ORIGINAL ARTICLE

Biofiltration of air contaminated with methanol and toluene

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Abstract

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Biofiltration of air contaminated with methanol and toluene
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Biofiltration of air contaminated with VOCs is inexpensive compared with the conventional techniques and very effective for treating large volumes of moist air streams with low concentrations of VOCs. In this study, biofiltration for the purification of polluted air from methanol, a hydrophilic VOC, and toluene, a hydrophobic VOC, was investigated. The experiments were operated using three separated stainless steel biofilters, for methanol, toluene, and a mixture of methanol and toluene, respectively. Biofilter consisted of a mixture of palm shells and activated sludge as a filter-bed material. Only the indigenous microorganisms of the bed medium without any addition of extra inoculum were used throughout the whole process. The polluted air inlet concentration was varied from 0.3-4.7 g/m^3 with flow rates ranging from 0.06-0.45 m^3/h, equivalent to the empty bed residence times of 9-71 sec. Polluted air was successfully treated by biofiltration, 100% removal efficiencies would be obtained when the air flow rate was lower than 0.45 m^3/h. The presence of toluene did not affect the removal rate of methanol while the removal rate of toluene was decreased with the presence of methanol in air stream according to the competition phenomenon.

Key words : biofiltration, biofilter, VOC, methanol, toluene

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บทคัดย่อ

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การกรองชีวภาพของอากาศปนเปื้อนด้วยเมทานอลและโทลูอีน

ว. สงขลานครินทร์ วทท. ธ.ค. 2548 27(ฉบับพิเศษ 3) : 761-773

การกรองชีวภาพของอากาศปนเปื้อนด้วยสารอินทรีย์ระเหยเป็นกระบวนการที่ไม่แพงเมื่อเปรียบเทียบกับกระบวนการที่ใช้โดยทั่วไป และมีประสิทธิภาพสูงในการบำบัดอากาศชื้นปริมาณมากที่ปนเปื้อนด้วยสารอินทรีย์ระเหยความเข้มข้นต่ำ ในการศึกษานี้ ได้ทำการค้นคว้าทดลองการกรองชีวภาพสำหรับทำความสะอาดอากาศปนเปื้อนด้วยเมทานอลซึ่งเป็นสารไฮโดรฟิลิคและโทลูอีนซึ่งเป็นสารไฮโดรโฟบิค การทดลองดำเนินการโดยใช้เครื่องกรองทางชีวภาพ 3 คอลัมน์ที่สร้างจากเหล็กกล้าปลอดสนิม คอลัมน์หนึ่งสำหรับบำบัดเมทานอล คอลัมน์หนึ่งสำหรับบำบัดโทลูอีน และอีกคอลัมน์สำหรับบำบัดของผสมเมทานอลและโทลูอีน คอลัมน์บรรจุด้วยกะลาปาล์มผสมกับตะกอนเร่งสำหรับใช้เป็นตัวกรองโดยไม่มีการเติมเชื้อเพิ่มเติม ใช้เฉพาะจุลินทรีย์ที่มีอยู่ในตัวกรองตลอดกระบวนการ อากาศปนเปื้อนสารอินทรีย์ระเหยความเข้มข้นในช่วง 0.3 -4.7 กรัม/ลบ.เมตร อัตราการไหลในช่วง 0.06-0.45 ลบ.เมตร/ชั่วโมง ซึ่งเท่ากับเวลาที่อากาศอยู่ในระบบ 9-71 วินาที ระบบกรองทางชีวภาพสามารถบำบัดอากาศปนเปื้อนได้ดีที่อัตราการไหลของอากาศต่ำกว่า 0.45 ลบ.เมตร/ชั่วโมง สามารถบำบัดได้ 100% โทลูอีนไม่มีผลต่อการบำบัดเมทานอล ในขณะที่อัตราการกำจัดโทลูอีนจะลดลงเมื่อมีเมทานอลอยู่ในอากาศด้วย ทั้งนี้เนื่องจากปรากฏการณ์การแข่งขันระหว่างสารประกอบทั้งสอง

ภาควิชาวิศวกรรมเคมี คณะวิศวกรรมศาสตร์ มหาวิทยาลัยสงขลานครินทร์ อำเภอหาดใหญ่ จังหวัดสงขลา 90112

Industrial plants and processes use and emit various types of volatile organic compounds (VOCs), which consequently become atmospheric pollutants. Methanol is a hydrophilic VOC (with a water solubility of 1,000 g/l at 25ºC) while toluene is a hydrophobic VOC (with a water solubility of 0.53 g/l at 25ºC). They both are hazardous air pollutants (HAPs) listed in Title III of the 1990 Clean Air Act Amendments (CAAA90) proposed by the US Environmental Protection Agency (EPA) (Spicer *et al*., 2002).

The presence of VOCs in air emissions has been subjected to the recent environmental regulations of which the industry is required to apply an appropriate technology to reduce such emissions. The current control technologies for VOCs (e.g. thermal incineration, wet scrubbing, and adsorption onto activated carbon) are costly, especially in cases where there are low concentrations of the pollutants (Mohseni and Allen, 2000).

Biofiltration of air containing VOCs is a relatively new application of bioprocess engineering in waste management. It applies microorganisms, attached to the porous support media, for breakdown of VOCs. This approach has a potential application to a number of industries. Biofiltration is inexpensive compared with the conventional techniques and very effective for treating large volumes of moist air streams with low concentrations of the biodegradable pollutants. In addition, the treatment is environmental friendly, as the treatment could be performed at an ambient temperature and it does not generate nitrogen oxides or secondary waste streams. Pollutants are generally converted to carbon dioxide under the action of growing or resting microorganisms (Deshusses, 1997).

