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Biofilter for treating toluene vapors: Performance evaluation and microbial counts behavior

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Abstract: A lab-scale biofilter packed with mixed packing materials was used for degradation of the toluene. Three gas flow rates, i.e. 0.1, 0.2 and 0.4 m³/h, were tested for inlet concentration ranging from 0.2 to 1.2 g/m³. Removal efficiencies ranging from 45.6 to 97.3% and elimination capacities ranging from 4.95 to 61.07 g/(m³ h) were observed depending on the inlet loading rates. Maximum elimination capacity of 35.95 g/(m³ h) occurred at inlet loading rate of 45.87 g/(m³ h). The lowest layer always had highest elimination capacity. Carbon dioxide concentrations and the microbial cell counts for bio-degraders followed toluene elimination capacities. Results of this study indicated that mixed packing materials could be considered as a potential biofilter carrier, with low pressure drop (less than 84.9 Pa/m), for treating air streams containing VOCs.

Keywords: Air treatment; Biofilter; Toluene; Empty bed residence time; Inlet loading rate

Introduction

Volatile organic compounds (VOCs) are emitted into atmosphere with large quantities from different resources, such as chemical, petrochemical, pharmaceutical, food processing, pulp and paper mills, color printing, painting works, vehicle exhaust, waste incinerators and composting facilities (Chen et al. 2005; Slominska et al. 2013; Yassaa et al. 2006). Among these VOCs, toluene is a hazardous chemical, which is highly toxic and mutagenic (Gallastegui et al. 2011), even when it was exposed to low concentration. Toluene is carcinogenic, the liver, kidney and the central nervous system could be damaged by it (Rene et al. 2005). According to the report of operating facilities in 2009, the rate of toluene emission into the atmosphere was 12.2 kt/yr in the USA, and 3.9 kt/yr in Canada (Gallastegui et al. 2011).

Biofilter is the most widely used bioreactor for odor and air pollution control (El-Naas et al. 2014; Maestre et al. 2007; Rahul et al. 2013b; Singh et al. 2010a). Compared to the conventional technology, the various advantages of biofilter are low capital investment and operating costs (Elmrini et al. 2004a), energy efficiency and no secondary pollutants produced. Biofilter is mostly used in air streams treating, particularly organic vapors, with high flow rate and low concentrations of pollutants (Rahul et al. 2013a; Rene et al. 2009) less than 1000 ppm (Delhomenie et al. 2003). There are numerous factors affect the performance of the biofilter. Henry's coefficient (Non-dimensional) is one of the main factors which impact the biodegradation. Although toluene, coupled with a Henry's coefficient of 0.25, is considered insoluble, many references show that it could be used as the biofilter's substrate (Aly Hassan & Sorial 2009; Cho et al. 2009; Xi et al. 2006). In the biofilter, the polluted air stream passed through the biofilter column, then flowed through the porous packed bed where immobilized by the pollutant-degrading organisms which attached to the surface of the packing material. There were two main processes occurred during this process: first, the contaminants in the air stream were left

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in the water or adsorbed on the surface; then the contaminants were utilized to synthesis the biomass or degraded into CO_2 and H_2O , by the organisms.

The packing materials are the place where physical, chemical and biological reactions occurred, thus the properties of them are highly concerned, such as high surface area and porosity for biofilm growth, suitable pH, acceptable buffering capacity (Mudliar et al. 2010; Zare et al. 2012) and benign water holding capacity (Anet et al. 2013). Peat, soil, compost, and wood chips are the commonly used organic packing materials (Lebrero et al. 2014). Inorganic packing materials are barks, perlite, vermiculite, glass beads, polyurethane foam, polystyrene, lava rock (Mudliar et al. 2010). The lifespan of organic packing materials, however, is often lower than that of inorganic (Dorado et al. 2010), and the inorganic packing materials have few nutrients and native microorganisms than the organic packing materials.

