Behavior of Field-Scale Biotrickling Filter under Nonsteady State Conditions

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Abstract: The performance of a field-scale biotrickling filter was investigated for the treatment of styrene vapors released from a bathtub manufacturing process. The two-stage biotrickling filter was operated in series with an average gas flow rate of 350 $m^3 h^{-1}$ corresponding to an overall empty bed gas contact time of 84 s. Daily average values of styrene removal efficiency varied from 40 to 90% with inlet concentrations ranging between 0.4 and 1.7 $g\,m^{-3}$. System performance was not significantly affected by changes in temperature and was moderately susceptible to 3-day starvation or complete system shutdown. After 7 months of styrene treatment, toluene contaminated air was fed to the system and experiments were performed in which styrene and toluene were fed alternately at 3-h intervals. While styrene elimination remained unchanged over the cycles, the elimination capacity of toluene increased with the number of cycles, indicating some adaptation of the process culture to the new contaminant. Overall, the results suggest that biotrickling filters for air pollution control can be successful even under greatly varying operating conditions.

CE Database subject headings: Filters; Biological treatment; Air pollution; Vapor.

Introduction

Biofilters and bioscrubbers are emerging biological techniques for air pollution control (Devinny et al. 1999). They work best for the treatment of large volumes of off gases that contain low concentrations of biodegradable contaminants (Fig. 1). Biofilters work by passing a humidified stream of air through a bed of porous material, generally compost and some bulking agent. There is very little or no free flowing water in biofilters. Bioscrubbing techniques, on the other hand, involve a free water phase and biodegradation of absorbed pollutants by either suspended or immobilized microorganisms. The most promising bioscrubber setup is the biotrickling filter, where both absorption and biodegradation are combined into one bioreactor, usually a packed tower equipped with either random or structured packing on which pollutant-degrading bacteria are allowed to grow (Cox and Deshusses 1998).

The advantages of biofilters include a simple configuration, ease of operation, and lower installation and operational costs. Problems associated with biofilters include lower volumetric performance and difficulties in controlling pH and nutrients in the packing material because of the absence of a free liquid phase. In the case where chlorinated or sulfur-containing compounds are treated, hydrochloric acid or sulfuric acid is produced, which exhausts the buffering capacity and reduces the pH of the packing material. In biotrickling filters, such acidic by-products can easily be flushed out of the system, and optimum pH and nutrient concentrations can be maintained over time by controlling the composition of the recirculating water (Gabriel and Deshusses 2003). However, because conditions (nutrient feeding) in biotrickling filters promote biomass growth, clogging of volatile organic compound (VOC) degrading biotrickling filters by excess biomass has been reported in a number of laboratory studies (see, e.g., Smith et al. 1996; Weber and Hartmans 1996; Cox and Deshusses 1999a,b). This has been a major obstacle to the deployment of biotrickling filtration for VOC control in the field. However, most studies of biotrickling filters for air pollution control have been performed in the laboratory with high pollutant concentrations. Only limited data exist for the operation of biotrickling filters in the field with low pollutant concentrations and transient condi-
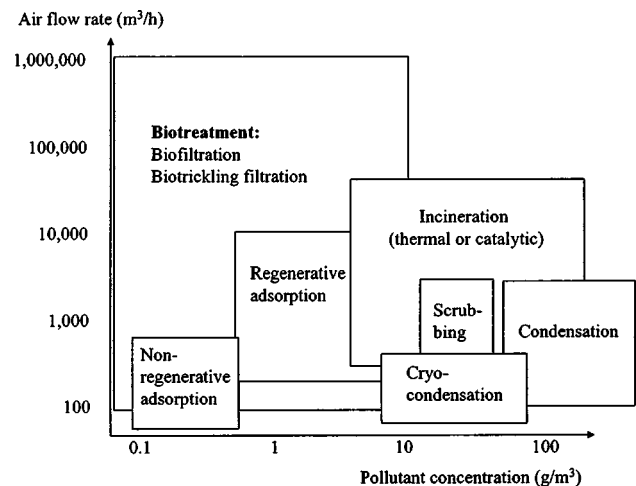
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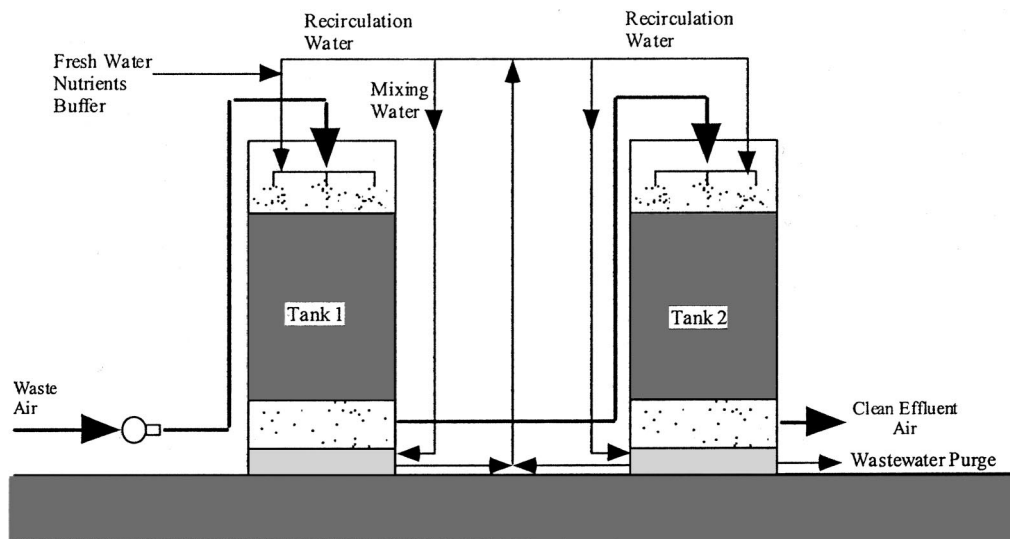**Fig. 1.** Applicability of biological techniques for VOC control

