# Granular Activated Carbon (GAC) Biofilter in Water and Wastewater Treatment

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# CERTIFICATE

I certify that this thesis has not already been submitted for any degree and is not being submitted as part of candidature for any other degree.

I also certify that the thesis has been written by me and that any help that I have received in preparing this thesis, and all sources used, have been acknowledged in this thesis.

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# Nomenclature

A = the specific surface area of the media

ATP = adenosine triphosphate

$BOD_5$ = biochemical oxygen demand

$b_{tot}$ = Total biofilm loss efficient ($s^{-1}$)

b*tot* = the total biofilm loss coefficient

C = the liquid phase concentration.

$C_0$ = initial TOC concentration of synthetic wastewater (mg/L)

$C_e$ = the equilibrium organic concentration (mg/L)

COD = chemical oxygen demand

CFU = Number of Colony forming units

$D_{ax}$ = the axial dispersion coefficient

DBPs =disinfection by products

$D_f$ = the molecular diffusivity within biofilm

$D_L$ = axial dispersion coefficient

DNA = deoxyribonucleic acid

DO = Dissolved oxygen

DOC = dissolved organic matter

$D_s$ = diffusion coefficient (, $m^2/s$)

$D'$ = the dispersion coefficient ($m^2/s$)

EBCT = empty bed contact time

EPS = extracellular polysaccharides

GAC = granular activated carbon

*H* = Henry law's constant

HAAs = haloacetic acids

HLR = Hydraulic loading rate

$J_f$ = the flux of substrate into the biofilm ($mg/m^2/s$)

$k$ = the maximum rate of substrate utilization (mg/mg/s)

K = reaction constant

$K_d$ = Decay coefficient ($s^{-1}$)

$K_f$ = film mass transfer coefficient (m/s)

$K_S$ = Solid phase mass transfer

$Ks$ = the Monod half-velocity coefficient (mg/L)

$K_{max}$ = Maximum rate of substrate utilization

$k_P$ = the particle phase mass transfer coefficient

$L$ = reaction constant

LDFA = Linear driving force approximation

$L_f$ = the biofilm thickness

$L_{f0}$ = Biofilm thickness (m)

$M$ = the weight of used GAC (g)

MW = Molecular weight (Dalton)

NDIR = Non-dispersive infrared gas reactor

N = The adsorbate uptake rate per pellet

$P_n$ = the number of characteristic packing spheres.

NOM = natural organic matter

q = the adsorbed-phase concentration

$q_m$ = saturation amount of organic adsorbed (mg/g)

$q_S$ = the value of q at pellet surface

RNA = ribonucleic acid

$S$ = substrate concentration (mg/L)

TOC = Total organic carbon (mg/L)

TC = Total carbon (mg/L)

TIC = Total inorganic carbon (mg/L)

THMs = trihalomethanes

$u$ = the interstitial velocity

$V$ = the volume of solution (mL)

VSS = volatized suspended solids

$x$ = the distance along the biofilter length (m)

$X_f$ = the cell density of biofilm

$X_{susp}$ = the suspended cell concentration (mg/L)

Y = Yield coefficient (mg/mg)

$\beta$ = the filtration efficiency

$\sigma$ = the biofilm shear loss coefficient ($s^{-1}$)

$\varepsilon_b$ = the bed porosity

$\mu$ = specific growth rate

$\mu_m$ = the maximum growth rate

$\theta$ = the empty bed contact time

$v$ = fluid velocity (m/s)

$\psi$ = organic concentration spreading parameter

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# Abstract

Biofilters can effectively remove organic matters from water and wastewater. Basically, the success of operation of a biofilter depends mainly on the activities of the microbial community in the filter. Organic substances are adsorbed onto filter media and then biodegraded by those microbes.

The experimental investigation of the granular activated carbon (GAC) adsorption indicated that GAC exhibited a high organic removal rate by adsorption in both the batch and column experiments. The GAC adsorption equilibrium with synthetic wastewater fitted better with association theory (Talu) than with Freundlich and Sips models. Adsorption kinetics and fixed bed of GAC with organic matter was well described and predicted by the Linear Driving Force Approximation (LDFA) model. Long term GAC biofiltration was mathematically described using a simple model which incorporated both adsorption and biodegradation. The model was mainly successful in describing the biological phase.

Biomass accumulation onto GAC was evaluated in terms of dry mass and number of viable cells both in the batch and column experiments. The attachment of micro-organisms onto the GAC surface depended on the hydraulic loading rate and influent concentration. The amount of dry mass retained on GAC in the column was double than that in batch test in the steady state with 44mg/g GAC and 85mg/g GAC respectively. More viable cells were enumerated from GAC in the experiments with synthetic wastewater than with river water. The prominent bacteria isolated from GAC biofilter for synthetic wastewater and river water included: *Pseudomonas Aeruginos, Pseudomonas Alcaligenes, Brevibacterium Otitidis, Enterobacter Cloacae, Enterobacter Aerogenes, Staphylococcus Epidermidis and Kocuria Rosea*.

The long term performance of the GAC biofilter with synthetic wastewater which has similar characteristics to biologically treated sewage effluent and river water was experimentally evaluated. The result showed that the GAC biofilter could maintain high

organic removal efficiency after a long filtration time without any regeneration of activated carbon. Even after 42 days of continuous run, the biofilter of very short depth of 15cm GAC bed depth maintained a consistent organic removal efficiency of 40-50% with synthetic wastewater and 55% with river water. GAC biofilter also removed 60-98% of total coliforms from synthetic wastewater and river water. Especially, no fecal coliforms were detected in effluent from the GAC biofilter for river water.

The daily backwash adopted to avoid the physical clogging of the biofilter did not have significant effect on the performance of the filter. The change of filter bed depths, filtration rates and influent concentrations affected the performance of the GAC biofilter. Total organic carbon (TOC) removal efficiency in the higher filter bed was significantly better than that with shallow bed depth (60% TOC removal with a 30cm- bed depth while only 30% with a 5 cm - bed depth). As expected, the efficiency in organic removal decreased with an increase in the filtration rate. The difference was more significant in the lower bed. TOC removal efficiency of the biofilter was also affected by the concentration of influent. It increased with the increase in the concentration of influent, but the TOC removal pattern with time was almost the same. Further, the increase of organic removal rate was not proportional to the rise of influent concentration.