Many researchers focused their studies on performance of biofilters. R.S. Singh et al. (Singh et al. 2010b) evaluated the performance of a biofilter treating toluene packed with polyurethane foam operated for 168 days. Removal efficiency ranging from 68.2 to 99.9% and elimination capacity ranging from 10.85 to 90.48 $\text{g}/(\text{m}^3 \text{ h})$, relying on inlet loading rates. Eldon R. Rene et al. (Rene et al. 2005) investigated a compost-based biofilter unit to treat toluene vapors from a synthetic and real gas stream. The biofilter was continuously operated for 8 months. Removal efficiencies ranging from 40 to 95% which the elimination capacities ranging from 3.5 to 128 $\text{g}/(\text{m}^3 \text{ h})$ was observed. Compared to the mixed gas stream, toluene removal efficiencies of 60 to 90% and benzene removal efficiencies of 60-80% were achieved. However, there were few researchers focused on the behaviors of different layers contributed to the overall performances, the relations between the microbial counts and the inlet loading rate were not clear up to now.

The main objective of this research was to determine the removal efficiency and elimination capacity of different layers as a function of inlet loading rate and empty bed residence time in a lab scale biofilter. The production of carbon dioxide and the microbial counts of three layers were also evaluated. And the variation of the pressure drops was observed.

Materials and method

1. Inoculum and packing material

The inert material employed in the biofilter was invented by this lab (China invention patent, CN103041695A), and it was mixed by compost, cement, perlite, CaCO_3 , plant fiber, etc. Sodium silicate which was used as adhesive was extruded and granulated. The physical properties were summarized in Table 1. Fresh activated sludge was used as the inoculum source for the biofilter, which was obtained from a municipal wastewater treatment plant in Zhengzhou, China. Microorganisms in the activated sludge were acclimated to toluene in order to accelerate the adaptation period. For acclimation, one liter of the activated sludge was placed in an aerated tank and diluted with 3 L of nutrient solution (Amin et al. 2014). The composition of nutrient solution per liter of distilled water was: K_2HPO_4 -0.11g, KH_2PO_4 -0.04g, NH_4Cl -0.54g, MgSO_4 -0.067g, CaCl_2 -0.036g, FeCl_3 -0.25mg, MnSO_4 -0.03mg, ZnSO_4 -0.04mg, $(\text{NH}_4)_6\text{Mo}_7\text{O}_{24}\cdot 4\text{H}_2\text{O}$ -0.03mg.

Table 1 Physical properties of the mixed packing materials

Parameter	Units	Mixed packing materials
Equivalent diameter	mm	10-12
Bulk density	kg/m ³	471.0±0.8
Specific surface area	m ² /g	3.91±0.20
Void space volume	%	38-41
Water holding capacity	%	52

2. Biofilter setup and operation conditions

The biofilter was constructed from Plexiglas cylinders with an internal diameter of 105mm, and a total bed height of 90mm, which was divided into three same sections. The total bed volume was approximately 8.24 L. Figure 1 shows the diagram of the biofilter system. Toluene (Kemel, 99.5% AR Grade, China) was stripped with compressed air. The biofilter was operated in an up-flow mode at room temperature. The concentration of pollutants was fixed by means of flowmeters (all from Yuyao Kingtai instrument CO., China).

