



Biofilter in leachate treatment processes

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ABSTRACT

Landfill leachate is produced when rainwater infiltrates into the landfill and permeates through the decomposing waste within the landfill leaching out with it contaminants and pollutants. Untreated leachates can permeate ground water or mix with surface waters and contribute to the pollution of soil, ground water and surface water. In this study, biofilter was evaluated in treating synthetic landfill leachate. The total organic carbon (TOC) removal efficiency (of landfill leachate) was evaluated by sequential adsorption/biosorption (on granular activated carbon [GAC]). Experiments were conducted at high velocity (2 m/h) and at low velocity (0.2 m/h) to simulate *ex-situ* and *in situ* biofilter treatment respectively. GAC biofiltration led to a consistent TOC removal even after a long period of operation without the need to regenerate the activated carbon. Even after 15 days of continuous running, the effluent from the GAC biofilter was approximately 60% of the influent quality (i.e. 40% removal). A mathematical model was developed to simulate the organic removal efficiency of the GAC biofiltration system. In this simulation model, the performance can be described in two stages: adsorption during the initial stage and biodegradation in the latter stage. The biofiltration process was modelled and compared with the experimental data.

Keywords: Landfill leachate; Biofilters; Adsorption; Modelling

1. Introduction

Australia is among the highest producers of waste in the world [1]. It generates waste at a rate of 2.25 kg per person per day, the majority of which ends up in landfill [2]. During 2002–2003 over 17 million tonnes of waste was disposed in landfills in Australia. There are approximately 650 licensed landfills in Australia.

Rain water percolating through a landfill leaches with it the decomposing organic matter, inorganic ions and heavy metals. This contaminant-laden concentrated effluent from the landfill is called landfill

leachate. Landfill leachate can be regarded as a high strength wastewater with acute and chronic toxicity. Organic matter (biodegradable and refractory organics), ammonia and heavy metals are the three principal contaminants of the leachate. Its composition will vary from site to site, depending on many factors including the nature of the waste in the landfill, the filling method, the level of compaction, the engineering design of the landfill, the rainfall of the region and the stage of decomposition of the waste [3,4]. Untreated leachate can permeate ground water or mix with surface waters and can contribute to the pollution of soil, ground water and surface water. Depending upon the ratio of biodegradable and

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refractory organic compounds, the landfills can be classified as young, mature (intermediate) and old (stabilised) landfills.

The successive phases of biodegradation cause reductions in chemical oxygen demand (COD) and biological oxygen demand (BOD) rendering the leachate with non-biodegradable or refractory compounds. Generally, a high COD (3,000–60,000 mg/L), high BOD5/COD ratio (>0.3) and a high fraction of low-molecular organics characterise leachate produced from young landfills (<1–2 years old) (Table 1). By contrast, moderate COD (100–500 mg/L), low BOD5/COD ratio (<0.3), and a high fraction of high-molecular weight organics characterise mature leachate coming from old landfills [5].

A range of biological and chemical treatment processes for landfill leachate have been studied. Biological processes are more effective for young leachate with a high BOD5/COD ratio [6]. Biological processes are less effective in treating leachate from intermediate or stabilised landfills with a low BOD5/COD ratio, or with high concentrations of toxic constituents. To treat these, physicochemical processes appear viable and include evaporation, sedimentation, flocculation/precipitation [7,8], ion exchange, activated carbon adsorption [9], chemical oxidation [10–12], membrane filtration including reverse osmosis (RO) [13], and nano filtration (NF) [14]. They are applied as either pre/post-treatments or complete treatments.

An emerging technology is biofilters which was found to effectively remove organic matters from water and wastewater [15]. In biofilters, microbial communities establish and grow on the filter media such as granular activated carbon (GAC). Organic substances are first adsorbed onto filter media and then biodegraded by the microbial communities. Results of long-term performance of the GAC biofilter with synthetic wastewater which has the similar characteristics as biologically treated sewage effluent showed that the GAC biofilter could maintain high organic removal efficiency for a long filtration time without any regeneration of activated carbon. Even after 42 days of continuous run, a bio-filter of very short depth of 15 cm GAC bed depth maintained a consistent organic removal efficiency of 40–50% with synthetic wastewater [16]. GAC biofilter also removed 60–98% of total coliforms from synthetic wastewater and river water. No fecal coliforms were detected in the effluent from GAC biofilter for river water. GAC bio-filter can be used as an economical treatment system in removing organic matters and pathogens from biologically treated wastewater and surface water. The merits of GAC biofilter are the consistent total organic carbon (TOC) removal efficiency, long life cycle and simplicity in operation.

