

THE INFLUENCE OF AIRFLOW ON INDOOR AIR BIOFILTRATION: ELIMINATION OF TOLUENE AND METHYLETHYLKETONE

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ABSTRACT

Biofiltration is a promising alternative to ventilation for maintaining indoor air quality. Researchers at the University of Guelph have developed a thin-layer trickle-bed biofiltration system that is capable of removing a variety of volatile organic compounds (VOCs) at very low concentrations and high airflow rates. This study investigated the removal of toluene and methylethylketone (MEK) at airflows from 0.025 to 0.4 m s⁻¹. Influent VOC concentrations followed a diurnal pattern; ranging from 10 to 80 ppbv, the resulting loading rates (per VOC) ranged from 0.5 to 10 g m⁻³ hr⁻¹. The packing materials consisted of a range of synthetic polyester fibre materials and a coconut fibre mat.

The biofilters operated under first order kinetics for MEK elimination over the range of airflow. However, the biofilters switched from first order to zero order toluene elimination at airflows over 0.1 m s⁻¹. This suggests that, under higher loading rates, toluene elimination became microbially limiting whereas MEK was substrate limited. The biofilters attained maximum elimination rates of 4.0 and 1.6 g m⁻³ h⁻¹ for MEK and toluene respectively. In recirculating air streams, elimination rates are more critical than single-pass removal efficiency. Hence, airflow should be adjusted towards attaining maximum elimination rates. These results will help define the optimum engineering specifications for incorporation of this technology into conventional building air handling systems.

INTRODUCTION

With Americans spending more than 90% of their time inside (Office of Air and Radiation 1989), indoor air quality (IAQ) is a growing concern. Indoor air quality is traditionally maintained through ventilation. The displacement of 'stale' indoor air with 'fresh' outdoor air dilutes the contaminants that tend to accumulate in the indoor air space. However, modern buildings are being increasingly sealed from the outdoors as an energy conservation strategy. This is especially true in climates of extreme heat and

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cold, where conditioning the incoming outdoor air is a substantial component of building operating costs. The consequence of 'air tight' buildings is that contaminants such as volatile organic compounds (VOCs) can accumulate to levels that may affect occupant well-being. In fact, the US EPA lists indoor air quality as one of the top 5 health concerns facing Americans (Jenkins, Phillips, Mulberg et al. 1992).

Indoor air characteristically contains a large consortium of VOCs, ranging widely in spatial and temporal concentrations (Godish 1991; Maroni, Seifert and Lindvall 1995). These contaminants arise from a wide variety of sources including building materials, cleaning supplies, electronics and occupant activities (Otson and Fellin 1992). The VOCs associated with poor quality indoor air also range widely in physical properties such as solubility and biological activity. Single VOC concentrations are typically less than 10^{-7} to 10^{-5} g m⁻³ and mixed VOC concentrations less than 10^{-5} to 10^{-3} g m⁻³ (Godish 1991). Many common indoor VOCs have been linked to acute and chronic health conditions. However, no single compound is typically present in concentrations high enough to influence occupant health. Rather, it is the combined synergism of a broad range of VOCs that leads to acute and chronic health concerns. Short-term symptoms include dizziness, fatigue, mucous membrane irritation, shortness of breath, headaches and irritability. Long-term exposure has been linked to asthma, organ and tissue damage, birth defects and cancer (Office of Air and Radiation 1989).

Biofiltration has been proposed as an alternative to ventilation for maintaining IAQ. Conventional, compost-based biofilters are a proven, energy-efficient technology for treating a wide variety of industrial waste gas streams. However, conventional biofilters are designed to treat 10^5 to 10^8 times higher VOC concentrations than are typical of an indoor space (Arcangeli and Arvin 1995; Deviny, Deshusses and Webster 1999). In addition, an indoor air biofilter must treat a recirculating air stream, that is the exhaust from the biofilter is directly returned to the occupied space which forms the influent for the filter. In this scenario, removal efficiency loses importance to the biofilter's overall impact on the indoor air mass (i.e. elimination capacity). Furthermore, effluent quality is influenced by additional parameters such as temperature, humidity and bioaerosols. These parameters must also be within acceptable levels to maintain occupant comfort.

