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Performance of Granular Activated Carbon (GAC) Adsorption and Biofiltration in the Treatment of Biologically Treated Sewage Effluent

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Abstract: In this study, the performance of GAC adsorption and biofiltration systems in treating biological treated sewage effluent (BTSE) was evaluated in terms of organic removal efficiency, organic fractions, and molecular weight distribution (MW) of organic matter (OM) removed. The GAC biofilter removed 23.5% and 61% of the hydrophobic fractions and hydrophilic fractions of OM in the BTSE respectively. MW distribution studies of GAC filter and GAC adsorption revealed the following: Hydrophobic fraction of the effluent showed a peak at 345 dalton after GAC biofiltration and 256 dalton after GAC adsorption, whereas, with hydrophilic fractions, peaks at 46,178 and 345 daltons were observed after GAC biofiltration and peaks at 46,178 and 256 daltons after GAC adsorption. Transphilic fraction showed the peaks at 12,783 dalton with GAC biofiltration, and 1,463 dalton with GAC adsorption. The performance of the GAC biofilter was successfully mathematically modelled.

Keywords: Biologically treated sewage effluent, effluent organic matter, GAC adsorption, GAC biofilter, molecular weight distribution, hydrophobic and hydrophilic fraction

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INTRODUCTION

Granular activated carbon (GAC) adsorption is widely used as an advanced treatment in wastewater treatment plant (1). Adsorption is very effective in removing refractory or hydrophobic organic substances which are not completely removed by the biological treatment process. Arana et al. (2) showed that adsorption removed 53% of dissolved organic carbon (DOC) from the BTSE taken from Universidad de Las Palmas de Gran Canaria sewage treatment plant (STP), Spain. Abdessemed and Nezzal (3) reported 71% of COD removal from the biologically treated sewage effluent (BTSE) of Staoueli STP, Algeria. Vigneswaran et al. (4) observed 72% removal of DOC from the BTSE of Gwangju STP, Korea by using GAC adsorption.

When the GAC adsorption column is operated for a long period of time, the adsorption capacity of GAC is gradually reduced with its operational time depending on the organic and hydraulic loading rates through the column. However, the adsorbed organics on the surface of the GAC leads to the growth of microorganisms on the surface of the adsorbent, which can further assimilate and degrade the organic matters. The fixed bed system with attached microorganisms on the surface of the adsorbent is referred to as biofilter. Biofilter can produce a stable high quality effluent, which can be reused for various purposes. Kim et al. (5) found that GAC biofilter combined with sand and anthracite media removed 64% of DOC from the BTSE of a sewage treatment plant, Singapore.

EfOM fractions in BTSE can be categorized into six classes: hydrophobic acids, bases and neutrals and hydrophilic acids, bases and neutrals. Dissolved organic matter (DOM) fraction distribution varies substantially depending on the kind of wastewater and the type of treatment process employed. In particular, the hydrophilic fraction was found to be the most abundant fraction in all effluents, constituting 32–74% of the TOC. Hydrophobic acids were the second most dominant portion, accounting for 17–28% (6). Jarusuthirat et al. (7) reported that BTSE consists of the majority of hydrophobic and hydrophilic fractions. This is due to microbial origin and refractory organic compounds since it is what remains after extensive biodegradation. These finding suggests that hydrophobic neutral and transphilic fraction may be easily biodegradable and adsorbed by activated sludge.

It is also important to investigate the range of MW of organic matter removed from the effluent organic matter (EfOM) so that removable and non-removable EfOM during GAC biofiltration can be defined. This will further assist in the selection of suitable treatment methods (8) and to optimize the biofiltration process. In addition, information by MW distribution provides advantages including:

- (i) a more fundamental understanding of the complex interactions that occur in the unit operations and treatment process,
- (ii) process selection and evaluation to develop improved technique (9).