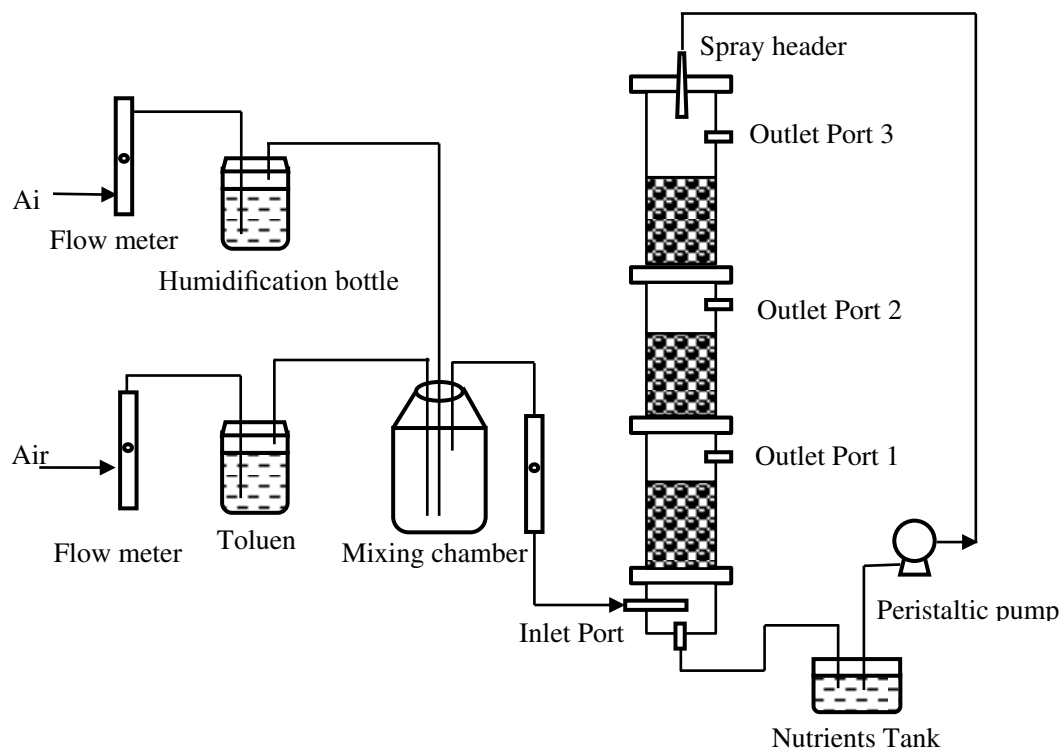


Figure 1 Diagram of the biofilter system

During the study of degrading toluene, different ILRs, 4.95 ± 0.96 , 15.24 ± 1.81 , 25.58 ± 2.92 , 34.42 ± 1.97 , 44.51 ± 1.46 and 61.07 ± 5.02 g/(m³ h), were set up at the EBRT of 74.2s. Experiments at EBRTs of 148.3s and 49.4s were also carried out, which ILRs were 24.35 ± 2.92 and 25.33 ± 2.63 g/(m³ h), respectively. At each different stage, the toluene concentration of the inlet load was kept relatively constant, and the operation time of the biofilter was carried out until the steady state, which was lasted at least five days. The microbial cell counts and carbon dioxide concentrations measured simultaneously. The operating conditions of the biofilter are summarized in Table 2. To order to insure satisfactory conditions of moisture and nutrients for microorganism activities, nutrient solution, about 20 ml/min, was continuously sprayed every day for 30 min on the top of the packing media through the nutrient distribution system using a peristaltic pump.

Table 2. Operating conditions of the biofilter

Phase of operation	Gas flow rate (m ³ /h)	Inlet concentration (g/m ³)	EBRT (s)	ILR (g/(m ³ h))	Operation times (days)
Phase I	0.2	0.10 ± 0.02	74.2	4.95 ± 0.96	7
		0.31 ± 0.04		15.24 ± 1.81	7
		0.53 ± 0.06		25.58 ± 2.92	7
		0.71 ± 0.04		34.42 ± 1.97	7
		0.92 ± 0.03		44.51 ± 1.46	8
		1.26 ± 0.10		61.07 ± 5.02	10
Phase II	0.1	0.53 ± 0.08	148.3	24.35 ± 2.92	7
	0.4	0.36 ± 0.05	49.4	25.33 ± 2.63	10

3. Analytical methods

Toluene concentration in the biofilter was measured using a gas chromatograph (GC1120; Sunny Hengping, China) equipped with a flame ionization detector (FID) and a FFAP chromatographic column (30m×0.25mm×0.25μm; Nanjingjianuo, China); Nitrogen was used as the carrier gas at a flow rate of 50 ml/min. The oven, injector and FID detector were maintained at 65°C, 150°C and 250°C, respectively.

