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Biofiltration to Treat Cyclic Air Emissions Produced in the Forest Products Industry

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Biofiltration to Treat Cyclic Air Emissions Produced in the Forest Products Industry

Final Project Report

SFM Network Project: Biofiltration of Gaseous Emissions from Forest Products Manufacturing

by

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EXECUTIVE SUMMARY

This research project investigates the use of biofiltration as an economical and energy efficient method to treat gaseous emissions from pulp and paper and wood processing operations. Biofiltration is an air pollution control technology that involves passing the polluted gas stream through a packed bed containing microorganisms that degrade the pollutant. It is a technology that is well suited to treating low concentration emissions where combustion or adsorption technologies are not appropriate. In addition, biofiltration may have a significant advantage in the wood processing industries since there is the potential to use waste wood as the packing that would be available on site. Although biofiltration is utilised to control a variety of continuous emissions involving odour and volatile compounds, its application to periodic emissions has not been systematically studied. Air emissions from certain processes (eg. brownstock washers, tank vents, and press vents) are cyclic in nature, having pollutant concentrations varying on time scales of the same order of magnitude as the residence time of a biofilter. Fluctuating air emissions at longer time scales (i.e. on the order of hours and days) are characteristic for industries having emissions only during operating hours and are on the same time scale as the doubling time of microorganims.

The objectives of this research are to examine the impact of cyclic loading on biofilter performance. The experiments involved running three biofilters in parallel with all conditions the same except the inlet concentration of the pollutant. One biofilter (control) received a constant concentration stream; the other two biofilters had the inlet pollutant concentration varying in a sinusoidal fashion with a given period and amplitude. All three biofilters received the same average pollutant loading. A mathematical model of the dynamic process has also been developed.

At the given set of operating conditions with a cycle frequency of 10 minutes, all three biofilters had similar removal rates and were able to achieve >80% removal with a loading rate of 30 g α-pinene$\cdot m^{-3}$ bed$\cdot h^{-1}$. These results indicate that steady state and cyclic operation are similar at this cycle frequency and that periodic operation is not a problem for a biofilter. These results are consistent with a mathematical model of the process, with the assumption that the kinetics of the microbial community are the same for cyclic or steady state operation. This indicates that results obtained from steady state studies can be directly applied to periodic emissions such as emissions from press vents. A maximum elimination capacity of 40 g α-pinene$\cdot m^{-3}$ bed$\cdot h^{-1}$ was observed across the first section of the biofilters.

As the cycle periods increases, biofilter performance begins to decrease. Biofilter performance at the beginning of the 'on' cycle is poorer in comparison to performance at end of the cycle period. Decreases in removal efficiencies of 4% and 13% were observed for cycle periods of 24 hours and 6 days, respectively, where the average α-pinene loading rate during the 'on' cycle was 50 g$\cdot m^{-3}$ bed$\cdot h^{-1}$. In both experimental runs nutrient and humidity requirements played an important role in maintaining high levels of α-pinene removal.

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INTRODUCTION

Through Title III of the Clean Air Act Amendments of 1990, the US Environmental Protection Agency has classified 189 Hazardous Air Pollutants (HAPs) targeted for reduction by the year 2000, methanol being one of the pollutants on this list. Methanol is a compound that is the main HAP that is emitted in the pulp and paper industry. There are many other air emissions of concern produced throughout the forest products industry. Kraft pulp mills emit odorous Total Reduced Sulphur (TRS) compounds such as hydrogen sulphide, dimethyl sulphide, dimethyl disulphide, and methyl mercaptans. Pulp and paper mills in general are also emitters of Volatile Organic Compounds (VOCs) including methanol, and terpenes such as α-pinene.

The types of emissions at a wood products facility in the manufacture of particle board and other laminated wood products depend upon the types of woods processed. During the manufacturing of oriented strand board (OSB) and particleboard from softwood (e.g. pine), phenol and formaldehyde (both HAPs), are typically emitted during press operations along with VOCs such as terpenes (e.g. α- and β-pinene). In the manufacture of hardwood products, primarily methanol and formaldehyde are emitted (Togna et al. 1997). From an OSB mill (source material is Aspen) in Alberta, Canada, the major toxic components, as listed by the US EPA's CAAA, identified were: methanol, phenol, and formaldehyde. Other compounds identified in these emissions include: straight-chain alkanes (C_{16-25}); formic, acetic, butyric, benzoic and propionic acid; aldehydes; and furans. Some of these compounds were emitted at concentrations that exceeded their threshold limit values (TLVs). Since many of the pollutants come directly from the wood, the type of wood stock used will determine the specific makeup of the emission to some extent. However, some of the compounds in press vent emissions are also due to the resins and press release waxes used during the process (Coleman and Dombroski 1996).

It is interesting to note that in the two year period between 1993-1995 an estimated $394 million was spent by Canadian mills on Air Pollution Control (APC) (Sears et al. 1995). As environmental regulations move to tighten air emission limits, industries will continue to invest in a variety of technologies to treat their waste gases. Biofiltration is a promising alternative to other gas treatment technologies, particularly for distributed emission sources of dilute compounds where collection and combustion is not cost effective.

Biofiltration Technology

Biofiltration is an air pollution control technology that uses microorganisms to biologically degrade pollutants. A biofilter is essentially a packed bed reactor containing microorganisms growing in an active biofilm (Figure 1). This biofilm is supported on the reactor bed material (filter medium) usually consisting of some type of compost, peat, or soil material. The contaminated air stream passes through the filter medium where the pollutant is transferred from the vapour phase to the biofilm. The model pollutant being used in this study is α-pinene, a volatile organic compound

that is naturally present in softwood species and is emitted during various wood processing operations. Alpha-pinene has a low water solubility and degrades more slowly than more water-soluble compounds such as methanol (Mohseni and Allen 2000). It is therefore considered a good model compound representative of others with low water solubility such as dimethyl disulphide.

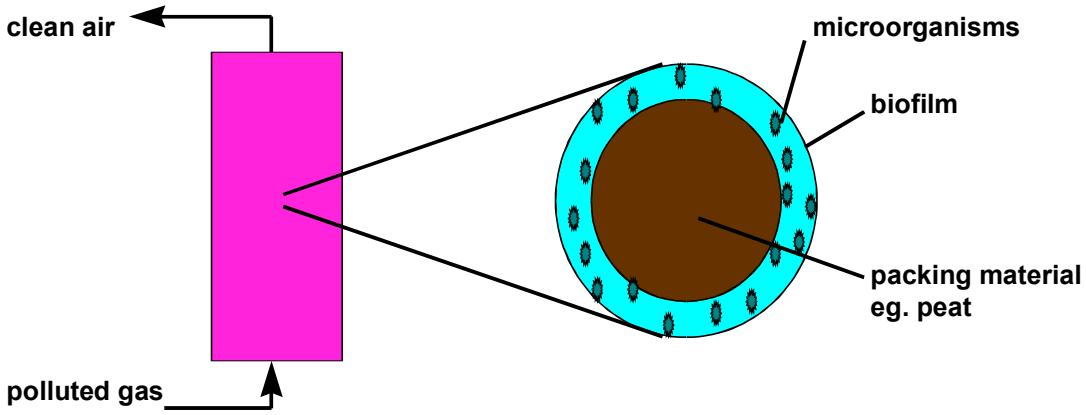

Figure 1 - Biophysical model for the biofilter

Biofiltration has shown promise for treating large volumes of air emissions containing low concentrations of pollutants typical to pulp and paper industries. Biofiltration has been considered as a potential technology to be considered for Best Available Control Technology (BACT) for wood panelboard furnish material dryers (Lamb 1994). Although biofiltration has achieved widespread acceptance and successful application throughout Europe as a preferred method of treating contaminated, dilute waste gases, North American industries have been slow to follow this trend. One reason for this is that insufficient knowledge and experience exists on the different processes that occur during steady state and transient operation of the biofilter. Biofilter performance is dependent on many factors, including, the microbial community, contaminants to be degraded, and reactor operating conditions such as moisture content, pH, temperature, pressure, residence time, and nutrient availability. Microbial degradation kinetics play a vital role in the design and scale up of a biofilter and therefore, it is imperative that these be well understood and defined if biofiltration is to be used successfully in industrial applications.

Motivation for Research

Air emissions from certain processes such as brownstock washers, tank venting, and press vents in the wood processing industries are periodic in nature, and as such, the pollutant concentrations vary with time. This is due to the periodic nature of the process or unit operation producing the emission. Fluctuations can be the result of seasonal changes in operation, daily variations in operating conditions, and hourly and/or minutely variations due to process conditions and operation. It is difficult, if not impossible, to control the amounts of pollutants in the off gases and it is therefore impractical to control the concentration of contaminants in the air emission. Fluctuating concentrations may have a significant impact on biofiltration when one considers that the residence time, τ, in biofilters is short (on the order of one minute) and on the same time scale as the

concentration fluctuations. This is quite different in comparison with wastewater treatment where the residence time is much greater (on the order of days); in this case rapidly changing inlet concentrations may not affect treatment system performance since they are damped out in the reacting vessel. To date there has been no dedicated study in the area of high frequency, cyclic, concentration fluctuations on biofiltration performance.

Research Approach

The thrust of the work focuses on the impact of the cyclic nature of waste gas emissions on biofilter performance (degradation kinetics and transient response). We are particularly interested in examining how exposure to cyclic feeds influences the biofilter's capability to handle such variations in feeds in comparison with a biofilter that has run in steady state. It is quite conceivable that acclimation of the microbial population to cyclic operation is important for handling periodic episodes of high concentration and so this may also have application to situations where periodic excursions from 'steady state' occur.

The overall goals of this research project were to consider the impact of cyclic pollutant loading on biofilter performance and to develop a kinetic model to describe the cyclic behaviour and transient response of the biofilter. These objectives were carried out by running two long-term biofiltration experiments treating an α-pinene laden waste gas in a biofilter packed with wood chip media. This was achieved by running three laboratory scale biofilters in parallel, two of which were treating a cyclically fluctuating pollutant gas stream and the third biofilter treating a constant concentration stream.

The first experimental run was carried out to monitor the performance of a cyclically operated biofilter at high cycle frequency and compare it to the performance a constant biofilter. The variation in the feed concentration cycled with a consistent frequency in the range encountered in many practical semi-batch operations (e.g. peak loads every few minutes as per a press vent). All three biofilters were receiving the same averaged pollutant loading (g of pollutant$\cdot m^{-3}$ bed$\cdot h^{-1}$). Over the course of the experimental run, the total inlet loading to the biofilters was varied. In this way, the affect of cycle amplitude on cyclic biofilter operation was also studied. The biofilter packing material used for this run consisted of a wood chip/compost mixture.

The second experimental run was set-up to look at the affect of low cycle frequency and its affect on biofiltration operation. Again, cyclic behaviour was compared to a control biofilter operating at constant concentration. Two different cycle periods were investigated. A 24-hour cycle period was used to mimic an intermittent emission characteristic of an industry operating on 12-hour workdays. A 6-day cycle period was chosen to resemble a typical weekend shutdown period. The biofilter packing material used for this run consisted of a bark chip/compost mixture.

The unsteady state model for the cyclic biofilter was based on an existing steady state model and expanded to include the time derivative. As a first approximation, steady state kinetic parameters from the constant concentration biofilter where used in the unsteady state model.