Fig. 2. Schematic of field-scale biotrickling filter setup

tions (Loy et al. 1997; Webster et al. 1999; Gabriel and Deshusses 2003). When a large biotrickling filter was operated in an industrial setup (Webster et al. 1999), the problems were very different from those observed in the laboratory. It was difficult to maintain a high-density process culture, most probably because of the cyclic operation of the plant and the low concentrations at which the reactor was operated. Before generalizing such observations, additional field studies are needed. In particular, issues related to biomass growth at low concentrations and to the treatment of rapidly changing contaminated air streams (pollutant nature and/or concentration) require further investigation. A cyclic starvation experiment (8 h/day) in a biotrickling filter treating polyalkylated benzenes showed a rapid increase of elimination capacity (EC) within the first 6–7 h after each starvation period (Hekmat et al. 1997). Hekmat et al.'s experiments approximate real systems more closely. Zuber (1995) investigated the dynamic behavior of a laboratory scale biotrickling filter for methylene chloride removal. He demonstrated that the system responded quickly to rapid changes in inlet concentration with steady state performance generally established within 1 h. He also showed that effective treatment was reestablished 2–4 days after complete starvation for an extended period of time. Similar observations were made by Gabriel and Deshusses (2003) for a high performance, full-scale H_2S-degrading biotrickling filter. They found that the biotrickling filter was robust, and was able to successfully treat a wide range of rapidly changing H_2S concentrations.

Still, there is lack of data on the operation of field biotrickling filters for air pollution control, in particular data acquired under a variety of greatly changing conditions. Further research with field-scale biotrickling filters is needed to identify possible issues specific to field conditions and to give further evidence that biotrickling filtration works under a wide range of operating conditions. Hence, the purpose of this work was to observe the behavior of a field biotrickling filter treating styrene releases from a bathtub manufacturing process under highly nonsteady conditions. The performance and the dynamic response of the biotrickling filter when subjected to selected perturbations (variable inlet concentration, cyclic pollutant feeding, starvation, etc.) were investigated.

Materials and Methods

Site and Biotrickling Filter Reactor

A two-stage pilot biotrickling filter was used for the study. The biotrickling filter was located at a fiberglass bathtub manufacturing facility (LASCO Bathware, Anaheim, Calif.) where it had been operated for 5 months prior to the experiments reported in this paper. Styrene vapor was emitted from the plant during the manufacturing process through a stack at a flow rate of 51,000 $m^3 h^{-1}$ at concentrations of up to 0.8 $g\,m^{-3}$. The biotrickling filter was located outdoors and it treated a side stream taken directly from the stack. Except for styrene, the air did not contain any other contaminants. All particulates were removed prior to entering the stack by a filter screen. The plant generally operated continuously Monday morning through Saturday morning. It was shut down for 48 h over the weekend. A detailed analysis of the biotrickling filter operation at this site was reported earlier (Webster et al. 1999). For selected experiments presented in this paper, tighter control of the pollutant inlet concentration was desired. In these cases, the biotrickling filter was disconnected from the stack and ambient air spiked with a metered stream of styrene or toluene was treated instead.

