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Development of Aerobic Biofilter Design Criteria for Treating VOCs

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INTRODUCTION

With the enactment of the 1990 amendments of the Clean Air Act, the control of volatile organic compounds (VOCs) in contaminated air streams has become an important issue.[1] The concept of using biological processes for controlling undesirable compounds in different kind of wastes has increasingly been applied. With respect to the purification of polluted air, biofiltration is a potential cost effective process for treatment of large gas flows contaminated with low concentrations of biodegradable VOCs, especially in comparison to the conventional VOC control technologies such as incineration and carbon adsorption.[2] The low operating cost is due to the use of microbial oxidation, which is achieved at low operating temperatures, rather than thermal or chemical oxidation. Essentially, biofiltration operates at ambient temperatures, and is a self regenerating, enzymatic catalytic process. It consists of contacting a contaminated air stream with a moist film of microbes attached to a stationary synthetic or natural support material. VOCs are oxidized to simple end products such as H_2O and CO_2. More recently, biofiltration as a hazardous VOC control technology has been the subject of extensive research, and the design criteria have been identified.[2-6]

In bench scale research, the use of trickle bed biofilters has become popular. This type of biofilter allows for more uniform surface area and gas distribution, resulting in better pressure drop control, as well as more consistent operation due to better nutrient and pH control. Such biofilters consist of a filter containing microbes supported on an inert material, with nutrients applied at the top at a minimal liquid flow rate. Effluent recycle is typical, both for pH control as well as microbe reseeding, and an effluent purge removes excess salts and biomass. Kirchner *et al.*[7] investigated the effect of contaminant solubility by examining the treatment of acetone, propionaldehyde, naphthalene, and toluene in a trickle bed biofilter. Diks and Ottengraf [8] found that a recirculating trickle bed biofilter using saddle packings was effective in treating methylene chloride. They directly correlated the amount of sodium hydroxide added to the recycle stream for pH control to the elimination capacity. Cocurrent flow air and recycle liquid flow avoided stripping of VOCs at the exit of the filter although the difference appeared to be minimal. Hartmans and Tramper [9] investigated the treatment of methylene chloride in a trickle bed biofilter and concluded that recirculated biofilters were superior to compost beds for treatment of halogenated compounds because pH and salt were more easily controlled. Utgikar *et al.*[10] proposed the use of activated carbon packed trickle biofilters for treatment of landfill leachate offgasses for adsorbing the substrate, that is not consumed by the microbes, on the support media.

This paper reports preliminary results of studies performed utilizing trickle bed biofilters with monolithic channelized microbial support for the treatment of VOCs typical of landfill leachate stripping. For the initial studies, toluene has been used for the purpose of characterizing the trickle biofilter apparatus, to be followed with ethylbenzene, chlorobenzene, trichloroethylene, and methylene chloride. The objectives of the experiment are to investigate

the use of such biofilters, both cocurrent and countercurrent, for treating these compounds with high removal efficiency at inlet concentrations which are high, relative to most biofilter research to date. The further research objective is to reduce to practice biofiltration for the treatment of such VOC containing air streams.

MATERIALS AND METHODS

Experimental Apparatus

The experimental apparatus consists of two independent, parallel, trickle biofilters designated as biofilter "A" and biofilter "B". Each biofilter is made of 304 stainless steel, and has a square cross-section with internal dimension of 5.75" and consists of the following sections from top to bottom:

1) a 4" module for nutrient and buffer addition, and for air inlet (or outlet);
2) a 12" module for housing the nutrient and buffer distribution apparatus;
3) four 12" modules containing the biofilter biological attachment media; the media used is Corning Celcor® channelized media (a magnesium aluminosilicate material) having 7.75 channels per cm^2.
4) a 4" module for waste water outlet, and air inlet (or outlet).

In order to minimize condensation and to maintain a constant temperature, the biofilters are insulated and temperature controlled with external cooling coils.

The air supply to the biofilters is purified with complete removal of water, oil, CO_2, VOCs, and particulates, especially microbes. After purification, the air to each biofilter is split off, humidified, externally heated to assist vaporizing the injected toluene into the air stream via a syringe pump, and finally fed to the biofilters. The air is mass flow controlled. A schematic of the experimental apparatus is shown in Figure 1.

Each biofilter is equipped with separate systems for feeding 20 L/day of a nutrient solution containing all necessary macro-, micro-nutrients and buffers. The compositions of the mixed nutrient solutions (consisting of trace salt, salts and vitamin solutions) are presented in Tables I, II and III. The daily recipe consists of 9.0 mL stock salt solution, 2.3 mL vitamin solution, 4 mL of 0.01M $FeCl_3$ solution, and 5 mL of nutrient spike solution (2M NH_4Cl, 0.5M NaH_2PO_4). One molar sodium bicarbonate was used as a buffer (45 mL). Since observing that most of the ammonia in the feed is converted to nitrate, the nitrification inhibitor TCMP (2-chloro-6-(trichloromethyl)pyridine) is added to the nutrient formulation. Each biofilter is also equipped with effluent recirculation in order to provide even distribution of the biomass throughout the attachment media. Biofilter "A" is operated in a cocurrent mode, top to bottom, with the air flow, nutrient, and effluent recycle flows directed downwards. Biofilter "B" is operated in a countercurrent mode, with the air flow directed upwards and the nutrient

and effluent recycle flows directed downwards.

Materials

Reagent grade Toluene (99.9%, Fisher Scientific Co., Inc., Fair Lawn, NJ) was used as the target VOC contaminant in this study.