SUMMARY OF DATA ANALYSIS

Experimental Methodology

The experimental objectives were accomplished by setting up and operating a biofiltration system capable of generating a synthetic waste gas with cyclic concentration fluctuations. Three biofilters were operated in parallel. Two of the biofilters were operated cyclically with both biofilters running with the same cycle amplitude and frequency (replicates), and the third biofilter served as a control and was treating a constant concentration stream. All three biofilters received the same averaged pollutant loading. Figure 2 shows a schematic of the biofiltration set-up.

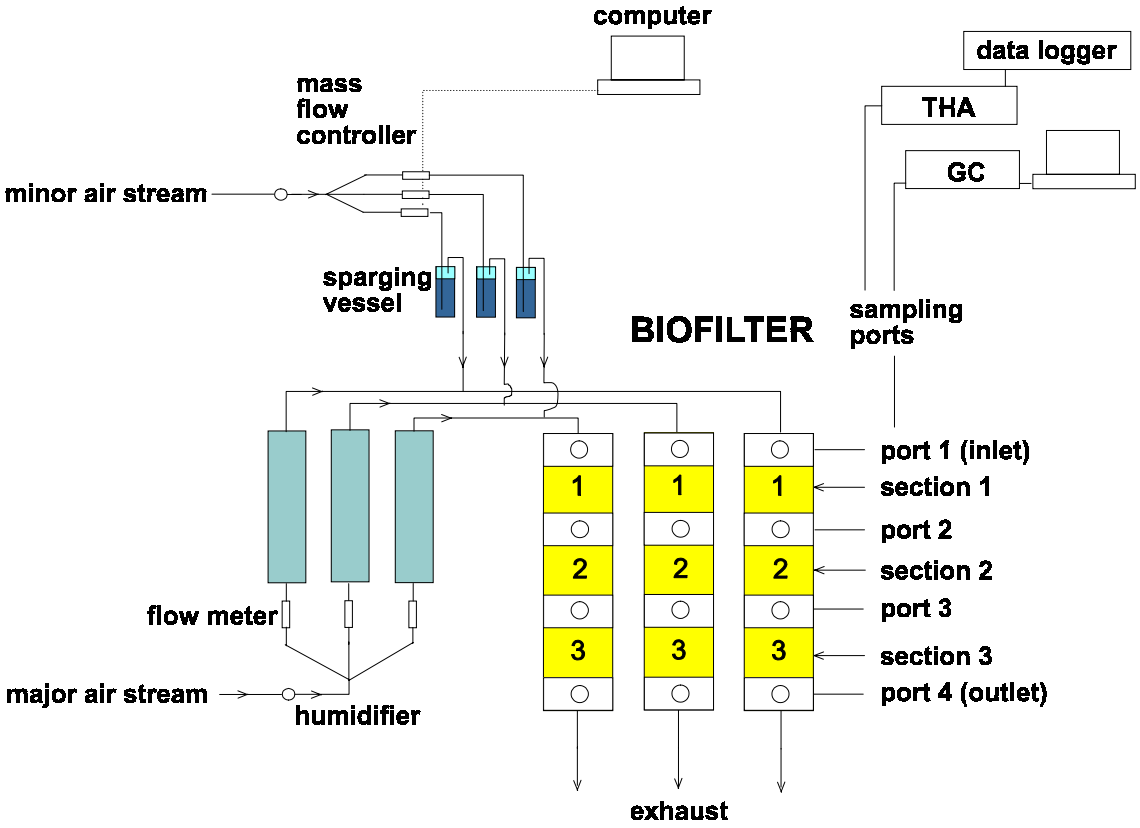

Figure 2 - Schematic of experimental biofiltration set-up

A synthetic waste gas was generated by bubbling air through liquid α-pinene (minor air stream) and mixing it with a humidified air stream (major air stream) before entering the biofilter. Gas phase α-pinene concentrations were measured at the inlet, outlet, and along the axial length of the biofilter using a total hydrocarbon analyzer (THA; MSA Model 8800, Baseline Industries, Inc., Lyons, CO). The inlet concentration to the biofilter was controlled by controlling the air flowrate through the sparging vessels. Mass flow controllers (MKS Type 1179A, Andover, MA) were used to vary the flow rate through the sparging vessels and these controllers were linked to a computer system where a computer program was used to adjust the air flow rate setpoints. Table 1 lists the

operating parameters over the course of the experiment. A detailed description of the experimental apparatus and set-up has been described elsewhere (Dirk-Faitakis 2000).

Table 1 - Biofilter operating conditions for experimental run

Variable	Parameter	Value	Units
A	Cross-sectional area of biofilter	177 (0.0177)	cm^2 (m^2)
H	Height of packing	34	cm
V_{total}	Total (empty) volume of biofilter	9.6	dm^3
V_{packed}	Packed volume of biofilter	6.0 (0.006)	dm^3 (m^3)
ε	Porosity of packing material	0.5	------
d_p	Average particle diameter	0.8	cm
U_g	Superficial gas velocity	8.96×10^{-3}	$m \cdot s^{-1}$
$T_{biofilter}$	Temperature of biofilter	25	ºC
% R.H.	Relative humidity of inlet air stream	> 95%	------

Summary of Experimental Results

Experimental run#1 - biofiltration of high frequency cyclic emissions

The biofilters were operated continuously for a period of 271 days during which time they were treating loadings of up to 30 g α-pinene$\cdot m^{-3}$ bed$\cdot h^{-1}$. Both the cyclically operated biofilters (BF1 and BF2) and the biofilter treating a constant concentration stream (BF3) achieved 80% removal efficiency at this loading while running at an empty bed retention time (EBRT) of 40 seconds.

Nutrient limitations of wood chip packing

The experiment was started with an average α-pinene inlet concentration of 15 ppmv and an empty bed retention time EBRT of 40 seconds, corresponding to a loading rate of 7 g α-pinene·m^{-3} bed·h^{-1}. Biofilters 1 and 2, running at the same cycle amplitude and frequency, saw inlet concentration fluctuations of 10 and 20 ppmv over a cycle time of 10 minutes while Biofilter 3 had a constant inlet concentration of 15 ppmv. After a 30 day start-up period, a non-inoculated biofilter composed of an 80/20 wood chips/compost mixture, to which no supplemental nutrients had been added, showed no signs of pollutant removal (Figure 3). On day 34, nutrients were added to all three biofilters in the form of a controlled release fertilizer and, subsequently, removal increased several fold.

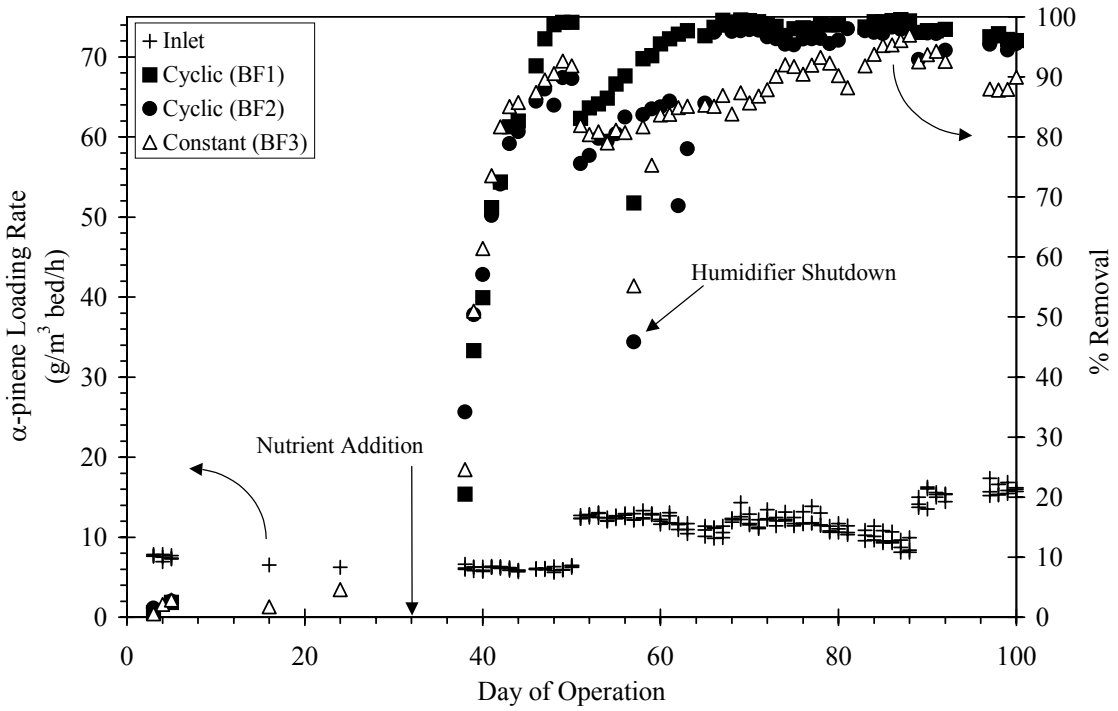

Figure 3 - Performance from day 0 to day 100; crosses (+) represent the averaged inlet loading rate (left axis), solid symbols represent the % Removal for the cyclically operated biofilters (BF1 ■ and BF2 ●) and the hollow triangles (Δ) represent the % Removal for the biofilter (BF3) with constant concentration loading (right axis).

Overall biofilter performance

Figure 4 shows the performance of all three biofilters from days 30 to 270 of operation. By day 50 all three biofilters had achieved 90% removal efficiency. On day 50, the average loading rate to all three biofilters was increased from 7 to 12 g α-pinene·m^{-3} bed·h^{-1} with an average concentration of 25 ppmv. By day 88 both the cyclically operated biofilters (BF1 and BF2) and the biofilter treating a constant concentration stream (BF3) were achieving >95% removal efficiency. The loading rate was further increased to 30 g α-pinene·m^{-3} bed·h^{-1}. Taking into consideration the

variability in the data, it can be seen that all three biofilters exhibited similar α-pinene removal efficiencies. At the given set of operating conditions with a cycle frequency of 10 minutes, cycle amplitude of 10 to 60 ppmv α-pinene, and a loading rate of 30 g α-pinene·m^{-3} bed·h^{-1}, there does not appear to be a significant difference in the removal efficiency of the three biofilters.