In this study, GAC adsorption and biofiltration processes were evaluated in terms of removal of dissolved organic carbon (DOC), different organic fractions, and molecular weight distribution. The experiments were conducted with BTSE from a wastewater treatment plant. BTSE was first used to study the adsorption equilibrium and kinetics of the GAC. The hydrophobic and hydrophilic fractions of EfOM were measured to investigate in detail how GAC adsorbs different components of wastewater. The analysis on different fractions was carried out in terms of MW distribution. The main objective of this study was

- (i) to evaluate the removal of different organic fractions,
- (ii) to find out the MW ranges removed by GAC adsorption and GAC biofiltration and
- (iii) to determine the coefficients of the adsorption equilibrium and kinetic equations and to mathematically model the biofilter.

EXPERIMENTAL METHODOLOGY

Bio-treated Sewage Effluent (BTSE)

The study was conducted with BTSE drawn from a sewage treatment plant. The wastewater treatment is a medium-sized activated sludge unit (25,000 m³/d). The characteristics of the BTSE used are presented in Table 1. The hydraulic retention time and the sludge age were 6 h and about 8 days, respectively.

Materials

In this study, GAC manufactured by Calgon Carbon Corporation, USA, was used in this study and its properties are shown in Table 2.

Adsorption Equilibrium and Kinetics Studies

Adsorption equilibrium experiments were conducted with 250 mL of BTSE in flasks, each with different but predetermined amounts of GAC. The amount of

Table 1. Characteristics of BTSE used

DOC (ppm)	BOD ₅ (ppm)	Ph	SS (ppm)	TN (ppm)	TP (ppm)	Conductivity (μ S/cm)
6.5–10.4	9.4–18	6.8–7.5	3.5–5.0	23.2–40	2.2–5	200–584

Table 2. Characteristics of granular activated carbon (GAC) (Calgon Carbon Corp., USA) used in this study

Specification	GAC
Surface area (m^2/g)	1001.2
Mean pore diameter (\AA)	22.55
Micropore volume (cm^3/g)	0.269
Mean diameter (μm)	750
Bulk density (kg/m^3)	600
Product code	F-400

GAC used for this study varied from 0.01 g/L to 3 g/L. These flasks were shaken continuously for 3 days at 130 rpm at 25°C. Adsorption kinetics experiments employed mechanical stirrers rotating at 100 rpm.

GAC Biofiltration System

Long term bioadsorption (biofiltration) experiments were conducted using a GAC biofilter column with BTSE influent. The column had ports for influent feeding, effluent collection, and backwashing. The column was packed with 20 g (to a bed depth of 7 cm in the column) of GAC (Fig. 1). The GAC bed was acclimated at a constant filtration rate of 1 m/h. The filter was backwashed for approximately 5 minutes every 24 hours of filtration run to eliminate excess biomass deposition which may lead to filter clogging. The backwash rate was controlled by allowing up to 30% bed expansion.

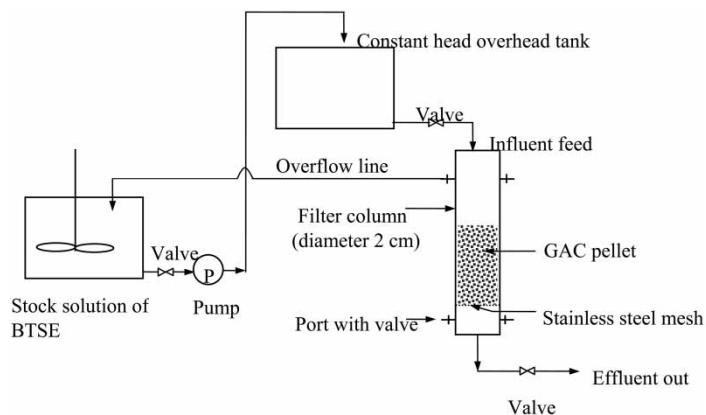


Figure 1. Schematic diagram of the biofiltration system.

EfOM Characterization Methods

Total Organic Carbon (TOC)

TOC was measured by using the Dohrmann Phoenix 8000 UV-persulphate TOC analyser equipped with an autosampler. All samples were filtered through a $0.45\text{ }\mu\text{m}$ membrane prior to the TOC measurement. Thus, the TOC obtained was, in fact, dissolved organic carbon (DOC) values.