Generally, a biofilter is a column filled with the porous and humid packing material and indigenous microorganisms or inoculated microorganisms that are able to degrade pollutants to the filter bed media. Biofiltration may be represented as a three-phase system that consists of (i) a gas phase which flows through (ii) a water-insoluble solid support which contains (iii) an aqueous static

phase where the soluble substrates (pollutants, oxygen) are dissolved and microorganisms grow. In this heterogeneous medium, complex heat (convective, evaporative, and conductive) and mass transport (diffusion, convection) associated with oxidation of the pollutant occur (Van Lith *et al.*, 1997).

Any porous material capable of adsorbing gaseous compounds and supporting biological growth can possibly be used as a packing material. The packing materials commonly used include natural materials such as peat, compost, soil, and sludge from sewage treatment plants and synthetic materials such as vermiculite, granular activated carbon, and extruded diatomaceous earth pellets (Aizpuru *et al.*, 2003a and Aizpuru *et al.*, 2003b).

The degradation of VOCs by micro-organisms is affected by various environmental factors such as moisture content, temperature, pH, VOC input rate, the kind of contaminant, and accessibility to the target substances (Yoon and Park, 2002). The effectiveness of the biofilter principally depends upon the solubility of the compounds in the liquid layer of the biofilm (Shareefdeen and Baltzis, 1994). The hydrophilic and hydrophobic characteristics of the pollutants discharged in air emissions may significantly influence their removal capacities in biofilters. Some researchers have studied the biofiltration of pure methanol (Mohseni and Allen, 2000; Shareefdeen *et al.*, 1993; Lee *et al.*, 1996) and pure toluene (Delhomenie *et al.*, 2002; Auria *et al.*, 2000; Acuna *et al.*, 1999; Morales *et al.*, 1998). However, no study has tried to examine the biofiltration of a mixture of methanol and toluene and to understand the interactions between the two compounds.

In this work, the feasibility of the biofilters consisted of a mixture of palm shells and activated sludge as a filter bed medium to treat air contaminated with methanol, toluene, and a mixture of methanol and toluene were studied. The effects of operating conditions, such as VOC input concentration, empty bed residence time, height of the column, and pressure drop on the treatment were

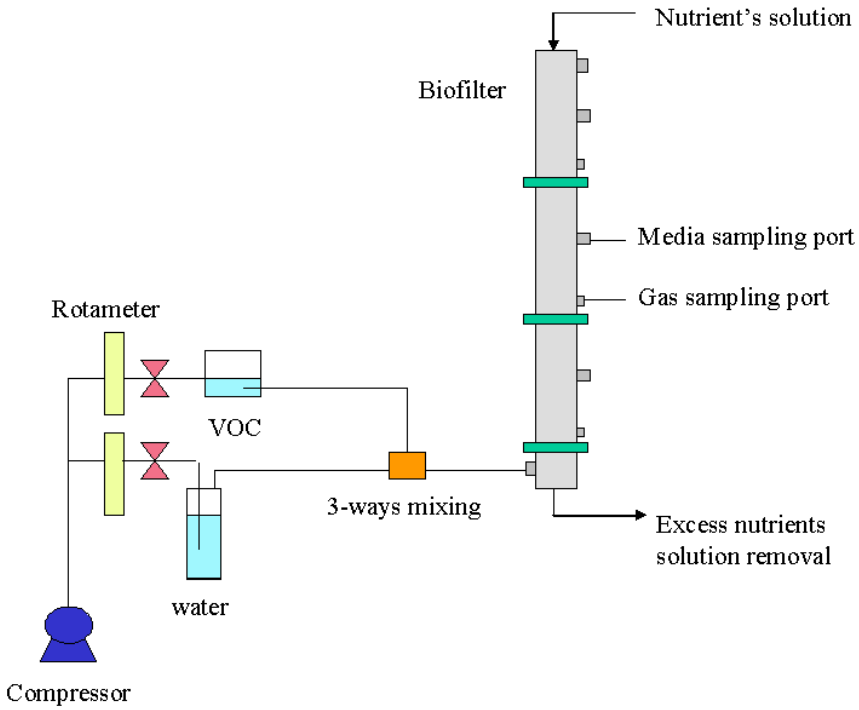

Figure 1. The experimental setup of the biofiltration system.

investigated. After 113 days of operation the optimum conditions were determined.

Materials and Methods

Volatile organic compounds

The VOCs studied were methanol (99.8%) and toluene (99.5%) obtained from Merck, Germany.

Equipments

Three separated identical bench-scale biofilters were used to treat air contaminated with VOCs, for methanol, toluene, and a mixture of methanol and toluene. The biofilters were made of stainless steel and each consists of three equal segments connected in series (Figure1). Each segment has a diameter of 5 cm and a height of 30 cm (being filled to a height of 20 cm with equal amounts of the prepared filter-bed material). In order to support the filter-bed and to ensure homogeneous radial distribution of the input gas, a stainless steel mesh was installed at the base of each section. These supports were reinforced with stainless steel rods in order to bear the weight of the wet filter material. Two ports were placed in each segment, one for gas sampling and another one for media sampling.

The biofilter was fed by airflow provided by a continuous compressed air source. The major portion of the air was passed through a water column in order to become fully saturated. A secondary fraction of the main air was directed to a bubbler unit containing the liquid VOC reagent. The previous separate gas flows were then mixed together and the resultant polluted humid input gas mixture was carried to the base of the biofilter.

Nutrient solution

The nutrient solution was periodically distributed over the bed upper-surface to maintain an adequate level of bed filling moisture content and to provide those nutrients necessary for the growth of microorganisms present in the biofilter. The composition of nutrient solution used is shown in Table 1.

Filter material

The biofilter media were a mixture of palm shells (0.5-1cm) and activated sludge (Kingfisher Holdings Ltd.) in the proportion of 1:2 by volume. A pH buffer ($CaCO_3$) was added to the filter media when necessary. The media were kept for one night before packing in order to prevent the expansion of palm shells in the biofilter.