Pressure drop at different height was measured by means of by means of testo 510 (Testo AG, Germany), and temperature was measured by testo 405-V1 (Testo AG, Germany). The Moisture Content (MC) of the biofilter packing materials was determined by weight loss method after drying 12 hours at 105°C.

Microbial cell counts of micro-organisms measured by taken 1 g of moist media materials from three different locations at each layer of the biofilter. After 9 ml sterile extraction buffer (0.9% NaCl) was mixed into each sample (Rene et al. 2009), the solution was shaken vigorously for 30 min and serially diluted with sterilized water, and then it was plated in a nutrient agar for isolation of bacteria (Saravanan & Rajamohan 2009). The colonies were incubated for 3 days at 30°C before the colonies were counted.

Carbon dioxide concentration (CO₂) in the gas phase was determined by capacity titration method. First, CO₂ gas was absorbed by Ba(OH)₂ solution (1.4 g/L), by using an atmosphere sampler (QC-2B; Beijing Municipal Institute of Labour Protection, China). Certain volume of the solution was titration by CH₃COOH solution (0.6 g/L), where phenolphthalein was used as indicator.

4. Performance evaluation

The performance parameters of the biofilter are illustrated in Table 3. The results from the biofilter are expressed in terms of toluene inlet loading rate (ILR), elimination capacity (EC) and removal efficiency (RE). Data of daily measurements were used to obtain the average values of the biofilter.

Table 3 Definition of biofilter performance parameters

Parameter	Definition	Units
Empty bed residence time	$EBRT = \frac{V}{Q}$	s
Inlet loading rate	$ILR = \frac{Q \times C_{in}}{V}$	$g/(m^3 \text{ h})$
Elimination capacity	$EC = \frac{Q \times (C_{in} - C_{out})}{V}$	$g/(m^3 \text{ h})$
Removal efficiency	$RE = \frac{C_{in} - C_{out}}{C_{in}} \times 100$	%

Results and discussion

1. Influence of toluene inlet concentration

During the phase I, in the biofilter operation time, the overall EC of toluene as a function of the ILR is illustrated in Figure 2. Initially, the biofilter was operated at a low concentration, which led to a low ILR. Then the ILR was gradually increased by the increase of the toluene inlet concentration. The ILR was gradually increased from 4.95 ± 0.96 to $61.07 \pm 5.02 \text{ g}/(\text{m}^3 \text{ h})$. The EC of the toluene biofilter was increased up to EC_{\max} of $35.95 \text{ g}/(\text{m}^3 \text{ h})$, which occurred at an ILR of $45.87 \text{ g}/(\text{m}^3 \text{ h})$. After that, with the increase of the ILR, the EC of the biofilter decreased slightly.

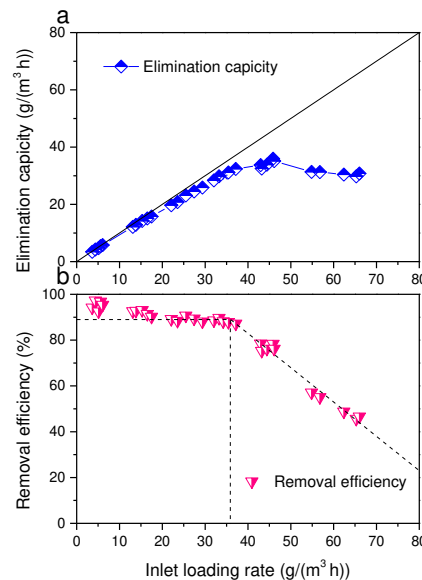


Figure 2 Influence of inlet loading rate on the elimination capacity (a) and removal efficiency (b) of the biofilter at an EBRT of 74.2s

