rate was maintained by a mass flow controller (Tyran Co., U.S.A.). The relationship among the feed rate of VOC solution to VOC mixing tank, gas flow rate, and the resulting concentrations of VOCs in the feed gas is shown in Table 1. The concentration of each VOC in the VOC reservoir was 200 mg l^{-1}.

Table 1. Generation of VOC mixtures of different concentrations

Feed rate of VOC solution to VOC mixing tank (ml h^{-1})	Air flow rate to the mixing tank (ml min^{-1})	VOC concentration in the feed gas (g m^{-3})
0.3	320	5
1.5	320	65
2.0	320	83
2.5	640	92
3.0	960	92

Variations of pH and moisture content in the column were minimized by supplying buffer solution (0.1 M Na_2CO_3) to the top of the column at a rate of 2 ml hr^{-1}. The moisture content was determined by drying the compost for 12 hrs at 105 °C [APHA, 1998]. Initial pH and moisture content of the compost were 7.1 and 61%, respectively. The columns were coiled with tygon tubings to control the temperature by circulating water from constant-temperature water baths. One column was operated at 25 °C throughout the 101 days of operation. The other column was operated at 45 °C from day 1 through day 79, and switched to 32 °C from day 80. Inlet concentration of VOCs was increased from 5 to 93 g m^{-3} in four steps (Fig. 2). Gas flow rate was increased in three steps (320, 640, and 960 ml min^{-1}), and corresponding empty bed residence time (EBRT) was 3, 1.5 and 1 min, respectively.

3. Analysis of VOC

The concentrations of VOC at the inlet, outlet and the four sampling ports of the column were determined by a gas chromatograph (HP 5890 series II, U.S.A.) equipped with flame ionization detec-

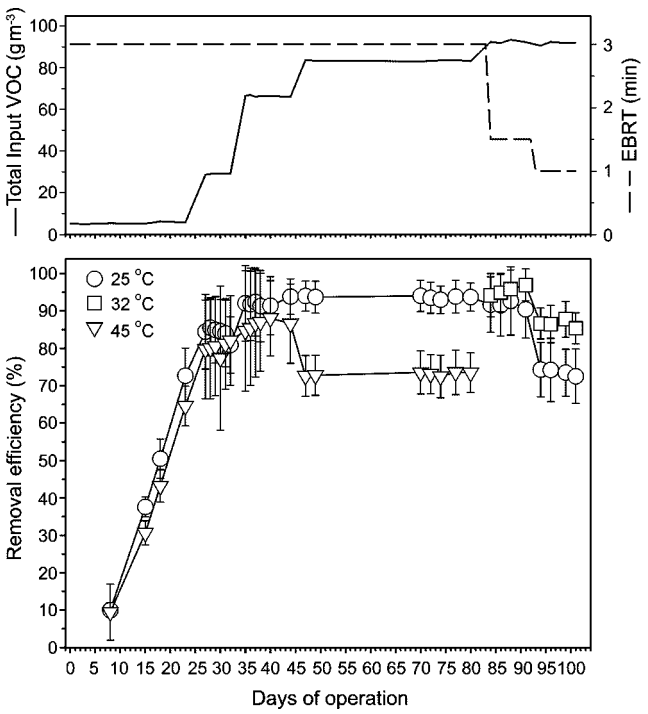

Fig. 2. Removal efficiency and total input VOC concentrations of the biofilter at different operation temperatures. EBRT: empty bed residence time.

tor (FID) and a 50 m-long Ultra-I capillary column (HP). The oven temperature was maintained at 32°C for 3 min and raised to 100 °C by 10 °C min^{-1}. Nitrogen (99.999%) was used as a carrier gas at a flow rate of 30 ml min^{-1}. One hundred μl of the sample was injected by using a 250 μl gas-tight syringe (Hamilton, U.S.A.). Retention time was used for material identification, and the amount of VOC was determined by peak area analysis using HP Chemstation program [Yoon and Park, 2002]. VOCs used in this study were reagent grade (Aldrich Chemical Co., U.S.A.) benzene, toluene, *m*-xylene, *o*-xylene, styrene, chloroform, trichloroethylene (TCE), isoprene, and dimethyl sulfide. Removal efficiencies of VOC were calculated as follows.

$$\text{Removal efficiency} = \frac{C_{in} - C_{out}}{C_{in}} \times 100\%$$

Where, C_{in}=incoming gas concentration, C_{out}=outgoing gas concentration.