The seed for the biofilters was an activated sludge acclimated to the target VOCs: toluene, ethylbenzene, chlorobenzene, trichloroethylene, and methylenechloride. A synthetic solution containing 2% by volume of each VOC in toluene was fed daily, batchwise, to a stirred, aerated reactor. The growth of the biomass was monitored indirectly by observing the volume of 10M NaOH required for daily pH adjustment.

Analytical Methods

Concentrations of toluene were measured by chromatographic separation on a 30-m megabore column (DB 624, J&W Scientific, Folsom, CA) using a gas chromatograph (GC) (HP 5890, Series II, Hewlett-Packard, Palo Alto, CA) equipped with a liquid sample concentrator (LSC 2000, Tekmar, Cincinnati, OH), and a photoionization detector (PID) (Model 4430, OI Corp., College Station, TX). The liquid sample concentrator was programmed according to USEPA Method 601, a Tenax trap was used with helium (He) purge flow of 40 mL/min. The GC oven temperature was programmed from 40 to 120 °C at 5 degrees/min with a 4-min hold at 40 °C and a 6-min hold at 120 °C. The carrier gas (He) flow rate was set at 8 mL/min and the PID detector was used with He make-up gas at a flow-rate of 20 mL/min, sweep gas flow (H_2) at 100 mL/min and base temperature of 250 °C.

Gas phase samples, for VOC analysis, were taken with gas tight syringes through low bleed and high puncture tolerance silicone GC septa (replaced every week) installed in the sampling ports at the gas inlet and outlet from the biofilters. Samples from the liquid phase, for VOC analysis, were taken out in a similar way from the liquid outlet from the biofilters. Both gas and liquid phase samples were introduced to the GC through the liquid sample concentrator accessory. The gaseous phase VOC analysis was conducted by introducing 5 mL of purged distilled deionized water into the purge vessel of the liquid sample concentrator prior to the injection of the gas sample.

Liquid phase samples were also analyzed for nitrate and ammonia concentrations by using the electrode method of analysis according to Standard Methods[11] 4500-D, and 4500-F, respectively. Samples were filtered through 0.45 μm nylon filters (Micron Separation, Inc, Westboro, MA) prior to analysis.

The pH determinations were conducted by using Fisher Accumet pH meter, Model 50 (Fisher Scientific Co., Inc., Fair Lawn, NJ). The pH meter was calibrated before use by using buffers (pH of 3.0 and 7.0) supplied by the manufacturer.

RESULTS AND DISCUSSION

Startup of each biofilter was with 50 ppmv toluene at a 12 minute residence time, and a nutrient solution feed 20 L/day. Each biofilter was maintained at a constant temperature of 14 ± 2 °C and 7.7 ± 0.2 pH range.

Biofilter "A": The mass flow of toluene was increased steadily up to 400 ppmv at a residence time of 12 minutes. On day 85 it was noticed that most of the ammonia in the nutrient solution went to nitrate formation instead of biosynthesis. A nitrate inhibitor was then added to the nutrient solution in order to minimize the nitrate production, in both the feed tank and the biofilter. The maximum percent removal of toluene that could be obtained at 400 ppmv was only 80%. In order to improve the performance, the effluent was recycled to provide even distribution of the biomass throughout the support media. On day 127, the recycle was started and the percent removal increased to about 90%. The mass flow of toluene was then increased to 500 ppmv at 12 minutes residence time. When the percent removal of toluene was stable at 99%, a residence time cycle test was performed, with the residence time being varied from 12 to 1 minutes and then back to 12 minutes. This was done while holding constant the total mass of toluene fed per day. Figure 2 shows the performance of the biofilter with respect to toluene removal. Note that during the second leg of the residence time cycle test, when the VOC concentration was being increased with each step, more time was required for the biofilter to achieve the same performance as on the first leg. Figure 2 also shows that the influent concentrations obtained from the GC analysis are in good agreement with the theoretical concentrations obtained from the flow rate of the syringe pump, used for injecting toluene to the biofilter system, and the air flow rate. Figure 3, showing the performance of the biofilter with respect to ammonia utilization, shows the amount of ammonia going to synthesis. This amount is indicated by the difference between influent ammonia and effluent ammonia plus nitrate formed. Figure 4 shows the performance of the biofilter during the residence time cycle test. From Figure 4 it is seen that the toluene removal stabilized at better than 96% up to 4 minutes residence time. At 2 minutes residence time the removal efficiency dropped to about 90% and the performance further dropped to 65% at 1 minute residence time. At 1 minute residence time the pressure drop was about 1 inch of water.

Biofilter "B": The mass flow of toluene was increased steadily up to 200 ppmv at a residence time of 12 minutes. The performance was very poor compared to biofilter "A". The removal of toluene did not exceed 70%, and after day 87 the performance in fact started to drop. On day 90 the nitrate inhibitor was added to the nutrient feed. On day 113 the removal of toluene dropped to about 57%, and at this point it was decided to introduce effluent recycle to the biofilter. On day 114 the recycle was started, and by day 122 the removal of toluene was 85%. At this point the mass flow of toluene was increased steadily until it reached 500 ppmv at a residence time of 12 minutes. The biofilter was maintained at these conditions until

a stable effluent was obtained. At this point a residence time cycle test was started, conducted in a manner similar to biofilter "A". Figure 5 shows the performance of biofilter "B" with respect to toluene removal. Note that during the second leg of the cycle test, when the VOC concentration was being increased with each step, more time was required for the biofilter to achieve the same performance as on the first leg. This behavior also was noticed previously with biofilter "A". Figure 5 also shows that the influent concentrations obtained from the GC analysis are in good agreement with the theoretical concentrations obtained from the flow rate of the syringe pump, used for injecting toluene to the biofilter system, and the air flow rate. Figure 6, showing the performance of the biofilter with respect to ammonia utilization, shows the amount of ammonia going to synthesis. This amount is indicated by the difference between influent ammonia and effluent ammonia plus nitrate formed. Figure 7 shows the performance of the biofilter during the residence time cycle test. From Figure 7 it is seen that toluene removal during the cycle test stabilized between 87 and 89% up to 6 minutes residence time, then 72% at 4 minutes, 45% at 2 minutes, and 30% at 1 minute. At a 1 minute residence time the pressure drop was about 1.75 inches of water.

