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Transient response of vapor-phase biofilters

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Abstract

Biofilters are a relatively inexpensive management method for biodegradable gas-phase contaminants, capable of meeting stringent air quality requirements. However, the nominally plug flow configuration of conventional unidirectional flow (UF) biofilters results in the microbial community having higher population densities and greater activity near the inlet. When transient loadings occur in conventional biofilters either the mass transfer capacity or the reaction capacity of the initial sections of the bed may be exceeded and contaminants move into the downstream sections where the microbial populations and reaction capacities are low and contaminant breakthrough may occur. Flow-directional-switching (FDS) operation incorporates the advantages believed to accrue from feast/famine operation of microbial processes and improved mass transfer characteristics of uniformly distributed biofilms. In this study, step function changes in toluene concentration were applied to UF and FDS laboratory reactors operated in parallel. Contaminant concentration was monitored at several points along the packed beds. FDS operation produced a more uniform (dispersed) distribution of biomass and microbial reaction capacity along the length of the packed bed without diminishing activity and removal capacity in the inlet section. Maximum mass removal rates in the FDS biofilter were approximately twice that of the conventional UF biofilter. In addition, FDS operation significantly improved biofilter response during an extended period of operation with intermittent feed and following a period of non-operation.

Keywords: VOCs; Biofilter; Biofiltration; Biological treatment; Vapor-phase contaminants; Air pollution control technology; Flow-directional-switching; Transient loading; Toluene

1. Introduction

Biological treatment is an attractive approach for removing volatile contaminants in gaseous waste streams. The most widely used biological air treatment process is the biofilter, a nominally plug flow, packed bed reactor in which a microbial community growing on the packing surface carries out the degradation of the contaminants. The first application of biological air treatment reported in the United States literature was by Pomeroy [1], who was conducting odor control experiments at municipal wastewater treatment plants. Use of biological processes for odor control has become fairly common and odor control remains the most widespread application of biofiltration [2,3]. Treatment of industrial and commercial waste air streams was initiated by Ottengraf and Vanden Oever in the 1980s and numerous examples of biological treatment for volatile organic compound (VOC) removal have been reported since that time [4–6]. Biological treatment systems can be significantly less expensive to build and operate than conventional treatment (e.g., combustion or adsorption), but the reactors tend to be larger. Issues that must be addressed for biological treatment to have widespread acceptance include questions about process reliability, the establishment of general or standardized design and operational parameters, and the absence of models that can predict steady-state and transient loading responses with a reasonable degree of accuracy. A major factor in process reliability is the ability of biological processes to respond to transient loadings.

Microbial population density in biofilters is related to the availability of substrate or nutrients, and has been shown to decrease by one to four orders of magnitude between the inlet and outlet when systems are operated under nom-

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inally steady-state unidirectional loading conditions [7–9]. The conventional system is inherently unresponsive to transient loadings because of the distribution of microbial activity along the length of the reactor. When large transient loadings occur, either the mass transfer capacity or the reaction capacity of the initial sections of the bed are exceeded and contaminants move into the latter sections where the microbial populations and reaction capacities are low and contaminant breakthrough may occur [10–12].

Most work to date has been focused on steady-state operating conditions. However, based on observations of both field and laboratory units, transient loadings are common and often result in increased bed penetration and contaminant breakthrough [13–25]. Typically, airflow rate is constant during the operation of vapor-phase biofilters and transient loading results from variation in contaminant concentration. However, applications where significant variation in contaminant concentration and gas flow rate do exist, as was the case at a soil–vapor extraction (SVE) operation in Hayward, CA, during the first 7-months of biofilter operation [13]. Transient loadings in biofilters have been studied by a number of researchers [8–11,13–42]; however, in most cases, the experimental conditions were limited and control strategies were addressed in only a few studies (discussed below).

The occurrence of transient loadings can be characterized with two parameters, regularity and frequency. For the purpose of this discussion regularity refers to occurrence on a cyclical or predictable schedule. Regular transients are the most common type due to normal cycles in process operations, overnight and weekend closures and scheduled maintenance activities. Normal cycles in process operations include intensification of work (e.g., higher rates of solvent use in a manufacturing process) and changes in operation throughout an operating shift of manufacturing and commercial operations such as paint spraying, baking, coating and chemical manufacture. Regular transients can occur on an infrequent basis due to quarterly, semi-annual or annual scheduled shut downs for plant maintenance and retooling. “Irregular” transient loadings result from random or erratic events such as spills and unplanned shut downs. Irregular transient loadings have been observed in emissions from wastewater treatment plants where relatively small diurnal transient loadings were overshadowed by occasional large spikes at intervals as long as several months [10]. Further characterization of transient loading can be accomplished by establishing a peak-to-baseline loading ratio and, for discontinuous feeding, length of feed-on and -off-time intervals, as illustrated in Fig. 1(a and b), respectively. For frequent transients, a peak-to-mean loading ratio parameter may be more appropriate than peak-to-baseline ratio, as illustrated in Fig. 1(c). The simplistic step-function patterns shown in Fig. 1(a–c) are used to illustrate the peak-to-baseline (or peak-to-mean) loading concept and have been used in laboratory studies, but they do not reflect the wide array of complex concentration patterns typically observed in field units.

Characterization of transient loading response and development of operating strategies to manage transient loadings to minimize contaminant breakthrough in biofilters is important because: (1) many potential applications for biofilters will be at facilities having inherent variation in waste gas characteristics, (2) appropriate performance monitoring requirements should reflect actual operating characteristics and (3) unsteady performance is a major drawback of biological

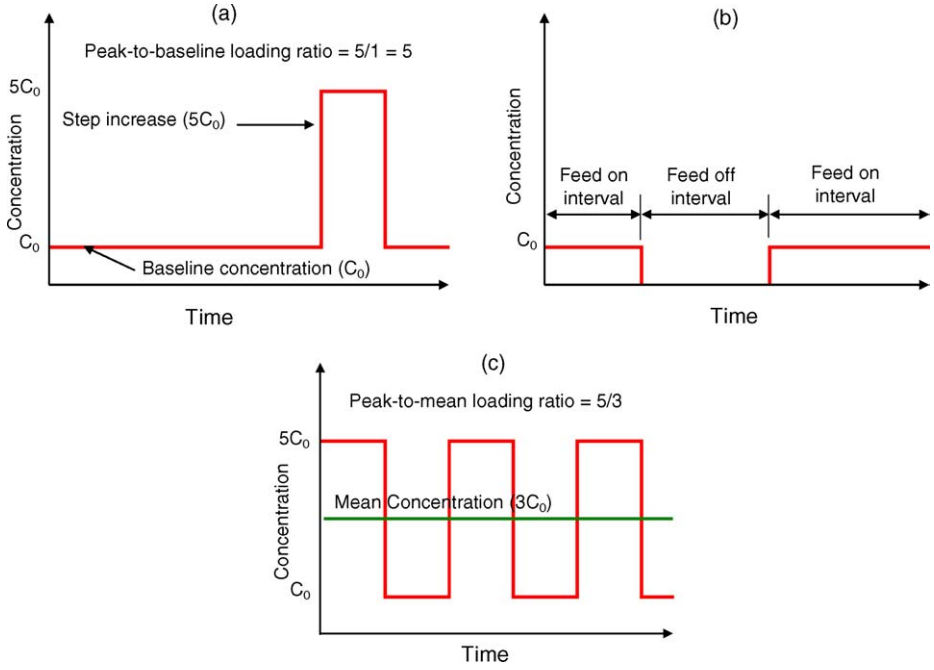

Fig. 1. Illustration of step-loading (a), off-time (b) and regular step-loading (c).

treatment relative to that of conventional physico-chemical treatment technology where performance is more stable and predictable.