Analytical methods

Gas samples were taken by 100% polypropylene bags (0.5 liter) at the different outlets of the filters. VOC concentration was analyzed by a gas chromatograph unit (HP 6890, Hewlett Packard) equipped with a flame ionization detector (FID) using a 30-m capillary column (HP-1, crosslinked methyl siloxane). For methanol and toluene measurements, the temperatures of the injection port, the oven, and the detector were maintained at 180, 70, and 200ºC, respectively. The flow rates of air and hydrogen for FID were 400 and 30 ml/min, respectively.

Gas pressure drop of the filter was measured by a U-tube manometer. Bed temperature and relative humidity were monitored via AP-104 (Sila Research Co., Ltd., Thailand) while pH of the filter media was measured by a pH indicator paper (Merck, Germany).

Samples for scanning electron microscopy (SEM) observation were fixed in 25% glutaraldehyde ($C_5H_8O_2$) in phosphate buffer for 1.5 h,

Table 1. Composition of one liter of the nutrient solution.

Composition	Amount
KH_2PO_4	0.91 g
$Na_2HPO_4.12H_2O$	2.39 g
KNO_3	2.96 g
$(NH_4)_2SO_4$	1.97 g
$NaHCO_3$	1.5 g
$FeSO_4.7H_2O$	0.2 mg
$MgSO_4.7H_2O$	2 mg
$MnSO_4.H_2O$	0.88 mg
$Na_2MoO_4.2H_2O$	1 mg
$CaCl_2$	3 mg

washed twice with the same phosphate buffer, and postfixed in phosphate buffer containing 1% osmium tetraoxide (OsO_4) for 1.5 hours. Subsequently, the samples were washed twice with the distilled water prior to dehydration in a graded water-ethanol (50-100%) for 15 min in each step. The samples were finally dried in a critical point dryer and gold coated by a sputter coater. Examination was carried out in a digital scanning electron microscope (JSM-5800LV, JEOL).

Operating conditions

To describe the mechanisms of biofiltration clearly, general terminology pertinent to the field should be well defined. Studies were performed on the level of the VOC inlet load (IL) and empty bed residence time (EBRT) while the pollutant degradation performance of the biofilter can be expressed in terms of the pollutant removal efficiency (RE) and the elimination capacity (EC). The definitions for these four parameters are given below:

$$IL = \frac{Q \times C_I}{V} \quad (1)$$

$$EBRT = \frac{V}{Q} \quad (2)$$

$$RE = \left(1 - \frac{C_0}{C_I}\right) \times 100 \quad (3)$$

$$EC = \frac{Q \times \left(C_I - C_0\right)}{V} \quad (4)$$

with C_I = VOC concentration at inlet (g/m^3), C_0 = VOC concentration at outlet (g/m^3), Q = volumetric gas flow rate (m^3/h), V = filter bed volume (m^3). All of these parameters were studied in accordance with the operating conditions, as summarized in Table 2.

Results and Discussion

Overall performance

For treatment of pure VOC, the whole experimental period (113 days) was split into six successive stages, i.e. A, B, C, D, E, and F. The results of methanol removal are presented in Figure 2. During stage A (air flow rate = 0.06 m^3/h), which was a start up period, the removal efficiency (RE) of the biofilter accounted for 100% on the first day of operation. The high value of 100% was due to the sorption of initial methanol on the wet filter material, regardless of the activity by microorganisms. Then the removal efficiency increased due to biodegradation and the steady state was reached 37 days after the start of the experiment. For stages B, C, D, E, and F, the air flow rate was maintained at 0.06, 0.12, 0.18, 0.24, and 0.45 m^3/h, respectively. The experiment for removal of toluene was operated in the same way and the steady state was reached 18 days after the start of the experiment (Figure 3). A shorter start up time for removal of toluene may be attributed to the inlet load of toluene being maintained lower than the inlet load of methanol during stage A.

Figure 2, along with Table 3, shows that during stages B, C, D and E the removal efficiencies of methanol were mostly maintained at 100%. In stage F (air flow rate = 0.45 m^3/h), the removal efficiency started to level off (<100%). The removal efficiency was highly influenced by the inlet load; this will be discussed further in the section "Influence of inlet concentration of VOC". For removal of toluene, 100% removal was also observed on the first day of operation and the removal efficiencies were almost maintained at 100% during stages B and C. Figure 3 and Table 4 show that during stages D and E, 100% removal can be obtained with the average inlet load lower than 160 and 148 g/m^3h, respectively. The toluene

Table 2. Biofilter operating conditions.

Filter media	Palm shell + activated sludge
Pollutant	Methanol and toluene
Microorganisms	Indigenous to filter media
Diameter of palm shell	0.5-1 cm
Bed height	3×20 cm
Column diameter	5 cm
Inlet concentration	0.3-4.7 g/m^3
Airflow rate	0.06-0.45 m^3/h
EBRT	9-71 s

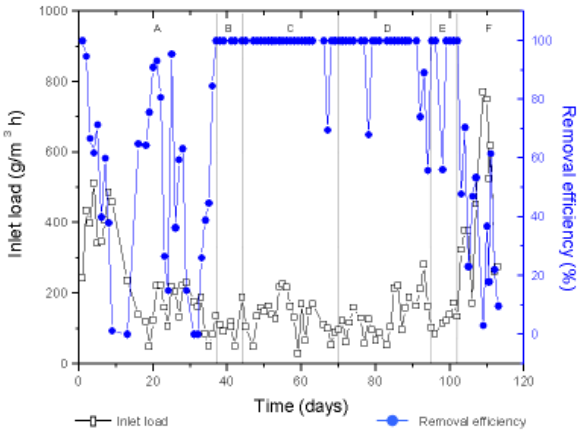

Figure 2. Overall performance of the biofilter for the removal of methanol from air contaminated with pure methanol.