In this study, the performance of biofiltration using GAC media was evaluated. Synthetic Landfill leachate was used as wastewater. A mathematical model was developed to simulate the long-term organic removal

Table 1
Composition of synthetic landfill leachate (SLL)

Component	Per liter	Composition of trace metal solution (TMS)	
Acetic acid	7 mL	FeSO ₄	2,000 mg
Propionic acid	5 mL	H ₃ BO ₄	50 mg
Butyric acid	1 mL	ZnSO ₄ ·7H ₂ O	50 mg
K ₂ HPO ₄	30 mg	CuSO ₄ ·5H ₂ O	40 mg
KHCO ₃	312 mg	MnSO ₄ ·7H ₂ O	500 mg
K ₂ CO ₃	324 mg	(NH ₄) ₆ Mo ₇ O ₂₄ ·4H ₂ O	50 mg
NaCl	1,440 mg	Al ₂ (SO ₄) ₃ ·16H ₂ O	30 mg
NaNO ₃	50 mg	CoSO ₄ ·7H ₂ O	150 mg
NaHCO ₃	3,012 mg	NiSO ₄ ·6H ₂ O	500 mg
CaCl ₂	2,882 mg	96% H ₂ SO ₄	1 mL
MgCl ₂ ·6H ₂ O	3,114 mg		
MgSO ₄	156 mg		
NH ₄ HNO ₃	2,439 mg		
Urea CO(NH ₂) ₂	659 mg		
Na ₂ S·9H ₂ O	Titrate to Eh: 120–180 mv		
NaOH	Titrate to pH = 5.8–6.0		
Trace metal solution	1 mL		
Distilled water	To make 1 L		

of the GAC biofiltration system. The performance of the mathematical model was assessed with the experimental data.

2. Experimental methodology

The study was conducted with synthetic landfill leachate (SLL). The composition of the SLL is given in Table 1 [17].

Long-term bioadsorption (biofiltration) experiments were conducted using a GAC biofilter column with SLL influent. In this study, GAC manufactured by Calgon Carbon Corporation, USA, was used as media in the biofilter and its properties are shown in Table 2. The column had ports for influent feeding, effluent collection and backwashing. The column was packed with GAC to a bed depth of 30 and 35 cm (Fig. 1, Table 3). The GAC bed was acclimatised at a constant filtration rate of 2 and 0.2 m/h. The filter was backwashed for approximately 5 min every 24 h of filtration run to eliminate excess biomass buildup which may lead to filter clogging. The backwash rate was controlled by allowing up to 30% bed expansion.

3. Results

3.1. Equilibrium and adsorption

3.1.1. Adsorption isotherm

A satisfactory description of the equilibrium state between two phases (solid and liquid) is important for a successful representation of the dynamic behaviour of the adsorption system. The adsorption equilibrium of solute (adsorbate) from the bulk solution onto the surface of an adsorbent (solid media) is quantified by adsorption isotherms.

Table 2

Characteristics of granular activated carbon (GAC) used in this study (Calgon Carbon Corp., USA)

Specification	GAC
Surface area (m ² /g)	1,001.2
Mean pore diameter (Å)	22.55
Micropore volume (cm ³ /g)	0.269
Mean diameter (µm)	750
Bulk density (kg/m ³)	600
Product code	F-400

Langmuir isotherm is valid for monolayer adsorption onto a surface with a finite number of identical sites and is given by the equation:

$$q = \frac{q_m b C}{1 + b C}$$

where, q is the amount of solute adsorbed per gram of adsorbent (mg/g), C is the equilibrium concentration of solute in the bulk of the solution (mg/L), q_m is saturation amount of organic adsorbed (mg/g) and b is a constant (L/mg).

Sips isotherm is the modified form of Langmuir isotherm and is given by the equation:

$$q = \frac{q_m b C^{1/n}}{1 + b C^{1/n}}$$

where, q is the amount of solute adsorbed per gram of adsorbent (mg/g), C is the equilibrium concentration of solute in the bulk of the solution (mg/L), q_m is saturation amount of organic adsorbed (mg/g) and n and b are constants (L/mg).