1.1. Transient loading management strategies

Microbial populations in biofilters will respond satisfactorily to transient loadings within certain boundaries of elevated concentration and length of off-time. However, at some ratio of peak-to-baseline (or peak-to-mean) inlet concentration, breakthrough can be expected. It should be noted that if a step increase in contaminant concentration is sustained, contaminant removal rates will increase over time as the population acclimates and increases in number. The process is slow and typically takes several days or weeks to complete [26–28]. A corresponding process exists with respect to downtime. When a unit is shut down for an extended period of time the microbial population begins to go dormant and increased penetration of contaminants will occur on start up. With time, days to weeks depending on the length of the shut-down period and other factors, performance returns to normal [28,35]. Short-term shut downs of a few hours do not appear to affect biofilter performance upon restart [8,9,29–34].

Strategies for managing transient loadings in biofilter systems include providing downstream polishing units; dampening variations in contaminant loading using sorbent material upstream or in the biofilter bed; supplemental feeding during extended periods of downtime; and maximizing reactor reaction capacity. The first three management strategies are given a brief introduction here and the fourth strategy (maximizing reactor reaction capacity) is discussed in greater detail here (and in Sections 3 and 4).

(1) *Downstream polishing units*. Inclusion of downstream polishing units (e.g., activated carbon) to remove fugitive emissions can be used to insure regulatory standards are met at all times. While increasing capital, operation and maintenance costs, this provision will improve system performance and system reliability.
(2) *Physical damping of contaminant concentration*. Physical damping can be accomplished by installing an upstream load-dampening unit containing absorbent liquid [36] or adsorbent solid [42,43], or by adding absorbent/adsorbent material as a partial or sole component of the biofilter packing [15,28,37–39,42]. Performance may become limited if granular activated carbon (GAC) is used with humid waste streams because capillary condensation occurs and GAC surfaces become coated with water that reduces contaminant adsorption capacity and slows contaminant mass transfer rates to micropore regions.
(3) *Feeding contaminant or surrogate compound as a supplement during extended "off-periods" or periods of low inlet concentration*. Feeding contaminant or a surrogate compound to the biofilter as a supplement during "off-periods" has been accomplished successfully [40] and the approach may be a reasonable if the substrate compound(s) can be stored and fed to the system economically.
(4) *Increasing the average reaction capacity of the biofilter*. Several methods for increasing biofilter reaction capacity have been proposed, for example, those found in Refs. [8,44–46]. In addition to those methods (and others), the average reaction capacity of a biofilter may be increased by reversing the direction of flow through the media bed at regular intervals [41].

Maintaining a more uniform microbial population throughout the media bed by alternating the direction of flow would be expected to provide higher levels of reaction capacity along a greater fraction of the bed's length by increasing the microbial population density in the downstream half of the bed and maintaining that population in an active state. Song and Kinney [41] demonstrated that both biomass and biomass activity decreased along the length of a unidirectional flow (UF) biofilter and were maintained relatively constant in a flow-directional-switching (FDS) biofilter. A similar finding was observed in this study (discussed in Sections 3 and 4). The benefits derived from FDS should be realized for all types of transient loading, including, variation in contaminant loading and restart following extended periods of downtime. In both cases, relative to conventional unidirectional biofilter operation, additional reaction capacity is available whenever contaminants penetrate deeper into the bed. Park and Kinney [32] demonstrated some improvement in transient loading response of a FDS biofilter with the addition of a slip-feed system.

1.2. Research objectives

The primary objective of this study was to provide quantitative information on the extent of the benefit of flow-directional switching. Three aspects of the benefit were investigated: (1) response to step-loading (major emphasis), (2) response following down time (minor emphasis) and (3) long-term stability during regular on/off cyclical loading (minor emphasis). Step function increases in contaminant concentration pattern were applied to FDS and conventional UF biofilters operated in parallel. Following those experiments both biofilters were subjected to a shut-down period, then a feed-on/-off regular transient loading pattern for an extended period of time. For all loading cases, toluene was used as the model compound and response was assessed by monitoring toluene vapor concentration at several points along the packed beds. Secondary objectives of the study were to determine the distributions of biomass and moisture content in the packed beds.

Increasing our understanding of transient loading response and development of operating strategies to minimize breakthrough will allow more extensive application of vapor-phase biofiltration technology. Information developed in this study should provide a more complete basis for establishing monitoring regulations for vapor-phase biofiltration systems.

2. Materials and methods

A schematic representation of the experimental biofilter system is shown in Fig. 2. Laboratory compressed air was filtered through two microfiber filter-regulators in series, humidified, then passed through a rotameter to measure and regulate gas flow rate. A syringe pump was used to deliver liquid toluene to a glass–wool wick, where it evaporated into one air-stream which then combined with a second air-stream containing 10 μm aerosolized nutrient solution generated by a Heart nebulizer™ (Vortran Medical Technology Inc., Sacramento, CA). The aerosolized nutrient solution supplied inorganic nutrients and moisture to the biofilter beds. Nutrient solution consisted of a custom recipe (Table 1) in which major and minor nutrients were assumed to be present in excess. Pressure gauges were located at the outlet of the rotameter and the inlet of the Heart nebulizer™. The combined air-stream (containing toluene vapor and nutrient aerosol) was conveyed to the inlet of the FDS biofilter packed bed (top or bottom depending on the cycle phase) using a double solenoid, four-way, five-port valve and electronic controller. The other combined air-stream was conveyed to the top of the UF biofilter bed. Biofilter columns were constructed of 15-cm i.d. stainless steel pipe sections. Each column consisted of four 25-cm long media bed sections in series separated by 5-cm deep plenums. Media bed sections were supported by perforated plates. Sample ports fitted with Teflon-lined septa were located in the plenums. The media beds consisted of a mixture of new media (approximately two-thirds of the bed, volume basis) and used media (originating from a FDS biofilter used in prior experiments). The packing media was 0.64-cm diameter rigid mineral (extruded diatomaceous earth) cylindrical pellets (Celite R-635, Janus Scientific Inc., Fairfield, CA).

Table 1
Nutrient solution recipe[a]

Mineral salt	Concentration (mg/L)
KH_2PO_4	1390
K_2HPO_4	1710
$NaNO_3$	12500
$MgSO_4{\cdot}7H_2O$	460
$CaCl_2{\cdot}2H_2O$	17.6
$FeSO_4{\cdot}7H_2O$	1.0
H_3BO_3	0.30
$CoCl_2{\cdot}6H_2O$	0.20
$ZnSO_4{\cdot}7H_2O$	0.10
$MnCl_2{\cdot}4H_2O$	0.03
$Na_2MoO_4{\cdot}2H_2O$	0.03
$NiCl_2{\cdot}6H_2O$	0.02
$CuCl_2{\cdot}2H_2O$	0.01

[a] Recipe from Professor Kate Scow of the Department of Land, Air and Water Resources, University of California, Davis, CA, except concentration of phosphate buffer and form and concentration of nitrogen were modified. Mineral salts were dissolved in NANOpure™ or deionized water.

