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PERFORMANCE OF TRICKLE-BED AIR BIOFILTER: A COMPARATIVE STUDY OF A HYDROPHILIC AND A HYDROPHOBIC VOC

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Abstract. Two lab-scale trickle-bed air biofilters were operated for investigating the difference in performance between a hydrophilic and a hydrophobic volatile organic compound (VOC). Methyl isobutyl ketone (MIBK) and styrene were selected as a model hydrophilic and hydrophobic VOCs, respectively. Effects of loading rates, biofilter re-acclimation, removal profile along biofilter depth, nitrogen consumption, and CO_2 production were compared under three operating conditions, namely, backwashing and two non-use periods (starvation and stagnant).

Consistent over 99% removal efficiency up to loading rates of 3.26 kg COD/m^3-day was obtained for the MIBK biofilter at 0.76 min empty bed retention time (EBRT) and 1.5 L/d nutrient flow. A similar performance for the styrene biofilter was obtained for loading rates up to 1.9 kg COD/m^3-day at 2.02 min EBRT and 2.4 L/d nutrient flow. The MIBK biofilter required only an initial acclimation period of 16 days while styrene biofilter required 46 days. Non-use periods can be used as another means of biomass control for both biofilters when the employed loading rate did not exceed 1.27 and 2.17 kg COD/m^3-day for styrene and MIBK biofilters, respectively. The re-acclimation of both biofilters was delayed with increase of loading rate. MIBK biofilter re-acclimated in 90 min, while styrene biofilter re-acclimated in more than 600 min. Under similar loading rates, MIBK biofilter utilized less biofilter depth than styrene biofilter. Nitrogen consumption behaviors were apparently different between the two biofilters. Styrene biofilter had higher CO_2 production than MIBK biofilter and its CO_2 production was closely related to the theoretical complete chemical oxidation.

Keywords: biofiltration, trickle-bed air biofilter, CO_2 production, hydrophilic, hydrophobic, nitrogen consumption, VOCs

1. Introduction

Biofiltration is a promising air pollution control technology for the removal of volatile organic compounds (VOCs) from waste gas streams. There are mainly two types of biofilters – traditional biofilters packed with natural organic media and trickle-bed air biofilters (TBABs) packed with inert synthetic media. TBABs facilitate more consistent operation than do traditional biofilters via better control of overall pressure drop, nutrient concentration, and pH.

In a gas phase biofilter, the VOCs transfer process from the gas phase to the aqueous phase is a key factor for the overall biofilter performance. Hydrophilic

VOCs have high solubility in water and low Henry's law coefficient (<0.01 dimensionless) and they include alcohols, ethers, and ketones. Hydrophobic VOCs have low solubility in water and high Henry's law coefficients and common examples are aromatics and alkanes. A comparative research between hydrophilic and hydrophobic VOCs is rather scarce. Deshusses and Johnson (2000) compared 18 VOCs with a wide range of Henry's law coefficients under transient biofilter performance. The critical loading rate was defined as the maximum loading rate beyond which elimination deviates significantly from 100%. Eighteen VOCs critical loading rates and maximum performance (EC_{max}) were obtained. They concluded that the Henry's law constants had great effect on removal of VOCs in biofilter. Zhu *et al.* (2004) investigated the effects of substrate Henry's law coefficient on biofilter performance. They concluded that the transfer of VOCs between the vapor and liquid phases was a rate-limiting step for VOCs with high Henry's law constants; and O_2 transfer may become a rate-limiting step in treating VOCs with low Henry's law constants.