RESULTS AND DISCUSSION

1. Overall Biofilter Performance

Biofiltration was performed for 101 days using variables such as gas residence time, inlet VOC concentration, and column temperatures (Fig. 2). During the 32 day initial period, removal efficiencies of VOCs for the 25 °C and 45 °C columns steadily increased up to 85.0% and 82.0%, respectively, when the inlet concentration of VOCs was increased from 5 to 30 g m^{-3} at an EBRT of 3 min. When inlet concentration of VOCs was raised to 66 g m^{-3} from 35th day, removal efficiency of the 25 °C column (92.1%) was better as compared to the 45 °C column (86.4%). At a higher inlet VOC concentration of 83 g m^{-3} removal efficiency of the 45 °C column dropped to 73.1%, while the 25 °C column maintained high removal efficiency (93.7%). This indicates that 45 °C can possibly be used as an operating temperature if the feed VOC concentration is below 66 g m^{-3}.

From this point the temperature of the 45 °C column was switched to 32 °C. When VOC input rate was increased further on the 84th day (inlet VOC concentration of 92 g m^{-3} and EBRT of 1.5 min), VOC removal efficiency of the 32 °C column (95.5%) was better as compared to the 25 °C column (91.6%). Upon further decrease in EBRT to 1 min at the same inlet VOC concentration, VOC removal efficiency for the 32 °C column (86.6%) was better as compared to 25 °C column (73.6%).

We showed that VOC removal efficiency was best at 32 °C, and lower at 45 °C as compared to 25 °C and 32 °C. This result was similar to the literature reports that the temperature range for good microbial activity was 20 to 40 °C [Leson and Winer, 1991; Yoon and Park, 2002], and that the optimum temperature was 30 to 35 °C, and near 30 °C for aromatic compounds (benzene, toluene, ethylbenzene, and *o*-xylene) supplied at 52 and 143 g m^{-3} h^{-1}, respectively [Lu et al., 1999].

2. EBRT and Inlet VOC Concentration

VOC removal rates were compared when EBRT was shortened stepwise from 3 min to 1.5 min and to 1 min, and when inlet VOC concentration was increased from 83 g m^{-3} to 92 g m^{-3}. At an EBRT of 3 min and inlet VOC concentration of 83 g m^{-3} VOC removal efficiency was 93.7% for the 25 °C column as compared to 73.1% for the 45 °C column. Removal efficiency for the 32 °C column (95.5%) was higher as compared to 25 °C column (91.6%) at an EBRT of 1.5 min and inlet VOC concentration of 92 g m^{-3}. Upon further decrease of EBRT to 1 min at the same inlet VOC concentration, it was clear that removal efficiency at 32 °C (86.6%) was much better than that of 25 °C (73.6%).

This work showed that gas residence time is one of the important operating variables, and optimum EBRT was 3 min for the 45 °C column (inlet VOC concentration of 66 g m^{-3}), and 1.5 min for the 25 °C and 32 °C columns (inlet VOC concentration of 92 g m^{-3}). It is presumed that this decrease of removal efficiencies for a shorter EBRT was due to limited microbial activity on the VOCs. An increase in VOC concentration is also known to decrease the removal efficiency [Wiggins and Alexander, 1988; Kim et al., 2002].

3. VOC Removal Rate

VOC removal rate per unit time and the unit volume of the biofilter was calculated from the difference between the inlet VOC concentration and outlet VOC concentration, residence time and biofilter volume (Fig. 3). At all three temperatures VOC removal rate increased along with VOC input rate. However, linear relationship held up to 1,251 g m^{-3} h^{-1} at 45 °C, and removal efficiency was 86.4%. Removal efficiency decreased to 73.1% at the input rate of 1,571 g m^{-3} h^{-1}. At 25 °C and 32 °C the removal rate was proportional to the input rate up to 3,486 g m^{-3} h^{-1} and removal efficiencies were 91.6% and 95.5%, respectively. Removal efficiencies decreased to 73.6% and 86.6%, respectively, at VOC input rate of 4,850 g m^{-3} h^{-1}.

VOC removal rate increased in proportion to VOC input rate probably because a higher VOC input rate increased the transfer of VOCs to the biofilm. However, when VOC input rate was higher than the critical value for each temperature, the removal rate could not reach the value predicted by a linear relationship. This result indicates that the biofilter had a limited VOC removal capacity specific at each temperature. The maximum removal rate was 3,561, 4,196,