The biotrickling filter reactor included two beds in series with a bed volume of 4.1 m^3 each. Each reactor had an internal diameter of 1.6 m and height of 2.4 m (Fig. 2). The packing material consisted of 9-cm diam Jaeger PP TriPack spheres with a 95% void space and a surface area of 125 $m^2\,m^{-3}$. The biotrickling filter was operated at an average air flow rate of 350 $m^3 h^{-1}$, which corresponds to and an empty bed residence time (EBRT) of 42 s/reactor. Contaminated air and recycled liquid were fed cocurrently to the top of the biotrickling filter. Liquid was recycled at average flow rates of 13.8 $m^3 h^{-1}$ for each reactor. During normal operation, an average of 20 $L\,h^{-1}$ of recycled liquid was replaced by fresh mineral medium solution. It served for drainage of potential metabolite and suspended biomass to make up for evaporative losses and to supply the necessary nutrients.

Fresh liquid fed to the biotrickling filter consisted of a concentrated nutrient mixture with trace elements, diluted with tap water to concentrations of 0.7, 1.2, and 0.8 $g\,L^{-1}$ of N, P, and K, re-

spectively (Webster et al. 1999). The pH of the recirculated water ranged from 6.1 to 7.2 and averaged 6.8, and was maintained using a 5% NaOH solution. The reactor setup included a programmable logic controller (PLC) unit with LabView software (National Instruments, Austin, Tex.) to monitor and control the system. Contaminant concentrations, air and liquid flow rates, head losses, water conductivity, temperature, reactor weight, pH, and water levels were continuously monitored at 3-min intervals. Further details of the reactor design and operation (Webster et al. 1999) or treatment filter cost analysis (Deshusses and Webster 2000) are discussed elsewhere. After 7 months of operation treating styrene exhaust from the manufacturing process, an experimental protocol was developed to study the effect of alternating toluene and styrene on treatment performance. For these studies, the biotrickling filter was disconnected from the plant stack, and synthetic waste air was produced by pumping either styrene or toluene directly into the inlet air. Each cycle consisted of an average of 0.690 $g\,m^{-3}$ of styrene supplied to the inlet stream for 3 h, a 1-h resting period with no VOC, a 3-h period in which an average concentration of 0.580 $g\,m^{-3}$ of toluene was supplied to the biotrickling filter, and a final 1-h resting period. Each complete cycle took 8 h of operation, and 16 consecutive cycles were completed. The above concentrations of toluene and styrene were selected as representative of the median concentration of styrene in the exhaust from the bathtub manufacturing facility.

Analysis

Total gas phase contaminant concentrations were measured with a flame ionization detector (FID) (SRI Instruments, Torrance, Calif.). The air sampling system incorporated six solenoid valves, a diaphragm air pump, a pressure relief regulator, and a flow rate control system. In routine sampling mode, the system collected air samples from air sampling lines upstream of reactor 1 (inlet), between reactors (outlet 1), downstream of reactor 2 (outlet 2), and from ambient air for 3 min at each sampling location. After completion of each cycle, the system backflushed the sampling lines for 3 min, then started the cycle again. Fifteen minutes was needed to complete the sequence. When continuous monitoring of one sampling location was required, automatic switching of the sampling ports was halted, and one sampling location was manually selected for continuous analysis. The FID was calibrated with either styrene or toluene standards before each experiment.

Results and Discussion

Biotrickling Filter Performance Treating Styrene from Manufacturing Discharge

The concentration of styrene in the off gases from the bathtub manufacturing process fluctuated cyclically during normal operation. The inlet concentrations in a specific day varied from 0.03 to 0.83 $g\,m^{-3}$ (Fig. 3). The average and standard deviations for inlet, between tank (outlet 1) and final (outlet 2), concentrations were 0.58 ± 0.22, 0.27 ± 0.11, and 0.10 ± 0.04 $g\,m^{-3}$, and their median concentrations were 0.66, 0.31, and 0.10 $g\,m^{-3}$, respectively. The normal probability plot for inlet and outlet concentrations on the same day showed that outlet 2 concentrations were relatively stable compared to the highly varying inlet concentrations (Fig. 4). The inlet distribution plot reflects the intermittent emission pattern. While the biotrickling filter received virtually no pollutants 10% of the time, concentrations usually ranged from 0.6 to
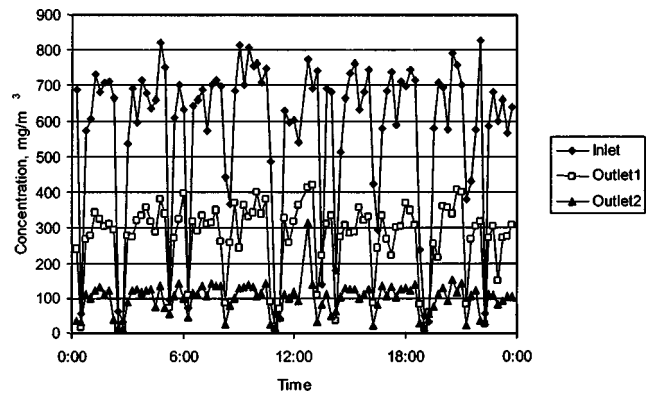