In order to achieve consistent high biofilter performance meeting the set regulations, the effect of non-use periods on its performance should be known. The authors in their previous studies (Cai *et al.* 2004; Cai *et al.* 2005; Kim *et al.* 2005a,b) showed that non-use periods (starvation and stagnant) had different effects on VOCs removal for different VOCs, and could be treated as another means of biomass control up to a critical loading rates of 5.63 kg COD/m^3-day for methyl ethyl ketone (MEK), 3.26 kg COD/m^3-day for methyl isobutyl ketone (MIBK), 1.90 kg COD/m^3-day for styrene, and 1.41 kg COD/m^3-day for toluene. Moe and Qi (2004) studied the behavior of a fungi biofilter packed with cubed polyurethane foam media during intermittent loading. They concluded that long-term loading shutdown required longer recovery time than shorter periods of shutdown, and furthermore, loading shutdown had varying effects for different compounds. Martin and Loehr (1996) reported biofilter re-acclimation after restart-up from two non-use periods (starvation and stagnant). The results showed longer periods of non-use required longer periods of re-acclimation. Shorter re-acclimation periods were required for no chemical loading than those for no flow. Togna and Frisch (1993) reported styrene re-acclimation periods of five to eight hours after two or more days without chemical contact.

The primary objective of this study was, therefore, to investigate the difference in performance between a hydrophilic and a hydrophobic VOCs under three different operating conditions, namely, backwashing and two non-use periods (starvation and stagnant). The specific objectives are focused on the following issues:

1. effect of loading rate on biofilter performance,
2. initial acclimation and re-acclimation periods after backwashing and non-use periods,
3. removal profile along the biofilter depth,
4. nitrogen consumption,
5. CO_2 production.

TABLE I

Physical properties of the selected VOCs (Verschueren 2001; Watts 1998)

	Boiling Point (°C)	Vapor Pressure (mmHg at 20 °C)	Solubility (g/L at 20 °C)	DimensionlessHenry's Law Constant (20 °C)
MIBK	116–119	15	19.1	0.00433
Styrene	145.2	5	0.23	0.125

2. Experimental Methods

2.1. EXPERIMENTAL METHODS

Two independent, parallel trickle-bed air biofilters were employed to compare the performance between hydrophilic and hydrophobic VOCs. Methyl isobutyl ketone (MIBK) and styrene were selected as the model hydrophilic and hydrophobic VOCs, respectively. Some of the physical properties of the two selected compounds are listed in Table I.

Each biofilter was constructed of seven cylindrical glass sections with an internal diameter of 76 mm and a total length of 130 cm. The sections were connected with high pressure clamps (75 psi). Each section was equipped with a sampling port that extended to the centre of the column. Each biofilter was packed with pelletized diatomaceous earth biological support media (Celite 6 mm R-635 Bio-Catalyst Carrier; Celite Corp., Lompoc, CA) to a depth of about 60 cm. Four gas sampling ports were evenly located axially along the media bed. The biofilters were operated at a constant temperature of 20 °C and in a co-current gas and liquid downward flow mode. The air flow and nutrient flow to the biofilters was initially set up at the rate of 3.6 L/min and 1.5 L/day, respectively. The feed nutrient solution was only used in a pass-through-then-discard mode. A spike solution of 2 M $NaNO_3$, and 0.22 M NaH_2PO_4 was added to the nutrient feed solution as the only nitrogen and phosphorous source to maintain an initial COD-to-nitrogen ratio of 50:1 and a nitrogen-to-phosphorous ratio of 4:1. $NaHCO_3$ was used as a pH buffer. The biofilters were initially seeded with activated sludge obtained from the secondary clarifier of an activated sludge system at a municipal wastewater treatment plant (Millcreek Wastewater Treatment Plant, Cincinnati, OH).