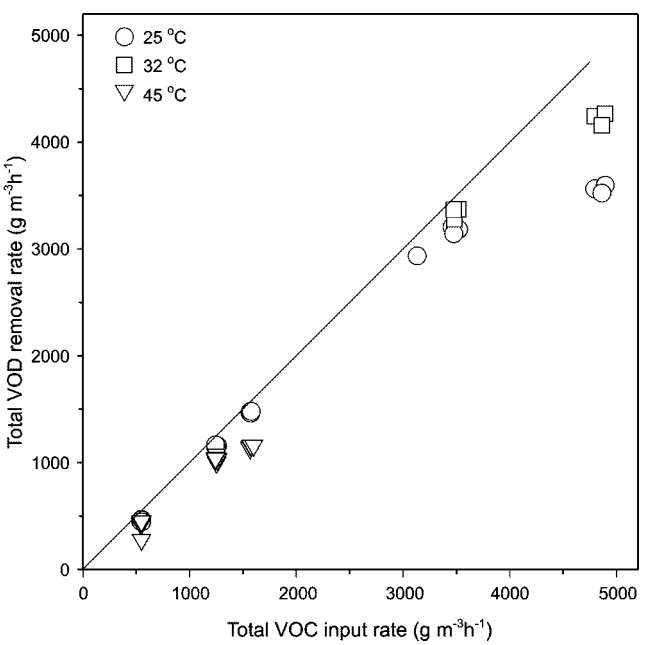

Fig. 3. Dependence of the VOC removal rate on the VOC input rate.

and 1,150 g m^{-3} h^{-1} at 25, 32 and 45 °C, respectively. The removal rate at 32 °C was 4,196 g m^{-3} h^{-1} at 1.5 min of EBRT and 92 g m^{-3} of VOC input concentration, which was one order of magnitude larger than the literature values [Jorio et al., 2000; Zill et al., 2000]. The maximum removal rate in this study was 4,196 g m^{-3} h^{-1}, which was 17 times higher as compared to the toluene removal rate (242 g m^{-3} h^{-1}) reported in the literature [Zilli et al., 2000]. Higher removal rate of our study can be explained if we compare the VOC inlet load and residence time. The inlet load of our study (92 g m^{-3}) was 23 times higher as compared to the literature value (4 g m^{-3}). EBRT of our study (1 min) was longer than the literature value (28 sec). Since our biofilter removed more than 90% of the incoming VOC, we can see that 17 times higher removal rate was obtainable.

4. Removal Efficiencies of Individual VOC Component

A mixture of aromatic and chlorinated compounds could be removed simultaneously. The removal efficiency of the aromatic and chlorinated compounds was 97.5% and 87.7%, respectively at 32 °C. Removal efficiency of each VOC was compared at 25 °C and 32 °C at the inlet VOC concentration of 92 g m^{-3} and the EBRT of 1.5 min (Fig. 4). Inlet concentration of each component in the VOC mixture was as follows (g m^{-3}): benzene 4.5, toluene 15, *m*-xylene 15, *o*-xylene 15, styrene 15, chloroform 7.5, TCE 10, isoprene 4.5, and DMS 5. For the 25 °C column, removal efficiency was the lowest (77.5%) for TCE, and highest for *m*-xylene (97.6%) and isoprene (97.1%). Aromatic compounds (benzene, toluene, and xylene) were removed by 92.0 to 97.6%. For the 32 °C column, removal efficiencies increased especially for chloroform (89.4%) and DMS (90.1%). Removal efficiencies of aromatic compounds also increased to 97.1% for benzene, 98.9% for toluene, 97.6% for *m*-xylene, 95.9% for *o*-xylene, and 96.2% for styrene. Higher removal efficiency for *m*-xylene as compared to *o*-xylene was consistent with the report of Jorio [2000] on xylene isomers. Our result at 25 °C for benzene, toluene, *m*-xylene was equal to that of Barker et al. [1987]. However, at 32 °C toluene showed the highest removal efficiency like the reports of Corsi and Seed [1995] and Lu et al. [1999].

In general, in our work the removal efficiencies were excellent for toluene, *m*-xylene, and isoprene. However, the removal efficiencies of TCE and chloroform were low. These results were the same as that of Todd et al. [1996] stating that aromatic compounds were degraded better as compared to chlorinated hydrocarbons. VOC degradation speed is known to be dependent upon chemical characteristics, cometabolism [Alexander, 1994; Cox et al., 1998], bioavailability of the compounds [Rasiah, 1992], and interaction of the substrates [Alvarez and Vogel, 1991]. In our work VOC degradation tendency was similar to the literature reports [Wiggins and Alexander, 1988; Cox et al., 1998] even though the number of target compounds was larger.