Fig. 3. Hourly variations of styrene concentration in biotrickling filter in typical day at bathtub manufacturing facility

0.8 $g\,m^{-3}$ when emission occurred (about 80% of the time). Distribution plots such as the one shown in Fig. 4 are useful for regulatory evaluation and for reactor design, and should be part of the problem definition during biotrickling filter evaluation. Distribution plots should be made for typical plant operation because they will enable sizing of the biotrickling filter for the most probable emissions, and/or help in identifying the need for adding some kind of buffering pretreatment (Al-Rayes et al. 2001) or polishing posttreatment, if intermittent operation is found to induce short, but unacceptable, emission breakthroughs.

The overall styrene removal efficiency in the biotrickling filter for this specific day ranged from -81 to 96%. These extreme removal efficiencies were not representative values, but rather a consequence of the 6-min lag between inlet and outlet FID analyses. While the inlet concentrations changed sharply when manufacturing processes were suspended or resumed, the FID was activated for only 1 min after a 3-min interval and resulted in some erroneous removal values (excessively high or negative values). As will be discussed later, in such cases of rapidly changing concentrations, the time-averaged removal or cumulative removal is a much better representation of the true performance of the biotrickling filter.

When considering daily averages over a 5-month period, the mean inlet styrene concentration was 0.48 $g\,m^{-3}$ with extremes ranging from 0.03 to 1.38 $g\,m^{-3}$ (Table 1). The daily overall
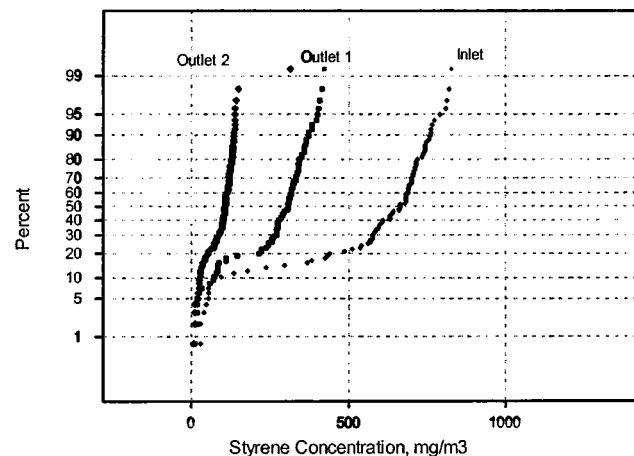

Fig. 4. Normal probability plot of styrene concentrations for day shown in Fig. 3

Table 1. Summary of Daily Performance of Biotrickling Filter after 5 Months of Operation

Parameter	Location	Mean	Median	Standard	Minimum	Maximum
Concentration (g m^{-3})	Inlet air	0.480	0.423	0.330	0.031	1.380
	Outlet 1	0.309	0.229	0.262	0.006	1.109
	Outlet 2	0.170	0.096	0.179	0.006	0.747
Loading rate (g m^{-3} h^{-1})	Overall	20.6	18.2	14.1	1.3	59.1
	Stage 1	41.2	36.3	28.3	2.6	118.3
	Stage 2	26.5	19.7	22.5	0.5	95.1
Elimination capacity (g m^{-3} h^{-1})	Overall	13.3	11.4	7.7	1.0	27.1
	Stage 1	14.7	13.4	8.4	1.5	30.5
	Stage 2	11.9	11.1	7.9	0.1	31
Removal efficiency (%)	Overall	68.9	70.9	13.4	43.2	89.9
	Stage 1	40.5	41.8	14.5	18.7	80.3
	Stage 2	49.4	51.0	12.8	23.3	73.3
Temperature (degrees C)	Inlet air	50.9	50.0	2.2	47.0	55.5
	Outlet 1	33.7	33.5	1.6	31.1	36.5
	Outlet 2	33.7	33.5	1.6	30.9	36.5
	Tank 1	33.5	33.4	1.6	30.8	36.3
	Tank 2	32.9	32.7	1.6	30	36.1

average, minimum, and maximum treatment efficiencies were 68.9, 43.2, and 89.9%, respectively. Other biotrickling filter parameters are given in Table 1. The daily average performance reported as the styrene elimination capacity (=inlet–outlet concentration×air flow rate/bed volume) as a function of the styrene loading (load=inlet concentration×air flow rate/bed volume) is shown for the first and second tanks and for the entire reactor in Fig. 5. Detailed examination of Fig. 5 reveals that at equal loading, the elimination capacity of styrene for the entire system is significantly higher than that of either tank 1 or tank 2. This is probably because treatment in two consecutive reactors allows better contact between the gas and the immobilized biomass, hence better overall treatment. The biotrickling filter could handle loads up to 15–20 g m^{-3} h^{-1} with a relatively high rate of removal, while higher loadings were only partially degraded. The maximum elimination capacities were 30.5 g m^{-3} h^{-1} for tank 1, 31.0 g m^{-3} h^{-1} for tank 2, and 27.1 g m^{-3} h^{-1} overall. These val-