The variations of EC and RE with respect to ILR of toluene are shown in Figure 2. When at relatively lower ILRs, from ILR 4.95 ± 0.96 to $34.42 \pm 1.97 \text{ g}/(\text{m}^3 \text{ h})$, the biofilter had an excellent performance and the RE was almost constant with the average value at 95.2, 92.0, 89.2 and 88.3%, respectively. As shown in Figure 2, the ECs and ILR had a near linear relationship and the EC very closely to the 100% conversion line. However, when ILR surpassed $34.42 \pm 1.97 \text{ g}/(\text{m}^3 \text{ h})$, the performance of the biofilter started to decrease, and the EC started to deviate the 100%

conversion line. And the average RE at ILR $44.51 \pm 1.46 \text{ g}/(\text{m}^3 \text{ h})$ were 76.9%, which was under 80% RE. When ILR surpassed to $61.07 \pm 5.02 \text{ g}/(\text{m}^3 \text{ h})$, the RE decreased heavily and the average RE was only 50.6%. Two distinct zones, mass-transfer controlling zone and bioreaction controlling zone, were observed in the RE versus ILR graph. The results got above were in agreement with Kiran Singh et al. (Singh et al. 2010a), Elmirini et al. (Elmirini et al. 2004b) and Kiared et al. (Kiared et al. 1997).

In the mass-transfer controlling zone, biofilter completely degraded the pollutants; and the dominant factor was the rate of the pollutants transferred into the biofilm where it could be degraded by microbial population, as EC was directly proportional to ILR. In the bioreaction controlling zone, biodegradation rate was the key step. Since the ILR was relatively high, the microbial population was incapable to degrade the pollutants completely. Thus, in this zone, EC was not directly proportional to ILR.

2. Effect of bed height

In the biofilter, the packing materials bed was subdivided into three identical layers, gas samples were collected from each part of the biofilter. In order to have a clear understand of the contribution of different layer to its overall performance, the RE of the three layers is shown in Figure 3. The corresponding EC of each layer with various ILRs, which may provide a further insight of the biofilter performance, is also shown in Figure 3.

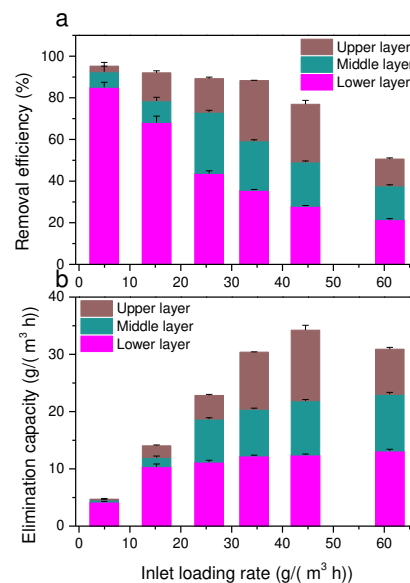


Figure 3 Comparison of removal efficiency (a) and elimination capacity (b) among the three layers at various inlet loading rates

Results illustrated that the contributions of the different layers changed depending on ILRs. As ILR increased, the RE of the lower layer decreased from 85.1 to 21.5%; while the middle layer improved from 7.5 to 29.4 (at an ILR of $25.58 \pm 2.92 \text{ g}/(\text{m}^3 \text{ h})$) then decreased to 16.2%; and the upper layer improved from 2.7 to 29.0 (at an ILR of $34.42 \pm 1.97 \text{ g}/(\text{m}^3 \text{ h})$) then decreased to 12.9%. At relatively lower ILRs, the contribution of RE was not even, but mostly by lower layer. For instance, at ILR of $4.95 \pm 0.96 \text{ g}/(\text{m}^3 \text{ h})$, the RE in the lower layer was 84.1 compared to 7.5 and 2.7% in the middle and upper sections, respectively; at ILR of $15.24 \pm 1.81 \text{ g}/(\text{m}^3 \text{ h})$, the RE in the lower layer was 68.2 compared to 10.4 and 13.5% in the middle and upper sections,