CONCLUSIONS AND FUTURE WORK

The removal efficiency for each residence time was similar, for both increasing and decreasing residence times. However, it appears that when reducing the residence time, which causes an increase in the VOC concentration at constant mass loading, more time is required by the biofilter to achieve maximum efficiency. Effluent recycle was necessary to achieve maximum efficiency.

Biofilter "A", which was operated cocurrently, showed the highest VOC removal efficiency. The efficiencies ranged from 99% to 65% for residence times of 12 and 1 minute, respectively. On the other hand biofilter "B", which was operated countercurrently, showed efficiencies less than 90%. The efficiencies ranged from 90% to 30% for residence times of 12 and 1 minute, respectively. The lower toluene removal efficiencies correlate with the substantially lower ammonia utilization, indicating a lower rate of cell synthesis in Biofilter "B".

The pressure drop for both biofilters were quite low, with a maximum of 1 and 1.75 inches of water for biofilters "A" and "B", respectively. This low pressure drop, even for the countercurrent mode, is very promising as it indicates that the major operating cost for this biofilter design, blower motor power, will be lower than for most typical biofilter designs.

Continuing work will include investigating the effect of the recycle flow rate on the performance of each biofilter. Investigation will also be made of increasing the mass loading to 500 ppmv at lower residence times. Xenobiotic VOCs will be tested for degradation after

initial characterization is complete.

REFERENCES

1. Lee, B. "Highlights of the Clean Air Act Amendments of 1990," *J.Air Waste Manage. Assoc.* 41(1):16 (1991).

2. Ottengraf, S.P.P "Exhaust Gas Purification," in *Biotechnology*, Vol. 8, Rehn,H.J.; Reed, G. Eds., VCH Verlagsgesellschaft, Weinham, 1986.

3. Hodge, D.S.; Median, V.F.; Islander,R.L.; Devinney, J.S. "Treatment of Hydrocarbon Fuel Vapors in Biofilters," *Environmental Technology* 12:655 (1991).

4. Leson, G., Tabatabal, F., Winer, A.M. "Control of Hazardous and Toxic Air Emissions by Biofiltration," Presented at the 85th Annual Meeting and Exhibition of the Air and Waste Management Association, Kansas City, Missouri, June 21-26, 1992.

5. Van Langenhove, J.; Lootens, A.; Schamp, N. "Inhibitory Effects of SO_2 on Biofiltration of Aldehydes," *Water, Air, & Soil Pollution* 47:81 (1989).

6. Van Lith, C.; David, S.L.; Marsh, R. "Design Criteria for Biofilters," *Trans. Inst. Chem. Eng.* 68B:127 (1990).

7. Kirchner, K.; Schlachter, U.; Rehm, H.J. "Biological Purification of Exhaust Air Using Fixed Bacterial Monocultures," *Appl. Microbiol. Biotechnol.* 31:629 (1989).

8. Diks, R.M.M.; Ottengrapf, S.P.P. "Verification Studies of a Simplified Model for the Removal of Dichloromethane from Waste Gases Using a Biological Trickling Filter (Parts I and II)," *Bioprocess Eng.* 6:131 (1991).

9. Hartmans, S.; Tramper, J. "Dichloromethane Removal from Waste Gases with a Trickle Bed Bioreactor," *Bioprocess Eng.* 6:83 (1991).

10. Utgikar, U.; Shan, Y.; Govind, R. "Biodegradation of Volatile Organic Compounds in Aerobic and Anaerobic Biofilters," in *Remedial Action, Treatment, and Disposal of Hazardous Waste*, Proceedings of the 17th Annual Hazardous Waste Research Symposium, Cincinnati, Ohio, 1991.

11. *Standard Methods for the Examination of Water and Wastewater*, 18th Edition, American Public Health Association, Washington, D.C. (1992).

Table I. Stock trace salt solution.

Component	Concentration, g/L
$(NH_4)_6Mo_7O_{24}.4H_2O$	2.08
$Na_2B_4O_7.10H_2O$	1.15
$MnCl_2.4H_2O$	4.74
$CoCl_2.6H_2O$	2.86
ZnCl2	3.27
$CuCl_2.2H_2O$	2.05

Table II. Stock salt solution.

Component	Concentration g/L
Trace salt solution	33.1 mL/L
$MgCl_2.6H_2O$	8.13
$NaH_2PO_4.H_2O$	8.28
$KH_2PO_4.H_2O$	13.6
NH_4Cl	49.2
$(NH_4)_2SO_4$	5.28
$CaCl_2.2H_2O$	4.44

Table III. Stock vitamin solution.