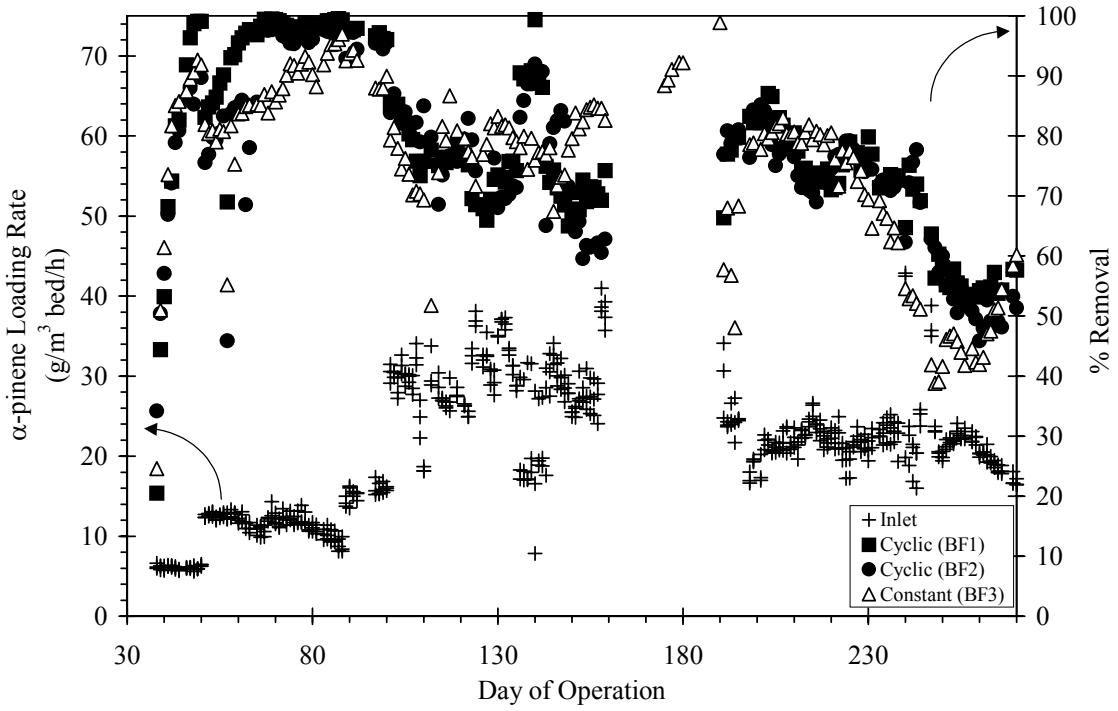

Figure 4 - Overall biofilter performance from day 30 to day 270; crosses (+) represent the averaged inlet loading rate (left axis), solid symbols represent the % Removal for the cyclically operated biofilters (BF1 ■ and BF2 ●) and the hollow triangles (Δ) represent the % Removal for the biofilter (BF3) with constant concentration loading (right axis).

Wood chip packing media performance

Wood chips served as a good packing material for the biofilters but were limited by moisture distribution problems. A decrease in the humidity of the entering air resulted in a decrease in microbial activity, but by restoring moisture to the biofilter this was reversed. On day 57 the humidifiers shutdown (due to pump failure for the water re-circulation) and the biofilters began to dry out (Figure 3). After less than 24 hours with an inlet air humidity of <15%, the removal efficiency of the biofilters decreased by as much as 45%. The packing material in the top sections of Biofilters 1 and 2 were mixed together and re-packed. The packing material in the top section of Biofilter 3 was allowed to recover on its own; moisture was restored to the media by adding 200 ml tap water to the top section of each biofilter on days 75, 78, 79 and 80. By day 88 all three biofilters had a removal efficiency of >90%.

Experimental run#2 - biofiltration of low frequency cyclic emissions

The biofilters were operated continuously for a period of 94 days during which time they were treating inlet loadings of 49 g α-pinene·m^{-3} bed·h^{-1}. The biofilters were operating on a 24-hour (12 hour ON/OFF) cycle from days 21-83 and on a 6-day (3 day ON/OFF) cycle from days 83-94. Overall performance indicates that both the cyclically operated biofilters (BF1 and BF3) and the constant biofilter (BF2) achieved comparable removal rates of α-pinene. Between 53-76% removal efficiency was achieved at an average loading rate of 49 ± 5 g α-pinene/m^3 bed/h and EBRT of 40 seconds.

Start-up and acclimatization

In this experiment all three of the biofilters experienced the same start-up conditions. All of the biofilters were operating with a constant concentration at the inlet from day 0 to day 21 of the experiment. The loading rate was ~47 g α-pinene/m^3 bed/h. This start-up was to allow for the growth and establishment of similar operating conditions (e.g. microbial community, biomass buildup, etc.) so that direct comparisons among the filters could be made once the ON/OFF cycle began. On day 21, two of the biofilters (BF1 and BF3) were switched to a cyclic inlet concentration, and the third biofilter (BF2) was maintained at constant inlet concentration. The same inlet concentration was maintained for all three biofilters and so during the ON part of the cycle, all three biofilters were treating the same pollutant loading. The "cyclic biofilters" had an off time of 12 hours while the "constant biofilter" kept running at the same loading all the time. Figure 5 shows the start up of the three biofilters for Experimental Run#2.

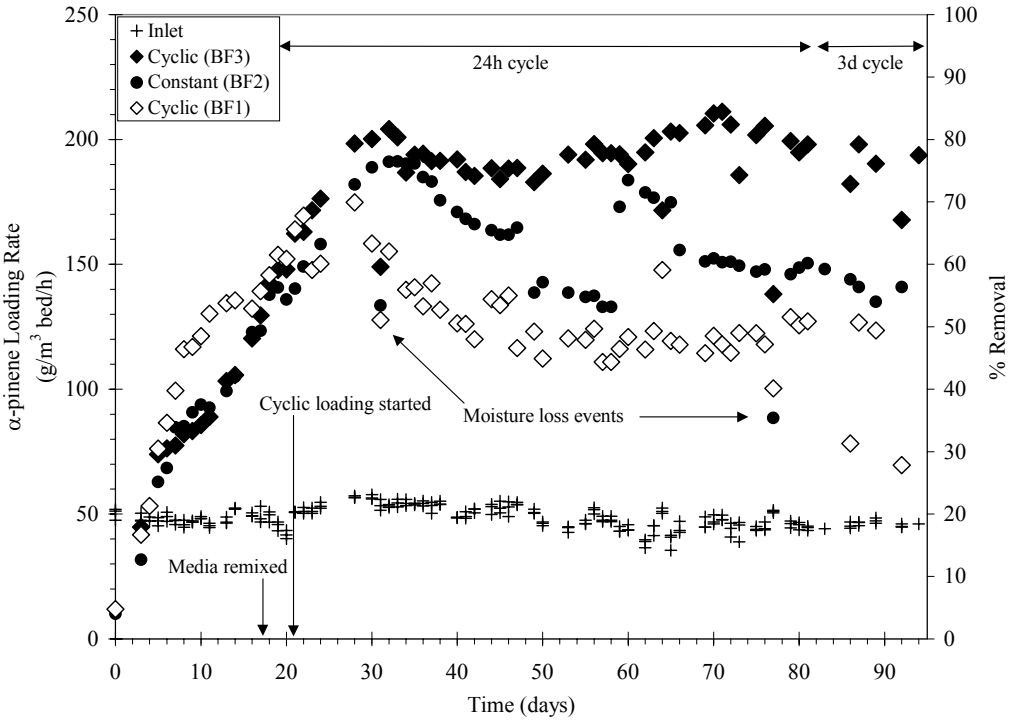

Figure 5 - Start-up period for Experimental Run#2. Cyclic loading to BF1 and BF3 was started on day 21 of the experiment.

Overall biofilter performance

The long-term biofiltration performance of Experimental Run#2 is shown in Figure 6. From day 28 to day 76 a pseudo steady state operation was reached. This was defined with respect to microorganism number. After a 4-week start-up period it was assumed that a stable, constant microbial community had been established and that therefore all reaction rates were constant over time. The average removal efficiency of α-pinene in the cyclic biofilters over this time period was 51 ± 6% (BF1) and 77 ± 4% (BF3). The removal efficiency of the constant biofilter was 65 ± 8%. Although there is a significant difference in the removal efficiency between the two cyclic biofilters, this has been attributed to operational problems experienced in BF1, specifically lack of moisture control occurring between day 30 and 31. The decrease in performance of BF1 after this time is attributed to this occurrence.

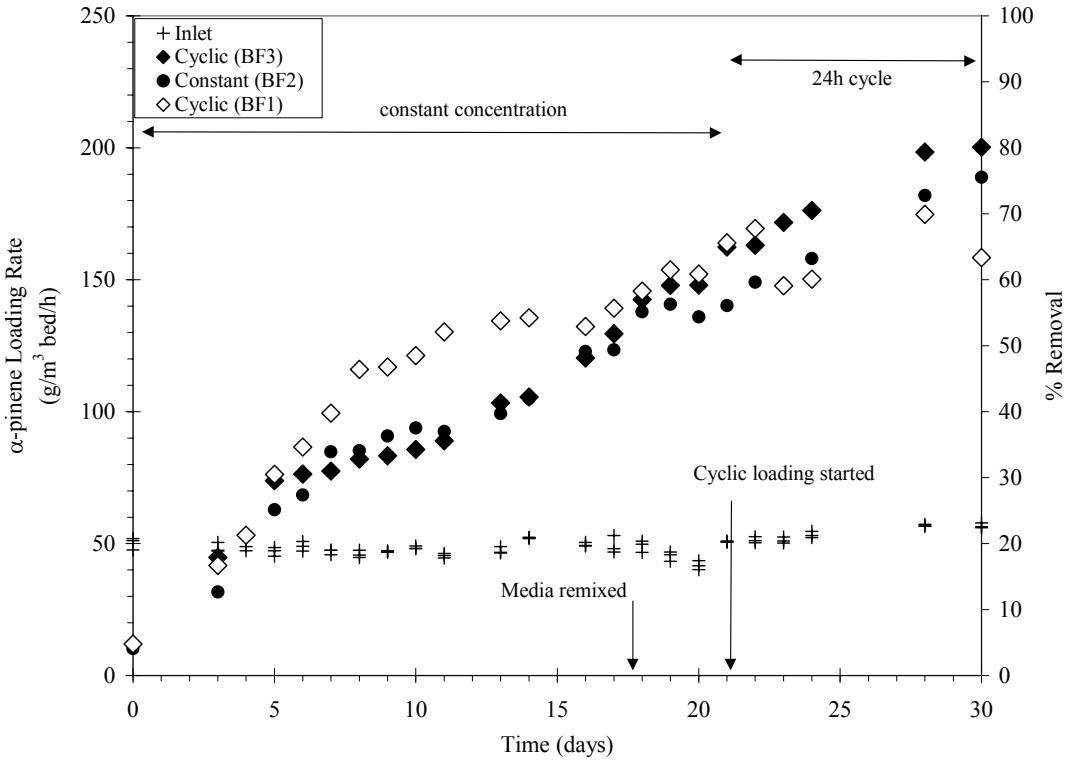

Figure 6 - Long term biofiltration performance for Experimental Run#2.

During the course of this experiment two moisture loss events, namely on day 31 and day 77, contributed to a significant decrease in overall biofilter performance. Of these two events, the loss of moisture control on day 31 had a serious and lasting affect on the biofiltration performance of the cyclic biofilter, BF1. On day 31 a decrease in performance was observed for all three biofilters due to water circulation problems in the humidifiers. After humidity control was re-established two of the biofilters (BF2 and BF3) regained previous levels of α-pinene removal efficiencies. Although the removal efficiency of Biofilter 1 increased after humidity control was re-established, previous levels of α-pinene removal for the first (inlet) section of Biofilter 1 were never achieved. Consequently, removal activity remained low in the first section of BF1 for the remainder of the

experiment. A reasonable explanation for this phenomenon is that irreversible moisture loss may have occurred from the packing material. Once compost and wood chips have dried out below a certain level, re-wetting through humidification alone will not increase the moisture content of the media, substantially. Only through direct water contact, like soaking the wood chips in water, can the media reabsorb water. A significant loss in moisture content translates into reduced microbial activity and an eventual loss in microorganism number.