Considering that standard ventilation rates range from 2 to 6 air changes per hour (ASHRAE 1989), an indoor air biofilter must be capable of treating extremely large volumes of air. The relatively low indoor VOC concentrations may be somewhat offset by the very large volumes of air to be treated. The practical solution, while minimizing the biofilter's footprint within the building, is to run air through the biofilter at a rapid rate. As a consequence, VOC loading rates may be comparable to conventional biofilters.

Researchers in the Controlled Environment Systems lab, Department of Plant Agriculture, University of Guelph have developed a thin-layer biofiltration system, designed to meet the constraints of small biofilter size capable of treating large volumes of air. These biofilters generally range from 1 to 5 cm thick and are made up of highly porous materials that allow high air fluxes. To date tested airflows have not exceeded 0.2

m s^{-1} and it is unknown how biofilters will respond above this rate. In addition, earlier work focused on the impact of loading rate on the removal of aromatics; this study has expanded that work to include ketones. Ketones have a simpler chemical structure, are significantly more soluble in water, and typically have higher biodegradation rates than aromatics (Devinny 1999). Toluene and methylethylketone (MEK) were chosen for this study due to their prevalence in indoor air (Kostiainen 1995; Mølhav, Clausen, Berglund et al. 1997).

Typical designs to date have been comprised of complex, botanically-based ecosystems (borrowing from phytoremediation concepts) growing in a porous, organic substrate base such as coconut fibre erosion matting. Green plants boost contaminant degradation (Shome, Darlington, Dixon et al. 2002) and are also included for aesthetic reasons. Although functional, the organic nature of the coconut fibre gives the material a relatively short life in the biofilter due to its own breakdown. Alternative materials need to be determined.

The purpose of this experiment was twofold:

1. Investigate whether synthetic media can act as a biofilter packing material (i.e. support a contaminant degrading population) for indoor air biofiltration.
2. Investigate the influence of airflow on contaminant degradation in indoor biofilters.

METHODS

Six individual, lab-scale biofilters were operated in a partially sealed, 65 m^3 laboratory at the University of Guelph, Ontario, Canada. Four of the biofilters were comprised of single layers of synthetic polyester weave material (3M Canada). These materials ranged in physical characteristics including coarseness, density, and porosity. The fifth biofilter was an open-weave, synthetic-coated coir (coconut) fibre material, typically used for particle filtration in air handling systems. The final biofilter packing material was coir fibre erosion matting. This material was much denser than the synthetic-coated coir as the fibres were compressed together to form the mat. Coir fibre mats have been

Material	Depth (mm)	Density (g/cm^3)	Displacement ($\mu\text{l/ml}$)
Synthetic 1	25	55.6 ± 2.1	37.6 ± 2.34
Synthetic 2	25	51.0 ± 1.34	45.2 ± 1.64
Synthetic 3	25	37.7 ± 1.04	38.3 ± 1.25
Synthetic 4	25	64.0 ± 3.42	51.0 ± 6.59
Synthetic-Coir Filter	40	28.7 ± 1.13	30.6 ± 1.08
Coir Fibre	20	57.1 ± 6.42	90.2 ± 2.73

Table 1. Physical properties of the six biofilter packing materials

previously used as a substrate for botanically-based biofilters (Darlington and Dixon 2002), and was considered as a control for this experiment. Each biofilter had a functional surface area of 0.075 m².