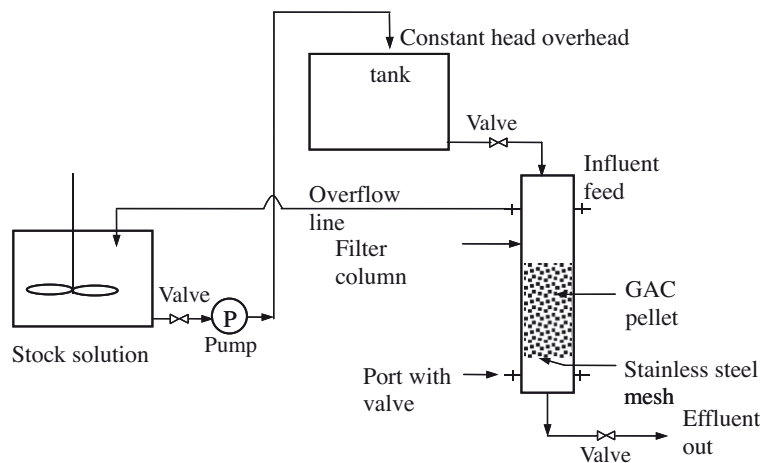


Fig. 1. Schematic diagram of the biofiltration system.

Table 3
Summary of column experiments

Parameter	Experimental 1	Experimental 2
Filter media	GAC	GAC
Bed depth (cm)	30	35
Filtration rate (m/hr)	2	0.21
Average TOC of influent (mg/L)	65	50
Backwashing expansion	30%	30%
Backwashing duration and frequency	5 min every 24 h	5 min every 24 h

The Freundlich isotherm describes heterogeneous surface adsorption. The energy distribution for adsorptive sites (in Freundlich isotherm) follows an exponential-type function which is close to the real situation. The rate of adsorption/desorption varies with the strength of the energy at the adsorptive sites. The Freundlich isotherm is expressed by Eq. (1):

$$q = k_F C^{\frac{1}{n}} \quad (1)$$

where, the constants k_F and n relate to sorption capacity of the adsorbent and the deviation from linear sorption.

The isotherm parameters were determined using the Langmuir, Sips and Freundlich isotherm model equations. These equations are shown in Table 4. The Langmuir and Sips isotherms were found to fit well with the experimental results (Fig. 2). The isotherm parameters are given in Table 4.

3.2. GAC biofiltration

In the biofilter column experiments, there is a short initial period of a few hours where organic removal occurs predominantly by the adsorption processes. After a few days of operation when biological activity is established, organic removal occurs through a biodegradation processes. Fig. 3 results show that the GAC biofilters led to a consistent TOC removal even after a long period of operation without the need to regenerate the activated carbon. Even after 50 days of continuous running, the removal efficiency by the GAC biofilter was 45–60%. This is due to the steady-state biological oxidation of organic matter in the biofilter. Initially, organic removal is a result of GAC adsorption. After this, the growth of microorganisms on the GAC establishes and the organic removal is principally the result of biomass activity. As can be seen the major advantage of biofiltration is that it can function without any need for regeneration of GAC. If

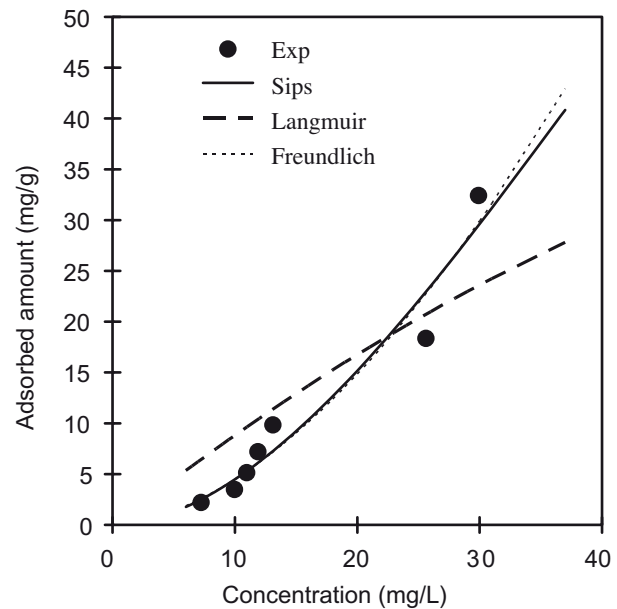


Fig. 2. GAC adsorption isotherm curves (SLL; contact time = 3 days; stirring speed = 130 rpm; initial DOC = 65 mg/L).

adsorption was the major contaminant removal process, it would have been exhausted after a few hours and the GAC would need to be replaced or regenerated.