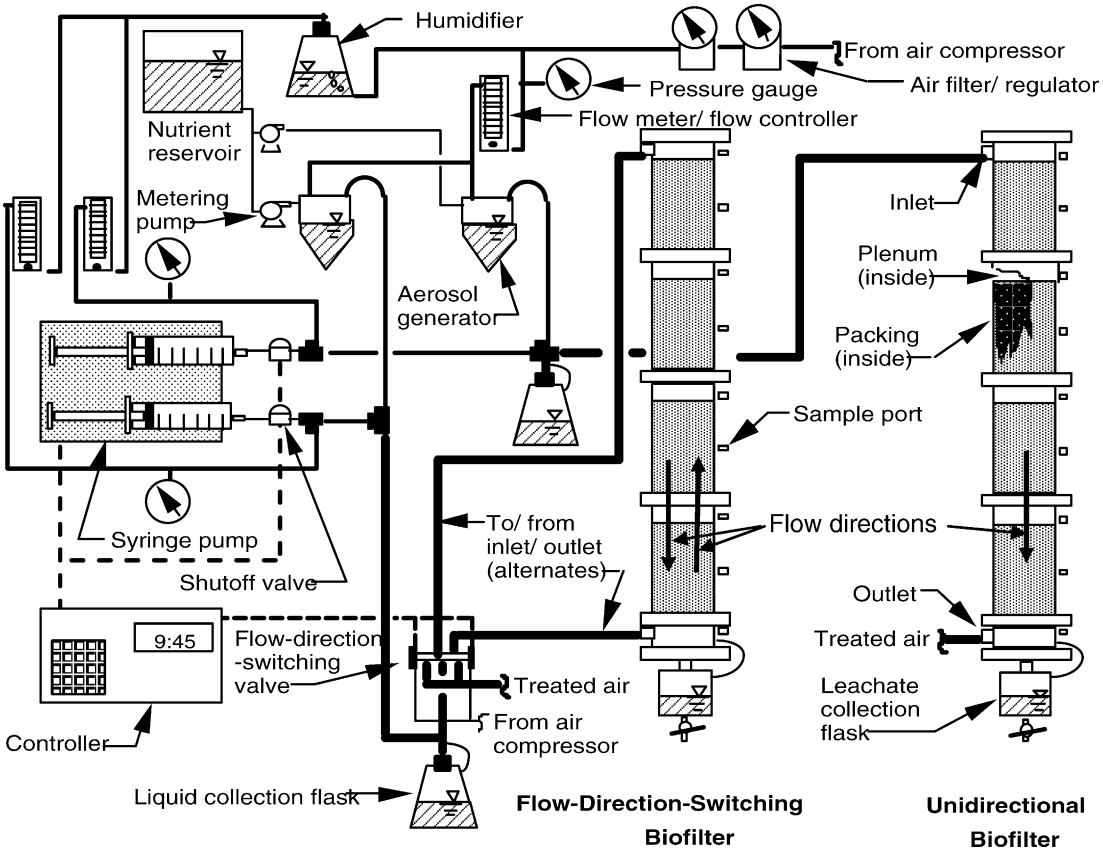

Fig. 2. Experimental biofiltration system schematic.

Measured pellet dimensions were highly variable with a median pellet length of approximately 0.8-cm.

2.1. Air-stream sampling and gas chromatography analysis

Flow-stream grab samples were collected from column ports at the inlet and at bed depths of 12.5, 25, 50, 75 and 100-cm (outlet) using 5-cc "gas-tight" Teflon Luer-lock syringes equipped with Mininert valves and Luer needles. Samples were analyzed for toluene concentration within 15 min by direct syringe injection into a Shimadzu 14A gas chromatograph equipped with a 0.5 mL sample loop, 30-m J & W Scientific DB-624 megabore column and flame ionization detector (FID). Blanks were used for quality control and toluene standards of 92.6 and 491 ppm_v (±2%) were used for toluene concentration determination (Scott-Marrin Inc., Riverside, CA).

2.2. Biofilter operation and maintenance

The biofilters operated with a nominal air-stream flux of 1 m^3/m^2 min, baseline inlet toluene concentration (C_0) of 107 ppm_v, empty-bed residence time of 1-min. and media bed temperature of 23 °C. The FDS biofilter operated on a 12-h FDS interval length throughout the study. In related experiments, it was found that a 12-h flow reversal interval was sufficiently short to maintain the toluene-degrading microbial community in a fully active state [47].

2.2.1. Operation and maintenance of step-loading experiments

Prior to initiating the experiments the packing media was washed with tap water, soaked in nutrient solution containing inoculum for 10 min, then brought to a pseudo-steady-state "mature" condition over a 13-day period with constant loading at the baseline inlet toluene concentration (107 ppm_v). The step-loading experiments were conducted over the next 87-days. For the purpose of this study, "mature" refers to a condition in which contaminant fractional removals in the first 25-cm of the reactor bed exceeded 85%. Nutrient solution was recirculated through the biofilter columns approximately once per week to maintain high removal efficiencies in the inlet region of the reactor bed. During step-loading events, the inlet concentration was increased for 1 h. Step concentrations of 2.5-, 4-, 5-, 10-, 20- and 50-times the baseline concentration were used. Three sets of baseline samples were obtained within a 2.5-h period prior to each step-loading event (the third set approximately 20 min prior to initiating the step) and three sets of transient loading samples were obtained during each step (approximately 6, 30 and 59 min after initiating the step). One additional set of samples was obtained for the 10-fold spike 30 min after the inlet concentration was returned to the pre-step baseline concentration.

2.2.2. Operation and maintenance of regular transient experiments

A cyclical feed-on/-off transient loading experiment was conducted on the UF and FDS biofilters operated in parallel to investigate long-term response to regular transient loading. The regular transient loading pattern used was 1-h-on/7-h-off, which represents a peak-to-mean loading ratio (PTMLR) value of 8 and cycle period length of 8 h. Air, toluene vapor and nutrient aerosol were passed through the biofilter during the "feed-on" periods. During "feed-off" periods air and nutrient aerosol flow was continued. For each regular transient-loading experiment, gas-stream samples were collected at the same point in time within the FDS interval each day. Nutrient solution was recirculated through the packing when performance in the inlet region of the bed deteriorated significantly.

2.3. Methods for biomass and moisture content determination

Gravimetric analyses for water content and biological (volatile) solids content were conducted on the packing media at 25-cm intervals along the length of the beds of both biofilters after several days of operation with constant loading. Samples of approximately 4–8 mL volume were obtained in triplicate, placed in weighing tins and analyzed for weight change after being subjected to drying and incineration. Drying was conducted at 104 °C for 24-h and incineration at 550 °C for 24 h. One empty weighing tin and three tins with new (clean) media samples were analyzed concurrently as a control. Water mass was defined as change in sample mass upon drying. Biological (volatile) solids mass was defined as change in dried sample mass upon incineration. Results were reported on the following basis:

$$\text{water content} = \frac{\text{mass of water}}{\text{total (bulk) volume}}\ (\text{kg/m}^3)$$

$$\text{biomass content} = \frac{\text{mass of biological (volatile)solids}}{\text{total (bulk) volume}}\ (\text{kg/m}^3)$$

Total (bulk) volume is the gross volume of space occupied by the packing material (i.e., the interstitial void space plus the volume occupied by the pellets) and was estimated as "total (bulk) volume (mL) = [mass of incinerated sample (g)] × [1.5 mL/g]." The 1.5 mL/g "specific volume" conversion factor for "mass of incinerated sample" to "bulk volume of media" is the inverse of dry packing bulk density and was obtained by measuring the bulk volume and mass of pellets in a 500 mL beaker.