On day 77, a short term loss in inlet humidity for 6 hours resulted in a decrease in biofilter performance, but after humidity was re-established all three biofilters recovered and were operating at their previous removal levels.

Effect of Cycle Period

Shortly after day 21, when two of the three biofilters were switched to a cycle period of 24 hours (12 hour ON/OFF cycles), the cyclic biofilters began to exhibit differences in performance over the duration of the ON cycle. The performance of the cyclic biofilters increased over the course of the ON cycle, such that the cyclic biofilters had a higher percentage removal of α-pinene near the end of the cycle when compared to α-pinene removal at the beginning of the ON cycle. This can be seen from Figure 7, which shows the α-pinene concentration at the exit of the biofilter over the course of the ON cycle. While the inlet α-pinene concentration remains constant over the 12-hour period the outlet concentration reaches a maximum of 31 ppmv and then slowly declines to a value of 27 ppmv.

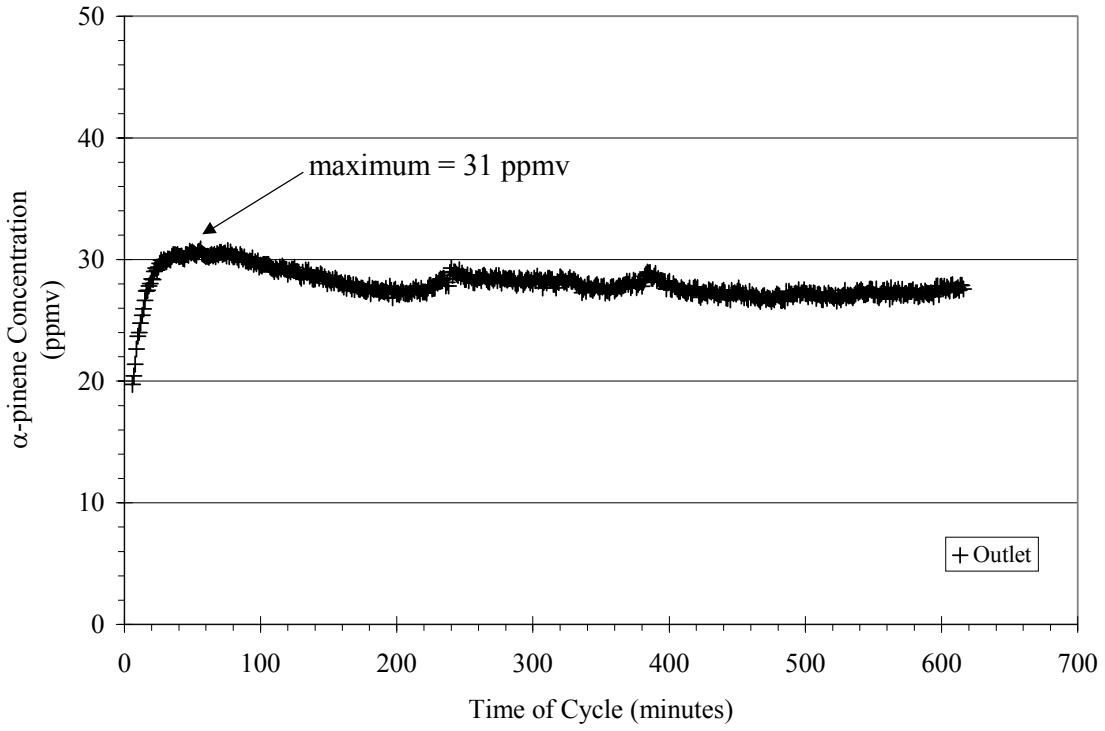

Figure 7 - Typical outlet concentration profile for 12 hour ON cycle for Biofilter 3 on day 48 of the experiment. Time zero represents the start of the ON part of the cycle.

The differences between morning and evening percentage removal efficiencies were compared for all three biofilters. As shown in Figure 8, the cyclic biofilters had removal efficiencies 4% higher at the end than at the beginning of the ON cycle. This 4% difference was statistically significant at the 95% confidence level based on a paired comparison and a sample size of 5 days. In comparison, the constant biofilter showed no significant difference in removal efficiencies between morning and evening performance. Although the cyclic biofilters exhibited somewhat poorer performance at the beginning of the ON cycle, this difference is likely too small to be of practical significance. An overall comparison shows that there no significant difference between the cyclic and the constant biofilters within the variability of the data.

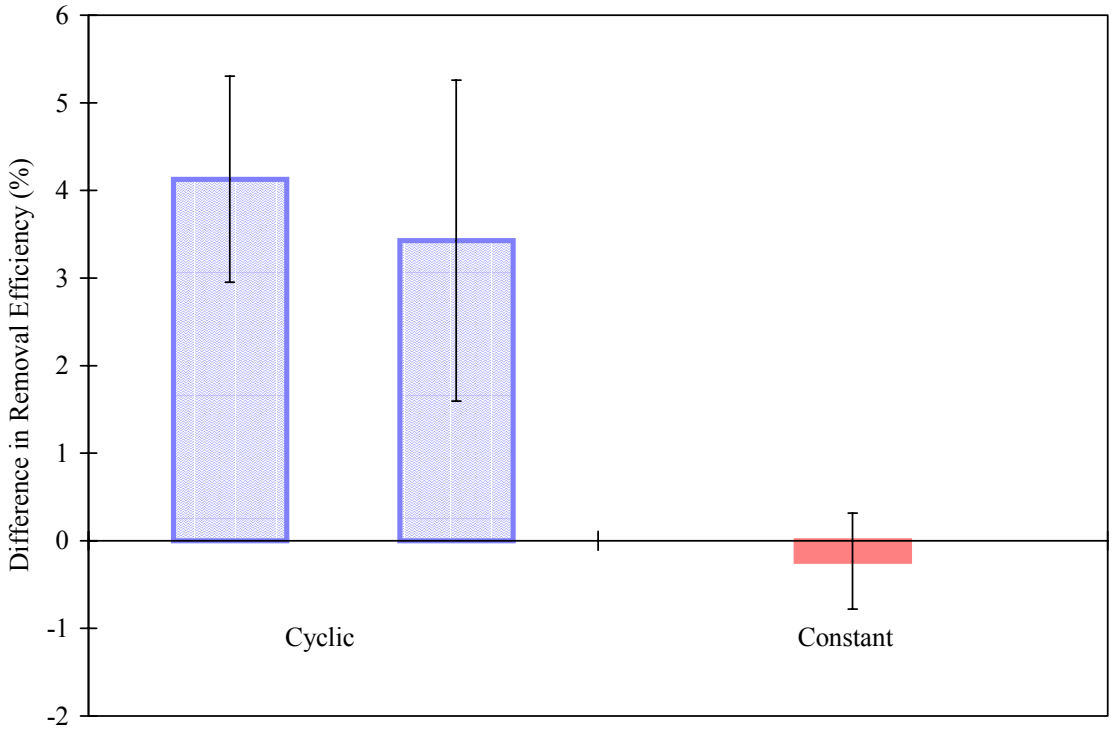

Figure 8 - Comparison of differences in removal efficiencies for morning and evening measurements. Error bars represent the standard deviation in the measurements.

This difference in removal efficiency during a cycle became more pronounced when the cycle period was increased to 6 days (3 day ON/OFF cycles). In this case, for an inlet concentration of 88 ppmv, a difference of 14% in removal efficiency was observed over the duration of the ON cycle. That is, the removal efficiency of the biofilter increased by 14% over the duration of the ON cycle. Figure 9 is a comparison of outlet concentration profiles during the ON cycle for Biofilter 3 for cycle periods of 24 hours and 6 days.

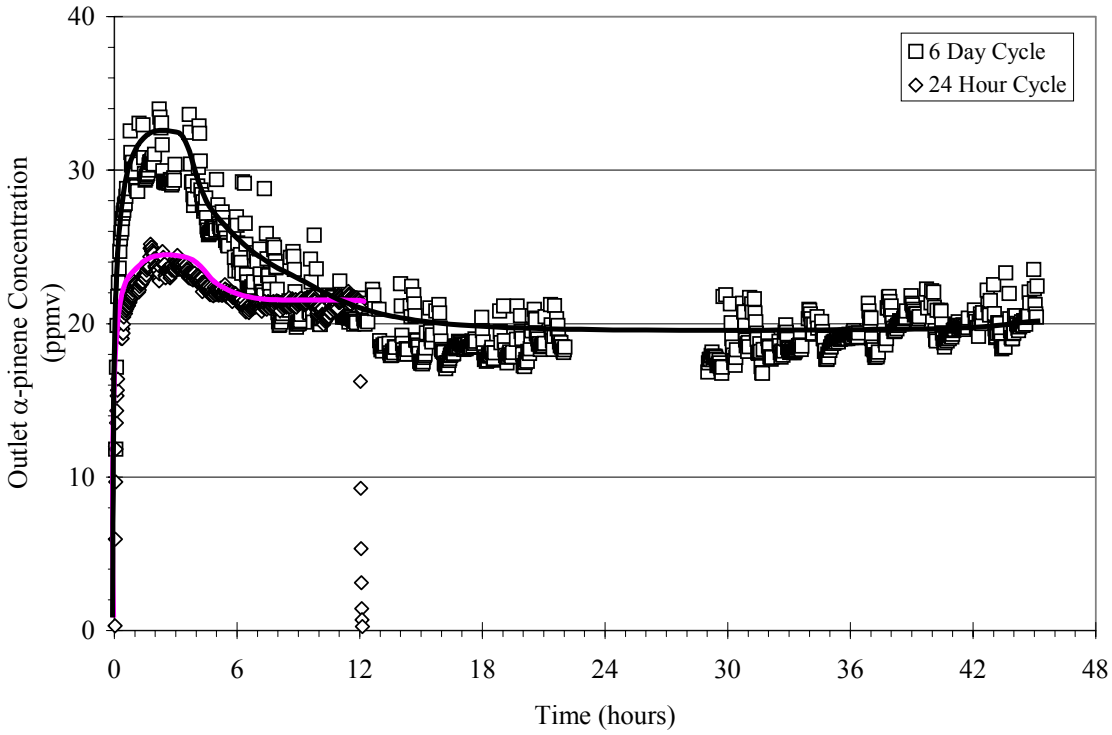

Figure 9 - Comparison of outlet concentration profile for Biofilter 3 during 6 day and 24 hour cycle periods.

It becomes apparent that as the cycle period is increased, the difference between beginning-of cycle versus end-of-cycle removal efficiency increases. The longer the period of cycle OFF time, or non-loading to the biofilter, the poorer the performance at the start of the ON cycle. From Figure 9 it can also be seen that the poorest performance occurs at approximately two hours after the beginning of the ON cycle. There is a short time lag for the inlet concentration to reach its steady state value. Since it takes less than 15 minutes for the inlet concentration to reach its steady state value, some other phenomena is occurring in the first two hours into the ON cycle. It is believed that adsorption of α-pinene to the tubing, biofilter walls, and biofilm is probably occurring at the beginning of the ON cycle and that is why the outlet maximum is not seen immediately after the cycle is started up again. From Figure 9, the recovery times of the biofilter to 95% of its previous removal efficiency can be estimated. The recovery time is less than 1 hour for a cycle period of 24 hours, and between 6-8 hours for a cycle period of 6 days.