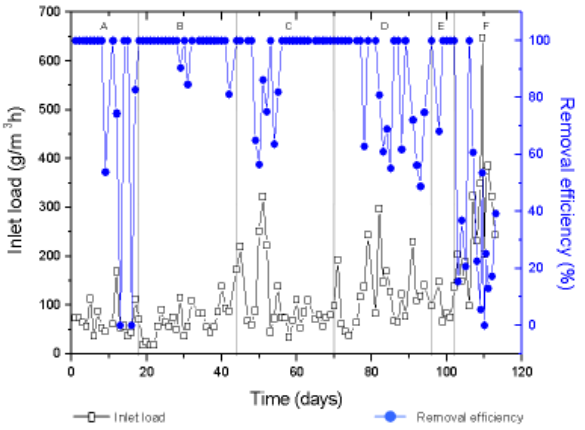

Figure 3. Overall performance of the biofilter for the removal of toluene from air contaminated with pure toluene.

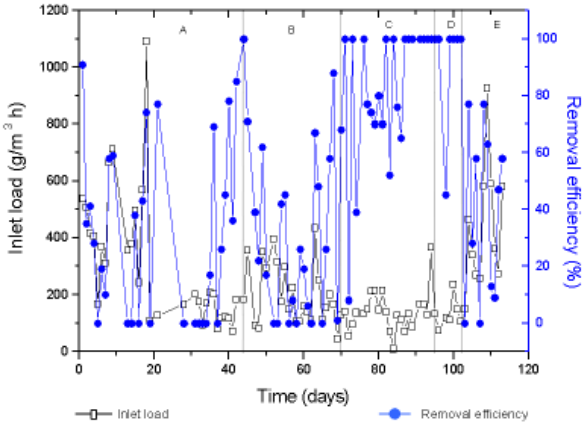

Figure 4. Overall performance of the biofilter for the removal of methanol from air contaminated with a mixture of methanol and toluene.

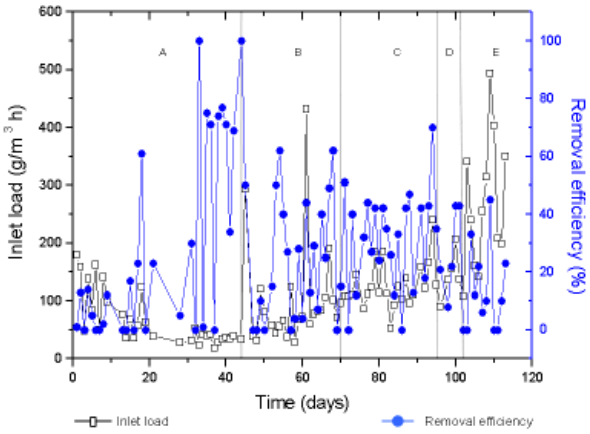

Figure 5. Overall performance of the biofilter for the removal of toluene from air contaminated with a mixture of methanol and toluene.

removal efficiency also decreased during stage F as a result of high inlet load.

Treatment of a mixture of methanol and toluene can be split into five successive stages, i.e. A (0.06 m^3/h), B (0.12 m^3/h), C (0.18 m^3/h), D (0.24 m^3/h), and E (0.45 m^3/h). For methanol treatment (Figure 4), the removal efficiencies varied during stages A and B corresponding to the acclimation period of the microbial population and the influence of the inlet load. The steady state was reached on day 71. This indicates that the start up period for treatment of a mixture of methanol and toluene was longer than the start up periods for treatment of pure methanol and pure toluene. Removal efficiencies as high as 100% could be obtained during stages C and D while the maximum removal efficiency of 77% was obtained during stage E. For removal of toluene (Figure 5), similar results

Table 3. Examples of the steady state results for the removal of methanol.

Day	Inlet conc. (g/m^3)	Air flow rate (m^3/h)	EBRT (S)	Outlet conc. (g/m^3)	RE (%)	EC (g/m^3h)
37	2.7	0.06	71	0	100	136
44	1.8	0.12	35	0	100	188
70	0.6	0.18	24	0	100	96
95	0.5	0.24	18	0	100	101
102	0.3	0.45	9	0	100	134
111	1.6	0.45	9	0.6	62	380
113	0.7	0.45	9	0.6	14	38

Table 4. Examples of the steady state results for the removal of toluene.

Day	Inlet conc. (g/m^3)	Air flow rate (m^3/h)	EBRT (S)	Outlet conc. (g/m^3)	RE (%)	EC (g/m^3h)
18	1.4	0.06	71	0	100	70
44	1.7	0.12	35	0	100	172
70	0.6	0.18	24	0	100	98
95	0.5	0.24	18	0	100	98
102	0.4	0.45	9	0	100	137
109	1.7	0.45	9	0.8	53	346
113	0.6	0.45	9	0.4	39	96

Table 5. Performance comparison between this work and other biofiltration studies.

Study	Contaminant	Biofilter medium	EC_{crit} (g/m^3h)	EC_{max} (g/m^3h)
Shareefdeen *et al*., 1993	Methanol	Compost/perite	50-80	100-120
Lee *et al*., 1996	Methanol	Compost/perite	10-20	301
Briggs, 1996	Methanol	Compost-based	42	N/A
Johnson and Deshusses, 1997	Methanol	Compost-based	30-35	70
Seed and Corsi, 1994	Toluene	Compost	30-40	45-55
Auria *et al*., 1996	Toluene	Peat	N/A	4-40
Johnson and Deshusses, 1997	Toluene	Compost-based	8	15
This study	Methanol[p]	A mixture of palm	90	230
	Toluene[p]	shells and	50	181
	Methanol[m]	activated sludge	90	309
	Toluene[m]		N/A	90

p = pure component in air stream
m = a mixture of methanol and toluene in air stream

were obtained, except that the removal efficiencies were lower. The maximum removal efficiency was 70% at stage C, 43% at stage D, and 45% at stage E.

