Evaluation of gas retention time effects on the bio-trickling filter reactor performance for treating air contaminated with formaldehyde

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The effect of different gas retention times (GRTs) on the efficiency of formaldehyde (FA) removal has been studied using a bio-trickling filter reactor (BTFR) for obtaining the optimal operating conditions. Mathematical models to determine the optimum process conditions of the BTFR system for FA removal from contaminated air are developed. Approximately 66% of the FA introduced into the BTFR treatment process dissolved in the nutrient solution, and about 34% of the residual FA was still present in the air. The predominant bacteria on the surface of supporting materials are identified as the five bacterial colonies Salmonella bongori, Salmonella choleraesuis subsp. arizonae, Salmonella typhimurium, Serratia entomophila and Serratia ficaria, and they have the ability to metabolise FA from two-phases (gas and liquid), as a source of carbon and energy. The optimum removal efficiencies of 450 mg FA L⁻¹ of contaminated air ranging from 95 to 99% are verified for GRTs ranging from 100 to 150 s. Exponential models are proposed as a new approach for determining the optimal operating conditions of the BTFR system and can make significant contributions to improving the air quality.

1 Introduction

Formaldehyde (FA) is commonly used as feedstock by a wide number of factories. Some of the FA used is released into the atmosphere every day. FA has been identified as a suspected carcinogen by the World Health Organization.¹ Because FA can harm DNA, it has been also classified as a mutagen.¹² Due to the low boiling point of FA, it is classified as a volatile organic compound (VOC). FA is a flammable, very reactive and colourless gas. It is soluble in water and can be rapidly absorbed in the respiratory and gastrointestinal systems of humans. The biological half-life of FA in the human body is exceedingly short, possibly less than 60 s.³ The metabolism of FA to formate takes place in all the tissues of the body, and formate is quickly removed by the supporting blood supply. FA can be converted to carbon dioxide and breathed out of the body. The typical standards for FA emissions are below 0.10 ppm according to the World Health Organisation and European E1 Emission Standard, and below 0.05 ppm in the State of California.⁴ The evidence-based FA exposure limit of 0.10 ppm for odour detection and sensory irritation was recommended as an indoor air level for all individuals.⁶

Many studies have been conducted on the application of biological–chemical methods for FA removal from waste gases. On the basis of the previous reports describing risk-based models applied around the world, the use of a bio-trickling filter reactor (BTFR) is a cost-effective and environmentally friendly process for the separation of air pollutants from waste gases.⁷–¹² Biological treatment processes do not yield any emissions of undesirable by-products, as those produced by chemical scrubbing or thermal waste gas treatment processes do.⁷ In the past, BTFRs were commonly used to remove odours by capturing the odour causing compounds in a media bed where they were oxidised by naturally occurring microorganisms. Nowadays, the bio-filter studies focus on the reduction of VOCs and inorganic compounds from a large volume of gases. Traditionally, biofilters are used to treat waste gases with VOC concentrations below 10 mg L⁻¹.¹³ It has been reported that a large number of fungi and bacteria can grow on the supporting materials in a biofilter, in which air contaminated with VOCs flows continuously.¹¹,¹⁴–¹⁶ Some studies have shown that the BTFR can operate effectively over a wide range of pH if the mixed culture of microorganisms is suitable for the reactor’s operating conditions.¹⁷,¹⁸ Generally, if the operation of BTFR system can be managed under favourable
conditions, the production of less biomass might decrease the pressure of gases passing through the biofilter fixed-bed. The kinetics of the microbial growth and biodegradation of methanol and toluene in biofilters have been studied for the removal of these two VOCs simultaneously. The use of a BTFR to remove FA, methanol, dimethylether and carbon monoxide from waste gases can achieve 100% efficiencies. A macro-kinetics model has been developed in a previous study by Strauss et al. to predict the performance of bio-filters.

Although several studies have been conducted on the removal of VOCs from waste gases in the BTFR system, the influence of the gas retention time (GRT) on the efficiency of the FA removal from a synthetic contaminated air stream (SCAS) is not fully understood. The objectives of this study are: (1) to assess the ability of the BTFR treatment processes depending on both the variations of the FA loading rate and GRT, (2) to identify the predominant bacteria colonies distributed in different parts of the biofilter bed, and (3) to establish mathematical models capable of determining the biochemical reaction rate coefficient and predicting the efficiency of FA removal by varying the GRT in the biofilter.