The concentrations of VOCs in the gas phase were measured by using a gas chromatograph (GC) (HP 5890, Series II, Hewlett-Packard, Palo Alto, CA) equipped with a flame ionization detector (FID) and 0.25 μm film narrowbore column (DB 624, J and W Scientific, Folsom, CA). Detailed description of the analytical method is provided by Cai *et al.* (2005) and Kim *et al.* (2005b). The detection limit for both MIBK and styrene was 0.5 ppmv. Effluent gas phase samples for CO_2 analysis were also taken by using gas-tight syringes through sampling ports in the biofilter. A GC (HP 5890, Series II, Hewlett-Packard, Palo Alto, CA) equipped with a thermal conductivity detector (TCD) was used for determining the CO_2 concentrations in the effluent gas phase. The detection limit was $0.001\%_v$ CO_2. Detailed description of the analytical method is provided by Cai *et al.* (2005) and Kim *et al.* (2005b). Liquid phase samples were analyzed for total carbon (TC), inorganic carbon (IC), and volatile suspended solid (VSS) concentration. TC and IC were determined by using a Shimadzu TOC 5050 analyzer (Shimadzu Corp., Tokyo, Japan). VSS concentrations in the effluent and backwashing water were determined according to Standard Methods 2540 G (Standard Methods, 1998). Aqueous samples for NO_3^-—N determination were analyzed by using a Shimadzu UVmini 1240 UV-Vis spectrophotometer (Shimadzu Corp., Tokoyo, Japan) according to Standard Methods 4500-NO_3^- B (Standard Metods, 1998).

2.2. Experimental plan

Comprehensive investigations were conducted on the biofilter system for studying the difference in biofilter performance between hydrophilic and hydrophobic VOCs. The investigations were conducted for determining their difference in the initial acclimation and re-acclimation periods after backwashing and re-start after non-use periods, removal profile along media depth, nitrogen consumption, and CO_2 production. Acclimation or re-acclimation was considered complete when over 99% removal efficiency was obtained. Inlet concentrations were varied from 50ppmv to the maximum allowed as determined in the study. For each employed inlet concentration, one biomass control operation, referred to as the "backwashing", and two non-use operations, namely "starvation" and "stagnant" were conducted.

2.2.1. *Backwashing Operation*

The backwashing was conducted while the biofilter was off line by recycling 18 L of the buffered nutrient solution at a rate sufficient enough to fluidize the media for a defined time period. Finally, the recycle was shut off and another 18 L of the buffered nutrient solution was passed through the column as a rinse. The backwashing duration and frequency were initially set at 1 hour duration per week for a period of three weeks for each inlet concentration.

2.2.2. *Starvation Operation*

This experimental operation involved the period without VOC loading, i.e., pure air with nutrient flow through the biofilter. The frequency for this operation was two days per week for a period of three weeks.

2.2.3. *Stagnant Operation*

This experimental operation reflected no flow, i.e., no nutrients, VOC, or air passing through the biofilter. The frequency for this operation was two days per week for a period of three weeks.

TABLE II

Biofilter operating conditions

MIBK	a	b	c	d	e	
Duration periods, days	1–70	71–126	127–215	216–244	245–302	
EBRT, min	0.76	0.76	0.76	0.76	0.76	
Inlet Concentration, ppmv	50	100	250	200	150	
Loading rate kg	1.09	2.17	5.43	4.34	3.26	
COD/m^3-day(g/m^3-h)	(16.7)	(33.4)	(83.5)	(66.8)	(50.1)	
Overall Removal	99.7%	99.3%	88.6%	94.8%	99.9%	
Liquid flow rate, L/day	1.5	1.5	1.5	1.5	1.5	
Styrene	**a**	**b**	**c**	**d**	**e**	**f**
Duration periods, days	1–46	47–92	93–155	156–195	196–259	260–316
EBRT, min	0.76	1.51	1.51	1.51	2.02	2.02
Inlet Concentration, ppmv	50	50	100	250	330	200
Loading rate kg COD/m^3-day	1.27	0.64	1.27	3.18	3.18	1.90
(g/m^3-h)	(17.4)	(8.7)	(17.4)	(43.5)	(43.5)	(26.0)
Overall Removal	83.7%	99.4%	99.2%	87.2%	90.6%	99.9%
Liquid flow rate, L/day	1.5	1.5	1.5,2.4	2.4	2.4	2.4