Theoretical Modelling of Biofiltration Process

Steady state model with zero order kinetics

The classical work of Ottengraf and van den Oever (1983) forms the underlying theoretical basis of all subsequent biofilter models. Ottengraf's macrokinetic model consists of a bed of solid

filter particles, surrounded by a wet, biologically active biolayer (also referred to as biofilm). Figure 10 shows a schematic representation of the macrokinetic model.

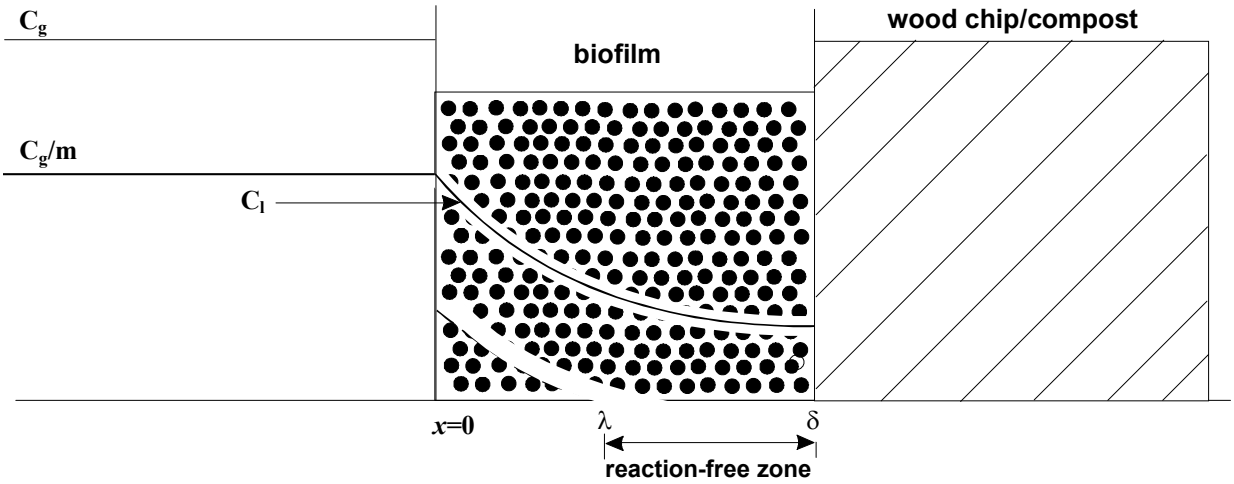

Figure 10 - Schematic representation of Ottengraf's macrokinetic model; x is the distance into the biofilm, δ is the biofilm thickness, λ is the "penetration thickness" of substrate within the biofilm.

For gas phase filter beds, interface resistance in the gas phase is neglected and it is assumed that the biofilm concentration at the interface is in equilibrium with the concentration of the bulk gas. It is also assumed that biofilm nutrients are transported by diffusion, quantified with an effective diffusion coefficient, D_e. Other assumptions made by Ottengraf and van den Oever include a biofilm thickness that is small compared to the diameter of the filter bed particle, zeroth order microkinetics of the substrate biodegradation reaction, and plug flow of the gas phase through the biofilter. The differential equation describing the concentration of the substrate (C_l) in the biofilm (based on reaction limitation) is given by:

$$D_e \frac{d^2 C_l}{dx^2} - k = 0 \qquad [1]$$

where: D_e = effective diffusion coefficient ($m^2 \cdot h^{-1}$)
C_l = liquid phase concentration ($g \cdot m^{-3}$ biofilm)
k = zeroth-order reaction rate constant ($g \cdot m^{-3} \cdot h^{-1}$)

With boundary conditions:

$$x = 0, \quad C_l = \frac{C_g}{m} \qquad [2]$$

$$x = \delta, \quad \frac{dC_l}{dx} = 0 \qquad [3]$$

The solution is given as:

$$\frac{C_l}{C_g/m} = 1 + \frac{1}{2}\frac{\phi^2}{C_g/C_{go}}(\sigma^2 - 2\sigma) \qquad [4]$$

where: $\phi = \delta\sqrt{\frac{km}{D_e C_{go}}} =$ the Thiele number

$\sigma = x/\delta =$ the dimensionless length co-ordinate in the biofilm

$m = (C_g/C_l)_{equilibrium} =$ the distribution coefficient

In the case of diffusion limitation within the biofilm, as shown schematically in Figure 5 by the dashed line, the biofilm is not fully active and substrate only diffuses to a depth of λ. In this case the boundary conditions are:

$$x = 0, \quad C_l = \frac{C_g}{m} \qquad [5]$$

$$x = \lambda, \quad \frac{dC_l}{dx} = 0 \qquad [6]$$

The solution of Equation [1] becomes:

$$\frac{C_l}{C_g/m} = 1 + \frac{1}{2}\frac{\phi^2}{C_g/C_{go}}(\sigma^2 - 2\sigma\frac{\lambda}{\delta}) \qquad [7]$$

Others have developed models, using Ottengraf's model as a starting point, and added complexity by separating the effects of contaminant adsorption and biological degradation, using different kinetics (*i.e.* first-order, Monod, Andrews), and modelling interactions of multiple substrates (Shareefdeen et al. 1993; Hodge and Devinny 1995; Mohseni and Allen 2000).

Unsteady state model with Monod kinetics

Other researchers have described dynamic modelling of the biofiltration process to try to explain the transient response of the biofilter due to a step-change in inlet loading. Deshusses and co-workers (Deshusses et al. 1995a; Deshusses et al. 1995b) used simulation software (SimuSolv) to simulate the biofiltration process for binary pollutant elimination. This model allows for storage of pollutant within a sorption volume after diffusion has occurred through the whole thickness of the biofilm. Nguyen and co-workers (Nguyen et al. 1997) modelled the biofiltration of TEX (toluene, ethylbenzene and o-xylene) compounds. Shareefdeen and Baltzis (Shareefdeen and Baltzis 1994; Baltzis et al. 1997) developed a model that allows for adsorption onto the packing material as well as within water retained on the packing material that was used to predict transient operation of the

biofilter. This model assumed a quasi-steady-state approximation for the biofilm. A further refinement of the model (Zarook et al. 1997; Zarook et al. 1998) includes axial dispersion along the length of the biofilter and a relaxation of the pseudo-steady state assumption for the biofilm. Other researchers have also included adsorption and neglected dispersion (Tang and Hwang 1997), or included dispersion in the gas-phase, but ignored diffusion in the biofilm (Hodge and Devinny 1995). The unsteady-state modelling work of these researchers has focused on long-term transients, i.e. those encountered in adsorption phenomena, start-up operation, and inhibition kinetics. There has been no work to date on the modelling of short-term, high-frequency transients; that is, transients that are on the same time scale as the residence time in the biofilter.

The general equation describing unsteady-state pollutant removal along the length of a packed-bed reactor with axial dispersion is given by:

$$\varepsilon \frac{\partial C}{\partial t} + U_g \frac{\partial C}{\partial h} = D_a \frac{\partial^2 C}{\partial h^2} + A_S D_e \left[\frac{dC_l}{dx} \right]_{x=0} \quad [8]$$

where: ε = porosity of the biofilter packing material

U_g = superficial air velocity of the bulk gas ($m \cdot h^{-1}$)

C = pollutant gas-phase concentration ($g \cdot m^{-3}$ air)

D_a = effective dispersion coefficient ($m^2 \cdot h^{-1}$)

D_e = effective diffusion coefficient ($m^2 \cdot h^{-1}$)

A_S = area across which diffusion occurs (m^2)

C_l = concentration in the biofilm (g α-pinene$\cdot m^{-3}$ biofilm)

The dispersion term was determined to be negligible based on qualitative observations of the concentration profile over the length of the biofilter (in the absence of reaction), and also from inspection of the Peclet number measured from tracer studies. Neglecting dispersion, Equation [8] reduces to:

$$\varepsilon \frac{\partial C}{\partial t} + U_g \frac{\partial C}{\partial h} = A_S D_e \left[\frac{dC_l}{dx} \right]_{x=0} \quad [9]$$

Equation [9] was then solved as an initial value problem, coupled with the following equation, which describes the time-varying pollutant gas-phase concentration within the biofilm:

$$\frac{\partial C_l}{\partial t} + D_e \frac{d^2 C_l}{dx^2} = \frac{\mu_m}{Y} X \left(\frac{C_l}{K_S + C_l} \right) \quad [10]$$

which is also a partial, non-linear differential equation and boundary value problem. Assuming dynamic equilibrium between the gas-phase and the surface of the biofilm, a pseudo-steady state assumption was made for the biofilm, and therefore, $\partial C_l / \partial t$ was set to 0. Equation [10] reduces to:

$$D_e \frac{d^2 C_l}{dx^2} = \frac{\mu_m}{Y} X \left(\frac{C_l}{K_S + C_l} \right) \quad [11]$$

A numerical finite difference scheme was employed to solve Equation [9] for the α-pinene gas phase concentration along the length of the column over time. Equation [11] was solved for the α-pinene

concentration in the biofilm using Newton's iterative technique (Mohseni Tonekaboni 1998). The partial differential terms in Equation [9] can be represented by the method of finite differences, using a backward difference scheme, as:

$$\frac{\partial C}{\partial h} \approx \frac{C(h,t) - C(h - \Delta h, t)}{\Delta h} \qquad [12]$$

$$\frac{\partial C}{\partial t} \approx \frac{C(h,t) - C(h, t - \Delta t)}{\Delta t} \qquad [13]$$

Substituting [12] and [13] into Equation [9], and solving for *C(h,t)* gives:

$$C(h,t) \approx \frac{A_S \left[\frac{dC}{dx}\right]_{x=0} + \frac{U_g}{\Delta h} C(h - \Delta h, t) + \frac{\varepsilon}{\Delta t} C(h, t - \Delta t)}{\frac{U_g}{\Delta h} + \frac{\varepsilon}{\Delta t}} \qquad [14]$$

A computer program was written in digital FORTRAN to solve Equation [14]. The existing code of Mohseni (1998) was used to solve Equation [11].

Summary of Unsteady State Modelling Results

Unsteady state computer simulations

The dynamic model was run on a Pentium II-MMX 233 processor with 64 MB RAM, running under Visual FORTRAN 5.0. The following table gives a list of parameter values used in the simulations. Other input values include: inlet gas-phase α-pinene concentrations over time (*i.e. C(h,t)* at h=0 and t>0), superficial gas velocity (U_g), and biofilter packing height (h=H).