The biofilters were supported on rigid plastic grid (15 mm openings) within a 40 x 30 x 60 cm, open-topped container that acted as a plenum (Figure 1). The grids were placed approximately 20 cm below the top of the container (giving a plenum depth of 40 cm) and situated on a 15° angle to facilitate drainage. The biofilters were kept moist through an overhead mist irrigation system. The misters would activate for approximately 5 of every 120s, delivering approximately 50 ml of water per cycle. Excess water would drain from the bottom of the plenum to individual, 4 L sumps. Sump water levels were maintained with deionized water by using float valves. The biofilter nutrient solution was comprised of: K₂HPO₄ (0.1 g L⁻¹), MgSO₄*7H₂O (0.1 g L⁻¹), Fe(SO₄)₃*7H₂O (0.005 g L⁻¹), NH₄NO₃ (0.2 g L⁻¹), CaCl₂ (0.1 g L⁻¹). The sumps were cleaned and the nutrient solution was replaced weekly.

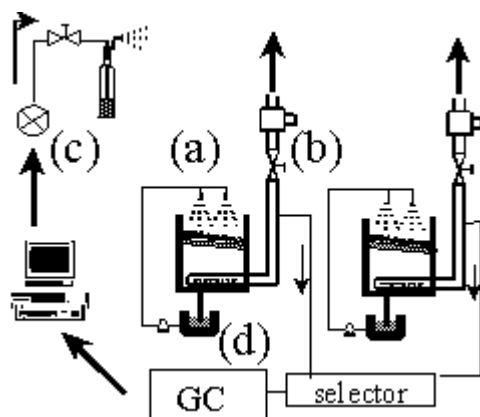


Figure 1. Schematic of biofilter apparatus (only two biofilters shown). a) misting system, b) effluent fan, c) VOC emitter, d) biofilter sump

Room air was drawn down through the biofilters with single-speed centrifugal blowers (Model 4C442, Dayton, Nile IL). Airflow was measured with a hand-held anemometer (Model 8346, TSI Inc., St. Paul, MN) and controlled by gate valves located in the ductwork upstream of the blowers. The biofilters were operated at flows of 0.025, 0.05, 0.1, 0.2, 0.3 and 0.4 m s⁻¹.

Airflow was randomized every other day in a Latin square design so that each biofilter operated at each flow rate once for a 2-day period. Once a biofilter has acclimated, no additional time is required for re-acclimation to changing airflows (Sorral, Smith, Suidan et al. 1997; Darlington and Dixon 1999).

A SRI model 310 gas chromatograph (GC) coupled to a custom gas sampling multiplexer was used to monitor VOC concentrations in the ambient air and the effluent stream of each biofilter (Figure 1). The GC operated in autosampler mode, measuring each biofilter effluent stream approximately once and the ambient air 5 times per hour. This provided real-time analysis of VOC removal. Biodegradation was measured as one minus the ratio of effluent concentration to a calculated, time-weighted ambient concentration (the removal efficiency). This removal efficiency could then be converted to mass loading and elimination rates with the inclusion of airflow and biofilter surface area or volume.

The GC was also used as the sensor in a feedback loop controlling the ambient VOC concentrations. Briefly, the GC would interface with a peripheral emitter system that could increase the ambient VOC concentrations if required, to desired levels by passing

metered volumes of air through impingers containing the specific pure solvent. VOC-saturated air would then be distributed into the room's air mass via the air handling system. Ambient VOC concentrations followed a diurnal profile ranging from 10 to 80 ppbv for MEK and toluene. This synthetic diurnal pattern reflects the fluctuations seen in indoor air.

RESULTS AND DISCUSSION

The 6 biofilters had acclimated to MEK and toluene about a week prior to the beginning of the experiment. Though the biofilters were not inoculated with VOC-degraders MEK, acclimation began within the first 2 days while toluene acclimation began by day 5 (data not presented). This phenomenon was consistent with previous indoor biofilters (Llewellyn, Darlington and Dixon 2000; Darlington and Dixon 2002).