However, the removal efficiency is an incomplete descriptor of biofilter performance because it varies with contaminant concentration, air flow, and biofilter size and only reflects the specific conditions under which it is measured. The elimination capacity allows for direct comparison of the results of different biofilter systems because the volume and flow are normalized by

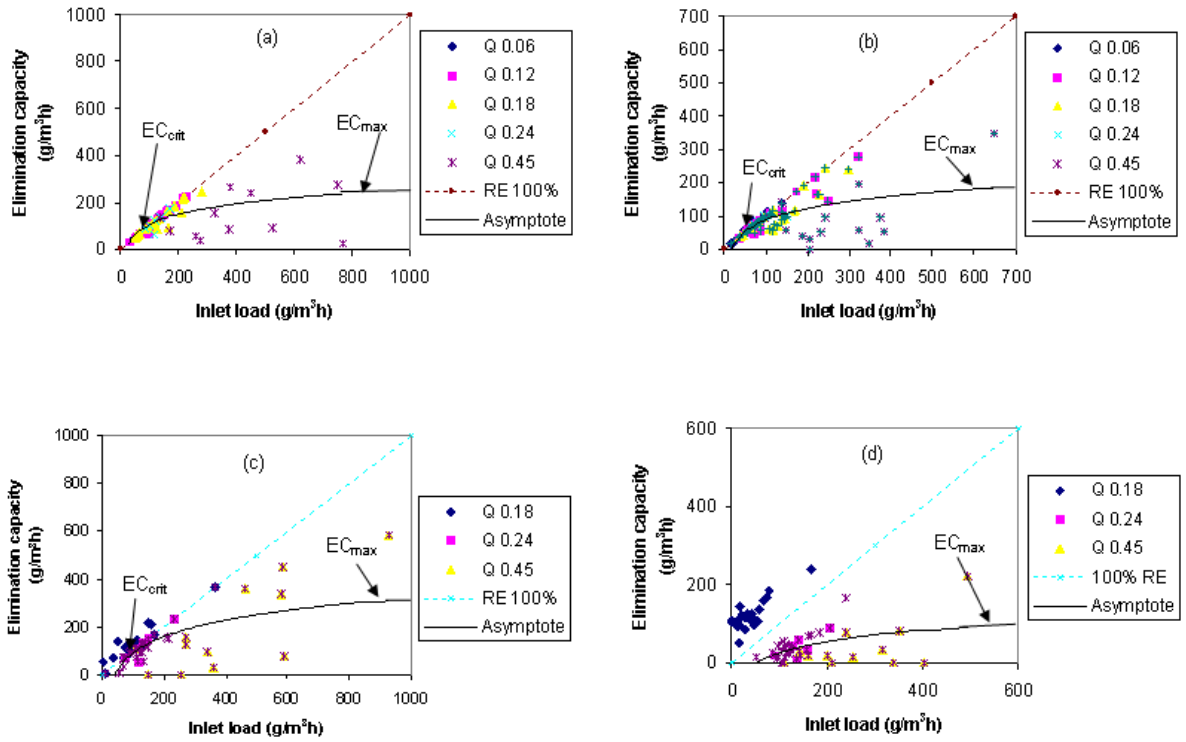

Figure 6. Elimination capacity and inlet load curve for methanol removal from air contaminated with pure methanol (a), toluene removal from air contaminated with pure toluene (b), methanol removal from air contaminated with a mixture of methanol and toluene (c), and toluene removal from air contaminated with a mixture of methanol and toluene (d).

definition. Figure 6 shows the elimination capacity and inlet load curve for the three biofilter systems. The critical elimination capacity (EC_{crit}) and the maximum elimination capacity (EC_{max}) for biofiltration of methanol were higher than those obtained for biofiltration of toluene. When biofilter was used to treat a mixture of methanol and toluene the EC_{crit} for methanol removal was the same as observed for pure methanol removal while the EC_{max} was higher. On the other hand, the EC_{max} for toluene removal from mixtures was lower than the EC_{max} for removal of pure toluene and the EC_{crit} for toluene removal from mixtures was not shown due to the variation of elimination capacity correlated to the inlet load. These indicated that the removal of methanol was not highly affected by the presence of toluene while there was significant effect on toluene removal with the presence of methanol. This can be probably explained by the competition phenomenon and their solubility and bioavailability. Methanol which has higher solubility was more easily biodegraded than toluene.

The removal rates obtained in this study were comparable to (or higher than) the results obtained by other researchers as shown in Table 5. This suggests that a mixture of palm shells and activated sludge can be used as the filter bed media for an efficient biofilter. High elimination capacities in this work probably due to bed temperature, pH, and relative humidity, which are the three most important parameters for an efficient biofilter (Devinny *et al.*, 1999), were maintained at the optimum conditions as can be seen in Figure 7 (bed temperature ≈30ºC, pH ≈7, and relative humidity