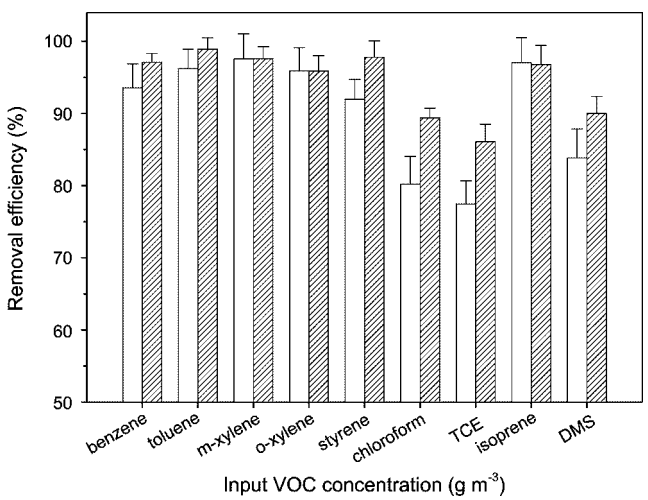

Fig. 4. Removal efficiency of individual VOC component at the inlet VOC concentration of 92 g/m³. Symbols: 25 °C (□), 32 °C (▨).

Table 2. Profiles of pH and moisture content in the columns

Height from the bottom (cm)	Days of operation	Moisture content (wt %)			pH		
		25 °C	32 °C	45 °C	25 °C	32 °C	45 °C
13	40	63.3	n.a.	65.5	7.1	n.a.	6.9
	80	63.3	n.a.	66.3	6.7	n.a.	6.5
	101	62.6	67.2	n.a.	6.5	6.3	n.a.
26	40	60.1	n.a.	61.4	6.9	n.a.	7.0
	80	62.2	n.a.	63.3	6.8	n.a.	6.7
	101	61.8	64.2	n.a.	6.9	6.5	n.a.
39	40	56.3	n.a.	57.4	7.0	n.a.	7.1
	80	58.2	n.a.	54.7	7.0	n.a.	6.8
	101	57.5	55.3	n.a.	7.0	6.6	n.a.
52	40	55.3	n.a.	55.5	7.1	n.a.	7.0
	80	53.2	n.a.	53.4	7.0	n.a.	6.9
	101	52.1	53.8	n.a.	7.2	6.7	n.a.

n.a. means not available.

5. Moisture Content and pH in the Column

Initial moisture content of the compost was 61% throughout the column. After 40 days of operation moisture content went up to 63.3% at the bottom (sp1) and decreased to 55.3% at the top (sp4) for the 25 °C column (Table 2). After 101 days of operation water content at the top and the bottom was changed to 52.1 and 62.6% for the 25 °C column and 53.8 and 67.2% for the 32 °C column, respectively. Even though the moisture content of the 32 °C column was relatively high (67.2%) after 101 days of operation, this value was within the optimum level (50-70%) reported in the literature [Ottengraf and Van, 1983].

pH was initially 7.1, but changed to 7.2 at the top (sp4) and 6.5 at the bottom (sp1) after 101 days in the 25 °C column (Table 2). pH was also high at the top and low at the bottom for the 32 °C column after 101 days and for 45 °C column after 80 days. pH difference between the top and the bottom was 0.4. For all the experimental conditions pH was 6.3 to 7.2, which was within the optimum pH (6-8) reported in the literature [Cox et al., 1993; Joseph et al., 1999; Oh et al., 1998].

Literature reported that it was important to maintain proper moisture content in the biofilter because low moisture content decreased microbial activity and VOC removal rate, and too much moisture content caused pressure loss, anaerobic environment, and a decrease in contact area between the VOCs and the packing material [Cox et al., 1993]. In this work moisture content could be maintained at a proper level by simply supplying the buffer solution for pH control. However, moisture content was higher at the lower part of the

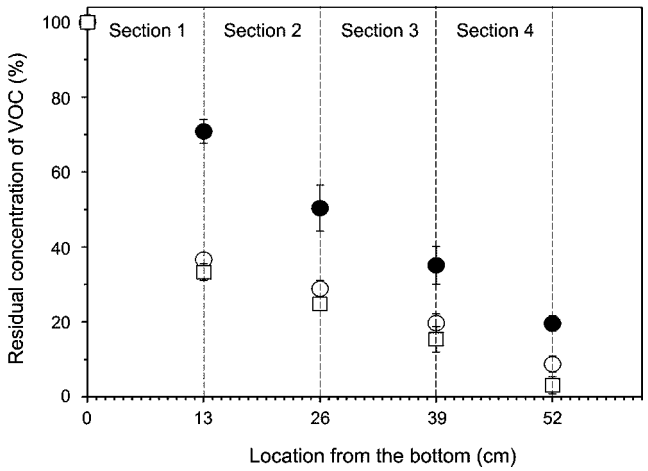

Fig. 5. Profiles of residual VOC (%) along the compost columns after 27 and 91 days of operation. Symbols: 25 °C, 27th day (●); 25 °C, 91st day (○); 32 °C, 91st day (□).