3. Results

Results of the transient loading experiments reported below indicate that, relative to conventional UF mode of

operation, FDS operation: (1) produced a more uniform distribution of biomass and microbial reaction capacity along the length of the packed bed without diminishing activity and removal capacity in the inlet section; (2) significantly improved biofilter response during step-loading events, following a shut-down period and during an extended period of operation with intermittent feed. These results are discussed further below.

3.1. Step-loading experiments

Prior to imposing step increases in contaminant concentration, the biofilter beds were brought to a pseudo-steady-state response condition at a nominal baseline inlet contaminant concentration (C_0) of 107 ppm_v. Response of the UF and FDS biofilters, operated in parallel, was assessed at baseline and 2.5-, 4-, 5-, 10-, 20- and 50-fold step increases in C_0. Variation of toluene concentration over time at the inlet and at five bed depths is given for a five-fold step in Fig. 3(a and b) for the UF and FDS biofilters, respectively. Inspection of Fig. 3(a and b) reveals that greater than 85% of the pre-step (baseline) toluene concentration was removed in the first 25-cm of bed depth and essentially 100% was removed by mid-depth (50-cm). When step-loading events were initiated, concentration profiles within the bed increased rapidly and achieved pseudo-steady-state response within 6 min, which represents the time between the initiation of the step and the collection of the first set of step-loading samples. Variation in syringe pump output resulted in some variability in inlet (feed) and response concentration profiles, as can be seen in the figures. During the five-fold step increase, toluene concentrations at mid-depth, 75-cm, and at the outlet were approximately the same value in the UF biofilter because reaction capacity of the second half of the bed was minimal and additional removal was not possible. In contrast, additional treatment was possible in the second half of the FDS biofilter media bed. Effects of toluene sorption and desorption within the packed bed during and following transient loading events was not evident within the time-limits/resolution of measurement procedures used in this experiment, which was 6 min at the start of the step (all experiments), and 30 min following the step (10-fold step only, data not shown). A lag in attaining elevated concentration profiles within the bed at the start of the step would be indicative of sorption effects and a lag in attaining lower concentrations in the bed following the cessation of the step would be indicative of desorption effects.

Profiles of average removal efficiency across the bed for both biofilters are shown in Fig. 4 for steady-state constant (baseline) loading and for several magnitudes of step-loadings. Results for the four-fold step experiment were not shown in order to improve figure clarity. The solid line in each plot represents steady-state loading response profiles. Mean values were calculated from the results of the six step-loading tests (three samples per test for a total of 18 samples for each bed depth). The dashed lines in each plot represent transient (step)-loading response profiles based on the average of three samples at each bed depth. Biofilter response profiles were similar in both biofilters during the 2.5-fold step with complete removal occurring within the first 75 cm of bed depth. Reaction capacity in the first half of the UF biofilter bed was exceeded during the four-fold step (not shown) and contaminants were carried deeper into the bed and emerged at the outlet (average value 56.6 ppm_v or 13.2% of C_0). In contrast, breakthrough was minimal in the FDS biofilter (average value 2.4 ppm_v or 0.6% of C_0). For the five-fold step, the response profile to the right of the 50-cm depth marker was relatively flat when compared to the FDS biofilter response profile because, as noted above, reaction capacity of the second half of the UF biofilter bed was minimal. In contrast, additional treatment was possible in the second half of the FDS biofilter bed, and the mean contaminant concentration at the outlet was minimal (2.6% of C_0) when compared to the UF biofilter

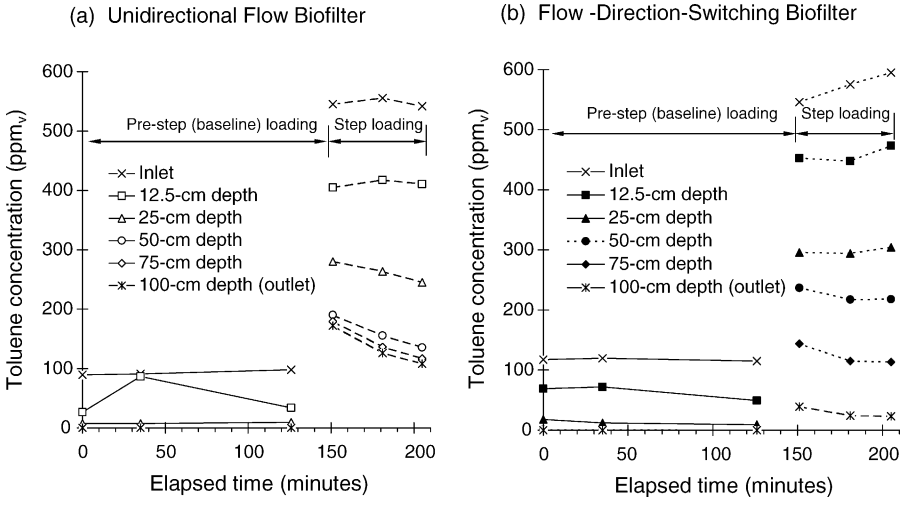

Fig. 3. Response of the conventional unidirectional flow biofilter (a) and the flow-directional-switching biofilter (b) to baseline and five-fold step transient loadings.

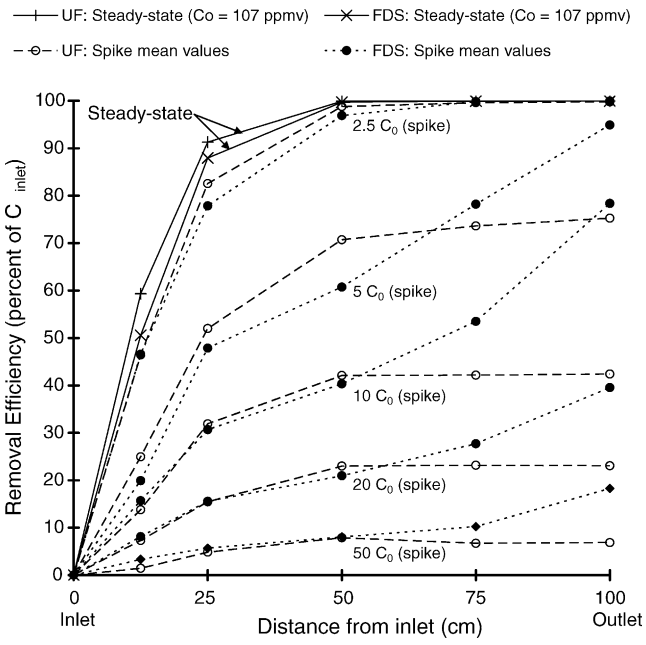

Fig. 4. Response of a conventional unidirectional flow (UF) biofilter and flow-directional-switching (FDS) biofilter treating toluene to steady-state loading and step transient loading as functions of bed depth. The solid lines represent steady-state response profiles in which mean fractional removal values at each bed depth were calculated from the results of six step-tests with three samples per bed depth per test (i.e., a total of 18 samples for each bed depth). The dashed lines represent step-loading response profiles in which mean values at each bed depth are connected. Mean values were calculated from the results of one step-test (a total of three samples for each bed depth).