Table 2 - Parameter values for model simulation

Variable	Parameter	Value	Units
C_{g0}	Initial α-pinene gas phase concentration	0.07 - 0.6	$g\ m^{-3}$
δ	Effective biofilm thickness[1]	0.0001	m
A_S	surface area to volume ratio of packing material[2]	600	$m^2\ m^{-3}$
m	α-pinene air/biofilm partition coefficient[3]	0.1	$\frac{g\ m^{-3} air}{g\ m^{-3} biofilm}$
D_e	Effective biofilm diffusion coefficient[4]	0.00000124	$m^2\ h^{-1}$
μ_{max}	maximum growth rate[5]	0.0055	h^{-1}
Y	cell yield coefficient	0.5	$g\ g^{-1}$ α-pinene
X	active cell density	100000	$g\ m^{-3}$ biofilm
K_S	half-saturation constant	1.246	$g\ m^{-3}$ biofilm
ε	porosity of the packing material	0.5	
Δt	differential time element	0.0183333	h
Δh	differential length along the column	0.0034	m
Δx	differential distance into the biofilm	0.0000005	m

[1] Taken from the literature (Shareefdeen et al. 1993)
[2] Estimated from packing material dimensions used in experiments
[3] Measured in the lab (Miller 1999)
[4] Estimated as 57% of diffusivity in water (Fan et al. 1990; Beyenal et al. 1998)
[5] Determined from steady state (constant concentration) biofilter data

Using inlet concentration data from BF1 on day 108 of the experiment, model predictions were calculated. Simulations were run over a time interval of two cycle periods (20 minutes) and results are shown in Figure 11.

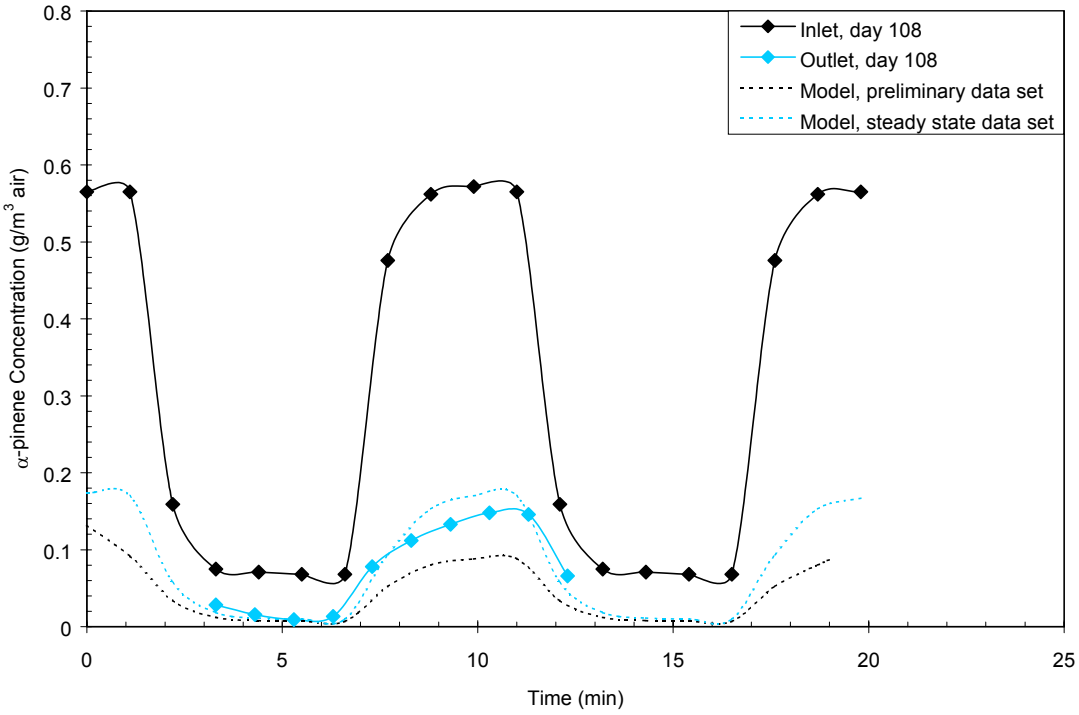

Figure 11 - Comparison of experimental data to simulation results using unsteady state model, based on a preliminary parameter set and kinetic data obtained from steady state biofilter.

Two sets of model results are presented in Figure 11, one based on the preliminary data set and one based on parameter values obtained from Biofilter 3 which was treating a constant concentration stream (referred to as the steady state data set). As can be seen from the graph, both model predictions give similar results when predicting outlet concentrations for a cyclically operated biofilter. The transient model is able to successfully predict the outlet concentration profile from the biofilter very well. The model reproduces the general shape and features of the output and the model predicted values compare reasonably well to actual output values. This is significant because it shows that, within the experimental error, steady state kinetic data can be used to predict unsteady state or transient data.

With the parameter values as given in Table 2, model predictions for cyclic operation and constant concentration loading of a biofilter show similar overall removal rates. This can be seen qualitatively in Figure 12. Figure 12 shows model predictions for a cyclically operated biofilter with inlet α-pinene concentration between 0.06 and 0.6 $g \cdot m^{-3}$ and for a constant biofilter with an inlet concentration of 0.31 $g \cdot m^{-3}$. The averaged removal efficiency for both biofilters at the outlet is essentially the same (i.e. 84.7 % for the cyclic biofilter and 86.7 % for the constant biofilter).

The model predicts that at very high loadings, when the cycle amplitude is very large, the overall removal efficiency for α-pinene would be less for a cyclic biofilter than that compared to a constant biofilter treating the same pollutant loading. This is what one would expect for a biological

system when pollutant removal is independent of concentration. For Monod kinetics, the removal efficiency of a cyclic biofilter and a constant biofilter would be the same as long as the inlet concentrations were within the first order (i.e. concentration dependent) range. As pollutant removal becomes zero order, and therefore, independent of concentration, one would predict the cyclic biofilter to have a lower removal efficiency. Furthermore, the difference in performance would be noticeable if the cyclic biofilter is operating at a true zero order rate (i.e. reaction-limited and not diffusion limited). In the case where the biofilter is diffusion limited the difference in removal may not be significant for a cyclically operated biofilter versus one operating at steady state. In the following section model simulations are used to examine how the kinetic parameters and physical properties of the biofilm affect cyclic and constant concentration.

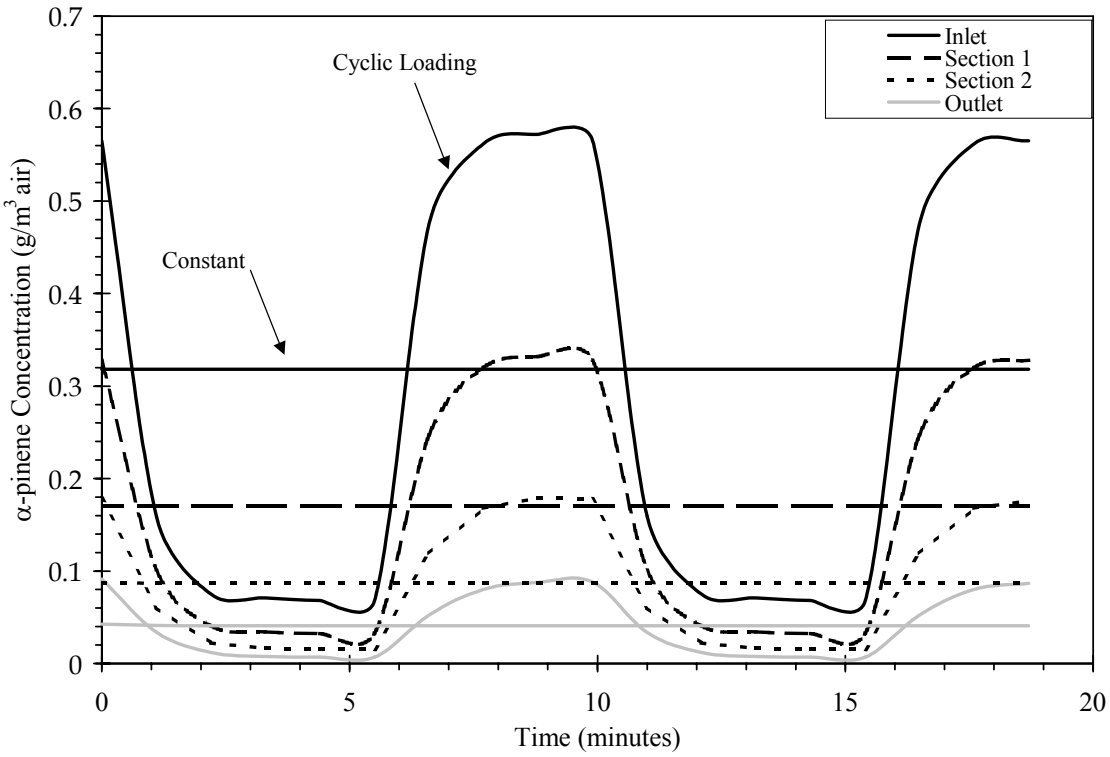

Figure 12 - Model Predictions for cyclic biofilter and constant concentration biofilter at an average α-pinene inlet concentration of 0.31 $g \cdot m^{-3}$.

Model simulation results

A parametric study was conducted using the unsteady state model to examine how biofiltration performance is affected by the kinetic constants and physical parameters of the biofilm for fluctuating concentration inputs. The parameter values in Table 2 are considered reference values and all relative values of model parameters are based on the table values (eg. $\mu_{max,\,rel} = \mu_{max}/\mu_{max,\,ref}$, where $\mu_{max,\,ref} = 0.0055\ h^{-1}$).

The effect of the kinetic parameters is shown in Figure 13. This graph shows how biofilter performance, measured as the average α-pinene removal efficiency, is affected by the maximum specific growth rate of α-pinene, μ_{max}, and the value of the half saturation constant (K_s).

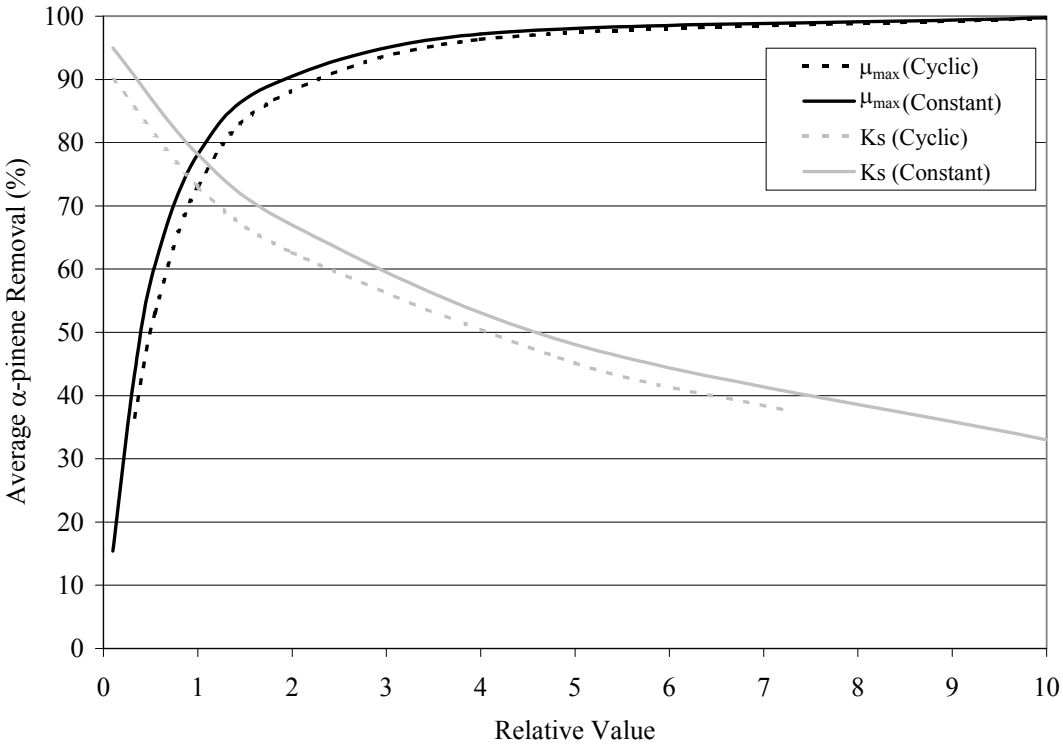

Figure 13 - Effect of kinetic parameters on biofilter performance. Dashed lines represent cyclic biofilter operating at a 10-minute cycle period, solid lines represent constant concentration biofilter.