2 Materials and methods

2.1 Pilot description and experimental procedure

This study used a BTFR with dimensions of 8 cm diameter, 66 cm height and 3.3 L volume, as shown in Fig. 1. The fragmented pieces of a polyurethane pipe were used as a supporting material. The SCAS, which can be ventilated using an air pressure generated by the compressor “Asian Star AP-1000”, was supplied from the gas washing bottle (GWB), containing a 37% (v/v) solution of FA dissolved in water, to the biofilter (see Fig. 1). The desired concentration of FA contained in the SCAS can be achieved by providing a fresh supply of FA solution in the GWB through performing repeated additions, learning largely through trial and error. A solution containing all the required nutrients for microbial growth was circulated using a peristaltic pump through the top of the biofilter bed during a 15-minute period every hour. Note that the formulation and preparation of the nutrient solution were as described in a previous study by Fulazzaky et al., and the solution was considered to have a sufficient quantity of minerals. The optimal pH of the nutrient solution to support bacterial growth was almost always near neutral pH of 7. An amount of 3 g L⁻¹ NaHCO₃ was used in the nutrient solution to add buffering capacity, while neutralising the water.

One litre of activated sludge was used as the inoculum for the start-up of the BTFR system, coming from a municipal wastewater treatment plant in Isfahan city, Iran. The fragmented pieces of a polyurethane pipe were used as a matrix to immobilise microorganisms; therefore, the activated sludge was circulated through the top of the biofilter bed over a period of two weeks. Biofiltration experiments were performed at room temperature for about 35 days after the completion of the microbial immobilisation phase, which took about 15 days. All the experiments were performed in triplicate. The installation of four sampling ports at the biofilter bed was important to collect usable samples. A high inlet FA concentration of 450 mg L⁻¹ was used to ensure the presence of FA in two phases, the nutrient solution and SCAS, during the entire experimental run at all the various conditions.

2.2 FA loading rate

The SCAS was fed from the GWB to the biofilter bed via an air temperature controller at a gas flow rate (Q) of 0.025 L s⁻¹. The BTFR system was in operation for four days until the efficiency of the FA removal achieved a steady-state level of less than 4%. The biofilter bed had four sampling ports of different heights (i.e., 5 cm – Part 1, 10 cm – Part 2, 10 cm – Part 3 and 15 cm – Part 4) and one final output (Port-5); therefore, five different volumes of the biofilter bed can be suggested for the purpose of operationally defining the variables, as shown in Fig. 1. Because the BTFR was operated at a Q of 0.025 L s⁻¹, the GRTs can be calculated as 10, 30, 50, 80 and 132 s at Port-1, Port-2, Port-3, Port-4 and Port-5, respectively. The FA loading rate was calculated using the equation: FLR = C_o × Q/V, where FLR is the FA loading rate (in mg L⁻¹ s⁻¹), C_o is the inlet FA concentration (in mg L⁻¹), Q is the gas flow rate (L s⁻¹) and V is the volume of the biofilter bed (L). The FA concentrations at each part of the biofilter bed and outlet of the BTFR were monitored using a hydrocarbon meter (model: HC-300). The sludge age is defined as the average biomass retention time in the BTFR treatment process and can be calculated as the ratio of the biomass concentration in the biofilter bed to the total solid load in the SCAS. Since the SCAS has no solid particles in the BTFR system without gas–solid separation, the sludge age and GRT are the same.

![Fig. 1](image-url) Schematic of the BTFR to remove FA from a SCAS.
2.3 Model development

Some limitations such as the concentration of FA in the SCAS, Q, thickness of the biofilm and operational time can be proposed as variables to develop mathematical equations for the BTFR treatment process. This study focused on the variability of the GRT due to the contact time between air-contaminated FA and biomass on the supporting material, affecting the efficiency of the BTFR treatment process which needs to be verified. The concern of this study was to understand the accuracy of the BTFR design to remove FA from contaminated air. The mass balance equation for the removal of FA from the SCAS by the BTFR process to the level of the system (macroscopic balance), at different parts of the process, is given by the following formula:

\[ \frac{dC}{dt} = -r \]  

(1)

where \( C \) is the FA concentration in the biofilter bed (in mg \( L^{-1} \)), \( t \) is the gas retention time (in s) and \( r \) is the FA removal rate (in mg \( L^{-1} s^{-1} \)).