(26% of C_0). For the 10-fold and larger steps, large fractions of the feed contaminant broke through both biofilters; however, the magnitude of breakthrough was significantly less in the FDS biofilter due to the larger reaction capacity of that unit. It is interesting to note that overall removal efficiency in the FDS biofilter during the 10-fold spike was larger than for the UF biofilter during the five-fold spike (78% and 75%, respectively). Mass removal in the FDS biofilter was 90% greater than that of the UF biofilter for the 10-fold spike and 71% greater for the 20-fold spike and 180% greater for the 50-fold spike. However, the comparison of UF and FDS biofilter performance for the 50-fold spike is tenuous due to a large degree of scatter in the UF biofilter data (percent removals of 15.3, 4.90 and 0.40 for the UF biofilter as compared to 21.6, 16.5 and 16.7 for the FDS biofilter). The scatter was believed to have been caused by unsteady syringe pump operation. A more complete characterization of the step-loading response data and additional details of the step-loading experiments can be found in Ref. [12].

Toluene removal efficiency profiles across the length of the UF biofilter bed were nearly identical to removal profiles across the FDS biofilter: (1) in the first half (50 cm) of the bed for all loading conditions (baseline and step) and (2) throughout the full bed (100 cm) for steady-state baseline loading at 107 ppm_v. Nearly identical removal profiles in the first half of the beds revealed that, relative to UF operation, operating with a 12-h FDS cycle did not diminish activity, i.e. a lag/re-acclimation period was not observed nor was removal capacity diminished in the first half of the bed. Clearly the FDS biofilter performance was superior to that of the conventional UF biofilter for spike magnitudes larger than approximately 2.5-times C_0 (as indicated by the larger fraction of contaminant removed in the FDS biofilter). Note that the decrease in toluene concentration in the first 25 cm of the FDS biofilter increases from approximately 95 ppm_v for the steady-state condition to approximately 340 ppm_v for the 20-times C_0 spike, although the fractional removal drops from 90 to 16%. Whether the first 25 cm became mass-transfer limited (including the possibility of oxygen transfer limitation) or reaction rate-limited cannot be determined from the data, however, Schroeder [20] suggested that toluene-treating systems were probably mass-transfer limited at lower concentrations.

Fractional removals at the biofilters' outlets are shown in Fig. 5 as a function of step-to-baseline concentration ratio. Each data marker represents the mean value of results of three sample measurements. Complete removal occurred for step-ratio values less than a threshold value and breakthrough occurred when that threshold was exceeded. Based on best-fit curves of post-threshold response data (power function for FDS data and logarithmic function for UF data), threshold step-to-baseline concentrations ratios for the UF and FDS configurations are approximately 3.3 and 4.2, respectively. For step-to-baseline concentrations ratios larger than a unit's threshold value, fractional removal declined in a non-linear manner with the UF biofilter response declining more rapidly than for the FDS biofilter.

Response of conventional UF biofilters to step-increases in toluene concentration has been documented previously [15–17,28,30,36], but published data on FDS biofilter response to step-increases in contaminant concentration appear to be limited to four studies by Song and Kinney [9,32,34,41]. A summary of results from other toluene step-loading studies is given below in Tables 2 and 3 for UF and FDS biofilters, respectively.

The two UF biofilter studies that used step-to-baseline concentration ratios exceeding ten (Tang et al. with a 37-fold

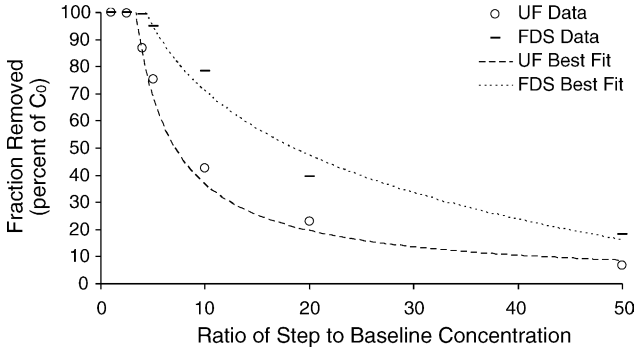

Fig. 5. Relationship between fractional removal and step-to-baseline concentration ratio. The baseline toluene concentration was 107 ppm_v. Each data marker represents the average value of data from three samples.

Table 2
Results from other UF biofilter toluene step-loading studies in which toluene was degraded

Study	Step-to-baseline concentration ratio	Fraction removed (%)
Al-Rayes et al. [36]	4	78
	4.3	83
Irvine and Moe [15]	10	73
Marek et al. [30]	3	76
	18	64
Métris et al. [16]	3.9	76
Moe and Irvine [17]	10	72
Tang et al. [28]	37	70
	1.8	50
	2.8	64

step and Marek et al. with an 18-fold step) resulted in remarkably large removal efficiencies (70 and 64%, respectively). In both cases, the pre-step baseline concentrations were relatively low (15 and 20 ppm_v, respectively). It is possible that, for the same step-to-baseline concentration ratio, lower values of pre-step baseline concentration results in larger fractions of the inlet concentration being removed during step-loading events than is the case for higher pre-step baseline concentrations. This could be the case if a larger fraction of the contaminant adsorbs to, or absorbs in, the packing at low contaminant concentration than is the case for higher concentrations. Other explanations are possible and additional study is needed to answer the question. A second possibility is that, prior to the studies, the biofilters had received higher concentrations of contaminant than the 15–20 ppm_v baseline concentrations indicated in the referenced article, which would have supported a larger population of toluene-degrading microorganisms with greater reaction capacity at the time the experiments were conducted. The response of the fungal biofilter in the Woertz et al. study to step-to-baseline concentration values ranging from 1.4 to 6.7 was remarkably similar to that of the FDS biofilter used in this study.

Table 3
Results from other FDS biofilter step-loading studies in which toluene was degraded

Study	Step-to-baseline concentration ratio	Fraction removed (%)
Park and Kinney [32]		
(Phase I)	1.2	100
	3.4	55
(Phase II)	1.5	100
	3.7	70
Song and Kinney [41]	1.75	[a]
Song and Kinney [9]	1.6	100
	3.6	74
Woertz et al. [34][b]	1.4	100
	6.7	82

[a] Breakthrough occurred in the UF biofilter but not occur in the FDS biofilter. The fractions removed were not reported.
[b] Fungal biofilter.

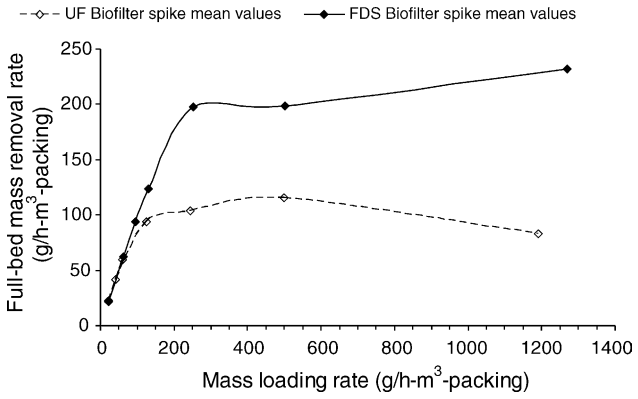

Fig. 6. Relationship between mass loading and mass removal rates in the full 100-cm of bed depth for step-to-baseline loading ratios of 1, 2.5, 4, 5, 10, 20 and 50. Each data marker represents the average value of data from three samples.