Component	Concentration, g/L
p-Aminobenzoic Acid	0.01
Biotin	0.0039
Cyanocobalamin (B12)	0.0002
Folic Acid	0.0039
Nicotinic Acid	0.01
Pantothenic Acid	0.01
Pyriodoxine Hydrochloride	0.02
Riboflavin	0.01
Thiamin Hydrochloride	0.01
Thioctic Acid	0.01

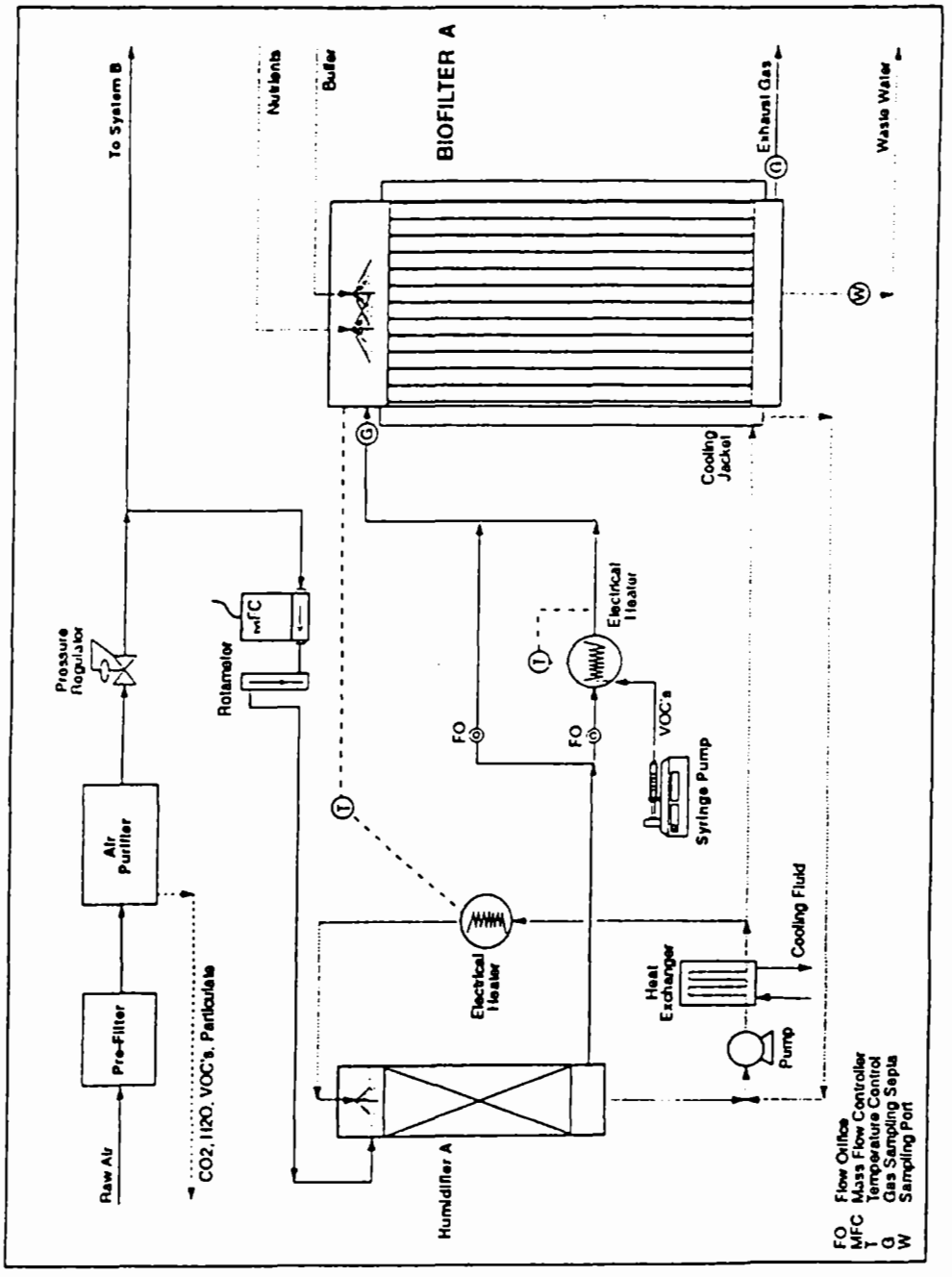

Figure 1. Experimental Schematic

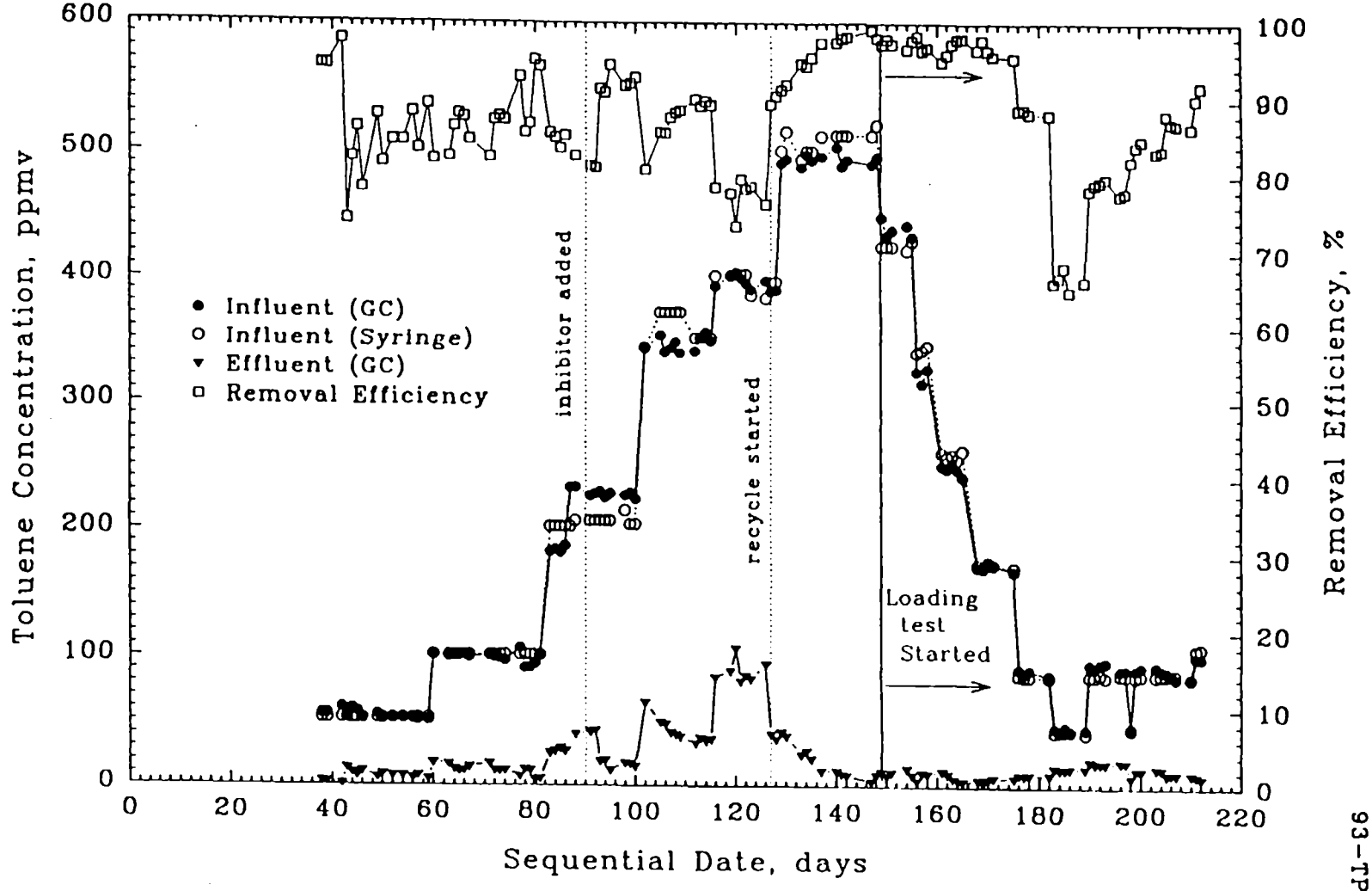

Figure 2. Biofilter "A" Performance w.r.t. Toluene Removal.

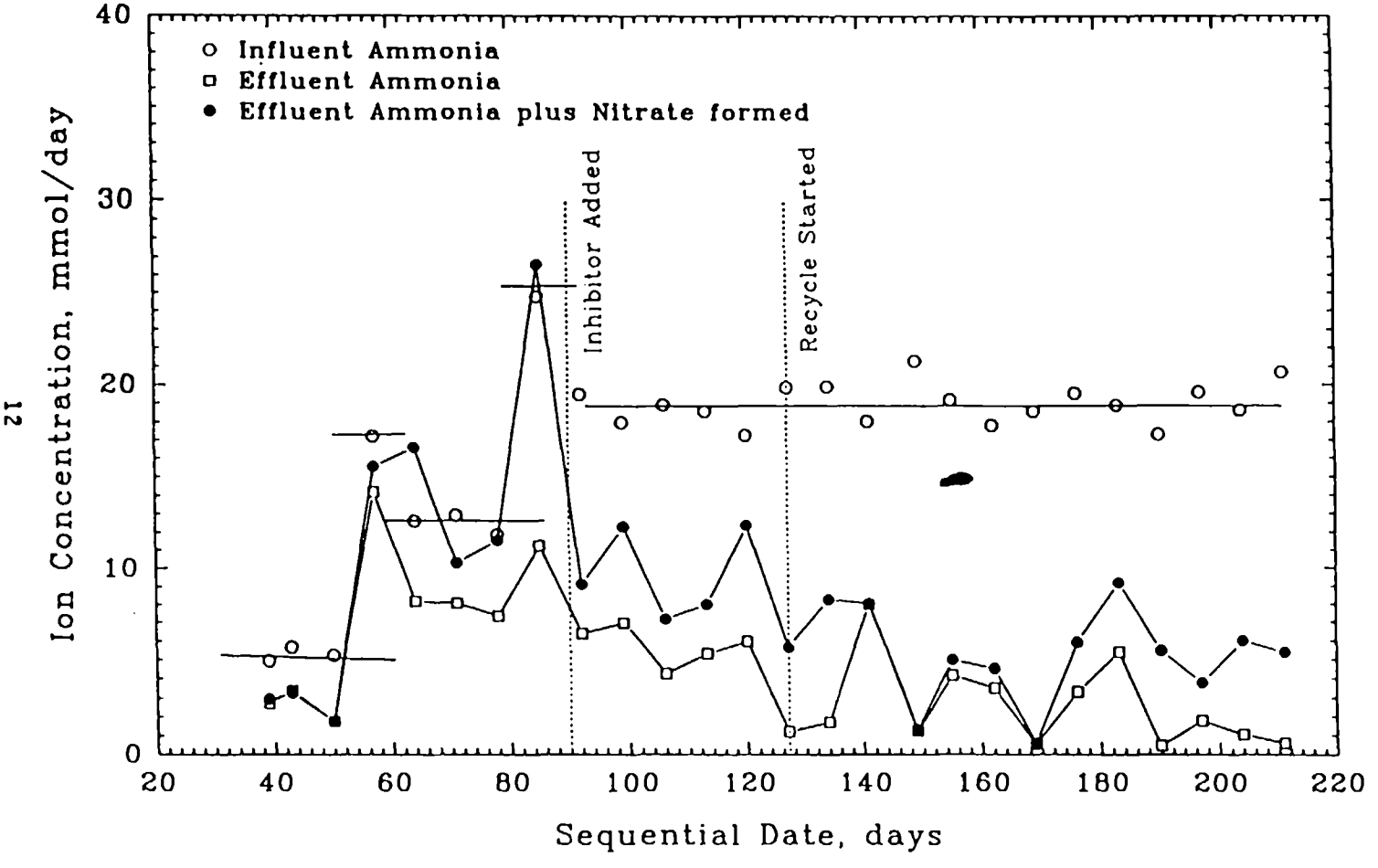

Figure 3. Biofilter "A" Performance w.r.t. Ammonia Consumption.