XAD Fractionation of EfOM

Organic fractions in BTSE can be classified as hydrophobic, transphilic and hydrophilic fractions. Figure 2 shows the isolation protocol of hydrophobic, transphilic, and hydrophilic fractions from BTSE. XAD-8 and XAD-4 resins were used for fractionating EfOM into hydrophobic EfOM (XAD-8 adsorbable; mostly hydrophobic acids with some hydrophobic neutrals) and transphilic EfOM (XAD-4 adsorbable; hydrophilic bases and neutrals) components. The remaining fraction escaping the XAD-4 was the hydrophilic component. The resin in the column was washed in order of pure water, 0.1 N NaOH, pure water, 0.1 N HCl and pure water. After filtering all the samples, they were then acidified to pH 2 due to reduction of HP interaction between EfOMs and resins. The acidified samples passed through the resins with low velocity (2 mL/min). The effluents which underwent the XAD-8/4 resins were decided as the HL fraction. The adsorbed HP and TP fractions on the XAD-8/4 were eluted with 0.1 N NaOH. The DOC was measured with the eluted effluents. The content percentage of each fraction was calculated by mass balance. The resins used were

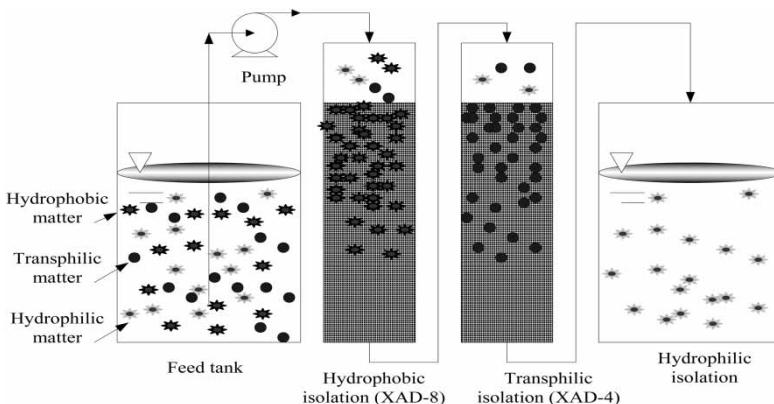


Figure 2. Schematic drawing of isolation for hydrophobic, transphilic, and hydrophilic components with XAD-8 and XAD-4 resins.

regenerated in methanol followed by acetonitrile with a Soxhlet extraction for 48 hours.

MW Distribution

BTSE after each pretreatment was subjected to measurement of the MW distribution. High pressure size exclusion chromatography (HPSEC) (Shimadzu Corp., Japan) with a SEC column (Protein-pak 125, Waters Milford, USA) was used to determine the MW distribution of organic matter. Standard solutions of different polystyrene sulfonates with known MW (PSS: 210, 1,800, 4,600, 8,000, and 18,000 daltons) were used to calibrate the equipment. The details of the measurement methodology are given elsewhere (10).

Colloidal Organic Fraction

The dialysis was performed with Spectra/Por-3 regenerated cellulose dialysis membrane bag (MWCO 3,500 daltons) (Fig. 3). The dialysis membrane was first soaked in pure water for 24 hours, and then the acidified wastewater sample (pH 1 with HCl) was placed in the pre-washed dialysis membrane bag. It was dialyzed for 8 hours (each time) against three 4 L portions of 0.1 N HCl to remove salts and low MW of organic matter. It was then dialyzed until the silica gel precipitate was dissolved against 4 L of 0.2 N HF. Finally, it was dialyzed for 12 hours (each time) against two 4 L portions of pure water. This was to

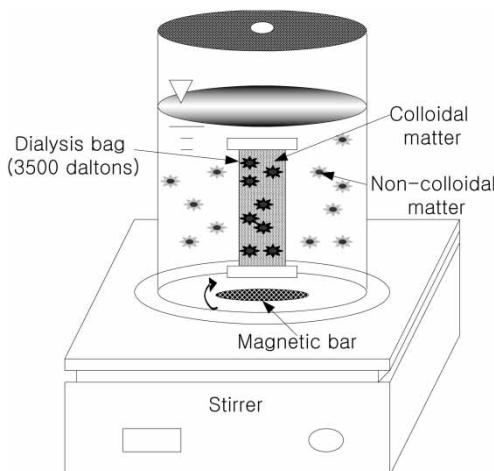


Figure 3. Schematic of separation for colloidal and non-colloidal fractions with Spectra/Por-3 regenerated cellulose dialysis membrane bag (MWCO 3,500 daltons).