A study with different filter media showed that GAC as filter media was superior to plastic bead, anthracite and sponge in terms of organic removal. In addition, GAC was combined with sponge or polypropylene as medium to enhance the effectiveness of biofilter in removing organic matter. The use of sponge and floating media can eliminate further 10-20% of TOC from the effluent of GAC biofilter with less effort in installation, operation and maintenance.

In summary, GAC biofilter 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 of TOC removal efficiency, long life cycle, and simplicity in operation. The combined system of GAC - sponge filter media biofilter can be an effective and a good practical solution for improving the organic removal from water and wastewater.

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# CHAPTER 1

# INTRODUCTION

## 1.1 Overview

Organic matter appears in all kinds of water, from natural waters to domestic and industrial wastewaters. Its presence in water poses many problems for both water and wastewater treatment processes. Organic removal is especially of concern in water and wastewater treatment. During treatment processes, organic matter can cause the problems such as filter clogging, membrane fouling, and increase of chemical dose usage etc. The effect of organic compounds extends to the post-treatment stage when water is in distribution systems. Organic contaminants can impair the color, odour and taste of water. They promote the regrowth of microorganism, including pathogens. Organic matter is the precursor of potential harmful disinfection by-products and causes pipe corrosion in distribution systems (Eikebrockk et al., 2001). Hence, a number of treatment systems have been developed for organic removal from water and wastewater.

Among the treatment processes, biological filtration (biofiltration) is widely applied for removing organic matter from water and wastewater. By utilizing activities of microorganisms fixed on filter media, a biofilter can effectively eliminate organic substances through the biodegradation process. Many studies show that a biofilter can remove the majority of organic matter from water and wastewater resulting in less operation and maintenance requirements (Clark and Boutin, 2001; McKay, 1996). Small fractions of organic matter which cannot be removed in other conventional treatment processes can also be removed by the biofilter. Moreover, biological filtration is economical and safe for the environment.

Biological filtration using granular activated carbon (GAC) is an efficient process in water treatment. Previous studies showed that biological filters using GAC have a great potential in removing disinfectant by-products, biodegradable organic and synthetic substances (McKay, 1996). Minear and Amy (1996) stated that enhanced coagulation and GAC were proposed as the best available technologies for precursor control. The use of GAC as biofilter media has several advantages. GAC possesses an extremely large and irregular surface of the order of several hundred $m^2/g$ of carbon that provides a large number of available sites for the adsorption of substrates and

microorganisms (McKay, 1996). The GAC structure can protect microbes from shear loss during biofilter operation.

A GAC biofilter is usually passed through the acclimation stage and steady state. In the initial stage of operating the biofilter, adsorption of substances and colonization of micro-organisms is the dominant activity while in later stages organic degradation by microbial activities is more important. Microbes growing on filter media include viruses, bacteria, and protozoa. Although there are many studies on biofilters, behaviours of microbes during the filtration have still not been explained clearly due to the variety of micro-organisms involved and the number of factors that can influence the biofilter performance.

In biofilter operation, the success of maintaining a stable microbial community determines the effectiveness of the biological filtration process. It was found that filter media, empty bed contact time, backwashing, feed water characteristics and temperature are among the most influential parameters of the organic removal efficiency of a biofilter (Huck et al., 1994; Servais et al., 1989; Wang et al, 1996; Hozalski, 1996; Liu et al., 2001).

## 1.2. Objectives

- Investigating the mechanisms of adsorption and biological oxidation in long term performance of GAC biofilter for organic matter removal;
- Investigating the community of micro-organisms in GAC biofilter;
- Comparing the applicability of GAC with three different kinds of biofilters in terms of organic removal; and
- Proposing and evaluating the combined systems of GAC and one other filter medium biofilter

## 1.3. Scope

The scope of this study is divided into two parts:

**Part 1:** Investigation of the behaviour and performance of GAC biofilter in water and wastewater treatment

- Study of the bioadsorption mechanisms of GAC. This was carried out through adsorption equilibrium, kinetics and fixed bed, biodegradation. This was later described by simple mathematical models;
- Study of the attached biomass on GAC (dry mass and viable cells), microbial activity and identification in biofilter;
- Evaluation of the long-term performance of the laboratory-scale GAC biofilter in terms of biomass growth, organic matter and pathogens removal;
- Investigation of the effect of the design and operational conditions such as filter bed depth, filtration rate, influent concentration, backwashing etc on the performance of GAC biofilter; and
- Comparison of GAC with other biofilter media (sponge, floating media) in terms of organics removal efficiency

**Part 2:** Evaluation of two different kinds of combined biofilter systems in water and wastewater treatment

- Evaluation of the combined system of GAC – sponge biofilter
- Evaluation of the combined system of GAC – floating medium biofilter

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# CHAPTER 2

# LITERATURE REVIEW

## 2.1 Organic matter in water and wastewater

### 2.1.1 General

Organic matter appears widely in water and wastewater both in the form of particulates and dissolved matter. In natural water, organic matter is usually defined as natural organic matter (NOM). It comes from various sources such as soils, diffused from sediment or released from plankton and bacteria, but the largest sources of organic matter are from ocean and soil (Simpson, 2002; Krasner et al., 1996). The release of extracellular products by living micro-organisms also contributes to the formation of organic matter in the environment (Parsons and Seki, 1970). Human activities in agricultures, chemistry, medicine, etc. also contribute to the existence of organic matter in water and wastewater. Many of organic compounds which come from anthropogenic source are toxic to environment and public health such as polyaromatic hydrocarbons, polychlorinated biphenyls and dioxin (Van Loon and Duffy, 2005).