3. Results and Discussion

3.1. Prformance and initial acclimation

The two biofilters were initially started up at 50 ppmv inlet concentration with EBRT of 0.76 min and liquid flow rate of 1.5 L/day. The operating conditions and overall performance are summarized in Table II. The detailed biofilters performance with respect to VOC removal at different loading rates has been presented in previous studies (Cai *et al.*, 2005; Kim *et al.*, 2005b). The initial acclimation of the MIBK biofilter to reach 99% removal at 0.76 min EBRT was 16 days. On the other hand, the styrene biofilter did not get acclimated under 0.76 min EBRT in 46 days (stage a) and the EBRT was then increased to 1.51 min (stage b). Furthermore for styrene in stage c, a higher nutrient solution feed rate 2.4 L/day was employed in order to obtain consistent high removal efficiency. In stage e, the EBRT was further increased to 2.02 min. It has been found that the non-use operation could maintain long-term 99% removal efficiency for loading rate up to 2.17 kg COD/m^3-day (stage b) for MIBK biofilter and 1.27 kg COD/m^3-day (stage c) for styrene biofilter, without any backwashing. For both biofilters, once the employed loading rates exceeded the critical loading rate (3.26 kg COD/m^3-day for MIBK and 1.9 kg COD/m^3-day for styrene), no acclimation to over 99% removal was encountered.

The difference in performance between the MIBK biofilter and styrene biofilter is speculated to be mainly controlled by the VOC mass transfer rate from the gas phase to the aqueous phase. The BOD_5 and BOD_{20} for MIBK are 4.4% and 56.6% theoretical oxygen demand (ThOD) (Ettinger 1956), respectively; and the BOD_5 and BOD_{20} for styrene are 65% and 87% ThOD (Bridie *et al.* 1979; Price *et al.* 1974), respectively. The BOD data for MIBK and styrene clearly indicate that styrene is more amiable to biodegradation than MIBK once it is in the aqueous phase. However, the biofilter performance in our study showed that the MIBK biofilter had higher elimination capacity and significantly shorter start-up period than the styrene. Hence, it's reasonable to contribute the reverse output of performance between MIBK and styrene to the difference of mass transfer from gas phase to aqueous phase. For the hydrophilic VOC-MIBK, the resistance of mass transfer was small due to its high solubility (see Table I) which in turn favored attaining steady stable 99% removal efficiency in a short time. It is worthwhile to note that the high mass transfer rate favored more biomass growth and accumulation, which might contribute to channeling and erratic behavior of the biofilter beyond the critical loading rate. On the other hand, for the hydrophobic VOC-styrene, a stable 99% removal efficiency was achieved at a much longer time as compared to MIBK because of the high mass transfer resistance from the gas phase to the aqueous phase. Hence, when the microbial community in the biofilter endured a change in loading, a delay in acclimation was encountered.

It is worthwhile to note that the chemical structure or the specific functional groups of a particular contaminant may also play an important role in its biodegradation behavior.

3.2. RE-ACCLIMATION AFTER BACKWASHING AND NON-USE PERIODS

Effluent gas samples were collected at prescheduled intervals to evaluate the biofilter response immediately subsequent to backwashing and non-use periods. Table III shows the biofilter re-acclimation with time under different operating conditions for the two concerned VOCs.

It is seen from Table III that when the loading rate did not exceed the biofilter critical loading rate (3.26 and 1.90 kg COD/m^3-day for MIBK biofilter and styrene biofilter, respectively), the performance of the hydrophilic MIBK biofilter recovered within 90 min to 99% removal efficiency, while the performance of the hydrophobic styrene recovered to the 99% level within 600 min. Furthermore, the biofilter re-acclimation was delayed with the increase of loading rate for both VOCs. The re-acclimation difference between hydrophilic VOC-MIBK and hydrophobic VOC-styrene is speculated to be due to reasons discussed in the previous section. For styrene, reacclimation after non-use was faster than after backwashing for all loading rates studied. For MIBK, reacclimation after non-use was faster than after backwashing only for loading rates up to 2.17 kg COD/m^3-day, while reacclimation after backwashing was faster than after non-use for higher loading rates. It is speculated that more active biomass during the non-use operation was available