3.3. Microbiological analysis

A quantitative microbiological analysis was carried out on GAC particle samples on a periodic basis. In order to estimate the number of viable bacteria in these samples, viable counts were carried out by spread-plate technique using a medium of nutrient agar. The composition of SLL used in these experiments was very rich in nutritional sources such as carbon and nitrogen and therefore a uniform increase in the number of viable bacteria was

Table 4
Summary of values of isotherm parameters

Isotherm	Equation	Parameters	Values
Langmuir	$q = \frac{q_m b C}{1 + b C}$	q_m	188.2
		b	0.0048E-03
Freundlich	$q = k C^{\frac{1}{n}}$	k	0.084
		n	1.726
Sips	$q = \frac{q_m b C^{1/n}}{1 + b C^{1/n}}$	q_m	212.2
		b	3.14×10^{-4}
		n	1.837

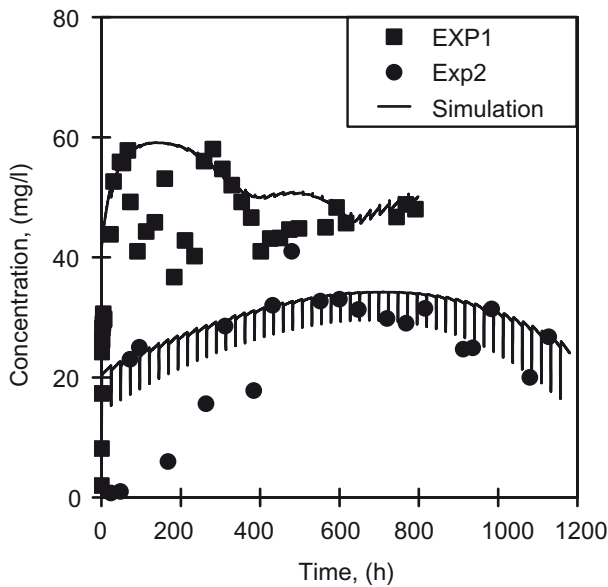


Fig. 3. Performance of GAC-packed biofilters with time for 60 days in SLL. Also shown is the fit between experimental data and model simulation in the long term for variation in bed depth. (Experimental conditions are summarised in Table 3, model parameters are summarised in Table 4.)

observed during the initial phase. In both GAC samples (concentration of 3 and 5%), the rate of growth appeared similar with few differences during the experiment. The viable numbers increased rapidly and reached stationary phase in a period of two weeks.

4. Mathematical model

A mathematical model was developed to simulate the organic removal efficiency of the GAC biofiltration system. In this simulation model, the performance can be described in two stages: adsorption during the initial stage and biodegradation in the latter stage. In practice, the prediction of the performance of a biofilter during the biological phase or steady phase is more important because the initial stage lasts only for a short duration of less than a day at the beginning of a run. Therefore, the simulation of the adsorption processed was simplified.

A systematic representation of the mass balance in a biologically activated carbon system and the biofilm on the activated carbon is shown in Fig. 4. The model is based on the fundamental mechanisms of transport of substrate in the bulk liquid, biofilm growth, transport and biodegradation within the biofilm and adsorption on activated carbon. The following

additional assumptions are made in relation to the modelling work.

- (1) The adsorbent particles are assumed to be spherical in shape and uniform in size and curvature effect of the adsorbent surface can be ignored. No biological reaction occurs inside the adsorbent particle.
- (2) The biofilm is thin relative to the radius of the adsorbent particle and can be modelled as a flat plate. The biofilm is homogeneous with respect to thickness, porosity, composition and density. The specific surface area and bed porosity are constant with biofilm growth. Any increase in biofilm thickness is due to the growth of biofilm.
- (3) The biological activity is assumed to be substrate limiting and can be represented by the Monod equation.
- (4) The Glueckauf approximation may be used to describe intrapellet diffusion.