respectively. Thus, in this case, the majority of the toluene was eliminated in the lower layer. In this case, only a small portion of toluene was available for the middle and upper layers and the overall RE was closed to 100%. However, for relatively higher ILR, the EC of different layers were relatively uniform; but the lower layer was still a little higher than other layers. This may be caused by that, at higher ILR, the contaminant injected into biofilter was very high which cannot be completely degraded by the microbial of the lower layer, the rest contaminant was flowed into other two layers. Then, with the increase of the ILR at other two layers, the microbial of other layers could gain more nutrients, i.e. toluene, which the utilization of other two layers was more than at lower ILRs. However, when the concentration of contaminant at other layers, especially at the upper layer, the exit concentration could be higher, which led to the decrease of the overall EC. The reason EC at lower layer was always the highest, may be due to higher microbial population and higher moisture content at the lower layer.

Eldon R. Rene et al. (Rene et al. 2015) studied the performance of a biofilter treating gas-phase mixture of benzene and toluene, and results showed that the elimination of toluene was mostly occurred at the topside of the biofilter which was not conformed to the results in this study. This may be due to the biofilter they used was first to treat benzene contaminant, whereas the biofilter in this study used was only to treat toluene.

3. Influence of gas flow rate

The biofilter performance at different EBRTs is illustrated in Figure 4. Three different EBRTs, i.e. 148.3, 74.2 and 49.4 s, represented low, middle and high gas flow rate, respectively. The corresponding ILRs was 24.35 ± 2.92 , 25.58 ± 2.92 and 25.33 ± 2.63 $\text{g}/(\text{m}^3 \text{ h})$, respectively. According to Figure 4, when compared to the EBRTs of 74.2 and 49.4s, the RE dropped heavily and the decline was around 18.2%. This decrease was primarily due to the reduction in the contaminant retention time in the bed, which could not provide sufficient time for the toluene to transfer from the gas phase to the biofilm. This led to the increased of the exit concentration which conducted to a reduction of RE.

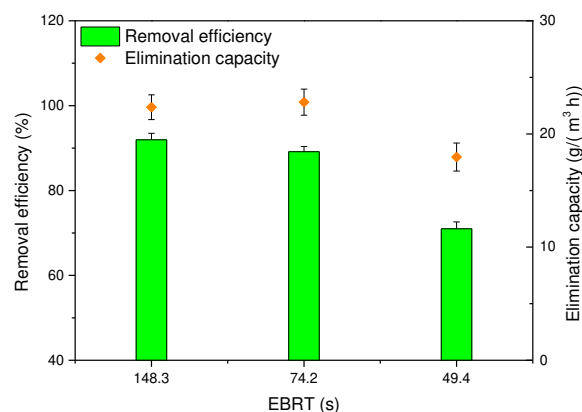


Figure 4 Influence of EBRT on removal efficiency and elimination capacity

However, compared to the EBRTs of 74.2 and 148.3 s, the RE was slightly improved, at an average RE of 89.2 and 91.9 %, respectively. Basically, decreasing EBRT means the increase of energy consumption, which would led higheroperation costs; high EBRT means the volume of the biofilter will be very large which needs a large plant and first investment capital is high. Thus, the objective is not only to maintain an acceptable biofilter performance, but also to reduce the costs

at the same time. Results showed that the EBRT of 74.2s was the proper operation condition in this biofilter.

The variation of carbon dioxide production of the total biofilter and different layers were calculated, as showed in Figure 5. In the biofilter, the toluene was degraded to water and carbon dioxide mostly by aerobic micro-organisms since the activation and cleavage of the aromatic nucleus require dissolved oxygen (Andreoni & Gianfreda 2007). Thus, to investigate the carbon dioxide concentration profile at different layers may provide a deep understanding of the biofilter performance. From the Figure 5, it was clear that the highest EBRT the highest carbon dioxide concentration, since the micro-organism at this moment could obtain large amounts of contaminants. The carbon dioxide generated by lower layer preceded the other two layers at the three EBRT, and this was accordance with lower layer had larger elimination capacity from the results got above.