From Figure 13 it can be seen that the percentage removal increases as the maximum specific growth rate increases. For the range of inlet gas phase concentrations considered, the model predicts a similar effect for both cyclic and constant biofilter operation. The value of the half saturation constant has the opposite affect on biofilter performance. As the value of the half saturation constant increases, the average α-pinene removal decreases. As the half saturation constant increases, the denominator of Equation [11] increases and the reaction within the biofilter becomes first order, concentration dependent. Conversely, as the value of K_s decreases, the term on the right hand side of Equation [11] reduces to zero order kinetics, independent of the gas phase concentration. Although barely distinguishable, the simulations for cyclic biofilter performance begins to deviate more from constant biofilter performance as K_s decreases to very small values.

The effect of the biofilm's physical properties is shown in Figure 14. Simulations show how the removal of α-pinene is affected by biofilm thickness, effective diffusivity, and microorganism density.

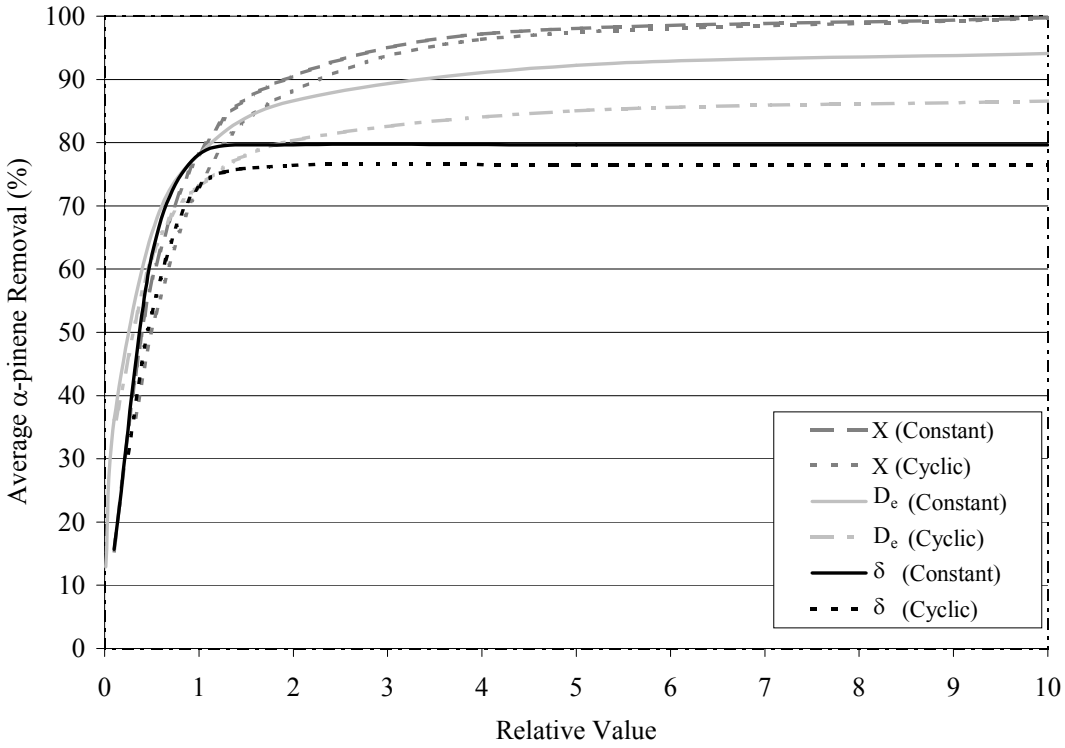

Figure 14 - Effect of the biofilm's physical properties on biofilm performance. Dashed lines represent cyclic biofilter operating at a 10-minute cycle period, solid lines represent constant concentration biofilter.

All three parameters, microorgansim density, effective diffusivity, and biofilm thickness have a positive affect on biofilter performance as their values increase. Microorganism density, X, because of its direct relationship to reaction rate (i.e. like μ_{max}), has the same affect on biofilter performance as μ_{max} does. For both cyclic and constant concentration simulations, as the microorganism density increases, so does the percentage removal of α-pinene. The effective diffusivity, D_e, also positively influences α-pinene removal, however less so for cyclic operation than for constant concentration biofiltration. As the effective diffusivity increases more α-pinene diffuses into the biofilm and is available for reaction. Although constant concentration biofiltration would appear to reach true zero order kinetics (i.e. no effect of concentration on removal rate), cyclic biofilter operation would still operate in the diffusion-limited regime during the low part of the concentration cycle. This would explain why cyclic biofiltration performance is less than that for constant operation at very high effective diffusivities. A doubling of the biofilm thickness, δ, shows a slight improvement in biofilter performance, however, practically, this increase is insignificant. A further increase in biofilm thickness has no effect on improving α-pinene removal. More importantly, biofilm thickness becomes significant to biofilter performance as it becomes increasingly thinner. A thin biofilm affects biofilter performance much in the same way as a low microorganism density would. Conceptually, a thin biofilm may have a smaller number of microorganisms capable of α-pinene degradation. As the thickness increases, there are more organisms present and therefore the removal of α-pinene increases. This would be significant at biofilter start-up, and could explain why performance is seen to slowly increase over this time period.

The existing model does not currently allow for development of the biofilm from thin to thick. This parameter affects cyclic and constant biofilter operation to the same extent.

A sensitivity analysis on the effect of the relative magnitude of the amplitude of cyclic fluctuations was also performed. For these simulations, the average α-pinene loading to the constant and cyclic biofilters, operating at a cycle period of 10 minutes, was the same. The relative value of the amplitude was calculated as $C_{g,max}/C_g$, where $C_{g,max}$ is the maximum inlet concentration to the cyclic biofilters during the cycle period, and C_g is the inlet concentration of the constant concentration biofilter. Figure 15 shows the effect of cycle amplitude on biofilter performance.

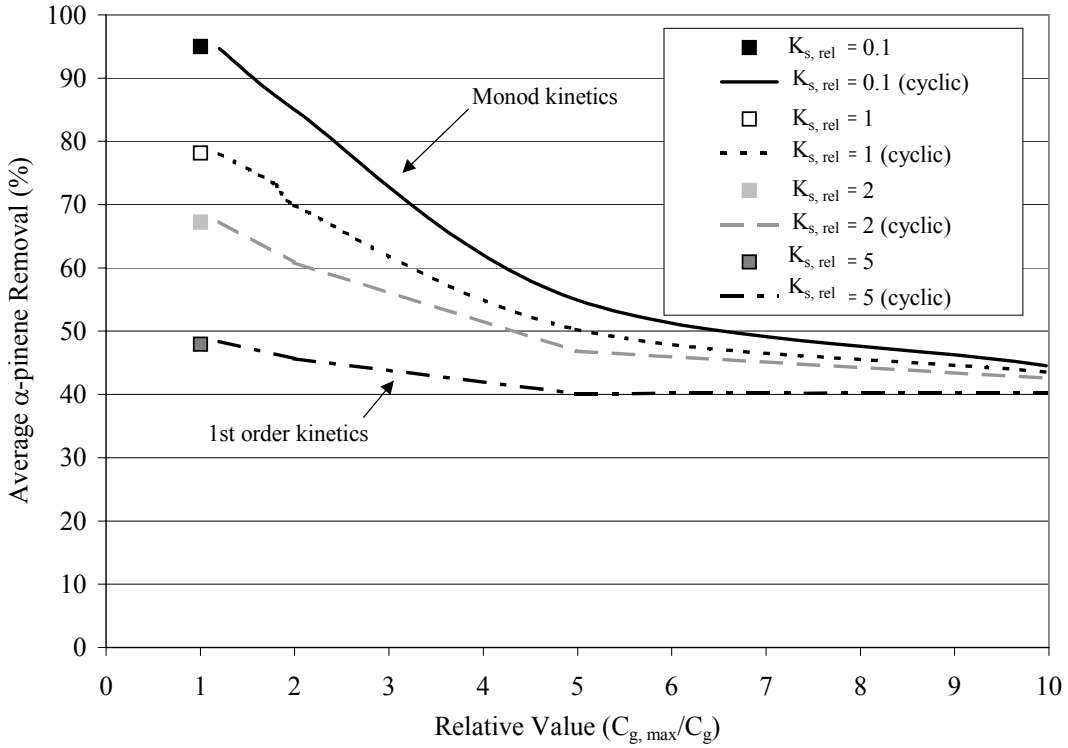

Figure 15 - Effect of cycle amplitude on cyclic biofilter performance for different values of the half saturation constant, K_s. Single data points correspond to constant concentration biofilter operating at same loading rate. The relative value of the amplitude is calculated as $C_{g,max}$ at the inlet of the cyclic biofilter divided by C_g at the inlet of the constant biofilter.

This graph shows how biofilter performance for cyclic biofilter operation decreases as the relative magnitude of the cycle amplitude increases. The four curves show how cyclic biofilter performance is affected by different operating values of the half saturation constant, K_s. For a high value of K_s, represented by the lowest curve on the graph, the removal of α-pinene approaches first order kinetics, and the affect on biofilter performance is not very significant. In this case, as the cycle amplitude increases (i.e. the relative value of $C_{g,max}/C_g$ gets very large), only a slight decrease in biofilter performance is anticipated. For the uppermost curve on the graph (K_s is very small), a doubling of the cycle amplitude would result in a decrease in removal efficiency by ~10%. As the cycle amplitude increases even further, poorer performance is expected for cyclic biofilter operation.

The reason for this effect has to do with the reaction kinetics within the biofilter. If operating under purely first order kinetics, cyclic fluctuations should not affect biofilter performance at all, as higher reaction rates would be observed during the high part of the concentration cycle and lower reaction rates would be observed during the low part of the concentration cycle. The average removal efficiency, however, would be the same as for a biofilter operating at constant concentration. For the case of Monod kinetics, operation is at times zero order (i.e. at the inlet of the biofilter), and can approach first order near the exit of the biofilter. As well, when operating at zero order, diffusion limitation may also be an important factor in the removal process. For very low values of K_s, where the biofilter is operating under zero order reaction kinetics, cyclic biofilter performance will be affected by the extent of diffusion within the biofilm. Figure 16 shows the effect of cycle amplitude on cyclic biofilter performance for different values of the effective biofilm diffusivity, D_e.