The relationship between mass loading and removal rates for the full bed (100-cm) and for the first 25-cm of the bed are shown in Figs. 6 and 7, respectively, for step-to-baseline loading ratios up to and including 50. Scales in the two figures differ because bed volumes used to calculate loading and removal rates differ by a factor of 4. Each data marker represents the average value of data from three samples. With the exception of the UF biofilter 50-fold step (in which unsteady syringe pump operation affected the results), mass removal rates increased with step magnitude and appear to asymptotically approach maximum values. It is possible that the functionality of the experimental apparatus was exceeded for step-to-baseline concentration ratios beyond 10 or 20. Maximum contaminant removal rates in the FDS biofilter were exactly twice as great as those observed in the UF biofilter (232 g/h m^3 for the FDS biofilter 50-fold step versus 116 g/h m^3 for the UF biofilter 20-fold step). In the first 25 cm of the bed mass removal rate curves were nearly identical with maximum contaminant removal rates of approximately 310 g/h m^3-packing (UF and FDS biofilters for 10- and 20-fold steps). This result shows that flow-directional-switching

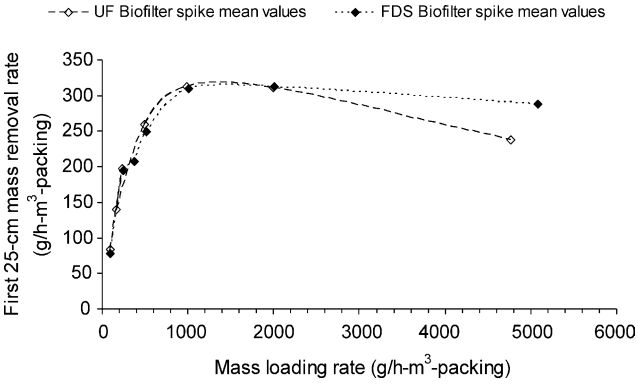

Fig. 7. Relationship between mass-loading and mass-removal rates in the first 25 cm of bed depth for step-to-baseline loading ratios of 1, 2.5, 4, 5, 10, 20 and 50. Each data marker represents the average value of data from three samples.

on a 12-h interval did not diminish reaction capacity in the inlet region relative to unidirectional flow.

3.2. Response to down time

Conventional UF and FDS biofilter response characteristics were monitored in the days prior to and following a 2.9-day down time event. Cumulative fraction of inlet toluene concentration (C_0) removed as a function of bed depth is shown in Fig. 8 for the UF biofilter (a, top) and the FDS biofilter (b, bottom). The units were operated with continuous feed (107 ppm_v) for 1.85-days after a nutrient recirculation procedure was performed, then shut down for 2.9-days. Continuous-feed operation was resumed on day 4. Data markers for the UF biofilter response at 50, 75 and 100-cm bed depths overlap at 100% removal prior to the shut down (as was the case in the FDS biofilter for the 75 and 100-cm bed depth markers). Fluctuations in C_0 produced variability in biofilter response, which was most pronounced near the inlet and attenuated with bed depth. The FDS biofilter 25-cm depth response was affected by switching the flow-direction (characterized by lower fractional removals in the period immediately following the switch and higher removals at the end of the switching interval). Overall (100-cm depth) response in the FDS biofilter was superior to that of the UF biofilter as evidenced by the large amount of contaminant that broke through the UF unit (40% 1 h after restart) relative to the significantly lower amount of contaminant that broke through the FDS unit (2.5% 1 h after restart). Performance improved with time in both units and complete removal was attained in the UF and FDS biofilters after approximately 12 and 3 h, respectively.

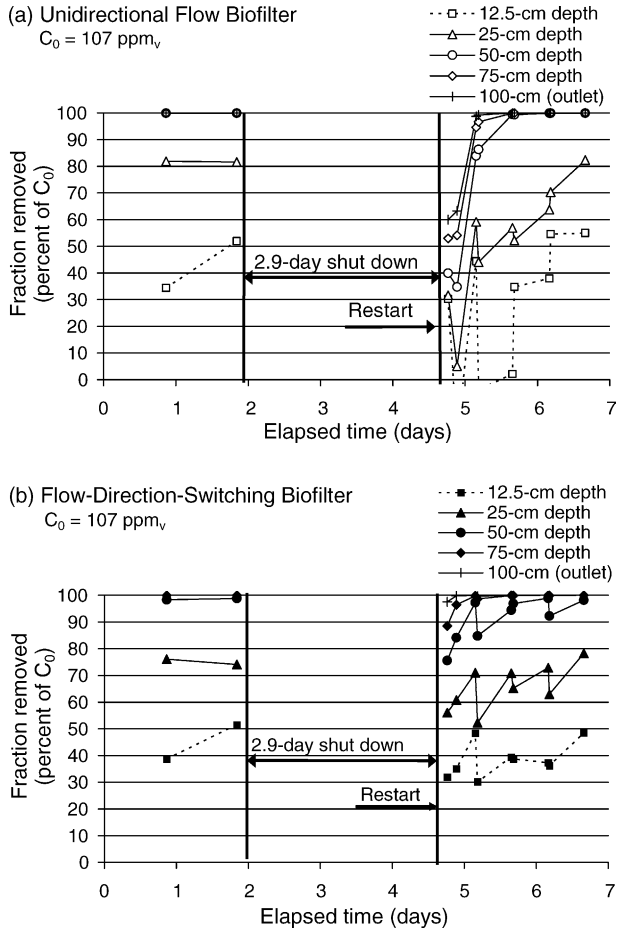

Fig. 8. Response of a conventional unidirectional flow (UF) biofilter (a, top) and flow-directional-switching (FDS) biofilter (b, bottom) treating toluene before and after a 2.9-day shut-down period.

3.3. Response to regular transient loading

A study investigating regular transient loading response was conducted by operating the UF and FDS biofilters in parallel on a regular 1-h-on/7-h-off transient loading pattern at a nominal C_0 value of 107 ppm_v. The biofilters had the same contaminant loading history prior to the experiment. The responses are shown in Fig. 9 for the UF biofilter (a, top) and the FDS biofilter (b, bottom). Complete removal occurred in the FDS biofilter and nearly complete removal occurred in the UF biofilter throughout the 26-day experiment. Following a brief decline in performance during the