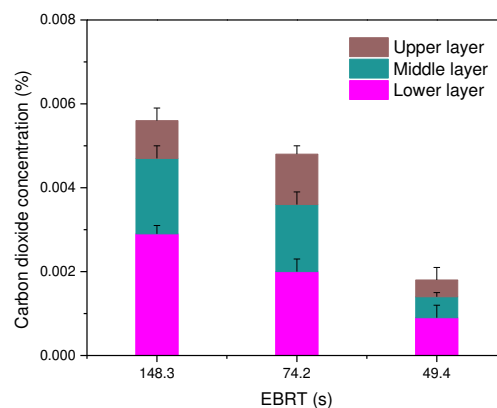


Figure 5 Carbon dioxide concentrations at the three layers versus various EBRT

4. Behavior of the microbial count

According to the results of the microbial cell counts, there were mainly three kind of micro-organism, one kind of fungi and two kinds of bacteria. The fungus was white and filamentous, and the microbial count versus time is shown in Figure 6. The microbial count of the two bacteria—one bacterium was pale yellow named bacterium-A, the other one was pinky named bacterium-B—versus time is shown in Figure 7.

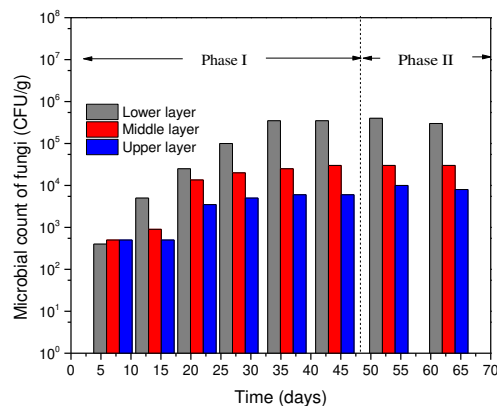


Figure 6 Microbial count of fungi at the three layers of the biofilter versus time

At the beginning of the operation, the microbial count of fungi at the three layers was at the same level which was less than 10^3 CFU/g. Then, gradually increased to about 3.5×10^5 CFU/g at the lower layer, 3×10^4 CFU/g at middle layer, 6×10^3 CFU/g at the upper layer, respectively, at the 36th day which the ILR was $44.51 \text{ g}/(\text{m}^3 \text{ h})$. The count of bacterium-A had the similar trend with fungi; however, the differences were the initial counts at the three layers were a little more than 10^4 CFU/g and the maximum number had occurred in the 28th day at an ILR of $34.42 \pm 1.97 \text{ g}/(\text{m}^3 \text{ h})$.

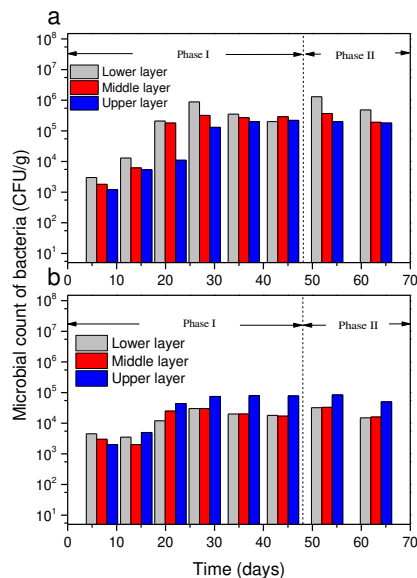


Figure 7 Microbial counts of bacterium-A (a) and bacterium-B (b) at the three layers versus time

However, when compared to bacterium-B, it showed some differences. Initially, the count of bacterium-B was close to bacterium-A, 4.5×10^3 CFU/g at the lower layer, 3×10^3 CFU/g at middle layer, 6×10^2 CFU/g at the upper layer, respectively. The count increased with the increase of the ILR, then achieved a relatively constant state, which was similar with fungi and bacterium-A. However, most of the time, the microbial count of the upper layer was higher than that of the other two layers, and a maximum value of 8×10^2 CFU/g was achieved at an ILR of $44.51 \pm 1.46 \text{ g}/(\text{m}^3 \text{ h})$.