TABLE III

Biofilter overall removal efficiency with time after different operating conditions (backwashing, starvation, and stagnant periods)

	Removal Efficiency (%)							
	MIBK				Styrene			
Time min	30	90	300	600	30	300	600	1200
	Inlet Concentration, 50 ppmv							
Loading rate	1.09 kg COD/m^3-day				0.64 kg COD/m^3-day			
Backwashing	99.5	99.9	99.9	99.9	62.5	95.9	98.3	99.5
Starvation	99.9	99.9	99.9	99.9	60.8	95.5	99.9	99.9
Stagnant	99.9	99.9	99.9	99.9	47.9	99.9	99.9	99.9
	Inlet Concentration, 100 ppmv							
Loading rate	2.17 kg COD/m^3-day				1.27 kg COD/m^3-day			
Backwashing	94.7	99.6	99.9	99.9	45.7	85.9	99.5	99.9
Starvation	98.3	99.9	99.9	99.9	64.9	94.4	99.9	99.9
Stagnant	93.2	99.9	99.9	99.9	82.2	99.9	99.9	99.9
	Inlet Concentration, 150 ppmv							
Loading rate	3.26 kg COD/m^3-day				–			
Backwashing	99.5	99.9	99.9	99.9	–	–	–	–
Starvation	–	–	–	–	–	–	–	–
Stagnant	–	–	–	–	–	–	–	–
	Inlet Concentration, 200 ppmv							
Loading rate	4.34 kg COD/m^3-day				1.90 kg COD/m^3-day			
Backwashing	95.0	98.2	97.4	96.0	99.9	99.9	99.9	99.9
Starvation	–	–	–	–	–	–	–	–
Stagnant	–	–	–	–	–	–	–	–
	Inlet Concentration, 250 ppmv							
Loading rate	5.43 kg COD/m^3-day				3.18 kg COD/m^3-day			
Backwashing	94.9	96.6	96.8	98.1	38.9	77.6	90.2	95.6
Starvation	91.2	91.4	88.6	88.4	84.2	93.4	96.1	98.1
Stagnant	78.5	87.8	83.1	91.3	96.0	95.7	89.1	91.3

than that during backwashing conditions. However, for the hydrophilic VOC MIBK, as the loading rate was increased beyond 2.17 kg COD/m^3-day, more accumulation of biomass during non-use operation led to channeling within the biofilter and eventually caused poor biofilter performance as compared to the backwashing strategy.

3.3. REMOVAL PROFILE ALONG BOFILTER DEPTH

One day following backwashing and non-use periods, gaseous samples were taken along the media depth of the biofilter to assess removal along depth and depth utilization for both MIBK and styrene biofilters. Figure 1 represents plots of removal profile along biofilter depth for the three conditions studied (backwashing and the two non-use periods) for both biofilters. The removal efficiency along the depth adopted in Figure 1 is the mean value from all cycles conducted for each loading rate and the operating condition. The error bars represent the standard deviation.