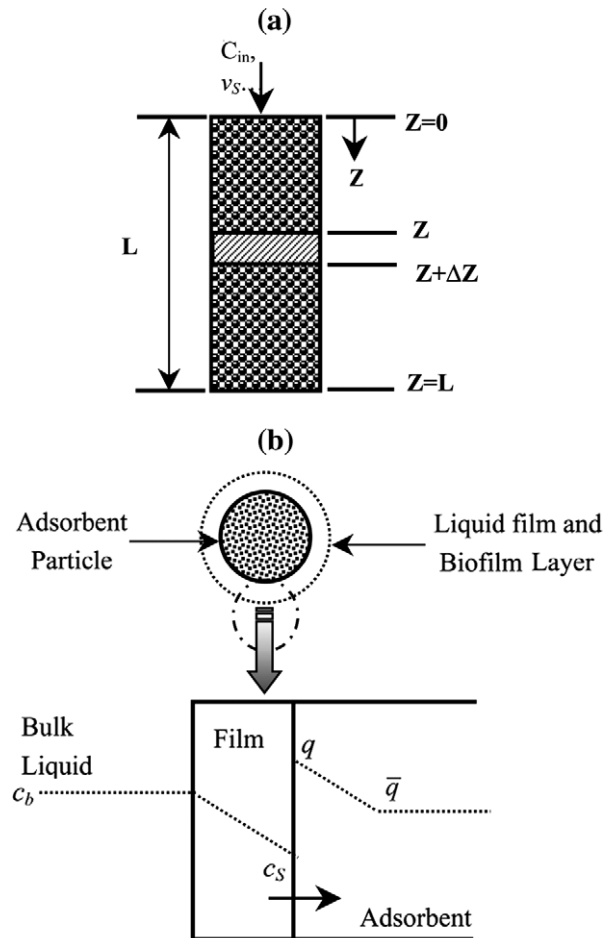


Fig. 4. Schematic representation of the mass balance in (a) bioactive GAC (b) and the biofilm on an adsorbent surface.

4.1. Substrate in the bulk liquid

The rates of removal of the substrate from the liquid phase by adsorption (γ_{ADS}) and biodegradation (γ_{BIO}) are given by:

$$\gamma_{\text{BIO}} = k_{\text{max}} \cdot \frac{C \cdot X_S}{K_S + C}, \quad \gamma_{\text{ADS}} = (1 - \varepsilon_b) \cdot \frac{3N}{4\pi R_p^3} \quad (2)$$

where N is the adsorbate uptake rate per pellet, ε_b is the bed porosity, R_p is the pellet radius, k_{max} is the maximum rate of substrate utilisation, K_S is the Monod half velocity coefficient, X_S is the suspended cell concentration and C is the liquid phase concentration [15].

The unsteady-state material balances on the substrate in the bulk liquid can be represented by the advection–diffusion equation with the inclusion of adsorption and reaction terms as follows:

$$\frac{\partial C}{\partial t} = D_{\text{ax}} \cdot \frac{\partial^2 C}{\partial z^2} - u \cdot \frac{\partial C}{\partial z} - \gamma_{\text{BIO}} - \gamma_{\text{ads}} \quad (3)$$

where D_{ax} is the axial dispersion coefficient and u is the interstitial velocity. The initial and boundary conditions are:

Initial condition, $C = 0$

Boundary condition at $z = 0$ is $C = C_0$, and at $z = L$ is $\frac{dC}{dz} = 0$.

4.2. Biomass suspended in the bulk liquid

Suspended biomass accumulates on the adsorbent due to deposition, growth, decay and shear loss. The equation for suspended biomass in the bulk liquid is as follows:

$$\frac{\partial X_S}{\partial t} = \left(Y \cdot \frac{k_{\text{max}} \cdot C}{K_S + C} - K_d - \frac{\beta}{\theta \cdot \varepsilon_b} \right) \cdot X_S + \frac{1 - \varepsilon_b}{\varepsilon_b} \cdot a_f \cdot X_f \cdot \sigma \quad (4)$$

where Y is the yield coefficient, K_d is the decay constant, β is the filtration efficiency, θ is the empty bed contact time, X_f is the cell density of biofilm, a_f is the specific surface area and σ is the biofilm shear loss coefficient.

The associated initial and boundary conditions are:

Initial condition, $X_s = X_{s0}$

Boundary condition at $z = 0$ is $X_s = X_{s0}$, and at $z = L$ is $\frac{dX_s}{dz} = 0$. Z and L are defined in Fig. 8.