It is well known that in chemical kinetics the rate of the chemical reaction effectively depends on the FA reactant, hence the value of the exponent is one.\(^{18,21}\) If it is assumed that \( r \) is first order,\(^{18}\) then the rearrangement of eqn (1) gives a continuous equation expressed as:

\[ \frac{dC}{dt} = -kC \]  

(2)

where \( C \) is the FA concentration in the biofilter bed (in mg \( L^{-1} \)), \( t \) is the gas retention time (in s) and \( k \) is the biochemical reaction rate coefficient (in \( s^{-1} \)).

By separating the variables, eqn (2) can be integrated in the form of:

\[ C = C_0 \exp(-kt) \]  

(3)

where \( C \) is the FA concentration in the biofilter bed (in mg \( L^{-1} \)), \( C_0 \) is the inlet FA concentration (in mg \( L^{-1} \)), \( t \) is the gas retention time (in s) and \( k \) is the biochemical reaction rate coefficient (in \( s^{-1} \)).

It is recognized that \( E \) is the concentration of FA which has been already removed from the SCAS to build a dedicated biomass on the supporting material (in mg \( L^{-1} \)) and \( L \) is the maximum concentration of FA removal in the biofilter bed (in mg \( L^{-1} \)) as theoretically schematized in Fig. 2. This gives:

\[ L - E = C \]  

(4)

The combination of eqn (3) and eqn (4) yields the following equation:

\[ L - E = C_0 \exp(-kt) \]  

(5)

Considering the fact that \( C_0 \) equals \( L \) (see Fig. 2), rearrangement of eqn (5) gives

\[ E = L - L \exp(-kt) \text{ or } E = L \{ 1 - \exp(-kt) \} \]  

(6)

where \( E \) is the concentration of FA which has been already removed from the SCAS to build a dedicated biomass on the supporting material (in mg \( L^{-1} \)), \( L \) is the maximum concentration of FA removal in the biofilter bed (in mg \( L^{-1} \)), \( t \) is the gas retention time (in s) and \( k \) is the biochemical reaction rate coefficient (in \( s^{-1} \)).
2.4 Analytical methods

Measurements of the nutrient solution pH were made using a portable pH-meter (Crison model 507). The levels of FA in the nutrient solution were estimated from the measurements of the chemical oxygen demand (COD) using a microwave digestion and spectrophotometric method. The measurements of FA in the SCAS at each port of the biofilter bed and the outlet of the BTFR system were made using a hydrocarbon meter (model: HC-300). The presence of microorganisms at the different parts of the biofilter bed was identified according to a conventional biochemical method. The biomass concentrations were measured in terms of mg L\(^{-2}\) of dried solids from each part of the biofilter bed, based on the selection of 10 pieces of the supporting material, dried at 104 °C for 24 h.

3 Results and discussion

3.1 Ability of the BTFR system to remove FA

In this work, the ratio of food to microorganisms can be calculated from each part of the biofilter bed using the algebraic equation approach of: 

\[ \frac{F}{M} = \frac{C_0^d}{X \times V} \]

where \( \frac{F}{M} \) is the ratio of food to microorganisms (in L\(^{-1}\) s\(^{-1}\)), \( C_0^d \) is the inlet FA concentration (in mg L\(^{-1}\)), \( X \) is the mean biomass present in each part of the biofilter bed in terms of dried solids (in mg) and \( V \) is the volume of the biofilter bed (in L). The ability of the BTFR treatment process to remove FA from the SCAS, as depicted in Table 1, is dependent on both the biomass and \( \frac{F}{M} \) ratio.

After 90 days of acclimatisation, feeding the BTFR system with FA dissolved in the nutrient solution, the biofilter became acclimatised to FA and experienced its effects more mildly, likely due to the high toxicity of FA that precludes its systemic use. Then, biofiltration experiments using air polluted with FA under steady-state conditions were carried out for 35 days at a Q of 0.025 L s\(^{-1}\) in a device with the BTFR design shown in the schematic of Fig. 1.