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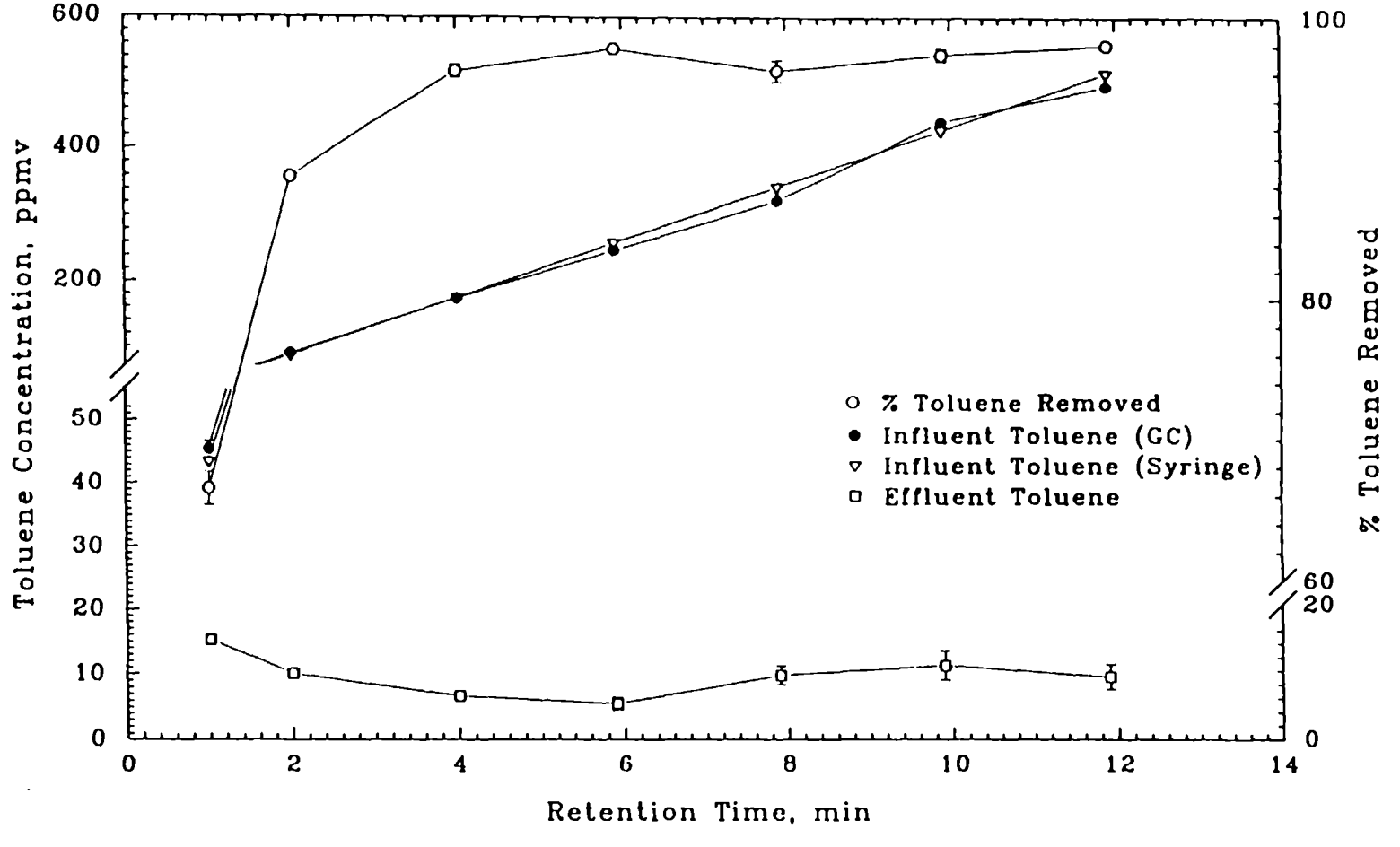

Figure 4. Biofilter "A" Performance w.r.t. Toluene Removal During a Residence Time Cycle.