ues are similar to those in some earlier reports and about 50% lower than in other reports on the removal of styrene in laboratory-scale biotrickling filters (Togna and Singh 1994; Pol et al. 1998; Sorial et al. 1998; Lu et al. 2001). The most probable reasons for lower performance are the highly transient environment in which the biotrickling filter operated compared to steady-state laboratory studies, and the fact that the packing used in this study (TriPack) may not be very suitable for biotrickling filtration. Regarding the latter issue, we observed (unpublished results) that the shape, and possibly the polypropylene material, of the TriPack packing did not favor microorganism attachment, hence achieving good coverage with biofilm was difficult, especially in low loading conditions.

The temperatures of the air and recirculation water also fluctuated during a typical day of operation, although to a lesser extent (Table 1). The inlet air temperature was always higher than that at any other location because it was heated both by the manufacturing processes and the air blower. Axial temperature profile measurements (not shown) revealed that the air was immediately cooled upon entering tank 1. Statistical analysis of performance and temperature trends revealed that fluctuations of temperature in the range observed did not significantly affect the performance of the biotrickling filter.

As discussed earlier by Webster et al. (1999) and illustrated in Fig. 5, the biotrickling filter experienced difficulties in reaching optimum pollutant removal. This was both because of frequent shutdowns of the biotrickling filter and fluctuating operating conditions that resulted in suboptimal biomass density immobilized on the packing, resulting in biological limitations. The manufacturing processes at the field site were operated 24 h a day with the usual weekend recesses (Webster et al. 1999), and further research on the effect of starvation and intermittent operation was warranted. The air blower and water pump were shut down for 3 days (full-system shutdown). Upon restarting the system, pure styrene was artificially fed into the biotrickling filter influent air in order to guarantee steady conditions and the response of the system was studied. Note that, because of experimental difficulties, the inlet concentration of styrene was lower than the steady value for the first 3–4 h after restarting the biotrickling filter. After restarting the system, the cumulative elimination capacity
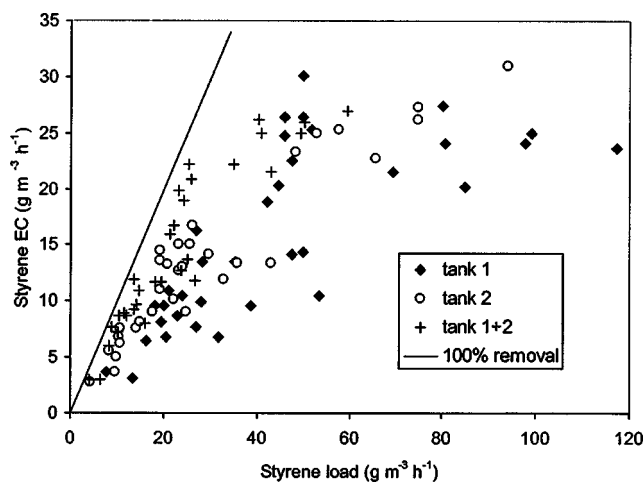

Fig. 5. Daily averages of styrene elimination capacity during routine operation in first and second tanks, and overall elimination capacity as a function of styrene loading. Air contact time in the system was 42 s/tank. Note that the highest loadings were obtained by injecting pure styrene into the air feed.
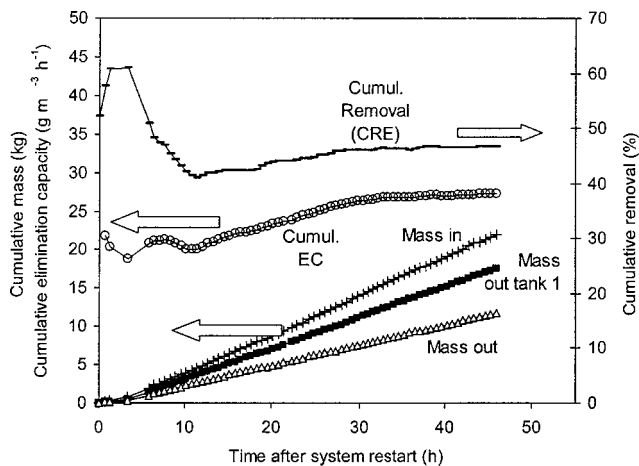
Fig. 6. Effect of 3-day system shutdown on the cumulative styrene masses, removal efficiency, and elimination capacity after restart at time zero; see Eqs. (1) and (2) for definitions of the cumulative parameters