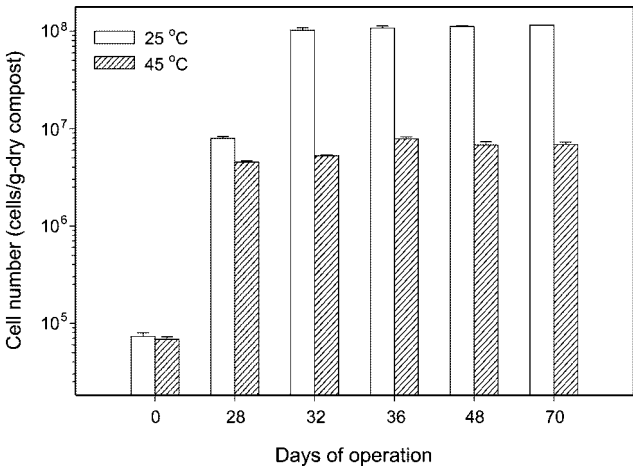

Fig. 6. Changes of the cell number in the compost columns.

column because of the gravitational force. Therefore, it is presumed that at the bottom part of the column microbial growth was more active, more acids were produced, and subsequently pH was lower.

6. Concentration Profiles of VOCs in the Column

VOC concentration profile in the 25 °C column was relatively linear along the column on the 27th day (EBRT 3 min). Twenty nine percent of VOC was removed in the first quarter of the column (0-13 cm), and the residual VOC concentrations were 50, 35, and 19% at 26, 39, and 52 cm, respectively (Fig. 5). VOC concentration profiles along the column supported the notion that microbial growth was more active at the bottom part of the column. On the 91st day the removal rate increased regardless of the location in the column, but VOC concentration profiles were exponentially decreasing because VOC removal at the inlet portion (0-13 cm) increased to 63% in the 25 °C column and 67% in the 32 °C column. This increase of VOC removal efficiency in the first section was probably due to enhanced microbial growth at higher moisture content. Residual VOC at 26, 39 and 52 cm locations were 29, 20 and 9%, respectively, for the 25 °C column, and 25, 15, and 3%, respectively, for the 32 °C column Our results were similar to the reports on VOC profiles in the literature [Zill and Converti, 2000; Cox et al., 1998; Ergas et al., 1994].

7. Changes in Cell Counts

Initially, the number of microorganisms in the 25 °C column (7.32×10^4 cells/g-dry compost) and in the 45 °C column (6.83×10^4 cells/g-dry compost) were similar. On the 28th day the cell number increased by 108 times (7.94×10^6 cells/g-dry compost) for the 25 °C column, and 66 times (4.53×10^6 cells/g-dry compost) for the 45 °C column. After 36 days of operation cell number reached a maximum of 1.08×10^8 cells/g-dry compost (Fig. 6). This cell number was 1,475 fold increase from its initial value, and 13.8 times higher as compared to the 45 °C column (7.85×10^6 cells/g-dry compost). Up to 36th day microbial growth (Fig. 6) and the increase of the removal efficiency (Fig. 2) showed the same trend. Thus, it is presumed that the increase in the removal rate was due to the increased number of microorganisms. From that time on the number of microorganisms remained relatively constant for both 25 °C and 45 °C columns. When the inlet VOC concentration was 66 g m^{-3} (for example, on the 36th day), the number of microorganism in the 25 °C column was 13.8 times larger as compared to the 45 °C column. At the inlet VOC concentration of 83 g m^{-3} (for example on the 70th day) the number of microorganisms at the two temperatures was similar. However, VOC removal efficiency increased from 92.1% to 93.7% for the 25 °C column, whereas the efficiency decreased from 86.4% to 73.1% for the 45 °C column. This was possibly due to higher substrate inhibition at 45 °C or a decrease in cell viability upon extended exposure to 45 °C.

CONCLUSION

The biofilter performance was best at 32 °C, and VOC removal rate (4, 196 g m^{-3} h^{-1}) was one order of magnitude larger than the literature values.

Our work also showed that aromatic and chlorinated compounds in a VOC mixture could be removed simultaneously at 97.5% and 87.7%, respectively.

Comparing the four sections of the column, VOC removal was most active at the bottom part of the column presumably because higher moisture content and higher VOC concentration promoted microbial activity. Accordingly, the pH was lowest at the bottom part of the column.

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