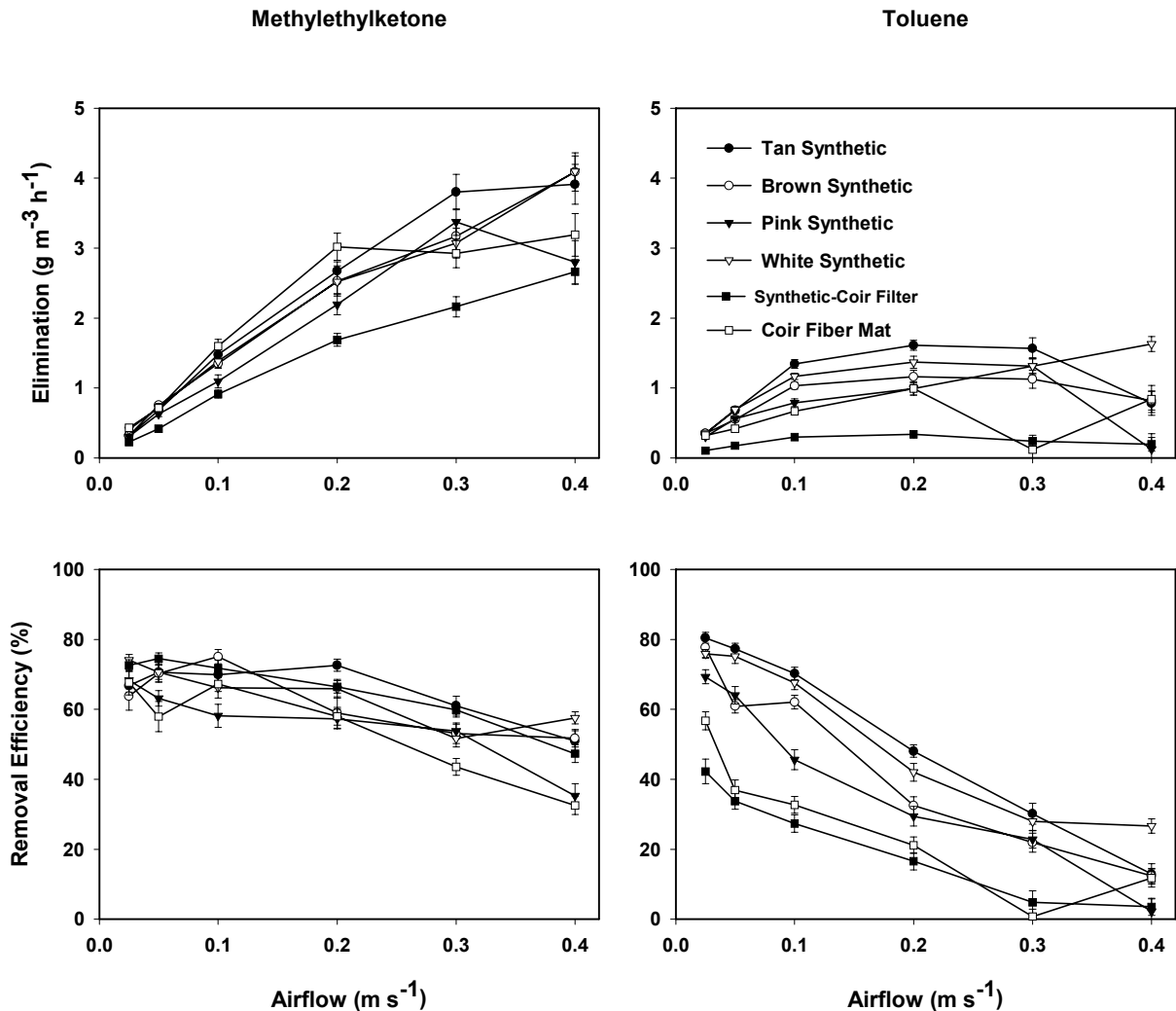


Figure 2. Removal efficiency and elimination of MEK and toluene under varying airflow rate. Each point represents the mean of 50 measurements over 48 h.