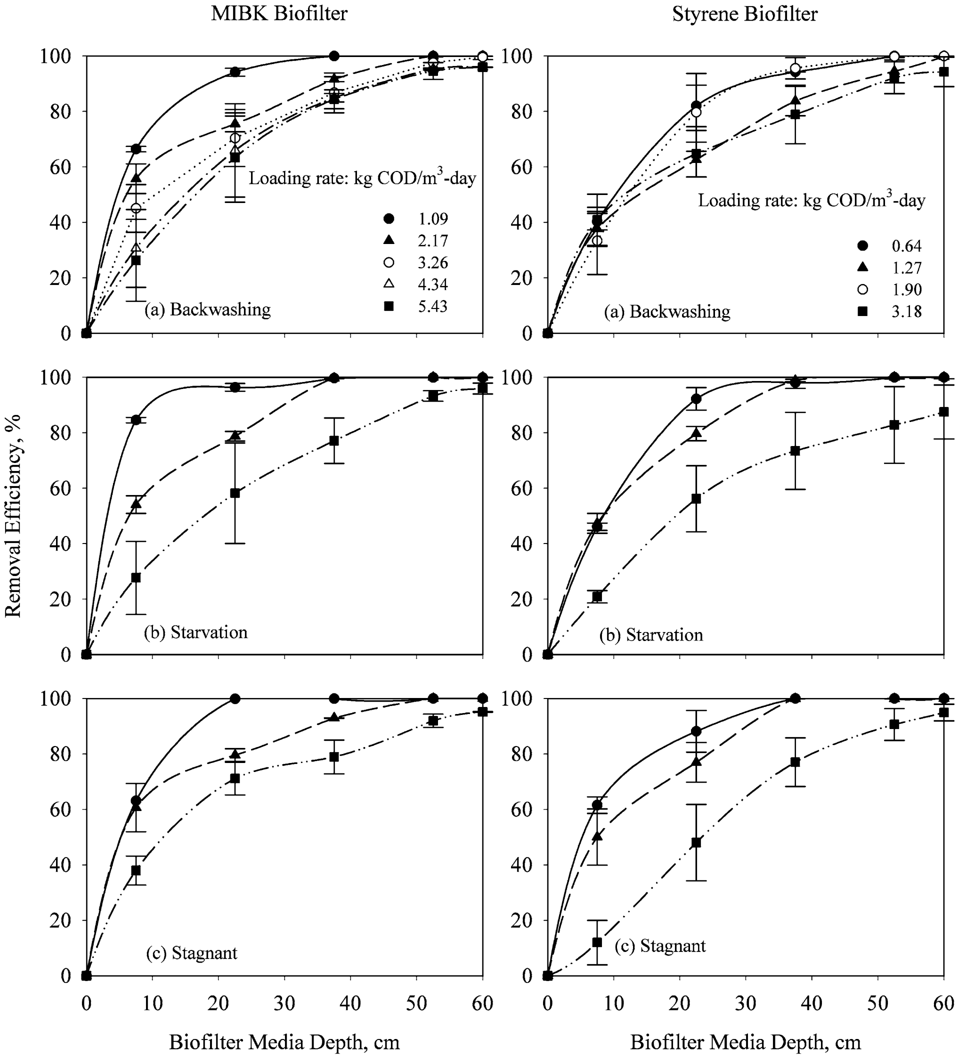

Figure 1. VOC removal along biofilter depth

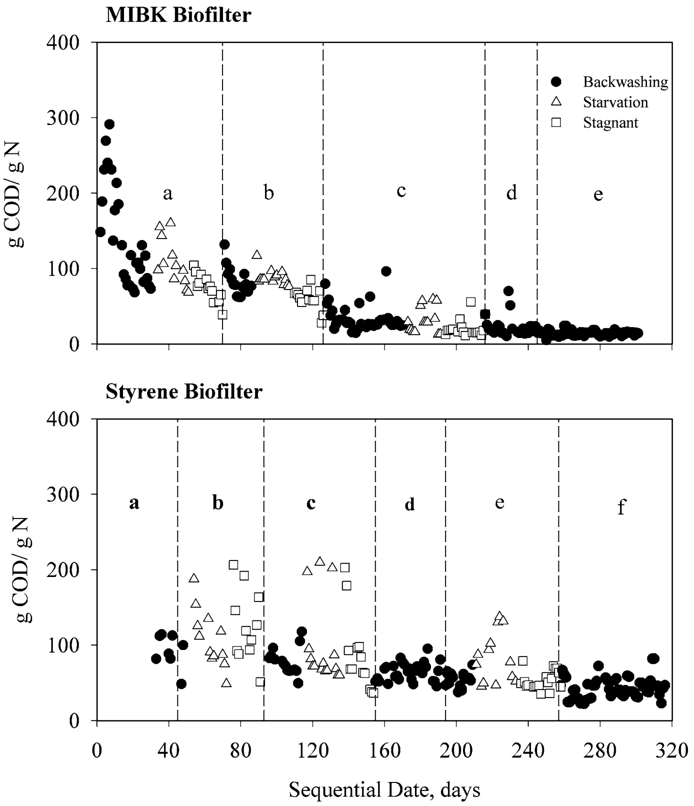

Figure 2. g COD/g NO_3^-—N with respect to time [MIBK biofilter loading rate (kg COD/m^3-day): a-1.09, b-2.17, c-5.43, d-4.34, e-3.26 (Cai *et al.*, 2005); Styrene biofilter loading rate (kg COD/m^3-day): a-1.27, b-0.64, c-1.27, d-3.18, e-3.18, f-1.90 (Kim *et al.*, 2005b)]

Figure 1 shows that the utilization of biofilter depth increased with the increase of VOC loading rate for both MIBK and styrene biofilters. It is speculated that the elimination capacity at a certain biofilter depth was reached for certain employed VOC loading rate, so when the loading rate was further increased, the utilization of biofilter depth increased correspondingly. Furthermore, it is seen that styrene degradation utilized more depth than MIBK under similar loading rate. This behavior further confirms the low critical loading rate of styrene as compared to MIBK.

3.4. NITROGEN CONSUMPTION AND VOC REMOVAL

Figure 2 shows the ratios of COD removed to NO_3^-—N consumption plotted against the sequential date for both biofilters. The nitrogen utilization behavior for MIBK biofilter and styrene biofilter is significantly different as seen in Figure 2. The ratios for MIBK biofilter show dependency on the employed loading rates and sequential time at low loading rates and as the loading rate was increased, the ratio of COD/N decreased and eventually was independent on time within a specific loading rate. On the other hand, the ratios for styrene biofilter are independent on loading rates and time for the backwashing operation, but the ratios are significantly dependent on loading rates and time for non-use operations. For both MIBK and styrene, the ratios that were dependant on time at a specific operational cycle (backwashing or the two non-use periods) showed a high ratio value on the first day after restart and then decreased with time to an asymptotic value.