In the natural water environment, the distribution of organic matter varies with water sources, depths, the seasons, etc. Menzel and Ryther (1970) found that in the ocean, particulate and dissolved organic matter concentrations at depths of 200 m and 300 m were 3-10 μg Carbon/L and 0.35-0.70 mg Carbon/L, respectively in all their sampling locations. In natural freshwater, organic carbon concentration ranges from 0.5 to 50mg/L (Steinberg, 2003). In the components of medium strength wastewater, organic compounds account of 75% of suspended solids and 40% of filtered solids (Tchobanoglous and Burton, 1991).

Many studies focussed on organic matter removal in water and wastewater treatment because of their important roles in water. Even though there is no direct evidence that organic matter has a direct influence on public health, consuming water containing organic matter may not be completely safe for consumers in the long term (Chaudhary, 2003). Humic acids, a component of natural organic matter, are thought to indirectly relate to testicular atrophy that reduces fertility in male mammals. Also, they are suspected to influence other diseases such as Kaschin-Beck and "black foot" disease (Steinberg, 2003). Humic substances can react with hydrophobic pollutants and heavy metals such as Cu, Pb, Hg, DDT, etc. to increase their bioavailability, mobility even toxicity that increases the human digestion of those substances (Steinberg, 2003). High

concentration of organic matter in discharged wastewater can cause anoxic condition that affects aquatic environment.

Moreover, organic matter has an adverse effect in water and wastewater treatment processes. In water treatment, the occurrence of organic matter is the source of precursors for forming disinfection by-products (DBPs) such as trihalomethanes (THMs), haloacetic acids (HAAs), etc. which are suspected to be carcinogen or mutagen sources. It also supports the regrowth of micro-organisms including opportunistic pathogens in distribution system by providing nutrients, thus impairing the taste and odour of water. Some researchers believe that organic matter may cause the corrosion in distribution systems due to its local accumulation (Eikebrockk et al., 2001). The presence of organic matter can significantly affect the performance of some treatment processes through filter clogging or membrane fouling. Organic matter can consume oxidants and coagulants during treatment processes, thus the chemicals dosage needed for effective treatment increases (Eikebrockk et al., 2001).

### 2.1.2 Organic matter characteristics

Organic matter consists of a wide range of organic substrates which possess different molecular sizes, structures, functional groups and polyelectrolytic characteristics. They vary from macromolecules such as humic substances, hydrophobic acids, carbohydrates to small molecular weight substances like amino acids, carboxylic acids, etc. (Tchobanoglous and Burton,1991).

Organic matter is usually comprised of carbon, hydrogen, oxygen and nitrogen. Other chemical elements such as sulphur, phosphorous and iron can also be found in organic matter components. The molecular weight distribution of dissolved organic carbon that was investigated by Marley et al (1996) was divided into two fractions with 44% at 30,000-3,000Da and 27% at 3,000-500Da. Krasner et al. (1996) in their study of natural organic matter characterization found that its composition comprised of humic acids, fulvic acids, the hydrophobic neutral fraction and hydrophilic acids with structural differences. Among them, fulvic acids account to about 46%; hydrophilic acids (which contain more sugars and amino acids) account to 24% of the dissolved organic matter (DOC). In other research by Thurman (1983), the organic composition of river waters

contains on average about 40% fulvic acid, 10% humic acid, 30% hydrophilic acids, 10% carbohydrates, 7% carboxylic acids, about 3% amino acids, and less than 1% hydrocarbons.

Tchobanoglous and Burton (1991) showed that in wastewater, organic matter is comprised of 40-60% proteins, 25-50% carbohydrates and about 10% oils and fats. Characteristics of those organic matters depend on the nature of the industry and can be removed before discharging (Eckenfelder, 1989).

Organic matter can be quantified by different measurements such as total organic carbon (TOC), dissolved organic carbon (DOC), UV absorbance ($UV_{254}$ or $UV_{285}$), biochemical oxygen demand ($BOD_5$), chemical oxygen demand (COD) and fluorescence (Krasner et al., 1996). By using bioassays, researchers also divided organic matter into biodegradable organic matters (that can be easily consumed by micro-organisms and are measured by BOD and COD), and non-biodegradable organic matters which is recycled or accumulated in aquatic ecosystems (Urfer et al., 1997).

The main components of organic matter include proteins, carbohydrates, fat, oils, grease and humic substances.

***Proteins***

Proteins have unstable chemical structure and high molecular weight, ranging from 20,000 to 20 million Daltons. They are the main sources of nitrogen in wastewater; the proportion of nitrogen in the structure of proteins can be up to 16%. In some cases, sulphur, phosphorous and iron are found in the composition of proteins. When proteins appear in a high concentration, they cause foul odour of water. (Tchobanoglous and Burton, 1991).

***Carbohydrates***

Carbohydrates also account for a significant proportion of organic substances in water. They include sugars, starches, cellulose and wood fibre. They can be decomposed to sugar and used during biological process (Tchobanoglous and Burton, 1991).

*Fat, oils and grease*

Fats and oils are stable organic compounds that are hard to be degraded by bacteria. Their presence in wastewater causes trouble both in sewers and wastewater treatment plants because they affect the surface of biological life and create floating matters and films. They usually come from the waste of the food and petroleum industry and domestic wastewater (Tchobanoglous and Burton, 1991).

*Humic substances*

Humic substances could be derived from lignin and the interaction between sugars and amino acids (Hood, 1970). In water, humic substances account for 30-50% of organic carbon. By using the diffusion ordered spectroscopy (DOSY) technique, Simpson (2002) found that humic substances are "pseudo high molecular weight materials" which results from the association of small molecular organic acids. Humic substances have polymeric structure where rings, chains and clusters form with the size ranging from 60-500 ´. Their molecular size ranged from 4.7 -33 Å in radius of gyration (Thurman et al., 1982). Composition of humic substances includes: 40-60% carbon, 30-50% oxygen, 4-5% hydrogen, 1-4% nitrogen, 1-2% sulphur and 0-0.3% phosphorous (Gaffney et al., 1996).