remove the residual HF and fluosilicic acid. Finally, the sample was taken out of the dialysis membrane from the last 4 L of dialysate of deionized water and measured for its TOC content. This represents the organic colloidal matter (with MW range from 3,500 daltons to 0.45 μm). In the context of wastewater engineering practice, organic matter of 3,500 daltons is too small to be called as organic colloidal matter.

RESULTS AND DISCUSSION

Adsorption Isotherm and Kinetics

In biofilter column experiments there is a short initial period of a few hours to days where organic removal occurs predominantly through the adsorption processes. After a few days of operation when biological activity is established, organic removal occurs through a biodegradation processes. Batch adsorption isotherm and kinetic experiments were conducted to evaluate the adsorption isotherm coefficients and the mass transfer rate from the BTSE solution to the GAC surface. These values were used to model the adsorption/biosorption process in the biofilter (11).

The EfOM removal by different GAC dosages was first measured in terms of DOC (Fig. 4). The removal of the EfOM in BTSE showed the optimal dose of GAC was at 1 g/L which resulted in 59.7% of DOC removal.

The isotherm parameters were determined using the Langmuir, Sips, and Freundlich isotherm model equations. These equations are summarised in

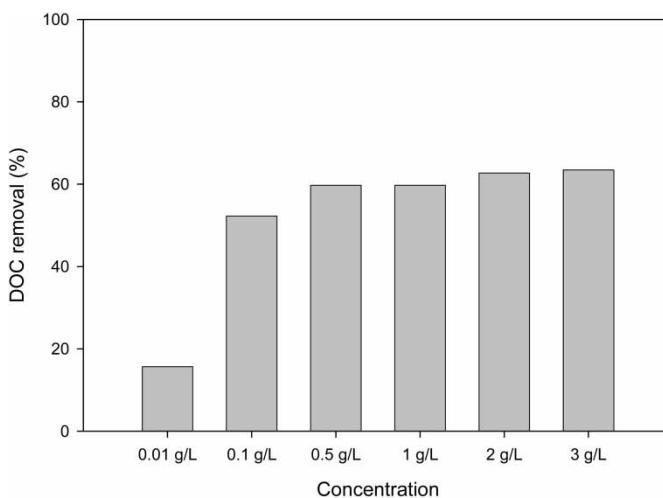


Figure 4. DOC removal of GAC at different concentrations in BTSE. (contact time = 3 days; stirring speed = 130 rpm; initial DOC = 6.5 mg/L).

Table 3. Isotherm parameters of GAC with BTSE

	q _m	b	n	K
Sips $q = \frac{q_m C^{1/n}}{1 + bC^{1/n}}$	2.100E + 1	1.676E - 8	6.750E - 2	
Freundlich $q = KC^{1/n}$			3.028E - 1	6.948E - 2
Langmuir $q = q_m C / (1 + bC)$	2.105E + 1	1.120E - 1		

Table 3. As can be seen in Fig. 5, the Sips isotherm was found to fit well with the experimental results although Freundlich isotherm is the commonly used model. The isotherm parameters are presented in Table 3.

Batch adsorption kinetic experiments were then conducted to evaluate the mass transfer rate of organics from the BTSE solution to the GAC surface. The overall mass balance in the batch reactor is given by the following equation.