Both the microbial counts of the fungi and the bacteria were depended on ILR, which demonstrated that the micro-organisms were fed on the contaminants. The trend of the micro-organisms at different layers under various ILRs was consistent with trend of RE and EC. According to G. Gallastegui et al. (Gallastegui et al. 2013), the microbial population and reaction capacity remained low at the lower layer, this was consistent with bacterium-B, however, was not consistent with the trends of the fungi and bacterium-A. In his study, the concentration of the contaminant could achieve to $8.72 \pm 0.78 \text{ g}/\text{m}^3$, because that the lower layer have the highest microbial population. The reason bacterium-B was higher at the upper layer may be that it was more sensitive to the concentration of the contaminant. Results of V. Saravanan and N. Rajamohan (Saravanan & Rajamohan 2009) showed that the removals were more efficient in the lower layer which was consistent with the results got here.

5. Behavior of the pressure drop

Pressure drop of the biofilter depends on many factors, which could be divided into three

categories: The first was gas flow rate which directly decides the velocity of the gas; the bigger the gas flow rate, the higher the pressure drop. The second was the media properties which include media size, porosity, depth and moisture content (Singh et al. 2006), et al. The last one was microbial community inhabited in the biofilter; biomass accumulation in the biofilter led to changes in media bed characteristics, which may cause channel diminished, thus increased pressure drop (Morgan-Sagastume et al. 2001). The pressure drop versus time is shown in Figure 8.

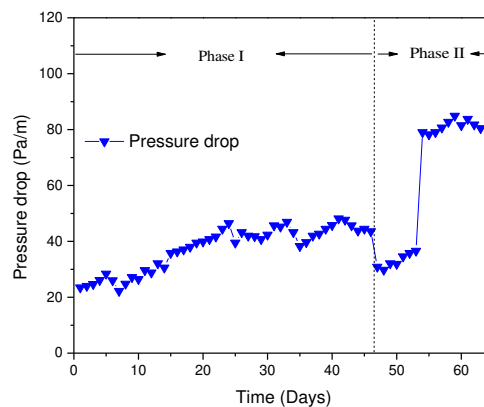


Figure 8 Pressure drop versus time at various phase

The initial pressure drop, during phase I, was about 20 Pa/m, then increased slowly with the operation time, and finally achieved a steady state about 43 Pa/m. During phase II, the pressure drop decreased to nearly 30 Pa/m with the doubled EBRT, then increased to 81 Pa/m at an EBRT of 49.4s. During phase I, the gas flow rate was maintained constant, and the increase of the pressure drops was mainly due to biomass accumulation. Then, during phase II, the change of the pressure drops were mostly influenced by the variations of the gas flow rate. In addition, the bed compaction and deterioration was observed negligible, which indicated the mixed packing material had a good mechanical strength. The maximum value of the pressure drops was 84.9 Pa/m, which was significantly advanced to some organic materials for wood chips with a pressure drop of 2600 Pa/m (Morgan-Sagastume et al. 2001), and matured compost with a pressure drop of 264.8 Pa/m (Delhoménie et al. 2003).

Conclusion

In this paper, toluene was treated with an up-flow lab scale biofilter filled with inert packing materials. The EC_{max} was observed at an inlet loading rate of 45.87 g/(m³ h), and mass-transfer controlling zone and bioreaction controlling zone were also observed. During the whole operation, the highest EC was appeared at the lower layer. The CO₂ concentration and the distribution of microbial populations in the biofilter were well correlated with the toluene removal efficiencies and elimination capacities, indicated that the toluene was biodegraded in the biofilter. The low pressure drop demonstrated that the packing materials were proper for biofiltration.

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