It is worthwhile to note that NO_3^- was the only nitrogen source for both biofilters and any nitrogen consumption is mainly utilized for biomass synthesis. It is speculated that the apparent difference in nitrogen consumption between MIBK and styrene could be related to the different microbial cultures. Microbial analysis in our future studies will challenge the "black box" for biofiltration. Bacterial community analysis and identification of dominant microorganisms for the two biofilters will aid in understanding the difference in nitrogen utilization for the two VOCs.

3.5. CO_2 PRODUCTION AND VOC REMOVAL

The molar ratio of CO_2 production to VOC removal with respect to time is presented in Figure 3 in order to assess the extent of strict aerobic biodegradation for hydrophilic MIBK and hydrophobic styrene. The experimental average ratio of CO_2 production to VOC removal is equal to 3.41 ± 0.84 for MIBK biofilter and 6.08 ± 1.00 for styrene biofilter. The stoichiometric ratios for complete chemical oxidation of these VOCs are 6 and 8 for MIBK and styrene, respectively. The discrepancy between these experimental values and theoretical values is typical during the process of biodegradation, since some of the removed carbon is converted into biomass for microbial growth. Nonetheless, the relatively small difference between the theoretical ratio and the experimental ratio for styrene indicates that the hydrophobic styrene was eliminated by aerobic biodegradation rather than by any other physical or chemical process. On the other hand, the relatively high discrepancy between these two ratios for MIBK indicates that the hydrophilic MIBK favored biomass growth and accumulation in the biofilter and a possibility of denitrification may have occurred (Zhu *et al.*, 2004). It is interesting to note that the change in CO_2 production at a particular loading rate is in the opposite direction as outlined in nitrogen utilization, i.e., a low CO_2 production on the first day after restart from backwashing or the two non-use periods was encountered and then increased with time till it reached an almost asymptotic value. The correspondence between CO_2 production and nitrogen consumption is speculated to be due to the synthesis of biomass immediately after backwashing or non-use periods by utilizing the VOCs as carbon source for buildup of biomass and later as an energy source for maintaining microbial viability.