Fig. 7. Weight of reactors after restarting the system (time zero) after 3-day system shutdown

(CEC) and cumulative mass removal efficiency (CRE) were calculated as a function of time as follows:

$$CRE(\%) = 100 \times (cumulative\ mass\ in - cumulative$$
$$mass\ out)/cumulative\ mass\ in\ (\%) \qquad (1)$$

$$CEC = (cumulative\ mass\ in - cumulative$$
$$mass\ out)/(bed\ volume \times time\ elapsed)\ (g\,m^{-3}\,h^{-1})$$
$$\qquad (2)$$

where the cumulative mass of styrene into or exiting the biotrickling filter was calculated by using the time-averaged concentration multiplied by the air flow rate. The use of cumulative values permits the assessment of the long-term effect of perturbation on the system.

Plotting the cumulative styrene performance (Fig. 6) after restarting showed that during the first 10 h of the transition period, a higher than usual percentage of removal was obtained. After about 30 h of treatment, a point of inflection was observed and cumulative removal stabilized at about 46%. In similar fashion, the cumulative elimination capacity reached about 27 g m^{-3}h^{-1}. The unusual pattern of removal deserves further discussion. Increases in performance after a starvation event have been reported before (Hekmat et al. 1997) in a cyclic starvation experiment (8 h/day) for polyalkylated benzene treatment in a biotrickling filter. Heckmat et al. observed a steep increase of elimination capacity within the first 6–7 h after each starvation period. This was presumably due to fast regrowth of the process culture under non-nutrient-limiting conditions after restarting, and is consistent with some of our observations published recently in which peak performance was observed immediately after startup, when plenty of nutrients were available (Cox et al. 1999). In a more recent paper, we investigated in detail starvation of toluene-degrading biotrickling filters (Cox and Deshusses 2002). Although the study involved toluene removal at relatively high concentrations, the findings are relevant to the present paper. It was found that the process culture was losing its toluene biodegradation potential (measured by the toluene-induced oxygen uptake rate) exponentially during pollutant starvation. After 2 days, about 75% of the original activity was lost, while after 5 days, the biodegradation
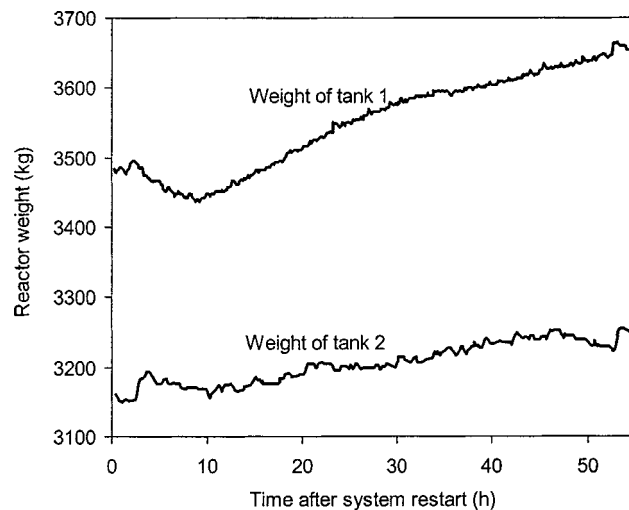
potential was essentially zero. After restarting, the biotrickling filters regained their full capacity in less than 20 h. Similar to what was reported by Heckmat et al. (1997) a peak in activity was observed immediately after restarting the biotrickling filters.

In the experiment presented in Fig. 6, the temporary high performance after restart may be due to more than one cause. It may be that excess nutrient was available in a way similar to that in the experiments of Hekmat et al. and Cox and Deshusses discussed above. However, examination of the cumulative elimination capacity reveals that the high removal did not coincide with the higher elimination capacity, so the nutrient effect, if present at all, must have been small. Another possibility is that some transient absorption of styrene in the recycling water and in the biofilm occurred, but calculation of the amount that can be absorbed based on Henry's law reveals that it can only account for a small fraction of the cumulative mass removed. Probably, the prevalent cause of the high removal after restarting was the lower loading experienced immediately after restart, which favored higher removal.