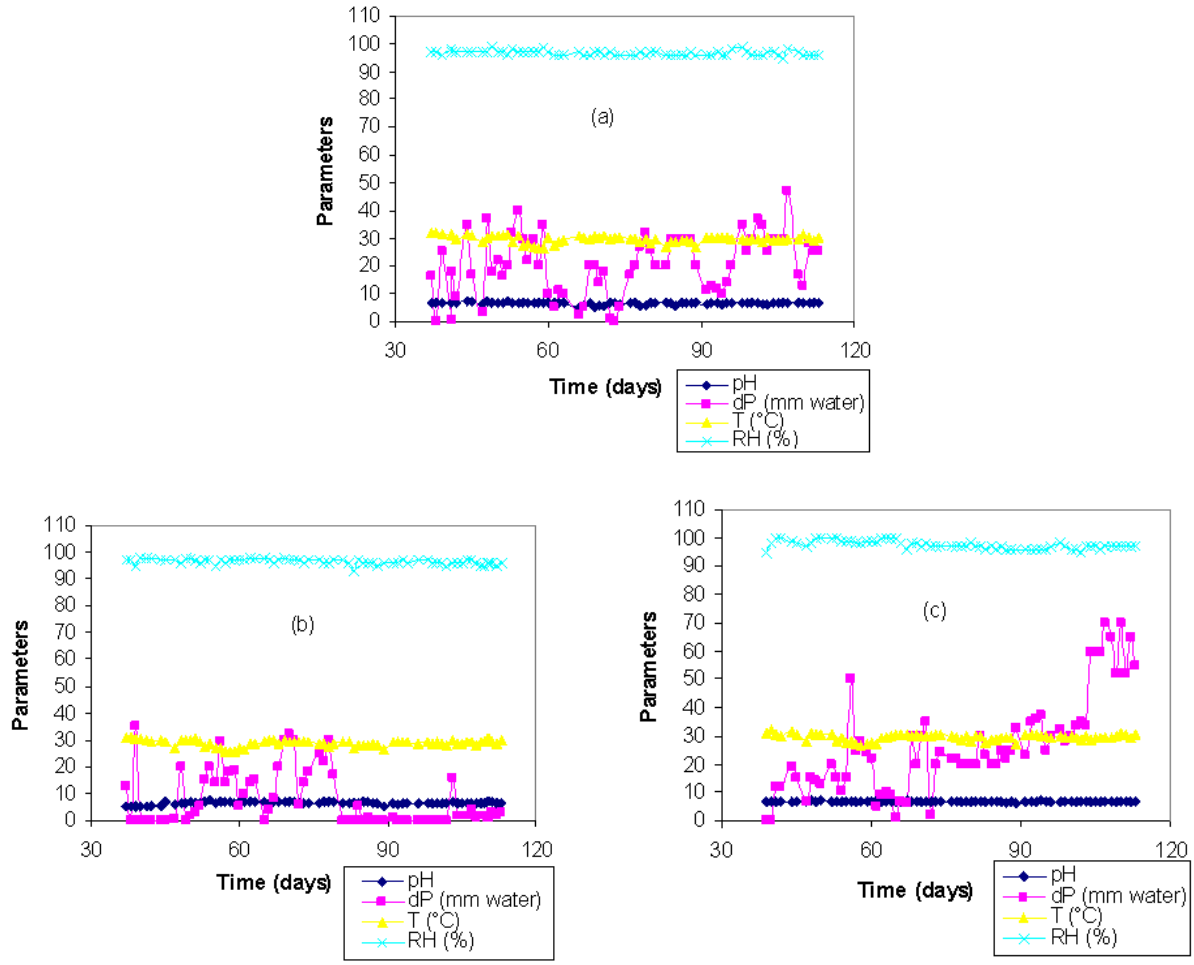

Figure 7. The measured values of pH, pressure drop (dP), temperature (T), and relative humidity (RH) of the biofilter for removal of methanol (a), removal of toluene (b), and removal of a mixture of methanol and toluene (c).

97%).

The medium bed pressure drops for methanol removal tend to be higher than the values for toluene removal; the maximum value was 47 mm H_2O on day 107 and 35 mm H_2O on day 39 for methanol removal and toluene removal, respectively. For treatment of a mixture of methanol and toluene, the maximum medium bed pressure drop was reached 70 mm H_2O on day 110. Delhomenie *et al.* (2002) stated that the medium bed pressure drop was related to the development of biomass accumulation in the biofilter column. This is in agreement with our result that the accumulation of larger amounts of biofilm was visually observed in the case of methanol removal. To reduce the pressure drop and to protect the bed-clogging problem the biofilter bed was periodically washed with water. The application of bed washing had almost no effect on the microorganisms viability, and satisfactory biofilter performance was soon reestablished (indeed by the next day). However, the bed clogging was still taking place resulting in pressure drop increases with time. Therefore, a reliable method for the prevention of the formation of excess biomass is required, especially in the case of removal of mixtures of methanol and toluene.

Influence of air flow rate

Figure 8 shows the impact of EBRT on the average removal efficiency. It can be seen that the removal efficiency increased with EBRT, especially EBRT in the range of 9-18 s. For long EBRT (71 s) corresponding to air flow rate of 0.06 m^3/h, high removal efficiencies (100% for methanol and 98% for toluene) were observed. High values of EBRT were favorable for the VOC degradation because the contact time between the microorganisms and VOC is increased. On the other hand, for short EBRT (9 s), and thus for correspondingly higher flow rates of 0.45 m^3/h, the removal efficiencies fell to values of less than 42%. Even if VOC flow through the interface was favored by the higher flow rate, the contact time between the micro-organisms and the VOC was too short and micro-organisms had insufficient time to perform the required degradation on the available amount of VOC.

Influence of inlet concentration of VOC

For removal of pure methanol and pure toluene, stage F (air flow rate = 0.45 m^3/h) was conducted to investigate the influence of inlet concentration of VOC on the removal efficiency. It was observed that removal efficiency was a decreasing function of the inlet concentration

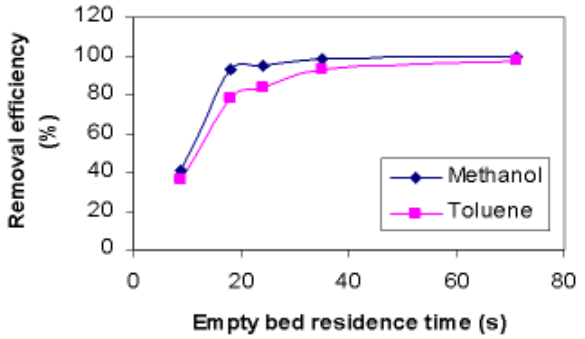

Figure 8. Influence of the airflow rate on the average removal efficiencies of the bio-filter.

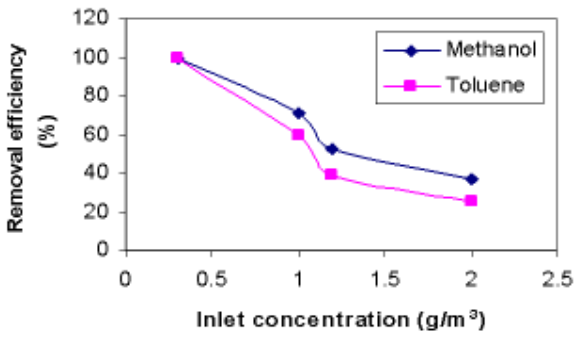

Figure 9. Influence of the inlet concentration of VOCs on the removal efficiency of the biofilter at constant air flow rate (0.45 m^3/h).