Generally, humic substances are divided into three categories: humic acids, fulvic acids and humin based on the traditional fractionation of soil humic substances and their solubility in base or acid environment (Steinberg, 2003). They share the following common characteristics: (i) high molecular weight substances (1000-10,000 Dalton) which vary with the sources of NOM, the seasons and analytical techniques (Simpson, 2002; Thurman et al., 1982); (ii) performs both aliphatic and aromatic characteristics; (iii) contain many functional groups and (iv) perform polyelectrolytes in solution (Daifullah et al., 2004 and Thurman et al., 1982).

In addition, surfactants, phenol, pesticides, agricultural chemicals and trace organic compounds are also found in water and wastewater.

## 2.3 Typical treatment technologies in removing organics from water and wastewater

### 2.3.1 Typical conventional treatment technology

*A. Coagulation and flocculation*

Coagulation and flocculation processes are usually applied to remove small particles and DBP precursors from water. Organic particles with the size less than 10mm cannot settle down by themselves but when they react with coagulant agents, they can form larger aggregates which can be steadily separated from water by settlement, floating and filtration processes. While coagulation refers to initial coalescing of colloidal particles, especially hydrophobic colloids, flocculation is the long term process of forming large particles and can be useful in removing hydrophilic colloids. In practice, aluminium and iron salts such as aluminium sulphate, sodium aluminate, ferrous sulphate, ferric chloride and ferric sulphate are usually used as coagulants (Percival et al, 2000).

Coagulation can reduce trihalomethanes formation up to 50% and organic carbon by 40-70% (Freese et al., 2001). Abdessemed et al. (2000) achieved a high removal of COD (86%) and turbidity (from 18 to 3.5 NTU) when they combined flocculation and PAC adsorption. Flocculation when combined with sedimentation can effectively remove organism up to 99.9% of poliovirus and 99.7% of bacteriophage (Percival et al., 2000). The performance of coagulation processes strongly depends on pH and coagulants dosage. Therefore pH adjustment and coagulant dosage can increase TOC removal and DBP precursor removal. Enhanced coagulation is less economical in large scale treatment plants because of expenses for removing coagulants and reducing organics content in water, but in small water works, it can be cost effective (Freese et al., 2001).

*B. Biological treatment*

Biological treatment involves activities of microorganisms to remove organic matters and other undesirable substances in water. It is widely applied in both water and wastewater treatment. In biological processes, both aerobic and anaerobic microbes in suspended or attached form can be used. They convert available biodegradable organic

substrates in influent into biomass and inorganic carbon through their metabolisms. Activated sludge is a typical example of suspended growth of microorganisms in aeration condition while biofilters are application of attached microorganisms (Tchobanolous and Burton, 1991).

Activated sludge is the most commonly used aerobic suspended growth treatment process. Maintaining an adequate dissolved oxygen amount throughout the reactor is a key factor in operating activated sludge system. Bacteria in an aeration reactor are provided air by diffusion or mechanical aeration which also maintains a completely mixed regime.

Biofiltration is characterized by the separation between microorganisms that are embedded onto filter media and liquid phase (Cohen, 2000). Microorganisms in a biofilter are fixed onto surface of filter media and the composition of biomass varies along the depth of biofilm while biomass in activated sludge is uniform throughout bioreactors (Grady et al., 1999). Biofiltration includes trickling filter, roughing filter, rotating contactor and fixed film nitrification reactors. Among them, the trickling filter is the most commonly used in organics removal and nitrification (Tchobanolous and Burton, 1991; Grady et al., 1999). Biofilter is an effective technique for removing organic matters and disinfection by-products. Moreover, biological filtration has proved to be effective in removing formaldehydes, glyoxal herbicides, some heavy metals (Krasner et al., 1993). The advantage of biofiltration is the low cost in comparison to other physical and chemical treatment methods. However, during the biofiltration process, bacteria in the filter may come into effluent and need to be removed or disinfected by other processes (Cohen, 2000).

### 2.3.2 Typical advanced water and waste water treatment technology

#### *Membrane*

Membranes are attractive advanced technologies in removing organic matter and controlling disinfection by- products. Reverse osmosis, nanofiltration, ultrafiltration and micro filtration membranes are the most common membrane processes used in water treatment. While nanofiltration membranes are specific in removing particles ranging from 200 to 1000 Daltons and divalent cations, ultrafiltration and micro-filtration

membranes are effective in removing pathogens and rather large particulates, about 1000-5000000 Daltons (Clark and Boutin, 2001). Membranes are highly effective in removing organic matter. Membranes are more suitable for small systems because the cost for operation and maintenance membrane system are relatively high.

## 2.4 Biological filtration

### 2.4.1 General

Biological filtration or biofiltration is one water treatment process that can effectively remove organic matter that is not able to be removed in conventional sewage treatment from water and biologically treated sewage effluent (Carlson and Amy, 1998). Biological filter works mainly rely on the activities of the community of micro-organisms that are attached onto filter media. The activities of microbes determine the performance of biological filtration. Microbes oxidize organic matters in water to produce energy therefore available nutrients sources in feed water is essential for their development. In addition, the parameters such as hydraulic loading rate, back washing techniques, temperature and pH etc. can affect the growth of biomass onto GAC in the biofilter. Moreover, biological filtration is economical and safe for environment. Therefore, biofiltration is more suitable than other treatment methods in terms of removing organic matter.

The biological filtration using granular activated carbon (GAC) is an efficient process in drinking water treatment. Many studies showed that GAC biological filter has a great potential in removing disinfectant by products, biodegradable organic matter and synthetic substances (McKay, 1996). The removal of organic matters in water impairs the regrowth of microbes in the distribution system, thus improving the quality of water in term of colour, odour and organic precursors. Minear and Amy (1996) proposed enhanced coagulation and GAC as the best available technologies for precursors control.