Monitoring of the reactor weight after restart shows that the biotrickling filters underwent significant changes as shown in Fig. 7. Immediately after restarting the system, the weight of the reactors decreased, most likely because of more enhanced detachment of some biomass from the packing after starvation. After about 10 h, the weight started to increase, consistent with active biomass regrowth after a short lag in time. Not unexpectedly, the rate of weight increase in reactor 1 was higher than that in reactor 2 because of higher styrene loading. Interestingly, calculation of the biomass yield based on the cumulative mass degraded and the weight increase (taken between 15 and 50 h) of the entire system resulted in an average yield of 29 kg of wet biomass/kg of styrene degraded. This value is about twice the expected value of 14 kg$_{ww}$/kg$_{styrene}$ assuming 5% dry weight in the biofilm, and a yield of 0.7 kg dry biomass/kg styrene degraded (Cox and Deshusses 1999a). The higher than expected value may be due to reattachment of lose biomass, or to growth of the process culture on decayed biomass after resuming the air supply.

As mentioned above, cumulative performance data allow the assessment of the long-term effect of perturbation on the system and may be useful for regulatory evaluations. From a pollutant emission standpoint, the rapidly stabilizing cumulative removal
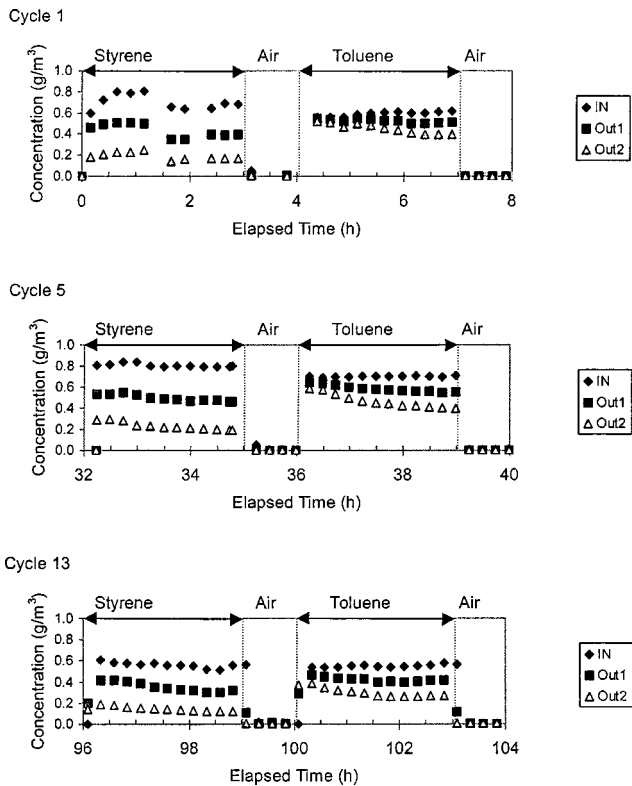
Fig. 8. Effect of alternating inputs of styrene and toluene on changes in concentration during different cycles

and cumulative elimination capacity trend shown in Fig. 6 suggest that the effects of starvation are negligible if the frequency of severe starvation is less than one event in 72 h.

Effect of Alternating Substrate Input

The effect of cyclically alternating inputs of styrene and toluene on biotrickling filter performance was investigated using a synthetic stream of air. Each cycle consisted of 3-h periods of toluene supply, 1 h of rest, a 3-h period of styrene supply, and a final hour of rest. Sixteen consecutive cycles were completed. The biotrickling filter had been exposed only to styrene prior to the experiment. The results are shown in Figs. 8–10. The patterns for styrene and toluene removal were quite different. The inlet, outlet 1, and outlet 2 concentrations for selected cycles (1, 5, and 13), are reported in Fig. 8. During the first cycle, the inlet concentrations fluctuated slightly as a result of adjustments made in the toluene and styrene pumping rates to achieve a target inlet concentration of approximately 0.6 $g\,m^{-3}$. The concentration data at the time of the step changes (up or down) show that sorption of the pollutant was minimal so that highly transient conditions lasted less than 1 h for both pollutants. Subsequent changes in performance were therefore most likely due to changes in biological activity. While styrene treatment rapidly reached a steady state, toluene treatment improved more slowly (see Figs. 8 and 9), but eventually reached a pseudosteady state during the 3-h step inputs.