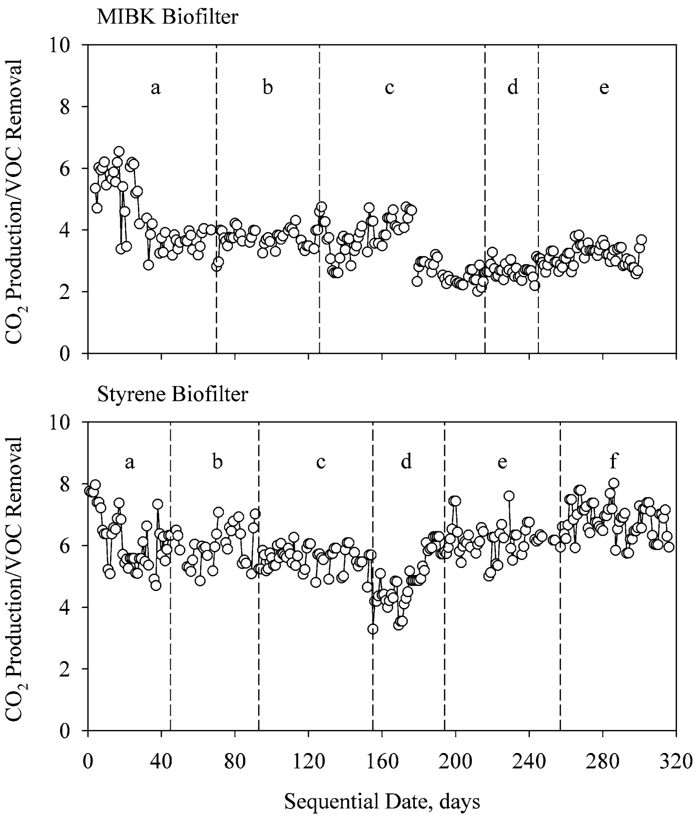

Figure 3. CO_2 production/VOC removal with respect to time [MIBK biofilter loading rate (kg COD/m^3-day): a-1.09, b-2.17, c-5.43, d-4.34, e-3.26; Styrene biofilter loading rate (kg COD/m^3-day): a-1.27, b-0.64, c-1.27, d-3.18, e-3.18, f-1.90]

Conclusions

This study compared the behavior of a hydrophilic-MIBK and a hydrophobic-styrene in a TBAB operated under three operating conditions, namely, backwashing and two non-use periods (starvation and stagnant). Long-term performance, analysis on nitrogen consumption, and CO_2 production revealed the following:

(1) Hydrophilic MIBK required much shorter initial acclimation periods than hydrophobic styrene. Furthermore, MIBK required lower EBRT to maintain long-term and stable 99% removal efficiency than styrene.

(2) Performance of styrene TBAB could be improved by increasing the nutrient flow rate.

(3) Non-use operations could be considered as another means of biomass control for both MIBK and styrene when the employed loading rate did not exceed their critical loading rates.

(4) Under similar loading rate, styrene utilized more biofilter depth as compared to MIBK.
(5) Nitrogen consumption for hydrophilic MIBK and hydrophobic styrene was significantly different which could be contributed to the different microbial communities within the two biofilters.
(6) CO_2 production with respect to VOC removal for hydrophobic styrene was closely related to the theoretical complete chemical oxidation value while high discrepancy was encountered for hydrophilic MIBK.

It is worthwhile to note that the results obtained for MIBK and styrene in this study cannot be generally expanded for all hydrophilic and hydrophobic VOCs. The chemical structure of the VOC may play an important role. For example, some VOCs with very low Henry's law constant can not be degraded in biofilter systems although they can be easily biodegraded in traditional activated sludge treatment systems e.g. methyl tertiary butyl ether (MTBE) (Salanitro *et al.*, 1994).

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