$$V \frac{dC}{dt} + M \frac{dq}{dt} = 0 \quad (1)$$

Where

V = Volume of the BTSE in batch reactor, L

M = Weight of the adsorbent, g

C = Bulk organic concentration, mg/L

q = amount organic adsorbed, mg/g

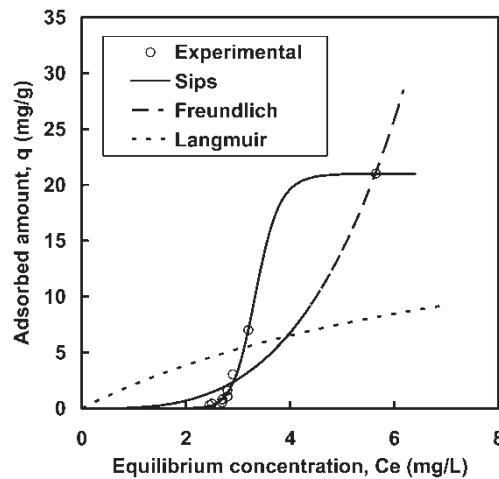


Figure 5. Equilibrium isotherm of EfOM with GAC from BTSE (contact time = 3 days; stirring speed = 130 rpm; initial DOC = 6.5 mg/L).

In this study, the mass transfer rate was described by linear driving force approximation (LDFA) model (12).

$$\frac{dq}{dt} = k_p(q_s - \bar{q}) = \frac{3 \cdot k_f}{R \cdot \rho_p} (C_i - C_s) \quad (2)$$

where q is the adsorbed-phase concentration, \bar{q} is the average concentration of q , q_s is the value of q at pellet surface, k_p is the particle phase mass transfer coefficient, ρ_p is the particle density of adsorbent, R is the radius of adsorbent particle, k_f is the external film mass transfer coefficient of organic, C_i is the initial organic concentration, and C_s is the saturation organic concentration.

It was selected because of its simplicity and the use of a lumped parameter such as TOC to represent the liquid phase concentration of the system. In the LDFA model it is assumed that the rate of adsorption of the adsorbate to a particle of adsorbent is linearly proportional to a driving force developed from the difference between the surface concentration and the averaged adsorbed-phase concentration. The mass transfer rate k_f in Equation (2) was calculated by fitting the experimental data with the model. The fluid mass transfer coefficient (k_f) in BTSE using 1 g/L of the GAC adsorbent was 2.11×10^{-5} m/s. The experimental results and the model prediction are shown in Fig. 6.

Biofiltration System

The experimental column packed with GAC fixed bed was operated for a period exceeding 2 months. The column was fed with BTSE at a filtration rate of 1 m/h.

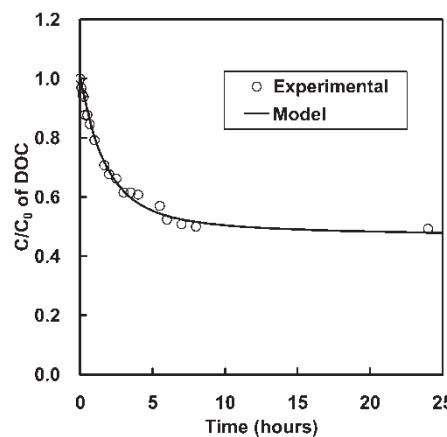


Figure 6. Mass transfer of batch adsorption of EfOM with GAC. GAC dose = 1 g/L; rotation speed = 100 rpm; initial DOC = 6.5 mg/L. $k_f = 2.11 \times 10^{-5}$ m/s.

The filtration rate was kept low as the GAC bed depth was shallow (7 cm depth which corresponds to 20 g of GAC). The organic removal capacity of the biofilters (in terms of the ratio of the concentration of TOC in the effluent to the concentration of TOC in the influent) is presented in Fig. 7.

These 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 60 days of continuous running, the effluent from the GAC biofilter was approximately 40% of the influent quality (i.e. 60% removal). Initially, in the first few days, organic removal is a result of GAC adsorption. Beyond that time with the growth of microorganisms on the GAC, the organic removal is principally the result of biomass activity.

The daily backwash adopted to avoid the physical clogging of the biofilter did not have any significant effect on the organic removal efficiency of the filter. Some of the biomass was lost during the backwashing of the filter but loss of biomass created more sites on the GAC for adsorption of microorganisms and organics and thus the impairment is balanced. This process however creates a slight fluctuation pattern as can be seen in the experimental data in Fig. 7.