Comparison of cycles 1, 5, and 13 (Fig. 9) shows that styrene elimination patterns were very similar in each cycle. This was expected because the reactor had been exposed to styrene for the past 7 months and short periods of starvation did not affect the process culture, as discussed earlier. On the other hand, toluene removal showed significant improvement over the series of
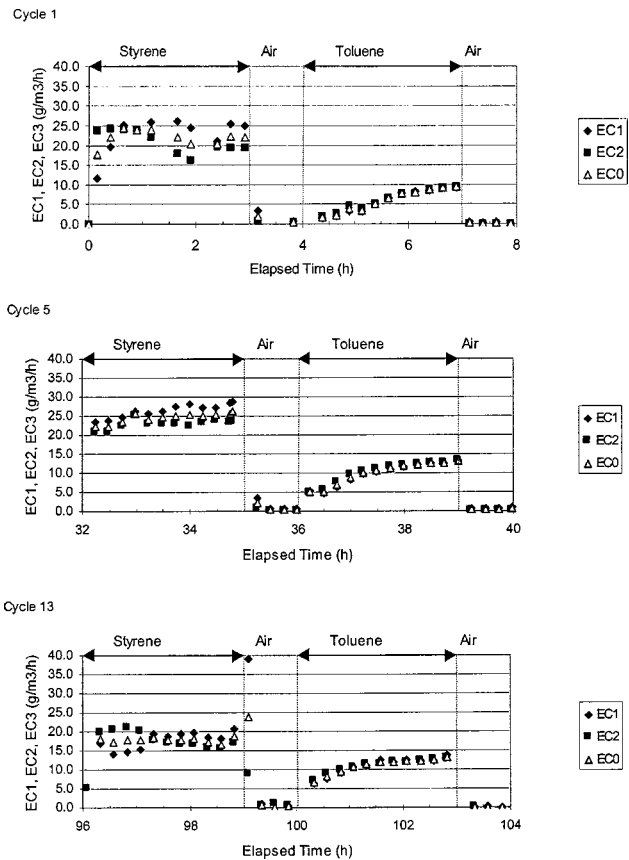

Fig. 9. Effect of alternating inputs of styrene and toluene on elimination capacity (EC1=elimination capacity of tank 1, EC2=EC for tank 2, EC0=overall EC)

cycles, in particular over the first five cycles (Figs. 9 and 10). Interestingly, the improvement over time was not so much in the ultimate value for the toluene elimination capacity (about 10–12 $g\,m^{-3}\,h^{-1}$) but rather in the time needed to reach a pseudosteady state with respect to treatment performance. This decreased from about 3 to less than 1.5 h in less than five cycles. The toluene elimination capacities of the first and second stages were not significantly different, although the toluene loadings were different.
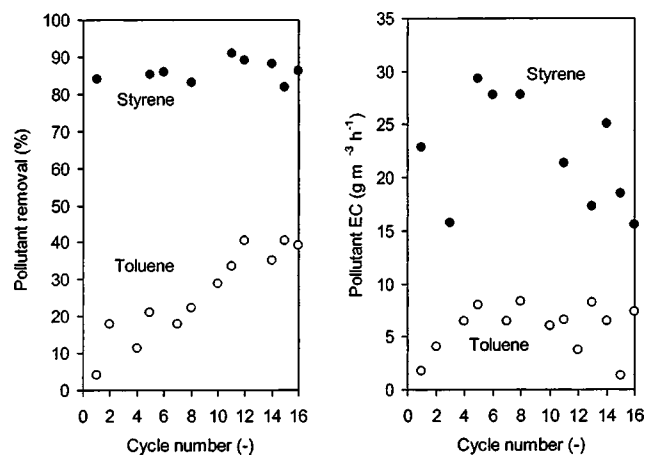

Fig. 10. Styrene and toluene removal (left) and elimination capacity (right) over different cycles during the alternating input experiment; some data are missing because of problems in analysis

Presumably, biodegradation kinetics had reached a zero-order regime and were no longer a function of the concentration.

The pattern for toluene treatment reflected two factors. As the number of cycles increased, the total amount of toluene-degrading biomass increased, causing improvement of the removal efficiency over the first five cycles. At the same time, the specific activity of the process culture varied during each cycle. The cells were losing their toluene-degrading enzymes during periods when toluene was not present, or synthesized more when toluene was present. However, as mentioned earlier, the variation of the toluene degradation activity during each cycle decreased over the cycles, thereby indicating some degree of adaptation to cyclic starvation.

The styrene-degrading biomass, in contrast, was present in adequate amounts because of the long history of styrene treatment before the cycling experiment was begun. Therefore, the styrene degradation capability did not decline during the toluene feed period and recover during the styrene feed. Any growth of biomass during the experiment did not provide an improvement in styrene treatment because the biomass density was not limiting and because the new biomass, if any, was primarily toluene degraders.

Conclusions

The operation of a field-scale biotrickling filter was studied under highly fluctuating conditions. Under these conditions, it was proposed to report performance as cumulative removal or cumulative elimination capacity, which better illustrates the long-term performance of the system. The two-stage biotrickling filter provided stable performance in nonsteady-state environments, and was not affected by temperature changes or 3-day periods of down time. When styrene and toluene were fed alternately at 3-h intervals, styrene elimination remained essentially unchanged, while toluene removal improved over the successive cycles as a result of acclimation of the process culture to the new contaminant. Overall, detailed monitoring and analysis of the biotrickling filter operation provided better understanding of the operation of biotrickling filters under field conditions.

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