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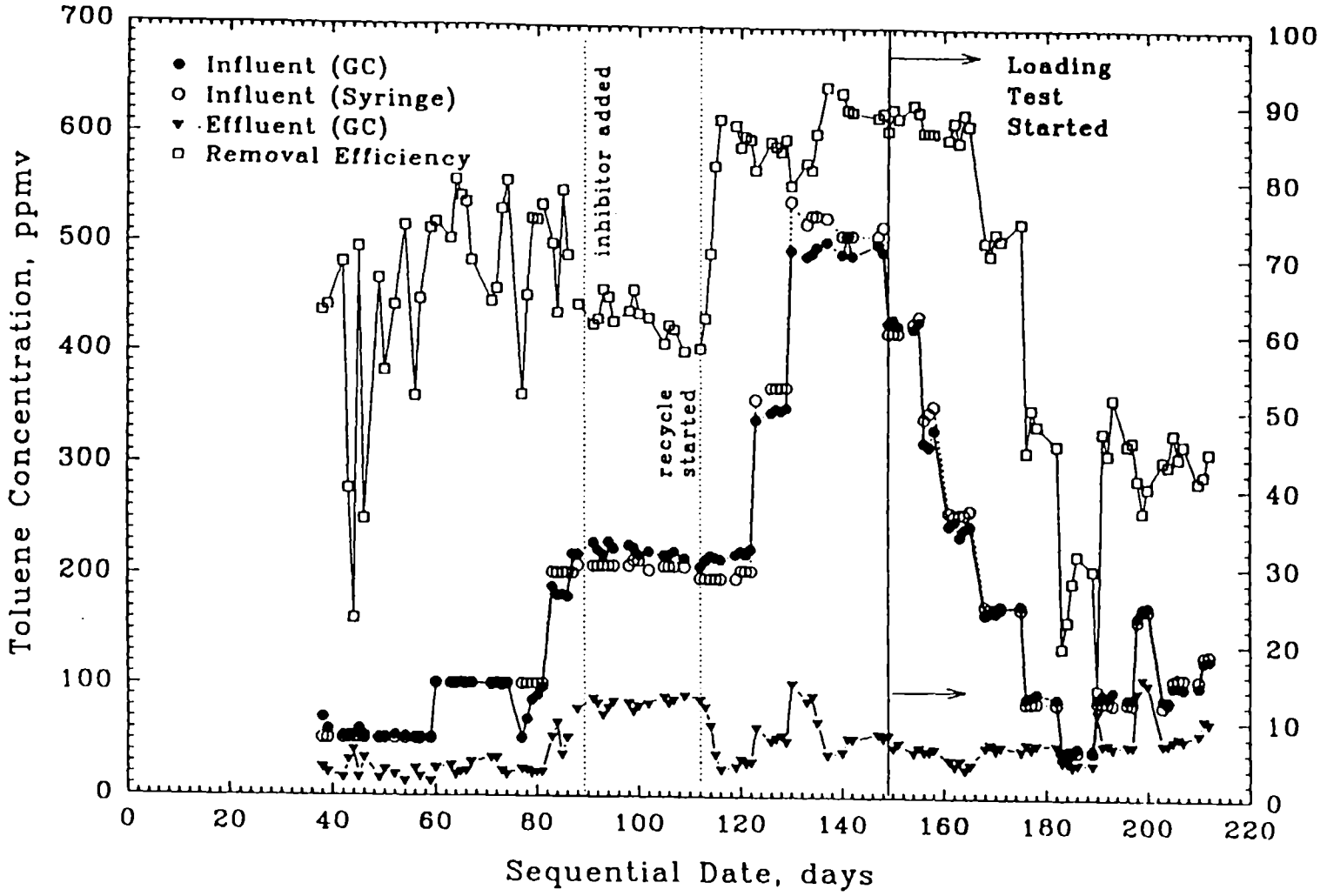

Figure 5. Biofilter "B" Performance w.r.t. Toluene Removal.

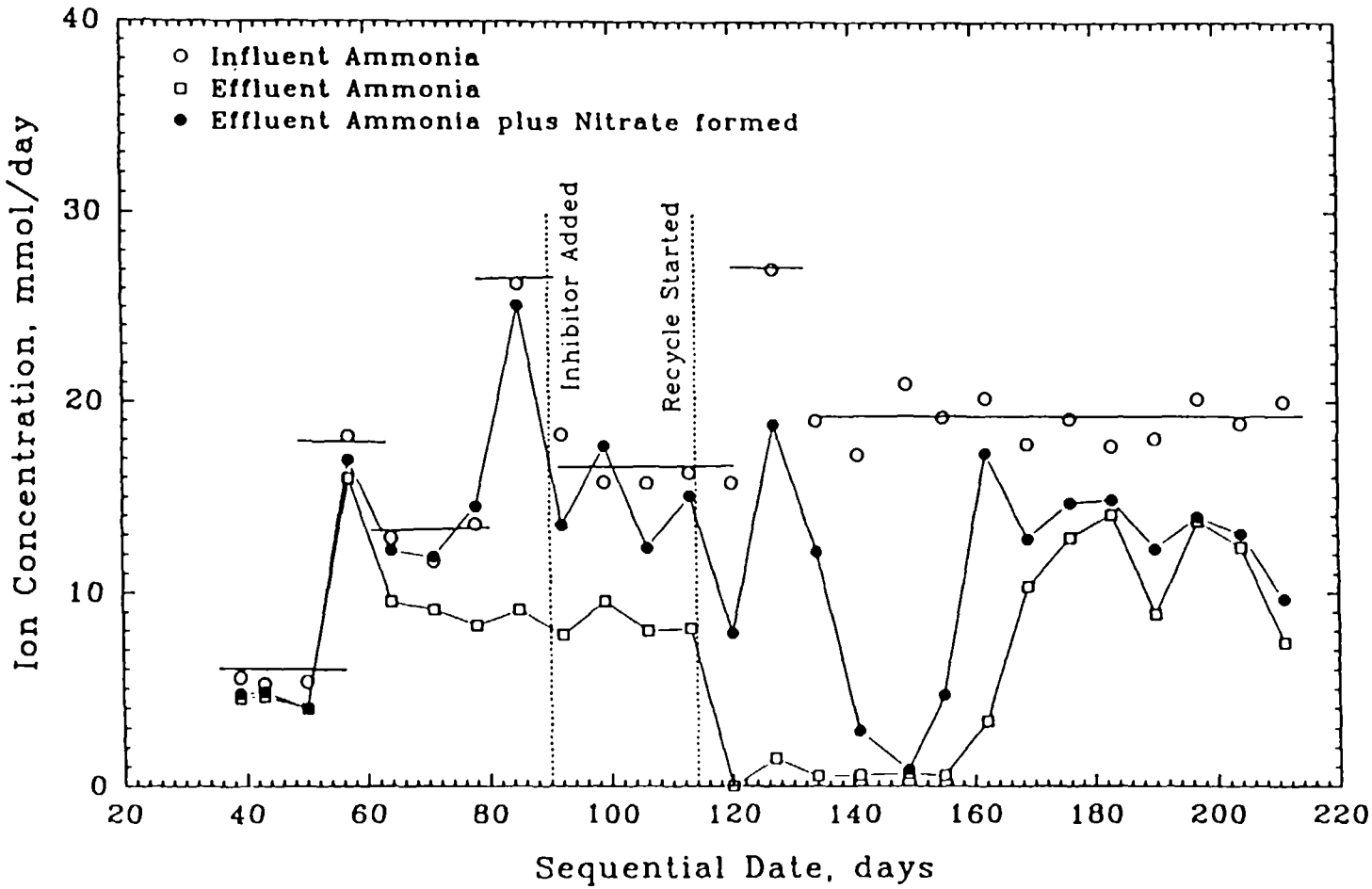

Figure 6. Biofilter "B" Performance w.r.t. Ammonia Consumption.