However, the rapid acclimation was surprising considering that the synthetic materials were seemingly abiotic environments. The most likely source of inoculum were microbes present within the indoor air stream.

It should also be noted that synthetic media offer little additional carbon source. Hence, the microbes appear capable of surviving on MEK and toluene as sole carbon sources. It has been stated that the organic base of the coir fibre may supplement the microbial growth in the biofilm (Darlington and Dixon 2002). However, the coir fibre biofilter did not perform significantly better than the synthetic media in terms of VOC removal (Figure 2).

All of the biofilters had similar removal efficiencies MEK over the range of airflow. MEK removal efficiency was relatively constant up to 0.2 m s^{-1} and only decreased by about 25 % when the flow was doubled to 0.4 m s^{-1} . As a result, MEK elimination capacities showed first order kinetics for all biofilter up to 0.2 m s^{-1} where it moved to half order up to 0.4 m s^{-1} . Furthermore, because the synthetic-coir filter was thicker, its relative elimination capacity was reduced compared to the other biofilters. This may be due to channelling as the pore size in this biofilter was much greater than the other materials. Alternatively, since MEK is so readily degraded, the majority of the removal may be occurring on the biofilter surface making the extra depth redundant.

The differences between biofilters with respect to toluene removal were much higher than MEK. For example, across the range of airflow, the tan and white synthetic materials had roughly double the toluene removal efficiency of the coir fibre mat and synthetic-coir filter. Toluene degraders may be more sensitive to their environment (i.e. physical characteristics of the different materials) than the MEK degraders. Further investigation into the impacts of these physical characteristics may be warranted.

All of the synthetic packing materials exhibited significantly greater toluene removal than the synthetic-coated coir and the coir fibre mat over the range of 0.025 to 0.3 m s^{-1} . The general reduction in toluene removal above 0.1 m s^{-1} may be due to bed channelling at elevated flows. With bed retention times of less than 0.1 s there may also have been inadequate contact between the airstream and the biofilm thus, diffusion becomes rate-limiting. Since toluene is much less soluble than MEK, it is not surprising that this phenomenon was observed only in toluene removal.

The average MEK and toluene eliminations of the synthetic biofilters in this study were 1.08 ± 0.117 and $1.32 \pm 0.082 \text{ g m}^{-3} \text{ h}^{-1}$ at 0.1 m s^{-1} . Shome et al. (2002) reported MEK and toluene elimination rates of 0.37 and $0.45 \text{ g m}^{-3} \text{ h}^{-1}$ at the same airflow for a 50mm thick biofilter of similar composition as the synthetics in this study. Darlington et al. (2001) reported toluene elimination rates of $1 \text{ g m}^{-3} \text{ h}^{-1}$ at airflows of 0.2 m s^{-1} in living moss-based biofilters. The synthetic biofilters compare favourably with average toluene elimination of $1.36 \pm 0.017 \text{ g m}^{-3} \text{ h}^{-1}$ at the same airflow. Hence, under similar loading conditions, the synthetic biofilters removed about 35 % more toluene than the moss biofilters. Darlington et al. (2001) also noted a 10 % increase in elimination between 0.1 and 0.2 m s^{-1} . The synthetic biofilters showed no significant change in toluene

elimination from 0.1 to 0.3 m s⁻¹ but 3 out of 4 exhibited a sharp decrease in toluene elimination at 0.4 m s⁻¹.

Conventional biofiltration research reports MEK and toluene elimination in the ranges of 40-120 and 4-100 g m⁻³ h⁻¹ respectively (Devinny 1999). Maximum MEK and toluene elimination in this study were about 4.0 and 1.6 g m⁻³ h⁻¹. These results compare very favourably with conventional biofiltration systems that typically have much higher inlet VOC concentrations and much longer bed residence times.

CONCLUSIONS

Synthetic polyester media were capable of supporting a VOC-degrading biofilm. In fact, the synthetic media performed as well as the coir fibre control in terms of MEK and toluene removal. Further research will investigate their use in botanically-based biofiltration systems.