Biofilter Mathematical Model

A mathematical model was developed to simulate the organic removal efficiency of the GAC biofiltration system. In this simulation model, the performance is described in two stages: adsorption during the initial stage and biodegradation in the latter stage. In practice, the prediction of

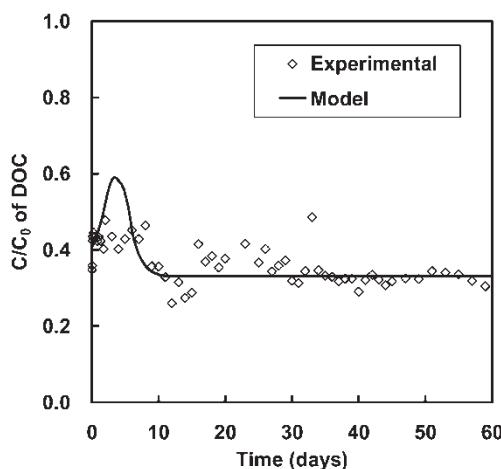


Figure 7. Performance of GAC-packed biofilters with time for 60 days in BTSE (velocity: 1 m/h, bed depth: 7 cm).

the performance of a biofilter during the biological phase or steady phase is more important because the initial stage only last for a short duration of 1–3 days at the beginning of a run (13, 14). Therefore the simulation of the adsorption process was simplified. The full details were previously presented in (14) and (13). Therefore only a cursory outline is presented.

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 biodegradation terms as follows:

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

where D_{ax} is the axial dispersion coefficient and u is the interstitial velocity. γ_{ADS} and γ_{BIO} are the rates of removal of the substrate from the liquid phase by adsorption and biodegradation respectively and are given by:

$$\gamma_{BIO} = K_{max} \cdot \frac{C \cdot X_S}{K_S + C}, \quad \gamma_{ADS} = \frac{(1 - \varepsilon_b)}{\varepsilon_b} \cdot a_f \cdot K_f \cdot (C - S) \quad (4)$$

where ε_b is the bed porosity, a_f are the pellet surface area and radius respectively, K_f is the film mass transfer coefficient, K_{max} is the maximum rate of substrate utilization, K_S is the Monod half velocity coefficient, X_S is the suspended cell concentration, S is the concentration of the substrate in the biofilm, and C is the liquid phase concentration. The linear driving force approximation model (LDFA) was used to describe the adsorption kinetics (12). The Sips adsorption isotherm was used to describe the overall adsorption of organics of BTSE. Details of parameters X_S and S are given in (13) and (14).

The initial and boundary conditions are

$$\text{at } z = 0 \text{ is } D_{ax} \frac{dC}{dz} = -v(C|_{z=0^-} - C|_{z=0^+})$$

$$\text{and at } z = L \text{ is } \frac{dC}{dz} = 0$$

In this study backwashing was not incorporated in the model as it did not affect the filter performance based on the experiments carried out in this study. The result of the modelling is shown in Fig. 7. The modelling parameters used are given in Table 4.

MW Distribution

The organic filter evaluation based on TOC removal alone is not sufficient and to improve the performance of adsorption and biofiltration it is important to know the range of MW distribution of EfOM so that the portion of the EfOM that can be removed by adsorption is able to be defined. The MW

Table 4. Physical parameters used for model simulation of GAC biofilter

Parameter	Value
Bed depth (cm)	7
Velocity (m/h)	1
TOC of influent (mg/L)	6.5
Diffusion coefficient, $D_f (\times 10^{-10})$ (m ² /s)	1.40
Film mass transfer coefficient, $K_f (\times 10^{-5})$ (m/s)	2.11
Axial dispersion coefficient, $D_{ax} (\times 10^{-7})$ (m ² /s)	1
Solid mass transfer coefficient, $K_s (\times 10^{-6})$	4.78
Maximum rate of substrate utilisation, $K_m (\times 10^{-5})$	5.77
Monod half velocity coefficient, K_p	1.11

distribution after GAC biofiltration for a different batch of BTSE is shown in Fig. 8. Figure 8 shows the MW distribution of biofilter after a few days of operation (1, 4, and 7 days of operation) where the predominant organic removal mechanism was biodegradation.