Even though it has high adsorption capacity, GAC can only maintain its adsorption for a short time of biofilter operation and then its adsorption capacity becomes exhausted, thus leading to lower treatment efficiency. To recover its capacity, GAC can be regenerated by different methods such as thermal, hydrothermal, chemical and ultrasonic regeneration. However, regeneration usually reduces GAC adsorption

capacity and requires high energy expense. Another way to extend GAC life is using exhausted GAC as support filter media for biological filtration. GAC provides its huge surface area for microorganism growth and development in the biofilter. In this case, both adsorption and biological degradation take part in treatment processes. Adsorption is more dominant in the first stage or acclimatized stage when GAC is in full adsorption capacity and microbes start to attach to surface of filter media and grow up. The latter stage or pseudo steady state was controlled by microbiological activity (Dussert and Van Stone, 1994). In this stage, biological degradation plays the major role in a biofilter; therefore maintaining sufficient biomass is very important. Applying backwash is an effective method to avoid the accumulation of excess biomass that can cause biofilter clogging. It is also useful in maintaining the balance of microbiological community in a biofilter by removing dead cells and end products that may poison the microbiological environment and create free sites for new organisms.

### 2.4.2 Adsorption mechanisms

Adsorption is a complex process that involves both physical and chemical mechanisms in which the donors provide electron from surface functional groups to the sorbate molecules. Therefore amount and position of those functional groups determine the bonds between adsorbent and adsorbate molecules which decide the type of adsorption, physisorption or chemisorption (McKay, 1996).

In physical adsorption, physical bonds such as Van der Waals force and electrostatic bonds between adsorbate molecules and adsorbent surfaces play a major role. There is no electron exchange between donors and acceptors thus the system is not stable. Physical adsorption can be reversible, depending on the environmental condition. The favourable condition for physical adsorption is at low temperature (McKay, 1996).

In contrast, chemical adsorption is specific and irreversible. Chemisorption involves ionic activities and chemical bonds formation to form monolayer. This adsorption process requires high energy but can occur in a wide range of temperatures (McKay, 1996).

Both physical and chemical adsorption processes take part in adsorption system but physical adsorption is more important in separation processes due to its capacity to form multilayers (McKay, 1996). Adsorption processes are characterized by adsorption equilibrium, adsorption kinetics and fixed bed adsorption.

In general, the adsorption is comprised of three steps: (i) external mass transfer of solute molecules from solution bulk to the sorbent particle surface by diffusion or turbulent movement, (ii) diffusion within particle internal structure to transport adsorbate molecules to the available sorption sites of adsorbents surface and (iii) rapid uptake (McKay, 1996). When the adsorption process occurs, it will continue until it achieves an equilibrium state, which depends on physical and chemical characteristics, concentration of adsorbate, temperature and subsequent interaction among adsorbates (McKay, 1996; Chaudhary et al., 2003). While some of the solute adsorbs to adsorbent, others will be desorbed and these processes repeat continuously in the equilibrium system. This phenomenon is called "seesaw behaviour"; an important characteristic of an equilibrium system (Sincero and Sincero, 2003).

#### 2.4.2.1 Equilibrium

Adsorption equilibrium is described by an equation relating the amount of solute adsorbed onto the adsorbent; the equilibrium concentration of the solute in solution at a given constant temperature is used to evaluate the effectiveness of an adsorption system (Sincero and Sincero , 2003; Chaudhary et al., 2003). Equilibrium adsorption is a most informative and important test for estimating adsorption capacity of an adsorbent including the amount of adsorbed substances in a given condition (concentration and temperature) and the competition among absorbable components in the case of multi-component adsorption (Suzuki, 1990). After the contact of adsorbents with fluid, the adsorption process starts to occur and reaches an equilibrium stage in which the rate of adsorption and desorption are equal. It is usually characterized by two parameters: solute concentration in the adsorbent (q) and final solute concentration (Ce). In order to describe and predict the adsorption process, many mathematical models have been put forward relating q and Ce. Among the different adsorption isotherms models,

Freundlich and Langmuir isotherms are the most popular because they are simple and can fit well to experimental data in most cases.

The Langmuir isotherm is usually used to illustrate the homogeneous adsorption in water. Langmuir adsorption is established based on four assumptions:

(i) adsorption happens in definite localized surface sites;

(ii) each site bind with only one molecule;

(iii) all identical sorption sites in adsorbent surfaces are energetically uniform and

(iv) no interaction occurs among adsorbed molecules (Cooney, 1999; McKay, 1996).

The rate of adsorption is assumed proportional to equilibrium concentration ($C_e$) and the fraction of the surface vacant ($1-\theta$) and can be mathematically expressed as the following equation:

$$\text{Rate of adsorption} = kC_e(1-\theta) \qquad \text{(Eq. 2.1)}$$

*where* $\theta$ is the fraction of the surface covered and k is a constant

While the Langmuir equation is quite suitable for adsorption of gas to solids, the Freundlich model is more appropriated to liquid-solid adsorption. Freundlich isotherm describes heterogeneous surface adsorption. Different to Langmuir isotherm, the Freundlich implies that the energy distribution for adsorptive sites is an exponential type which is close to the real situation. Thus the rate of adsorption/desorption changes with the strength of energy. The Freundlich isotherm is more accurate than the Langmuir isotherm but it does not satisfy Henry's law at low surface coverage (McKay, 1996). The Freundlich isotherm is usually expressed by Equation 2.2 with the assumption that energy distribution for adsorption sites is exponential.

$$q = KC_e^{1/n} \qquad \text{(Eq. 2.2)}$$

where *n* is a constant (usually greater than 1).