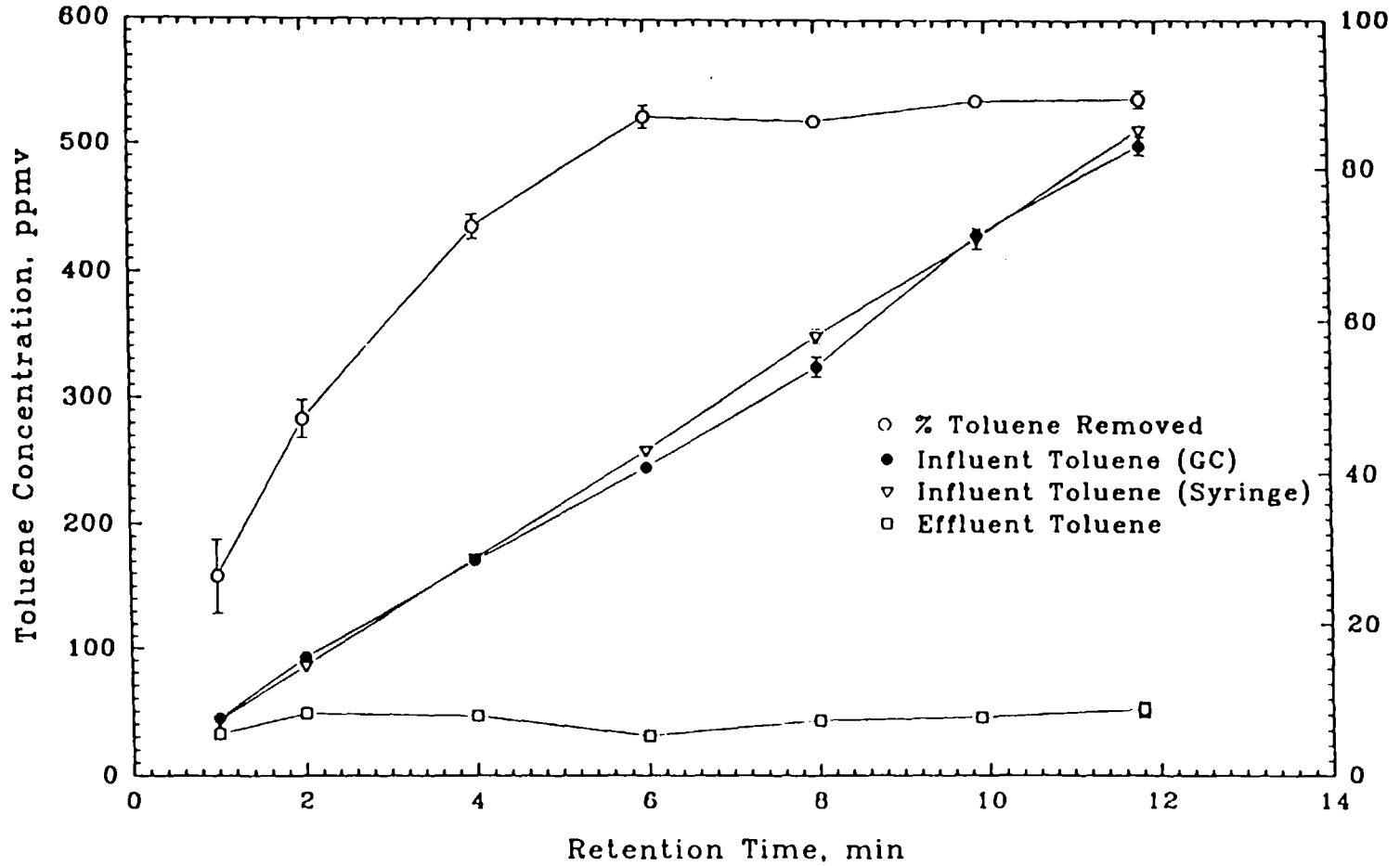

Figure 7. Biofilter "B" Performance w.r.t. Toluene Removal During a Residence Time Cycle.

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16. ABSTRACT

This paper reports preliminary results on the use of trickle bed biofilters with monolithic ceramic channelized microbial support structures for the treatment of VOCs typical of landfill leachate stripping. Toluene was used for the purpose of characterizing the trickle bed biofilter apparatus. The objectives of the experiment were to investigate the performance of such biofilters, with both cocurrent and countercurrent gas VOC and liquid nutrient/buffer flows, at inlet toluene concentrations that are high, relative to most biofilter research to date. A future research objective is to reduce to practice biofiltration for the treatment of air streams containing mixtures of VOCs, such as ethylbenzene, chlorobenzene, trichloroethylene, and methylene chloride in addition to toluene.

17. KEY WORDS AND DOCUMENT ANALYSIS

a. DESCRIPTORS	b. IDENTIFIERS/OPEN ENDED TERMS	c. COSATI Field/Group
Air pollution, Toluene	Volatile organic compounds, Trickle bed biofilter, Ceramic Monolithic channelized media	

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