It was possible to achieve removal efficiencies of 51, 66, 76, 87 and 99%, observed at the Ports 1, 2, 3, 4 and 5, respectively, after 10 days of running the experiments where the BTFR system was fed at FLRs of 45, 15, 9, 5.6 and 3.4 mg L\(^{-1}\) s\(^{-1}\), respectively (see Fig. 3). Evidence shows that the efficient operation of the BTFR system to remove FA from a SCAS strongly depends on the FLR.

A power-law model (see Fig. 3) can be established by plotting the variations of the FLR versus the percentage of FA removal by the BTFR process, such that: 

\[ RE = 135.1(FLR)^{-0.25} \]

where \( RE \) is the FA removal efficiency (in %) and FLR is the FA loading rate (in mg L\(^{-1}\) s\(^{-1}\)). The model shows that RE decreases with an increase of FLR. The prediction of RE to assess the performance of the BTFR system can be expressed on the basis of the variability of FLR. The expression shows a very good agreement with the experiments in describing the ability of the bio-trickling filter to remove FA from contaminated air (\( R^2 = 0.998 \); see Fig. 3), rather than monitoring the variable effect of the changes in the FA flow rate to predict the performance of the BTFR system.

A previous study has reported that the effect of \( t \) on the performance of a biofilter showed a linear increase of RE over time. A shift from zero- to first-order FA removal rate kinetics was observed when the biofilter was typically used to treat air contaminated with high concentrations of FA. The apparent first-order kinetic behaviour can be observed in the removal of FA by the BTFR treatment process. Fig. 4 shows that the

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Table 1 Efficiencies of the BTFR system monitored at the different ports

<table>
<thead>
<tr>
<th>Port</th>
<th>( X^d ) mg L(^{-1})</th>
<th>( \frac{F}{M}^b )</th>
<th>( C_0^d ) mg L(^{-1})</th>
<th>( C_e^d ) mg L(^{-1})</th>
<th>RE(^c ) (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Port-1</td>
<td>52.7</td>
<td>0.213</td>
<td>450</td>
<td>220.5</td>
<td>51</td>
</tr>
<tr>
<td>Port-2</td>
<td>157.5</td>
<td>0.071</td>
<td>450</td>
<td>153.0</td>
<td>66</td>
</tr>
<tr>
<td>Port-3</td>
<td>262.5</td>
<td>0.042</td>
<td>450</td>
<td>108.0</td>
<td>76</td>
</tr>
<tr>
<td>Port-4</td>
<td>476.0</td>
<td>0.024</td>
<td>450</td>
<td>58.5</td>
<td>87</td>
</tr>
<tr>
<td>Port-5</td>
<td>693.0</td>
<td>0.016</td>
<td>450</td>
<td>4.5</td>
<td>99</td>
</tr>
</tbody>
</table>

\( ^a \) \( X \) is the average weight of biomass in terms of dried solids in the biofilter bed. \( ^b \) \( \frac{F}{M} \) is the ratio of food to microorganisms. \( ^c \) \( C_0 \) is the inlet FA concentration. \( ^d \) \( C_e \) is the outlet FA concentration. \( ^e \) RE is the FA removal efficiency.
efficiency of the BTFR to remove FA depends on $t$. In general, the BTFR treatment process emits less and less FA monitored at its effluent with increasing $t$. The effect of a long $t$ in the settler on the FA removal from the SCAS is necessary to ensure the presence of microbial growth; however, it requires technical support from a large-scale reactor at the same time, meaning that the system is not economical. The nonlinear regression model can be proven by plotting the RE versus $t$, and gives a line of best fit with a strong correlation value of $R^2 = 0.998$ (see Fig. 4). Symbolically, this process can be expressed by the following power equation: $RE = 27.8(t)^{0.258}$, where RE is the FA removal efficiency (in %) and $t$ is the gas retention time (in s).