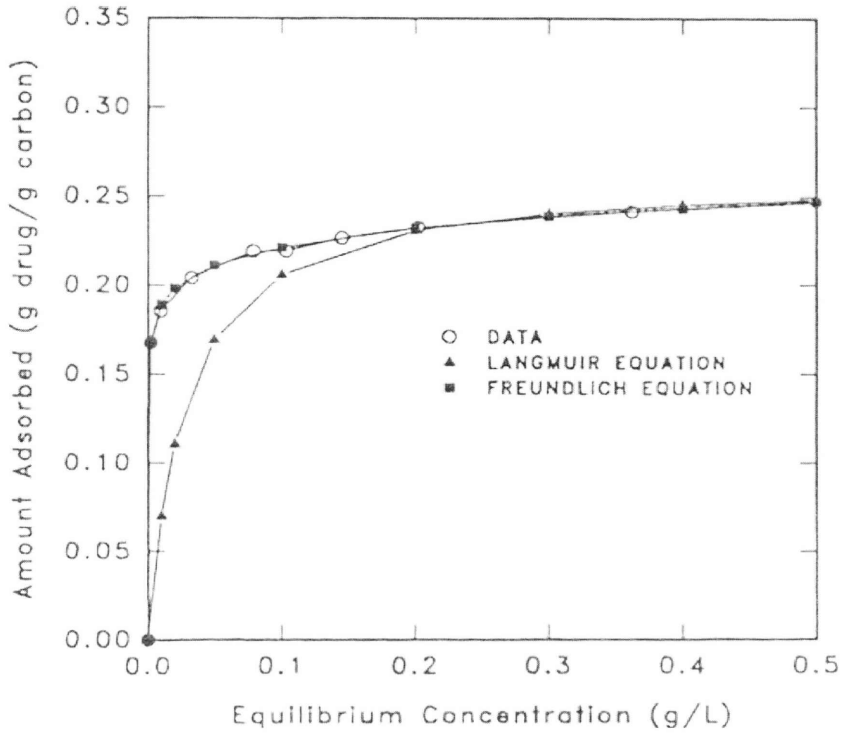


Figure 2.1 Comparison between Langmuir and Freundlich isotherms (Cooney, 1999)

Even though the Freundlich and other models are quite successful in describing and predicting adsorption behaviours of adsorbents, they require a complex process to estimate required parameters. Sips is another model that combines both the Langmuir and Freundlich model. The equilibrium curve obtained from Sips is similar to the Freundlich pattern if the initial solute concentration is low, while the Sips curve follows a Langmuir pattern for the higher solute concentration (Chaudhary et al. 2003). The Sips isotherm is expressed by Equation 2.3.

$$q = \frac{q_m C^{\frac{1}{n}}}{1 + K_S C^{\frac{1}{n}}} \qquad \text{(Eq. 2.3)}$$

where,

$C$ is equilibrium organic concentration (mg/L)

$q_m$ is saturation amount of organic adsorbed (mg/g)

$K_S$ is the Monod half velocity coefficient

Another isotherm model for multi-component systems is association theory. Talu and Meunier (1996) developed this theory to depict adsorption of water to hydrophobic surfaces based on adsorption thermodynamics. This theory is based on the characteristics of adsorption system including chemical equilibria, equation of state (EOS) and phase equilibrium (Chaudhary et al., 2003). Assumptions for association theory are (i) adsorption happens on active sites of GAC surface, (ii) dissolved organic matter forms clusters around active sites and (iii) adsorptive sites are limited by the shape and size of microspores. The association theory can describe the adsorption system by using only three parameters: saturation amount of organic matter adsorbed ($q_m$), reaction constant (K) and adsorption constant (H). The mathematical equation of association theory is shown in Equation 2.4:

$$C_e = \frac{H\psi . \exp\left(\frac{\psi}{q_m}\right)}{1 + K\ \psi} \qquad \text{(Eq. 2.4)}$$

where,

*Ce* is the equilibrium organic concentration (mg/L)

*H* is Henry law's constant

*L* is reaction constant

$q_m$ is saturation amount of organic adsorbed (mg/g)

$\psi$ is organic concentration spreading parameter

### 2.4.2.2 Kinetics of adsorption

Kinetics of adsorption is another key parameter in the estimation of the adsorption capacity of a system. The kinetic data is described by the effluent concentration as a function of time with the "concentration decay curve" (McKay, 1996). Process during kinetic adsorption is described as the diffusion of solute particles through the liquid to

reach and attach to adsorptive sites and then travel further into the particle to find vacant sites. The two main resistances to mass transfer are the external resistance in liquid phase and internal resistance in solid phase; but in practice solid phase resistance is usually greater than liquid phase resistance (Cooney, 1999). Batch adsorption kinetics are usually more suitable for GAC than PAC because PAC quickly reaches equilibrium while GAC takes a longer time or cannot attain equilibrium. The kinetics of adsorption influences the shape of adsorption profile. In slow kinetics, a breakthrough profile will be flat shape while fast kinetic adsorption results in a steep profile (Clark and Boutin, 2001).

Tien (1994) and Suzuki (1990) developed the linear driving force approximation (LDFA) model to describe adsorption kinetics. They assumed that there is a linear proportion between the rate of adsorption and driving force which was caused by the different concentration between adsorbent surface and adsorbed phase.

#### 2.4.2.3 Fixed bed adsorption

Fixed bed adsorption describes behaviour of an adsorbent in conditions which are close to practice. The adsorbent is packed in the column and fluid passes through the bed. It is a complex process controlled by many factors such as axial dispersion, fluid–to–particle mass transfer, intra-particle diffusion, reversible adsorption. In modelling fixed bed adsorption, the following assumptions are usually applied (Chaudhary, 2003):

(i) adsorbents are homogeneous and uniform;

(ii) column is uniform in adsorbent packing and influent distribution;

(iii) all mechanisms in the column are isothermal;

(iv) the flow through the column is axial;

(v) mass transfer rate coefficient is constant;

(vi) the flow rate and influent concentration are constant.

It was found that result of equilibrium and kinetic adsorption strongly influences the prediction curve of fixed bed adsorption because many parameters used in fixed bed adsorption are obtained from them. Therefore, it is important to obtain accurate parameters from equilibrium adsorption in modelling fixed bed adsorption.

### 2.4.3 Biological attachment processes

The attachment of microorganisms onto the surface of filter media to form a biofilm is a complex process. It has been studied by many methods such as scanning confocal laser microscopy, microbalance applications, microelectrode analysis, high resolution video microscopy, atomic force microscopy and scanning electron microscopy (Percival et al., 2000). There are several elements that take part in microbial attachment to a surface in which the strength of the attachment relies on environmental conditions, type of micro organisms, surface properties and fluid characteristics.

The attachment of microorganisms to the surface of bedding materials can be divided into five steps: development a surface-conditioning film, transportation of cells to a surface, adhesion, surface colonization and detachment (Percival et al., 2000). Figure 2.2 describes the attachment of microorganisms to the surface of supported media.