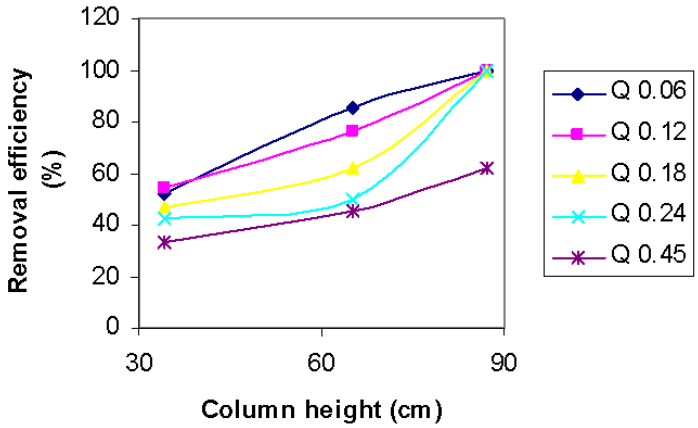

Figure 10. Influence of the column height on the removal efficiency of the biofilter for methanol removal, Q = volumetric gas flow rate (m^3/h).

(Figure 9). For VOC concentrations lower than 1.0 g/m^3, 60-100% of the VOC was eliminated. Over this concentration range, microorganisms were able to metabolize all of the available substrate. For higher concentrations, the level of microorganisms activity became the limiting factor for VOC elimination, the removal efficiency remained below 60% and 40% for methanol and toluene, respectively. However, it should be pointed out that decreasing of removal efficiency with inlet concentration was not observed at lower air flow rate (<0.45 m^3/h).

Influence of column height

Removal of methanol increased with the column height as shown in Figure 10. For removal of toluene and a mixture of methanol and toluene, the similar results were observed. This suggests that the removal efficiency depends on the filter bed volume (V) as $V = \frac{\pi D^2 \times H}{4}$, with D = diameter of biofilter column and H = height of filter bed.

Scanning electron microscopy observations

Figure 11a shows the palm shell structure before mixing with activated sludge. The samples from biofilter systems were taken on day 42 at mid level. A high microbial density was shown for each biofilter system. Diverse microbial morphologies, such as bacterial colonies, budding yeast, mycelial structures, and also some noncolonized regions on the surface of palm shell were observed (Figures 11b, c, and d). These observations revealed a heterogeneous biofilm, in contrast to that of a

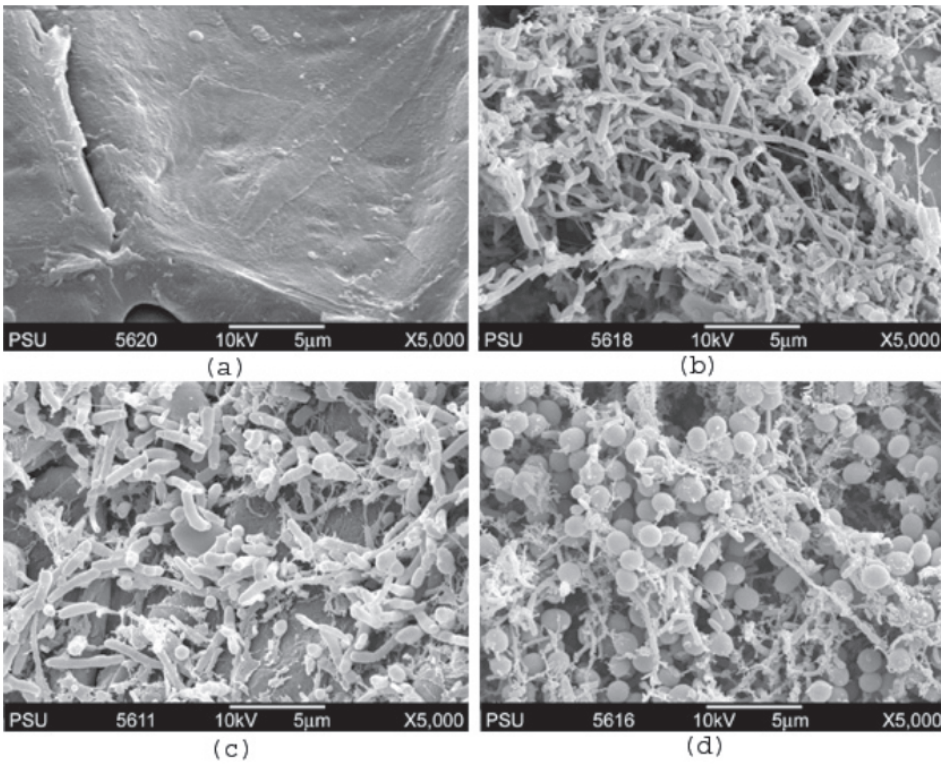

Figure 11. SEM microphotograph of the palm shell (a); samples of the biofilter media in the column for removal of pure methanol (b), removal of pure toluene (c) and removal of a mixture of methanol and toluene (d). The samples were taken on day 42 at mid level.

smooth and homogeneous biofilm which is generally assumed in the models used to describe biofilter behavior (Ottengraf and Van den Oever, 1983). In addition, the dominant microorganisms for removal of methanol (Figure 11b) were different from those observed for removal of toluene (Figure 11c). For treatment of a mixture of methanol and toluene, not only the microorganisms observed in those two previous biofilter systems were found but also the coccus microorganisms as seen in Figure 11d were observed. These results implied that the microorganisms indigenous to the bed medium can adjust themselves to be avail in each biofiltration system. Inoculation was not necessary in our study.