Fractions and Colloid before and after GAC Biofiltration

The influent and effluent from the GAC biofilter were then analysed for the hydrophobic, transphilic and hydrophilic fractions. The colloid and hydrophobic and hydrophilic fractions in the influent and effluent of the biofilter

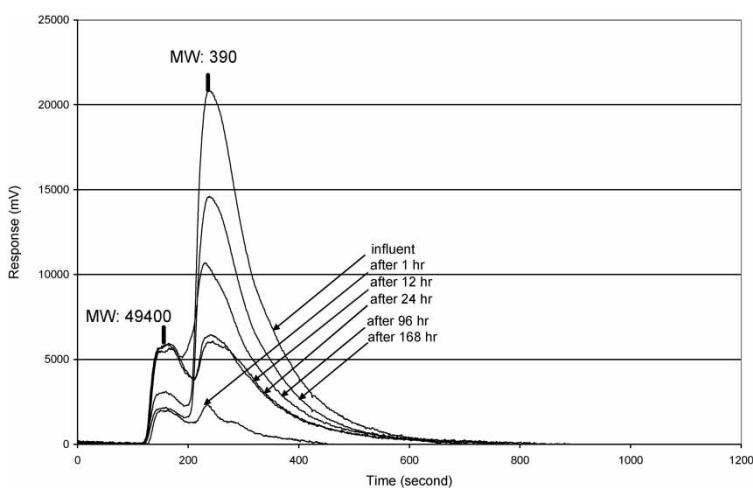


Figure 8. MW distribution of effluent following GAC biofilter treatment. BTSE used was different from that of Figure 8 (velocity = 1 m/h; bed depth = 7 cm, initial DOC = 6.5 mg/L).

Table 5. Organic colloidal portion (in DOC) in the BTSE and in the biofilter effluent after 30 days

	Colloidal portion of EfOM (rejection, %)
BTSE	4.04 mg/L
After biofiltration	0.96 mg/L (76.2%)

are presented in Tables 5 and 6. The removal of colloidal compounds by the GAC biofilter was 76.2%, indicating that the GAC biofilter was adequate in removing the relatively large MW organic compounds (above 3,500 daltons). The hydrophobic fraction removed by the GAC biofilter was 23.5% of DOC whereas the removal of the hydrophilic fraction was 61.1%. The removal of hydrophilic portion of organics could be attributed to biodegradation. It could also be (to some degree) due to the physical and chemical affinity between hydrophilic organic molecules and GAC (through Vander Waals, electrostatic forces and chemisorption) (15).

MW Distribution of Different Fractions and Colloidal Matter

The MW of the initial BTSE ranged from 300 daltons to about 46,178 with the highest fraction of 300–5,000 daltons. The MW distribution of hydrophobic, transphilic and hydrophilic fractions and colloids in the effluent by GAC adsorption and GAC biofiltration is presented in Fig. 9.

Hydrophobic fraction showed a peak at 256 daltons for GAC adsorption and at 345 dalton for GAC biofilter. This indicates that hydrophobic fractions consisting of smaller organics are found both after GAC biofilter and adsorption. In contrast, hydrophilic MW distribution showed a reversed order compared to hydrophobic fraction. Both GAC biofiltration and GAC adsorption showed peaks at large MW range of 46,178 daltons for the hydrophilic organic fractions. This means that GAC biofilter and adsorption do not remove the large

Table 6. Hydrophilic, hydrophobic, and transphilic fractions in the BTSE with biofiltration after 30 days

Fraction	DOC of BTSE (ppm)	DOC of the effluent after biofiltration (rejection, %)
Hydrophobic	4.98	3.81 (23.5)
Transphilic	1.68	1.42 (15.5)
Hydrophilic	3.19	1.24 (61.1)