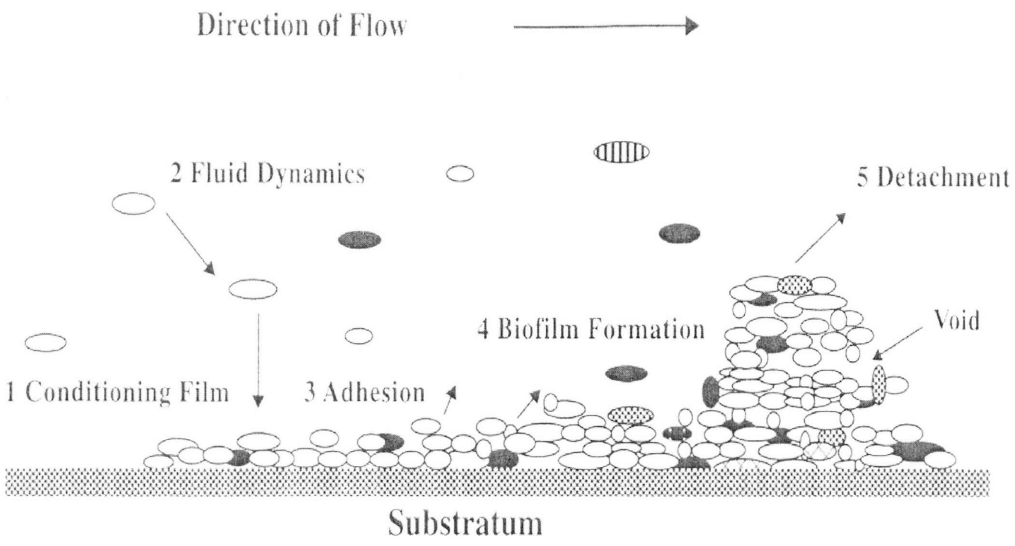


Figure 2.2 Schematic of biofilm formation (Percival et al., 2000).

*Surface-Conditioning Film*

When a clean surface is exposed to a bathing fluid in the first 15 minutes, it becomes conditioned with nutrients by the transport of organics and some microbial cells. Detailed information about this process is still unclear but in general, conditioning film is acknowledged as chaotic and dynamic. Surface-conditioning film contains glycoproteins, proteins and humic substances with the thickness ranging from 30 -80 nm. These components can react with surface appendages of some bacterial species in latter stages. It was supposed that conditioning film acts as controlling factors to adjust the amount of bacterial attachment to surfaces.

In biological adsorption process, the roles of surface-conditioning film include: modifying physico-chemical properties of substratum; acting as concentrated nutrient source; suppression of release of toxic metal ions; adsorption and detoxification of dissolved inhibitory substances and supply of required metal trace elements (Percival et al., 2000).

*Transport of cells to the surface*

Microorganisms in liquid reach and attach to surface of media by mass transport, thermal effects (Brown ion motion, molecular diffusion) and gravity effects (settling and sedimentation). Diffusive transport is a slow process occuring randomly among small bacteria and interfaces and which can be observed under the microscope. Diffusive transport is significant for the sedimentation of bacteria due to gravity in inactive condition while convective transports occur in flowing liquids. In this stage, some motile bacteria reach to a surface by active movement, which can happen by chance, or a response to any concentration gradient (Van Loosdrecht et al., 1990).

Mass transport is strongly influenced by fluid mixing (laminar or turbulent flow) and water flow rate (Percival et al., 2000). While laminar flow is slow and smooth with a little mixing, turbulent movement is random and chaotic which can increase the adhesion of microorganisms to surface, but the risk of detachment is also high due to the thick biofilm formed.

*Adhesion*

When bacteria reach the surface of media, initial adhesion will occurs by physicochemical processes. Bacteria still exhibit Brownian motion in this stage but they can be detached by shear or bacterial mobility. According to DLVO (named after Derjaguin, Landau, Verwey and Overbeek) theory, depending on the balance between electrostatic double layer forces and Van der Waals at different ionic strengths, initial adhesion can be reversible or irreversible (Loosdecht et al., 1990). Van der Waals attraction also relates to the effective size of bacteria but not including space occupied by appendages such as flagellum, pili, fimbriae and exopolysaccharides. The appearance of those appendages will form bridges that increase the effective distance (Percival et al, 2000). Electrostatic interactions with ionic and hydrogen bonding are not very strong individually but they can form a firm attachment when it appears in a large amount. These bonds will be stronger if the surface of the medium is positively charged because most microorganisms' surfaces are negatively charged (Cohen, 2000).

Busscher and Weerkamp (1987) suggested a three-point hypothesis where the distance between cells and surface can decide the kinds of interaction between them. Van der Waals attraction exists at distances greater than 50nm; within the distance of 10-20nm, both Van der Waals and electrostatic interaction occur and create reversible and irreversible adhesion; Van der Waals, electrostatic and specific interaction produce irreversible binding and form exopolysaccharides.

Firm attachment happens after bacteria deposit on the media and strong bonds between reactive groups on the surfaces of bacteria and media are formed. During this process, the occurrence of polysaccharides is necessary for the development of biofilm. Chemical and physical properties of the supported media have great influence to the adhesion of microbes into the surface. Normally, surface with high degree of hydrophobicity and roughness, carrying divalent cation (such as $Ca^{+}$ and $Mg^{+}$) will promote the attachment of bacteria to surface (Wuertz et al., 2003).

### *Surface Colonization*

The next step of microorganism attachment is surface colonization. The attached bacteria start to consume nutrients from water to grow and form colonies on the surface of supported media. Monolayer film is resulted from the development of bacteria which only attach to surface but not to other cells. If cells reversibly adhere to the surface and onto each other, exchange between attached and suspended organisms can occur. Biofilm is formed by irreversibly adhered organisms having strong link to media surface and other bacteria. (Loosdecht et al., 1990). When biofilm is formed, the proximity among immobilised microorganisms in biofilm becomes relatively close (Percival et al., 2000).