4.3. Biofilm diffusion and biodegradation

Andrews and Tien [18] proposed a conceptual model of biofilm and its growth in which they

assumed that the substrate diffuses through and is taken up by the biofilm. The diffusion of the substrate across the biofilm is accompanied by its biodegradation. The model equation for biofilm diffusion with Monod type is given by:

$$\frac{\partial S}{\partial t} = D_f \cdot \frac{\partial^2 S}{\partial x^2} - X_f \cdot \frac{k_{\text{max}} \cdot S}{K_S + S} \quad (5)$$

where D_f is the molecular diffusivity within biofilm and S is the concentration of substrate in the biofilm. However, it is assumed that the amount adsorbed into the particles is small and can be ignored. Further it is assumed that no biological reaction occurs inside the adsorbent particle. The associated initial and boundary conditions are:

Initial condition, $S = S_0$

Boundary condition at $x = 0$ is $S = 0$ at $x = L_f$ is

$$D_f \cdot \frac{\partial S}{\partial x} = k_f \cdot (C - S)$$

where k_f is the interphase mass-transfer coefficient from liquid to biofilm.

4.4. Biofilm growth and decay

Since the concentration profiles are expressed over a film thickness and the biofilm thickness varies with time, a complete description of the biofilm requires the knowledge of the film thickness as a function of time. The biofilm accumulation in the GAC biofilter due to biological activity, deposition, decay and shear loss at each time step may be written as follows:

$$\frac{dL_f}{dt} = \int_0^{L_f} \left(\frac{Y \cdot k_{\text{max}} \cdot S}{K_S + S} - b_{\text{tot}} \right) \cdot dr \quad (6)$$

where L_f is the biofilm thickness and b_{tot} is the total biofilm loss coefficient.

The initial condition is $t = 0$, $L_f = L_{f0}$,

$$L_{\text{fmax}} = R_p \left[\frac{1}{(1 - e_{\text{bo}})^{1/3}} - 1 \right]$$

4.5. Support-phase substrate balance

The linear driving force approximation (LDFA) model was used to describe adsorption kinetics in this study:

$$\frac{\partial \bar{q}}{\partial t} = k_p \cdot (q_s - \bar{q}) \quad (7)$$

where q is the adsorbed phase concentration, \bar{q} is average concentration of q , q_s is the value of q at pellet surface and k_p is the particle phase mass transfer coefficient.

4.6. Adsorption isotherm

Several isotherms are available in the literature [19]. Previous studies showed that the Sips adsorption isotherm could describe the overall adsorption of organics in wastewater effectively [15]. As can be seen from Fig. 2, the Freundlich adsorption isotherm was successful in describing the overall adsorption isotherm results of the wastewater system. Therefore, in this study, the Freundlich isotherm was used in the modelling of the biofiltration system.

4.7. Bed porosity and specific surface area

The growth of biofilm outside an adsorbent results in a change in the bed porosity and specific surface area. Alonso et al. [20] showed that the specific surface area could be calculated based on the consideration of the area and volume of biofilm lost in each contact point as compared with no contact point between solids. Then the specific area is given by:

$$a_f = \frac{3 \cdot (1 - \varepsilon_{b0})}{2 \cdot R_p} \cdot \left(1 + \frac{L_f}{R_p}\right) \cdot \left[(2 - P_n) \cdot \frac{L_f}{R_p} + 2\right]$$

where P_n is the number of characteristic packing spheres. Assuming L_f is small in relation to R_p then

$$a_f = \frac{3 \cdot (1 - \varepsilon_{b0})}{R_p}$$

The bed porosity with biofilm, ε_b , is given by:

$$\varepsilon_b = 1 - (1 - \varepsilon_{b0}) \cdot \left[\left(1 + \frac{L_f}{R_p}\right)^3 - \frac{P_n}{4} \cdot \left(\frac{L_f}{R_p}\right)^2 \cdot \left(2 \cdot \frac{L_f}{R_p} + 3\right) \right]$$