As can be observed during the experiment, the pressure drop of the BTFR system was less than 1 psi for each metre of increasing biofilter bed height. The aerobic bacteria in the biofilter bed preferred a neutral pH of 7 and most of them were not able to grow quickly on the supporting materials in the presence of FA. The thickness of the biofilm is an important parameter, because it is related to the rate of growth of the biofilm and the extent to which the biofilm interferes with the operational BTFR system. Though the thickness of biofilm was not uniform, the supporting materials in the biofilter bed can have a very high porosity at the beginning of the experiment. Several variables such as the moisture content, pH, nutrient solution and solubility affect the performance of a biofilter. Because the operating conditions of the BTFR can keep the moisture content of the biofilter fairly constant by periodically circulating the nutrient solution through the top of the biofilter bed using a peristaltic pump, the moisture content in the SCAS was not a limiting factor during normal operation of the BTFR (i.e., at pH 7 and room temperature). The balanced FA solubility seems to be a key factor for obtaining a good performance of the BTFR with effective microbial activity.25 This study has found that the BTFR process is one of the best techniques for removing FA from contaminated air, due to the fact that FA dissolves easily in water since they are both polar compounds.

3.2 Identification of bacterial colonies

The microorganisms identified in this study were gathered from biomass samples from different parts of the biofilter bed. To identify the bacterial colonies, the samples were cultured in a nutrient agar medium at 35 °C for 24 h. Even though many microorganisms can be grown in the culture medium, the predominant bacteria on the surface of the supporting material were from five colonies, which were identified by conventional biochemical methods as *Salmonella bongori*, more dominant in Part 1 of the biofilter bed, *Salmonella choleraesuis* subspecies *arizonae* in Part 2, as well as *Salmonella typhimurium*, *Serratia entomophila* and *Serratia ficaria* in Parts 3 and 4. The results of the microbiological tests are depicted in Table 2. It is suggested that each microorganism is adapted to its particular environmental niche in order to grow optimally. Knowing how to best grow these predominant bacteria using FA as the carbon source could increase the performance of the BTFR system. The biomass concentration of 210 mg of dried solids L$^{-1}$ was verified for the experiment with an effluent $t$ of 132 s.

### Table 2 Results of the biochemical tests for the identification of bacteria colonies

<table>
<thead>
<tr>
<th>Morphological and biochemical tests</th>
<th>Name of bacterial colonies</th>
</tr>
</thead>
<tbody>
<tr>
<td>Shape</td>
<td>Sb$^a$ Sc$^b$ St$^c$ Se$^d$ Sf$^e$</td>
</tr>
<tr>
<td>Lactose test</td>
<td>Rod Rod Rod Rod Rod</td>
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<tr>
<td>Indole test</td>
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<tr>
<td>Urea hydrolysis test</td>
<td>+ + + + +</td>
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<tr>
<td>Motility test</td>
<td>+ + + + +</td>
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<tr>
<td>H$_2$S test</td>
<td>+ + + - -</td>
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<tr>
<td>KCN growth test</td>
<td>- - - + +</td>
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<tr>
<td>Oxidase test</td>
<td>- - - - -</td>
</tr>
<tr>
<td>Gram test</td>
<td>- - - - -</td>
</tr>
<tr>
<td>Catalase test</td>
<td>+ + + + +</td>
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<tr>
<td>Lysine decarboxylase test</td>
<td>+ + + + +</td>
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<tr>
<td>Methyl red test</td>
<td>+ + - - -</td>
</tr>
<tr>
<td>Voges–Proskauer test</td>
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<tr>
<td>Citrate test</td>
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<td>Methyl red test</td>
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<td>Lactose test</td>
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$^a$ Sb is *Salmonella bongori*. $^b$ Sc is *Salmonella choleraesuis* subspp. *arizonae*. $^c$ St is *Salmonella typhimurium*. $^d$ Se is *Serratia entomophila*. $^e$ Sf is *Serratia ficaria*.

3.3 Profile of FA removal by the BTFR treatment process

Apparently, the use of the BTFR treatment process as seen in Fig. 5 can remove approximately 70% of FA at Part 1 (at 5 cm height) of the biofilter bed, because FA in the SCAS has already been partially dissolved in the nutrient solution at its highest concentration. With a vast network of tiny holes, the supporting material resembles a honeycomb and has an enormous surface area, covered by a thin biofilm on which certain gas molecules can adsorb.27 In this study, the supporting material was made from plastic with a polycarbonate chop thread, which offers the elasticity of rubber combined with the toughness and durability of metals to ensure no adsorption of FA on its surface. At a constant feed of
Q at 0.025 L s\(^{-1}\), increasing the height of the biofilter bed from 0 to 25 cm resulted in a linear increase in RE. From the extrapolation of that straight line, it can be found that approximately 66% of the FA introduced into the BTFR system could dissolve in the nutrient solution. Experimental data verification (Fig. 6) shows that the levels of COD in the nutrient solution can increase linearly with increasing time. The rate of dissolved FA in the nutrient solution can be defined as the gradient of the straight line and should be equal to 1.056 mg of COD L\(^{-1}\) min\(^{-1}\) (Fig. 6; see Fig. 6 caption). Evidence suggested that the five predominant colonies were able to metabolise FA from both the nutrient solution and SCAS as their only source of carbon and energy.