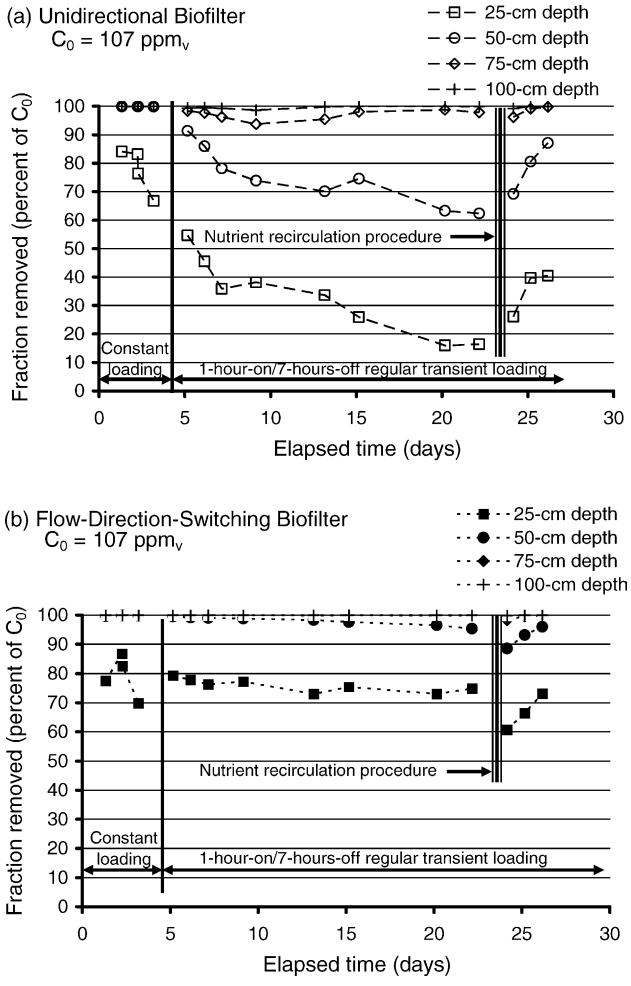

Fig. 9. Response (removal efficiency) of a UF biofilter (a, top) and the FDS biofilter (b, bottom) treating toluene to constant loading followed by 1-h-on/7-h-off regular transient loading.

constant loading period, the FDS biofilter exhibited stable response during the first 18 days of the regular transient loading period with fractional removals at a bed depth of 25 cm consistently above 70%. In contrast, fractional removals in the UF biofilter at a depth of 25 cm began at 55% at the start of the regular transient loading period and steadily declined to less than 20% over the next 18 days. A similar decline in removal efficiency with time can be seen at mid-depth (50 cm). A nutrient recirculation procedure was performed on day 23, during which time both biofilters were not operated for 24 h. A temporary drop in performance was observed within the FDS biofilter when feeding resumed (on day 24) as contaminant penetrated deeper into the bed. The performance of both units improved in the days that followed. The restorative effect of the nutrient recirculation procedure can be seen in the UF biofilter response on days 24–26. It should be noted that biofilter performance can be improved/restored with nutrient recirculation when using significantly shorter periods of recirculation times than the 12 h used for each unit here (e.g., a few hours). If the duration of the experiment were to have been longer without nutrient recirculation, the trend of declining performance would most likely have continued with contaminant breakthrough expected in the UF biofilter well before the FDS biofilter.

3.4. Biomass and moisture content

Results of biomass content and moisture content measurements are presented below in Fig. 10(a and b), respectively. Bed depth was measured from the top of each unit (depth = 0 cm) down to the bottom (depth = 100 cm). The analyses were conducted concurrently 34 days after the system had returned to normal operation following a media wash and mix procedure. However, the true operating time was significantly less than 34 days because the period included 10 days of down time (days 12–22) and four media recirculation procedures with a cumulative (total) down time of 83 h. Therefore, the effective operating time following the previous wash and mix procedure was approximately 21 days. Pressure drop across each unit was measured periodically and the maximum value measured was 0.4 in. of water column (and typically was much lower).

Inspection of Fig. 10(a) reveals that biomass (mass of volatile solids/bulk volume of packing) was concentrated near the inlet (0-cm depth) of the UF biofilter and was more uniformly distributed at much lower densities in the second half of the bed. The biomass profiles are in agreement with visual observation of biomass on the packing as it was removed from reactor sections when sampled. The presence of moderate levels of biomass in the second half of the UF biofilter is probably the result of the nutrient recirculation procedure which likely removed biomass from the inlet region and deposited in the latter half of the bed. The preceding conclusion is supported by the fact that, in the second-half of the UF biofilter bed: (1) biomass density was uniformly distributed with depth rather than decreasing with depth and (2) toluene removal rates were found to be minimal during the step-loading experiments. Both of these observations make it unlikely that the significant amount of biomass observed in the second-half of the UF unit originated (was grown) there. In contrast to the UF unit, the FDS biofilter biomass content profile was nearly symmetric with biomass concentrated near the ends of the media bed. The FDS biofilter 0-cm depth biomass value was greater than the 100-cm depth value and a similar result was observed by Song and Kinney [41]. This result (less biomass at the bottom of the FDS biofilter bed) may be real (for reasons that are not known), or it may be the result of sampling limitations and/or experimental error. Observations that support the latter conclusion include: (1) the packing at 100-cm depth was resting on a perforated plate (support grid) and, therefore, some of the pellet surface area was blocked from substrate flow which would have precluded biomass growth in that area of the bed and (2) 25-cm of packing had to be removed from the fourth column section in order to reach the 100-cm depth pellets resulting in mixing of pellets from different depths during the process (i.e., the samples may have been contaminated with pellets having lower biomass densities). Overall average biomass content values for the UF and

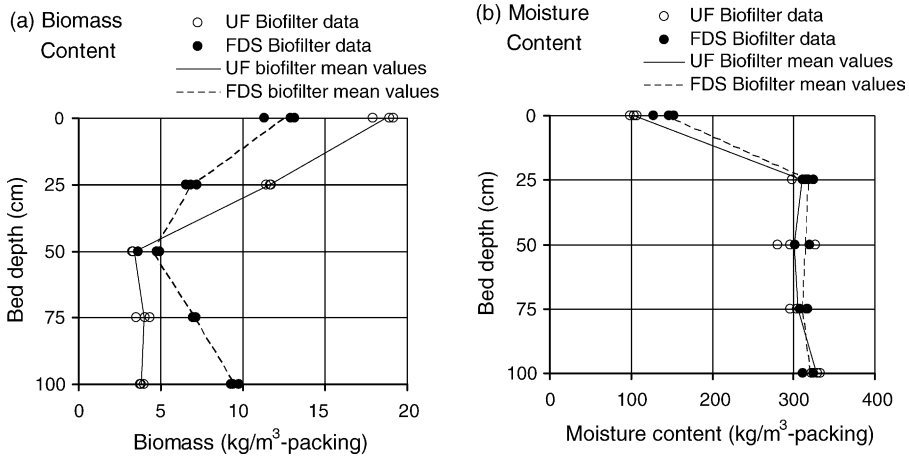

Fig. 10. Biomass content (a) and moisture content (pellets + biofilm) (b) profiles for the UF and FDS biofilters.

FDS biofilters, measured as volatile solids, were nearly identical at 8.3 and 8.1 kg/m^3-packing (or 146 and 142 g/unit), respectively. Song and Kinney made similar observations of nearly equal biomass levels in UF and FDS biofilters [9].