Design of biofilter

It is possible to design a biofilter system to treat air contaminated with methanol or toluene by using the elimination capacity obtained in this study. The minimal volume of filter bed can be calculated by $V_m = \frac{C_{Gi} \times Q}{EC_{max}}$, where V_m = minimal volume of filter bed, C_{Gi} = inlet concentration of VOC, and EC_{max} = maximum elimination capacity. If the calculated EBRT, $EBRT = \frac{V_m}{Q}$, is greater than or equal to the EBRT reported in this report, it may be appropriate for use. If not, the EBRT should be increased if a margin of safety is required.

Conclusions

Methanol, a hydrophilic compound, and toluene, a hydrophobic compound, were successfully treated in biofilter consisted of a mixture of palm shells and activated sludge as the filter bed media without inoculation. The biofilters removed as high as 230 g methanol/m^3 bed medium/h and 181 g toluene/m^3 bed medium/h. The presence of methanol in the system significantly decreased the removal rate of toluene while the removal of methanol was not affected by the presence of toluene.

The bed temperature, pH, and relative humidity should be maintained at the optimum conditions (bed temperature 30ºC, pH 7, and relative humidity 97%). The medium bed pressure drop should not be too high (< 35 mm H_2O) to protect the clogging problem.

For the air flow rate lower than 0.45 m^3/h, the inlet concentration of VOC did not have significant effect on the removal efficiency of the biofilter. However, when the air flow rate was equal to 0.45 m^3/h, the inlet concentration of VOC should be less than 1.0 g/m^3 to obtain the removal efficiency not less than 60%. The removal efficiency is proportional to the filter bed volume. As the volume of media increases, the overall target compound removal efficiency also increases. Since the cost of a biofilter system is also proportional to the volume of media used, a balance between system cost and system performance must be established.

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References

Acuna, M.E., Perez, F., Auria, R. and Revah, S. 1999. Microbiological and kinetic aspects of a biofilter for the removal of toluene from waste gases. Biotechnol. Bioeng., 63: 175-184.

Aizpuru, A., Khammar, N., Malhautier, L. and Fanlo, J.L. 2003a. Biofiltration for the treatment of complex mixtures of VOC influence of the packing material. Acta Biotechnol., 23: 211-226.

Aizpuru, A., Khammar, N., Malhautier, L. and Fanlo, J.L. 2003b. Biofiltration of a mixture of volatile organic compounds on granular activated carbon. Biotechnol. Bioeng., 83: 479-488.

Auria, R.M., Morales, M., Acuna, E., Perez, F. and Reva, S. 1996. Biofiltration of toluene vapors: start up and gaseous ammonia addition. USC-TRG Conference on Biofiltration (an Air Pollution Control Technology), The Reynolds Group, USA, 1996: 134-141.

Briggs, T.G. 1996. Biofiltration of air containing volatile organic chemicals from batch manufacturing operations. USC-TRG Conference on Biofiltration (an Air Pollution Control Technology), The Reynolds Group, USA, 1996: 69-76.

Delhomenie, M.C., Bibeau, L., Bredin, N., Roy, S., Broussau, S., Brzezinski, R., Kugelmass, J.L., and Heitz, M. 2002. Biofiltration of air contaminated with toluene on a compost-based bed. Adv. Environ. Res., 6: 239-253.

Deshusses, M.A. 1997. Biological waste air treatment in biofilters. Env. Biotechnol., 8: 335-339.

Devinny, J.S., Deshusses, M.A. and Webster, T.S. 1999. Biofiltration for air pollution control, Lewis Publishers, New York.

Johnson, C.T. and Deshusses, M.A. 1997. Quantitative structure-activity relationships for VOC biodegradation in biofilters. The 4th International *In Situ* and On-Site Bioremediation Symposium, Vol. 5, Battlelle Press, Columbus, USA, 1997: 175-180.

Lee, B.D., Apel, W.A., Cook, L.L. and Nichols, K.M. 1996. Effect of bed medium moisture on x-pinene removal by biofilters. USC-TRG Conference on Biofiltration, The Reynolds Group, USA, 1996: 214-222.

Mohseni, M. and Allen, D.G. 2000. Biofiltration of mixtures of hydrophilic and hydrophobic volatile organic compounds. Chem. Eng. Sci., 55: 1545-1558.

Morales, M., Auria, R., Perez, F. and Revah, S. 1998. Start-up and ammonia additions to biofilter for removal toluene. Biotechnol. Bioeng., 60: 483-491.

Ottengraf, S.P.P. and Van Den Oever, A.H.C. 1983. Kinetics of organic compounds removal from waste gases with a biological filter. Biotechnol Bioeng. 23: 3089-3102.

Seed, L.P. and Corsi, R.L. 1994. Biofiltration of BTEX contaminated streams: laboratory studies. The 87th Annual Meeting and Exhibition of the Air and Waste Mangement Association, Air and Waste Management Association, Pittshburgh, USA.

Shareefdeen, Z., and Baltzis, B.C. 1994. Biofiltration of toluene vapors under steady-state and transient conditions- theory and experimental results. Chem. Eng. Sci., 49: 4347-4360.

Shareefdeen, Z., Baltzis, B.C., Oh, Y.S. and Bartha, R. 1993. Biofiltration of methanol vapor. Biotechnol. Bioeng., 41 : 512-524.

Spicer, C.W., Gordon, S.M., Holdren, M.W., Kelly, T.J. and Mukund, R. 2002. Hazardous air pollutant handbook: Measurments, properties, and fate in ambient air, Lewis Publishers, New York.

Van Lith, C., Leson, G. and Michelsen, R. 1997. Evaluating design options for biofilters. J. Air Waste Manage. Assoc., 47: 37-48.

Yoon, I.K. and Park, C.H. 2002. Effects of gas flow rate, inlet concentration and temperature on biofiltration of volatile organic compounds in a peat-packed biofilter. J. Biosci. Bioeng., 93: 165-169.