3.4 Results of the model test

The use of eqn (7) to plot a straight line graph provides a very good fit to the data sets of \(E\) and \(t\) (\(R^2 = 0.9184\); see Fig. 7 and Fig. 7 caption) and gives new insight into the graphical prediction of the BTFR performance. The straight line relates to the equation: \(Y = a(X) + b\), where \(a\) is defined as the slope, \(b\) is the intercept of the curve \((t/E)^{1/3}\) versus \(t\), \(Y = (t/E)^{1/3}\), and \(X\) is \(t\).

With \(C_0 = 450\) mg L\(^{-1}\), values of \(a\) and \(b\) as high as 0.0024 L mg\(^{-1}\) and 0.3743 L s mg\(^{-1}\) were verified from the linear line (see Fig. 7), and then using eqn (8) permitted us to calculate \(k\), which was equal to 0.032 s\(^{-1}\). Using eqn (10) permits us to predict \(t\) at any RE targeted for the BTFR treatment process. The results (Fig. 8) show that the use of the BTFR treatment process, operated at a \(Q\) of 0.025 L s\(^{-1}\), can achieve more than 99% efficiency, while the optimal \(t\) of the biofilter process should be more than 150 s. Overall, the results of this study appear to be generally similar to a previous study,\(^{18}\) where an optimal \(t\) of 141 s for 100% efficiency of the FA removal was verified from BTFR experiments operated at a \(Q\) of 0.025 L s\(^{-1}\).

Evaluating the model performance, a fictitious curve (see Fig. 8) can be made based on the results calculated using eqn (10) for the BTFR system operated at a \(Q\) of 0.025 L s\(^{-1}\) and \(C_0\) of 450 mg L\(^{-1}\). The figure shows that the level of FA in the treated SCAS can be controlled to less than 22.5 mg L\(^{-1}\) at \(t\) of 100 s, representing a FA removal greater than 95%. The efficiency of the BTFR treatment process can increase progressively with the increase of \(t\); however, the optimal \(t\) ranges from 100 to 150 s. The general accepted value is an exhaust FA of 445.5 mg L\(^{-1}\) from a SCAS with \(t\) of 150 s, which is said to result in greater than 99% efficiency. The consideration of optimal \(t\) is a very important factor for the BTFR treatment process to achieve 95% efficiency.\(^{26}\) To achieve a range of efficiencies from 95 to 100%, an increase in \(t\) to more than 100 s is required when the use of the BTFR system cannot effectively remove FA from the contaminated air.

Some limitations of using the BTFR treatment process to remove FA from contaminated air include internal and external validity of the bio-trickling filter system, such that: (1) the BTFR system cannot be used on extremely hot air since
the efficiency of the FA removal can vary depending on the sensitivity of the microorganisms to temperature, (2) the collected waste nutrient solution may not be free of FA residues, hence there is a need for additional treatment before its authorised release into the environment, (3) the flexibility of the BTFR system is not generally applicable to all the VOCs present in contaminated air because the predominate bacterial colonies need a lag phase to allow acclimatisation to each possible compound.

4 Conclusions

This study used a BTFR to remove FA from contaminated air. Mathematical models were developed to determine the efficiencies of different parts of the biofilter bed to remove FA based on the variations of $t$. The verification of the RE of the BTFR treatment process, depending on the variation of either FLR or $t$, was formulated as a nonlinear equation. A linear model was used to calculate the value of $k$. The use of the model performance of the BTFR system on a fictitious curve was able to predict the efficiency of the FA removal for contaminated air using a variable $t$. A new method to determine the optimal operating conditions of the BTFR system related to the variations of $t$ is proposed to contribute to the air quality improvement analysis and advanced bio-trickling filter technologies.

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References