Assuming L_f is small in relation to R_p , then

$$\varepsilon_b = \varepsilon_{b0}$$

4.8. Backwashing system

The daily backwash adopted in the experimental study to avoid the physical clogging of the biofilter

was found not to have any significant effect on the organic removal efficiency of the filter. Further several investigators have examined the bed expansion due to filter backwash [21,22]. They found no major loss of biomass during backwash of the biofilter. Servias et al. [22] backwashed the GAC biofilter with air scour and water routinely every 50–100 h of continuous run, but observed no significant difference in vertical biomass profiles before and after backwash. After backwashing, the bed length was assumed to be constant; and the specific surface area and bed porosity were calculated as follows (Eqs. (8) and (9), respectively):

$$a_f = \frac{3 \cdot (1 - \varepsilon_{b0})}{2 \cdot R_p} \cdot \left(1 + \frac{L_f - L_{f-bw}}{R_p}\right)^2 \cdot \left(1 + \frac{L_f - L_{f0}}{R_{peff}}\right) \cdot \left[(2 - P_n) \cdot \frac{L_f - L_{f0}}{R_{peff}} + 2\right] \quad (8)$$

$$\varepsilon_b = 1$$

$$- (1 - \varepsilon_{b0}) \cdot \left[\left(1 + \frac{L_f - L_{f0}}{R_{peff}}\right)^3 - \frac{P_n}{4} \cdot \left(1 + \frac{L_f - L_{f0}}{R_{peff}}\right)^2 \cdot \left(2 \cdot \frac{L_f - L_{f0}}{R_{peff}} + 3\right) \right] \quad (9)$$

4.9. Model results and discussion

The GAC biofilter model was employed to predict the real performance. TOC influent concentration and filtration rate were varied to match the experimental conditions, see Table 5.

In this model, the Freundlich isotherm was used. In particular, the variable bed porosity and backwashing system were also employed in the theoretical model to fit the real data. Table 5 present the estimated value of physical and biological parameters, which were used in the modelling of the GAC biofilter. Several biological parameters including film and solid mass transfer coefficients were derived using a reliable optimisation technique, see Table 5. The Nelder–Mead simplex algorithm which is one of the well-known procedures for unconstrained optimisation was used to effectively determine the optimised values. Other biological parameters such as biofilm thickness, maximum growth rate, suspended cell concentration, etc. were obtained from previous studies [24,25]. The diffusion coefficient, D_s , and axial dispersion coefficient, D_{ax} were based on values in Shim et al. [16] and Chang and Rittman [26,27]. Physical parameters such as D_s and D_{ax} were kept constant while the filter bed depth was varied. Realistic values of coefficients were used in the model.

Fig. 3 shows the fit with experimental data with different filter velocities where the influent concentra-

Table 5

Parameters used for model simulation of GAC biofilter. The same parameters were used for all runs

Physical and biological parameter	Experimental 1	Experimental 2
Diffusion coefficient, D_f (m ² /s)	9.978×10^{-13}	4.339×10^{-13}
Film mass transfer coefficient, K_f (m/s)	9.86×10^{-6}	1.223×10^{-6}
Solid mass transfer coefficient, k_s (1/s)	9.68×10^{-6}	1.152×10^{-6}
Biomass density, X_f (mg/L)	8.53×10^3	2.532×10^3
Yield coefficient, Y (mg/mg)	0.086	0.086
Decay coefficient, K_d (s ⁻¹)	1.017×10^{-7}	6.157×10^{-7}
Biofilm thickness, L_{f0} (m)	1.0×10^{-6}	1.0×10^{-6}
Suspended cell concentration, X_s (mg/L)	1.0×10^{-08}	1.0×10^{-08}
Shear loss, σ (s ⁻¹)	2.346×10^{-7}	6.596×10^{-7}
Maximum rate of substrate utilisation, K_{max} (s ⁻¹)	1.605×10^{-4}	1.605×10^{-4}
Monod half velocity coefficient, K_s (mg/L)	0.238	0.238

tion was held constant. The model is able to predict the average long-term experimental results in the biological phase.

5. Conclusions

The GAC biofilter was found to remove a significant amount of organic matter from the diluted synthetic landfill leachate (which represents the contaminated groundwater by untreated landfill leachate). The experiments were conducted at low (0.2 m/h) and high (2 m/h) flow velocity through the GAC filter to represent in-situ and ex-situ biofiltration. The results show that the organic matter can be removed in a consistent manner for a long period of time. GAC biofiltration led to a consistent TOC removal even after a long period of operation without the need to regenerate the activated carbon. Even after 30–50 days of continuous running, the organic removal efficiency from the GAC biofilter was approximately 40% and 60% when high (2 m/h) and (0.2 m/h) low filtration velocities were used. It should be noted that the performance can be enhanced by using a larger filter depth which is the case in real situations. A comprehensive model derived from the fundamental adsorption and biodegradation mechanisms was used to successfully simulate the long-term biofilter data. This model is representative and can predict the long-term organic removal.

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