Profiles of media moisture content (mass of water/bulk volume of packing) for the UF and FDS biofilters in mature condition are presented in Fig. 10(b, right). As with the biomass profiles, lines in the figure connect mean values calculated from the results of three sample measurements. Biofilter water content profiles were similar and revealed: (1) a large amount of water held by the packing pellets, (2) pellets located on the top surface of the bed had significantly less water than pellets located below the surface and (3) water content was uniformly distributed within the bed below the 25-cm depth (and, though not measured, may have extended up to a depth near the surface). Average water content values for the UF and FDS biofilters were 270 and 281 kg/m^3-packing, respectively. Most of the moisture is located inside of the porous media and may not be associated with biomass. Thus, the low water content at the top of the beds does not appear to be reaction related (i.e., caused by reaction or affecting reaction rates) because water content of the FDS unit was low at the top of the bed and high at the bottom, but removals in the top and bottom sections were similar. It is possible that the lower water content at the top of the beds was due to drainage of unbound water or due to evaporation by inlet air that may have not been fully humidified.

4. Discussion

Evidence is mounting that biofilters subjected to regular transient loadings perform better during transient loadings than biofilters that have not [9,10,15,34,37,41,48]. In addition, flow-directional-switching appears to improve long-term operational stability, as was seen in this study and in the work of Song and Kinney [9]. Flow-directional-switching operation incorporates the advantages believed to accrue from feast/famine operation of microbial processes and improved mass transfer characteristics of biofilms that are more uniformly distributed (dispersed) throughout the length of the packed bed.

4.1. Feast/famine

Irvine and Moe [15] operated three UF biofilters in parallel with the same average loading rate, but one unit was fed continuously while the others were fed intermittently using a regular feed-on/-off transient loading pattern. The intermittently fed units responded better to shock-loading (higher removal efficiencies) than was the case for the continuously fed unit. Addressing one of two factors believed to be responsible for the improved performance, Irvine and Moe reasoned that intermittent feeding may alter the microbial community's physiological state, providing it with an enhanced ability to sorb and store (accumulate) contaminant or high-energy compounds during shock-loading events. The stored material is then utilized/degraded during rest periods. It is also possible that the resting period may allow for metabolic activities of the microbial ecosystem to "clean up" the resting biofilm, leaving it in an improved state when feeding resumes. Biofilm clean-up activities might include removal of waste products (by mass transfer and/or consumption by secondary organisms); replacement of depleted nutrients (by mass transfer and/or cell lysis during endogenous decay); increased level of cellular resources being directed towards cell maintenance and repair activities; or reduction of excess biomass (by endogenous decay and/or predation).

Flow-directional-switching is analogous to the intermittent (or sequencing batch) operation studied by Irvine and Moe in that alternating feast/famine sequences are introduced with downstream sections of the biofilter receiving virtually no feed half of the time and are thus allowed to "clean up." Thus, if physiological advantages result from feast/famine operation, FDS would be an appropriate management strategy based on simplicity. Results of this study (discussed above) and the work of Song and Kinney [9,41] appear to support this position. In the work of Song and Kinney [41], performance had deteriorated faster in a conventional UF biofilter treating toluene than in a 3-day FDS biofilter. Two reasons were cited: first, exposure to toluene inhibits microbial activity over time and the inlet section of the UF unit was exposed to toluene continuously, whereas each end of the FDS biofilter was exposed to toluene only half of the time (due to FDS operation). Second, biomass clogging occurred sooner in the UF biofilter than in the FDS unit (discussed below in the next section). In a subsequent study, Song and Kinney observed a rapid loss of biodegradation capacity and serious bioreactor instability when a FDS biofilter was operated on a 1-day FDS frequency, but greater stability in units operated on 3- and 7-day switching frequencies [9]. They concluded that frequent dynamic loading hindered biofilm development because, as concluded in their previous study, continuous or near-continuous exposure to toluene can inhibit microbial activity. If Song and Kinney's conclusion is correct (toluene exposure inhibits microbial activity over time), it may partially explain results of this study in which performance of the UF biofilter deteriorated over time faster than was the case for the FDS biofilter. Alternatively the result could have been due to biomass clogging (discussed below) and/or other factors such as differences in moisture content, pH, nutrient or oxygen availability, etc. In the present study, a 12-h direction-switching period resulted in stable operation, contrasted with the Song and Kinney studies, and was probably a result of frequent nutrient recirculation.

4.2. Mass transfer

In addition to improved physiological state of the microbial population, the improvement in transient loading response in intermittently fed and FDS biofilters may also be due to improved mass transfer characteristics of biofilms that are distributed throughout the bed. Although total biomass

levels measured in this study were approximately the same in both biofilters, contaminant-degrading capacity in the FDS biofilter was approximately twice that of the UF biofilter. The apparent dilemma may be explained by mass transfer considerations as follows: relative to the FDS biofilter, most of the active biomass in the UF biofilter was concentrated in a relatively small region of the bed (near the inlet) and the length of time that contaminant was in contact with active biofilm as it flowed through the bed was therefore limited. In contrast, active biomass in the FDS biofilter was more evenly distributed over the packing media throughout the length of the bed, which may have increased active biofilm/gas interfacial area (specific biofilm surface area) and certainly would have increased active biofilm/gas contact time in the bed. Both factors would increase overall mass transfer rates during transient loadings. Returning to the previous discussion of Irvine and Moe [15], the second reason cited for superior performance of intermittently fed biofilters is consistent with this position. They suggested that the contaminant mass flow rates used in their intermittently fed biofilters, which was greater than in the continuously fed biofilter, caused the growth of microorganisms to extend deeper into the bed. The presence of microorganisms deeper in the bed allowed contaminant removal to occur during shock loading that would otherwise pass through the bed untreated. Returning to the previous discussion of Song and Kinney [41], the second reason cited for the unidirectional-flow biofilter loosing performance faster than the FDS biofilter is also consistent with the position stated above. They suggested that excess biomass accumulation near the inlet end of the unidirectional-flow biofilter reduced the specific surface area of the biofilm and resulted in mass transfer limitations. In contrast, the distribution of biomass in the FDS biofilter was more uniform across the bed and, presumably, had superior mass transfer characteristics. Finally, Yang et al. [45,46] cited uniform distribution of biomass within the bed of a concentric layered media biofilter as a reason for increased reaction capacity and operational stability relative to that of a single-layer media biofilter. The latter was found to have poorly distributed biomass and significant gas-stream channeling.

5. Conclusions

The ability of conventionally configured UF biofilters to respond to transients is limited by the decreasing contaminant concentration and microbial activity as distance from the inlet increases. When large transient loadings occur, either or both the mass transfer capacity or the reaction capacity of the initial sections of the bed are exceeded and contaminants move into the downstream sections where the microbial populations and reaction capacities are low, resulting in potential breakthrough. The performance of biofilters can be improved by adopting design and operational strategies to manage those transients. Relative to UF mode of operation, flow-directional-switching produced a more uniform distribution of biomass and microbial reaction capacity along the length of the packed bed without diminishing activity and removal capacity in the inlet section. Operating in FDS mode incorporates advantages believed to accrue from feast/famine operation of microbial processes and improved mass transfer characteristics of biofilms that are uniformly distributed (dispersed) throughout the length of the packed bed. Dispersed biofilms are thought to have increased active biofilm-vapor interfacial area and potential mass transfer rates relative to biofilms that are concentrated near the inlet. The benefits of flow-directional-switching were demonstrated when; (a) maximum mass removal rates in the FDS biofilter were approximately twice that of the UF biofilter in step-loading experiments, (b) the FDS biofilter performed better than the UF biofilter following a 2.9-day shut-down period and (c) FDS operation resulted in greater long term stability relative to UF operation in regular feed-on/-off transient loading experiments.

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