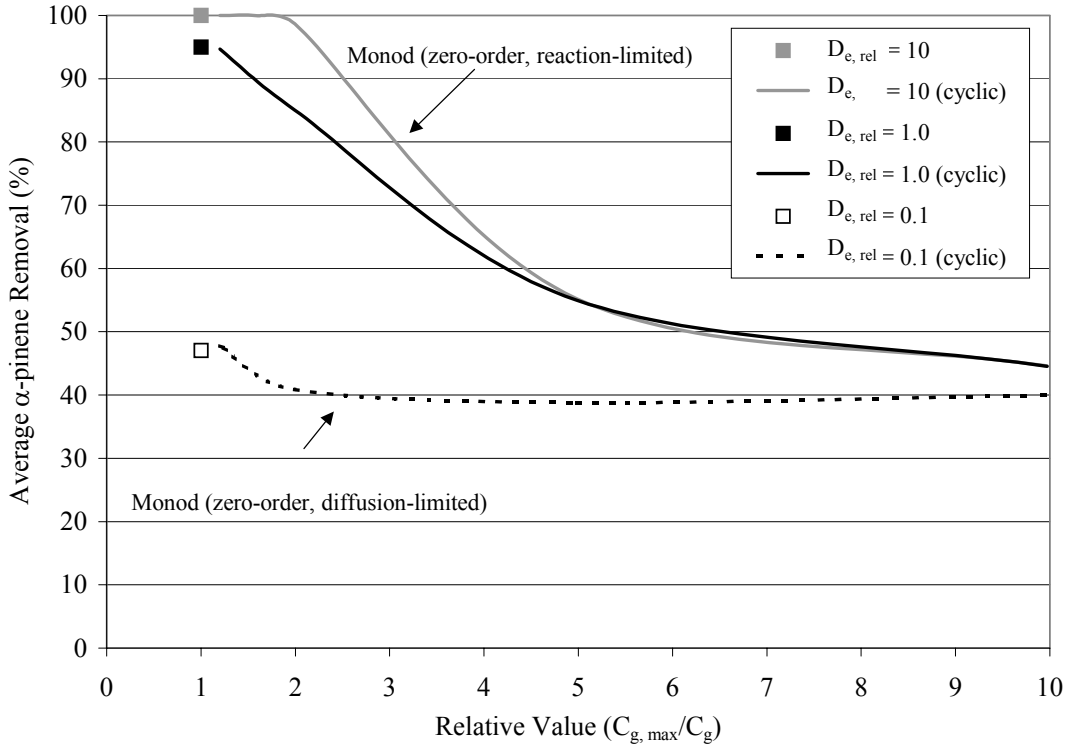

Figure 16 - Effect of cycle amplitude on cyclic biofilter performance for different values of the effective biofilm diffusivity, D_e. The relative value of the half saturation constant, $K_{s,rel}$, is $K_s/K_{s,ref}$ = 0.1. Single data points correspond to constant concentration biofilter operating at same loading rate. The relative value of the amplitude is calculated as $C_{g,max}$ at the inlet of the cyclic biofilter divided by C_g at the inlet of the constant biofilter.

For a biofilter that is operating at 100% removal efficiency (essentially an overdesigned biofilter that has excess removal capacity), an increase in the cycle amplitude has no affect on the overall biofilter's performance as can be seen in the horizontal plateau of the uppermost curve. However, as the cycle amplitude increases further, poorer biofilter performance is expected, as the high concentration of pollutant can no longer be treated along the length of the column. As the effective biofilm diffusivity decreases, α-pinene removal is characterized by zero-order diffusion limited kinetics. In this regime a decrease in biofilter performance occurs with increasing cycle amplitude, however, not as much as in the zero order, reaction-limited domain.

Simulations for different values of the effective biofilm diffusivity show how cyclic biofilter operation can sometimes have better α-pinene removal than for constant concentration operation. Figure 17 shows the effect of effective biofilm diffusivity on biofiltration performance for relatively high values of the half saturation constant, K_s. For large values of K_s, the biofilter tends to follow first order reaction kinetics. For a constant concentration biofilter, removal of α-pinene would be concentration dependent along the entire length of the biofilter column. However, for cyclic operation, as the cycle amplitude increases the inlet of the biofilter removes α-pinene under zero order (DL) kinetics and only shifts to first order kinetics at lower concentrations. In this way it is possible for a cyclically operated biofilter to outperform a constant concentration biofilter. This can be seen from the lower two curves of Figure 4.6 which show that for very low values of the effective biofilm diffusivity, increasing the cyclic amplitude actually increases the percentage of α-pinene removed.

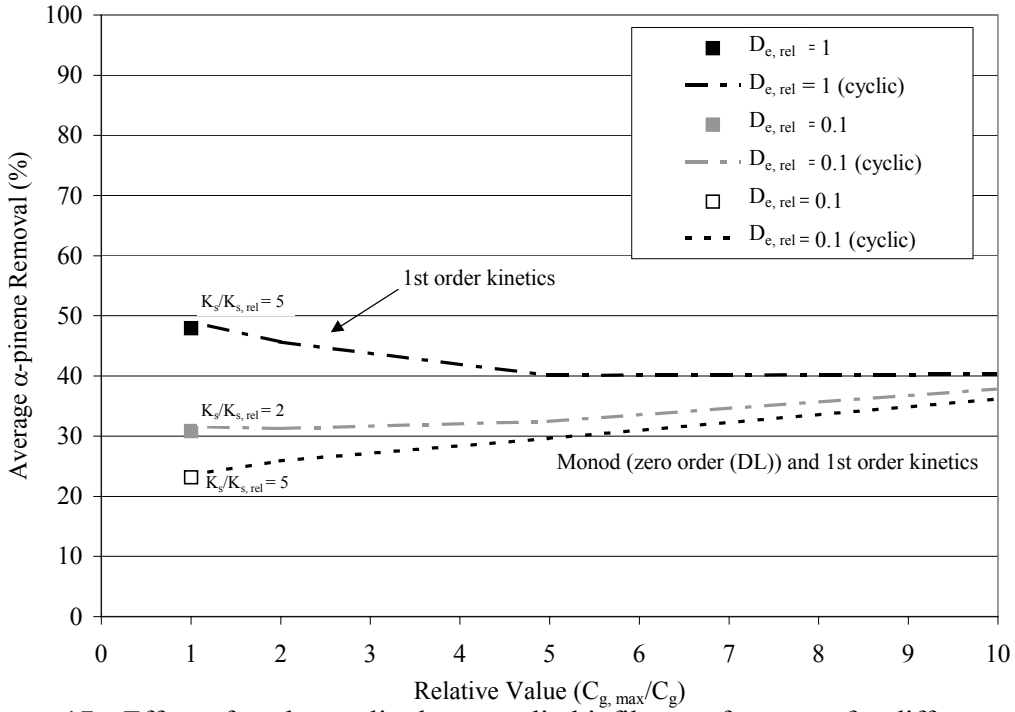

Figure 17 - Effect of cycle amplitude on cyclic biofilter performance for different values of the effective biofilm diffusivity, D_e, and half saturation constant, K_s. Single data points correspond to constant concentration biofilter operating at same loading rate. The relative value of the amplitude is calculated as $C_{g,max}$ at the inlet of the cyclic biofilter divided by C_g at the inlet of the constant biofilter.

From the simulation results it was shown how model parameters have a significant affect on the removal efficiency of α-pinene for both periodic and constant concentration biofiltration. Furthermore, the simulation results also show how different kinetic regimes within the biofilter influence biofilter performance and how the relative magnitude of the cycle amplitude effects biofilter performance. Table 3 provides a summary of how different kinetic regimes influence the removal efficiency of α-pinene for cyclic and constant concentration biofilters.

Table 3 - Comparison of performance of cyclic biofilter operation versus constant concentration biofiltration

Kinetic Parameter	**Cycle Amplitude**	
	$C_{g,max}/C_g = 2$	$C_{g,max}/C_g = 10$
Monod		
Zero order (RL and DL)[†] and first order	similar[‡]	poorer
Zero order (DL) and first order	similar	better
First order		
	similar	similar
Zero order		
RL	poorer	poorer
RL and DL	similar	poorer
DL	similar	similar

[†]RL (reaction-limited), DL (diffusion-limited)

[‡]similar behaviour is described as within 10% difference in removal efficiency

MANAGEMENT APPLICATIONS

This project involves the development of a technology that has the potential to help minimize the adverse effects of both the pulp and paper and wood products industries on the air quality of the environment. Biofiltration has the potential to treat distributed sources of volatile organic compounds and odour in a cost effective, efficient manner, utilizing natural and/or waste materials that are available on site and that can be returned to the environment once consumed.

The thrust of this work is to look at how fluctuating pollutant concentrations in waste gases can be treated in a biofilter. Specifically, biofilter performance in the removal of α-pinene over time is examined during the long-term operation of a biofilter. This work is relevant from an industrial standpoint because it will advance engineering design and operation principles for commercial applications. This work is particularly relevant to this industry since many of the sources amenable to a biofilter (eg. press vents, distributed sources such as tank vents) have non-steady emissions and yet the design is generally based on steady state results.

Experimental results show that short-term transients can be successfully treated in a biofilter. Biofiltration performance for a cyclic (or transient) air emission is comparable to a biofilter treating a constant concentration air emission. Modelling results confirm this view for short term transients. The simulation results further show that the key parameters influencing biofiltration performance for unsteady state operation are the relative magnitude of the concentration fluctuations, $C_{g,\,max}/C_g$, and the relative values of the kinetic constants in the Monod equation, namely, the maximum growth rate, μ_{max}, and the half saturation constant, K_s.

For longer cycle periods (ie. on the order of hours and days) it was shown that as the cycle period increases, biofiltration performance decreases. For a biofilter operating in this type of intermittent mode, a decrease in performance is to be expected each time that the biofilter is 're-started'. For an evening or overnight shutdown (i.e. 12 hours), the biofilter's recovery time is less than one hour. The recovery time for a cycle period of 6 days (i.e. three day shut-down) is between 6-8 hours.

CONCLUSIONS

From the experimental and theoretical results it has been shown that:

- There is no significant difference in the removal efficiency of a cyclically operated biofilter in comparison with a biofilter treating a constant concentration stream at the set of operating conditions in this study (cycle frequency of 10 minutes, cycle amplitude of 10 to 60 ppm α-pinene, and a loading rate of 30 g α-pinene·m^{-3} bed·h^{-1}).

- As a first approximation, kinetic parameters obtained from steady state (constant concentration) biofiltration data can be used in the dynamic model developed in this study to describe short-term transient behaviour of the biofiltration process.

- The model predicts similar removal efficiencies for cyclic and constant biofilter operation when inlet concentrations fall within the zero-order diffusion-limited and first-order kinetics regime.

- For biofilters operating on long cycle periods (i.e. hours and days), removal efficiency decreases after periods of non-loading (or non-use); the longer the period of non-loading, the poorer the biofilter's performance at the re-commencement of the 'On' part of the cycle. The recovery time for a cycle period of 24 hours is less than one hour. The recovery time for a cycle period of 6 days is between 6-8 hours.

- The moisture content of the bed and humidity of the inlet waste gas stream are important parameters in biofilter performance. Even after the humidity was restored to the entering waste gas, the biofilters continued to be dry and extra direct water addition to the bed was required before removal efficiencies returned to their previous levels.

- Nutrient addition is necessary to achieve and maintain high levels of α-pinene removal; a biofilter composed of an 80/20 wood chips/compost mixture has insufficient nutrients to sustain microorganisms capable of α-pinene degradation.

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