The biofilters responded differently to increasing loads of MEK and toluene. MEK elimination had not saturated at the highest airflow of 0.4 m s⁻¹ while toluene elimination peaked at 0.1 m s⁻¹. This may have been due to their relative solubilities and reduced bed residence time at high airflows. It appears MEK is usually substrate limited while toluene is microbially limited in thin-layer indoor air biofilters. However, airflows above 0.2 m s⁻¹ may induce channelling change the dynamic of biofilm/ VOC contact.

It was previously believed that airflow through these thin-layer indoor air biofilters should be as fast as possible in order to maximize VOC elimination. This study has shown that there may be a threshold beyond which elimination does not improve with increasing flux but the threshold is VOC specific.

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REFERENCES

- Arcangeli, J and Arvin, E (1995). Biodegradation rates of aromatic contaminants in biofilm reactors. *Water Science and Technology* **31**(1): 117-128.
- ASHRAE (1989). *Ventilation for Acceptable Indoor Air Quality*. Atlanta, GA,, American Society Heating, Refrigeration and Air Conditioning Engineers.
- Darlington, A and Dixon, M A (2002). Acclimation and nutrition of indoor air biofilters. *International Journal of Indoor Air Quality and Climate*, Monterey, California.
- Darlington, A B, Dat, J and Dixon, M A (2001). The biofiltration of indoor air: air flux and temperature influences on the removal of toluene, ethylbenzene and xylene. *Environmental Science and Technology* **35**: 240-246.

- Darlington, A B and Dixon, M A (1999). Acetone removal kinetics by an indoor biofilter. SAE Technical Series Paper **1999-01-2069**.
- Devanny , J, Deshusses, M. and Webster, T. (1999). Biofiltration for air pollution control. New York, Lewis Publishers.
- Devanny, J S, Deshusses, M A and Webster, T S (1999). Biofiltration for Air Pollution Control. New York, Lewis Publishers.
- Godish, T (1991). Indoor Air Pollution Control. Chelsea, Michigan, Lewis Publishers, Inc.
- Jenkins, P L, Phillips, T J, Mulberg, E J and Hui, S P (1992). Activity patterns of Californians: use of and proximity to indoor pollutant sources. Atmospheric Environment **26A**: 2141-2148.
- Kostiainen, R (1995). Volatile organic compounds in the indoor air of normal and sick houses. Atmospheric Environment **29**: 693-702.
- Llewellyn, D, Darlington, A and Dixon, M (2000). The biofiltration of indoor air I: A novel reactor for a novel waste gas stream. 2000 USC-TRG Conference on Biofiltration, Los Angeles California.
- Maroni, M, Seifert, B and Lindvall, T (1995). Indoor air quality: a comprehensive reference book. The Netherlands, Elsevier.
- Mølhave, L, Clausen, G, Berglund, B, Ceaurriz, J D, Kettrup, Lindvall, T, Maroni, M, Pickering, A C, Risse, U, Rothweiller, H, B. Seifert and Younes, M (1997). Total volatile organic compounds (TVOC) in indoor air quality investigations. Indoor Air **7**: 225-240.
- Office of Air and Radiation (1989). Report to Congress on Indoor Air Quality, U.S. Environmental Protection Agency: 4-14.
- Otson, R and Fellin, P (1992). Volatile Organics in the Indoor Environment : Sources and Occurrence. Gaseous Pollutants. J. Nriagu. Toronto, John Wiley and Sons Inc.: 335-421.
- Shome, U, Darlington, A, Dixon, M and Llewellyn, D (2002). Higher plants in indoor air biofilters. 2002 USC-TRG Conference on Biofiltration, Los Angeles, California.
- Sorial, G A, Smith, F L, Suidan, M T, Biswas, P and Brenner, R C (1997). Performance of peat biofilter: impact of empty bed residence time, temperature and toluene loading. Journal of Hazardous Materials **53**: 19-33.