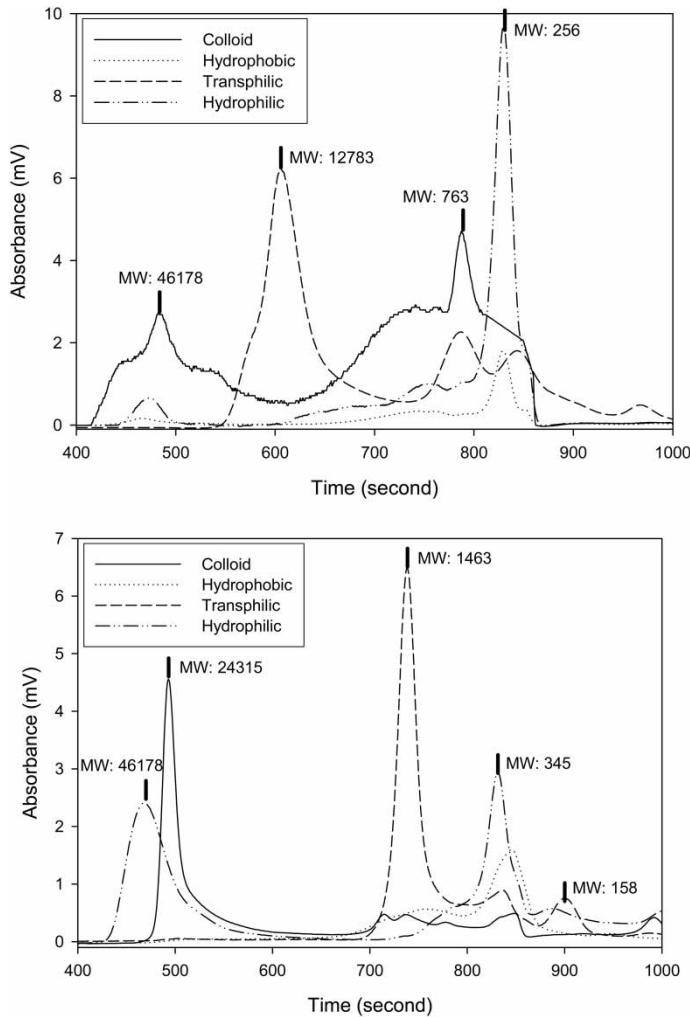


Figure 9. MW distribution of colloid, hydrophobic, transphilic and hydrophilic with BTSE (a) GAC adsorption and (b) GAC biofiltration after 30 days of operation.

hydrophilic MW. Transphilic fraction indicated the peaks at 1,463 daltons for GAC biofiltration and at 12,763 daltons with GAC adsorption.

CONCLUSIONS

This study investigated in detail the characteristics of the EfOM by GAC adsorption and GAC biofiltration. A detailed characterization of EfOM was

made in terms of the colloidal organic matter, hydrophobic and hydrophilic organic matter, and MW distribution of EfOM. Further the organic removal by GAC adsorption and biofilter was modelled. The results obtained lead to the following conclusions.

1. In the adsorption isotherm study, the Sips was found to fit well, on an overall basis, with the experimental results of GAC adsorptions of BTSE. The performance of the GAC biofilter was mathematically modelled. The modelling was limited to TOC removal only.
2. GAC adsorption could not remove the MW of 324 daltons. It could be due to the presence of recalcitrant carbohydrate, amino acid and fatty acid.
3. Removal of colloidal compounds was 76.2% by GAC biofilter. Hydrophobic fraction removal was 23.5% by the GAC biofilter. The removal of hydrophilic fraction was 61.1% by GAC biofilter. The removal of hydrophilic portion of organics could be attributed to biodegradation. It could also be to some degree due to the physical and chemical affinity between hydrophilic organic molecules and GAC through Vander Waals, electrostatic forces and chemisorption.
4. Hydrophobic fraction showed the peak of 345 daltons for GAC biofiltration, while GAC adsorption had the peak at 256 daltons. In contrast, hydrophilic fractions had a twin peaks compared to hydrophobic fraction i.e. GAC adsorption showed peaks of 46,178 daltons and 256 while GAC biofilter showed peaks at 46,178 daltons and 345. This shows the inability of the biofilter and adsorption in removing large MW hydrophilic fraction due to extracellular polymer substance. Transphilic fraction indicated the peaks of 12,783 daltons (GAC biofilter) and 1,463 (for GAC adsorption).

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