### *Detachment*

Detachment of biofilm is a common phenomenon, which always happens during biofilm formation process. Biofilm detachment occurs through the participation of different processes. Detachment of biofilm by abrasion, erosion, sloughing, occurs when there is shear stress, lack of nutrient and oxygen in the biological filter. Abrasion and erosion lead to the removal of small groups of cells from biofilm while sloughing results in the detachment of relatively large fraction of biofilm. Porosity and roughness of the biofilm supporting surface will play an important role to protect biofilm from the hydrodynamic shear and abrasion (Wuertz et al., 2003). Besides, human intervention can lead to detachment process. Predator razing is also another factor that causes biofilm detachment with the involvement of protozoa, snails and worms (Percival et al, 2000). The detachment process has a significant impairment to the distribution of microorganisms within biofilm and its structure. However, on the other hand, detachment removes dead microbes and creates free sites for new organisms attachment, thus microorganisms can quickly be replaced to retain the stability of the microorganism community and their activity (Percival et al., 2000; Wuertz et al., 2003).

## 2.5 Factors influencing biological filtration

### 2.5.1 General

Biological filtration is a highly complex system that involves activities of microorganisms immobilised on supporting media. Thus the filter media and factors related to the development of microorganisms will influence the performance of biological filters. While filter media aid the attachment process of microorganisms, the growth of microbiological community in the biofilter is influenced by influent characteristics (such as nutrients, toxics, pH) and temperature. In addition, operational conditions such as backwashing techniques, empty bed contact time, etc. will also affect the effectiveness of biological filtration in water treatment.

### 2.5.2 Filter media

Filter media plays an important role in designing biofilter because it affects the effectiveness of treatment and the cost implications in water and wastewater treatment. Porosity and surface roughness, adsorption capacity and surface chemistry are parameters which are concerned in evaluating a filter media (Dussert and Tramposch, 1996).

Dependent on adsorption capacity, filter media are divided into adsorptive media and non adsorptive media. Sand and anthracite are non adsorptive media which have been widely used in conventional biofilter for a long time. It is recorded that sand filter is quite effective in removing Cryptosporidium otocysts and Giardia cysts from water (Graham et al., 1996). Activated carbon was widely used as adsorbent for purifying purpose a long time ago but till the $18^{th}$ century, it was not studied in detail (McKay, 1996). Activated carbon, that is in particle form as GAC and in powder form as PAC, is used for various purposes such as in water and air treatment, solvent recovery, decolorizing, etc. They can remove phenolic compounds, benzene, heavy metals and industry solvents. In comparison with sand and anthracite, biofilter using GAC as supported filter media has better DOC, AOC removal efficiencies in pseudo steady state and shorter acclimation during starting up and after backwashing (Dussert and Tramposch, 1996; Uhl and Gimbel, 1996).

The influence of particle size of media on the effectiveness of biological filters has been reported by many authors. An investigation conducted by Van der Hoek et al. (1996) in Amsterdam Water Supply showed that a small grain size of sand (0.19-0.35mm) biofilter produced better quality of water than a big grain size (0.25-0.84mm) sand filter in terms of removing bacteria and turbidity, but the small grain size biofilter had shorter filter run length. Daifullah et al. (2004) also showed that activated carbon with finer particle size has higher capacity in removing humic acids than the larger particles of activated carbon while macroporous GAC has better capacity in supporting the growth microorganisms (Dussert and Tramposch, 1996).

McKay (1996) reviewed different kinds of adsorbent used in commercial and laboratory situations including peat, lignite, molecular sieves, silica gel, activated alumina, wood meal, bagasse pith and chitin. Peat and lignite are inexpensive, easy accessible and have high adsorptive capacity with trace and toxic metals. Lignite has strong affinity for basic dyes and can increase the organic removal rate when treated by calcium. Synthetic adsorbents like molecular sieves, silica gel are used for adsorption in both domestic and industrial sites. Molecular sieves are zeolite metal alumino – silicates, which have exact pore size and good performance in a wide range of temperatures. Silica gel is hard, white, glassy and highly porous. Silica gel is used to dehydrate air and gas. It can adsorb toluene and xylene quite well. Others adsorbents such as activated alumina and chitin, are specific to remove a certain type of pollutant. Activated alumina is used as a desiccant to adsorb hydrogen chloride while wood meal is good for basic dyes adsorption. Chitin is usually used to adsorb metal cation: copper, zinc, cadmium, and pesticide (McKay 1996).

Recently, several researchers investigated sponge as a biofilter media in sewage treatment. In their researches, sponge was designed as down flow hanging sponge cubes units with the characteristics of high removal rate of COD, BOD and suspended solids but low cost and easy to maintain (Machdar et al., 1997; Agrawal et al., 1997).

The combination of different kinds of biological filter media to take advantages of their strengths has been investigated by many researchers. LeChevallier et al. (1992) who studied GAC-sand and anthracite-sand filter which are called GAC sandwich at laboratory and pilot scale showed that GAC-sand filters were more effective in removing AOC, TOC and aldehyde than anthracite-sand filters. In addition, a GAC-

sand filter can also recover biological activities faster than anthracite-sand filter after a period of not being in operation. Bauer et al. (1996) succeeded in operating a full scale GAC sandwich biofilter (sand and GAC). This filter was successful in removing organics, DBPs, micro-pollutants and pesticides from Thames river water, which supplied water for 6 million customers in London.

### 2.5.3 Empty bed contact time (EBCT) and hydraulic loading rate

Empty bed contact time (EBCT) refers to the amount of time that water is in contact with particles (GAC, anthracite, sand, etc.) when influent passes through the filter bed. There are many studies emphasizing the importance of contact time on the effectiveness of biofilters. It is considered as the most crucial parameter for BDOC removal. Most of studies showed that the increase of EBCT results in a better removal rate of organic matters and DBPs (Huck et al., 1994; Servais et al., 1989; Wang et al, 1996). LeChevallier et al (1992), through their study on a GAC-sand filter following ozonation, observed an increase of TOC removal from 29% to 33% and 51.2% when EBCT was increased from 5 to 10 and 20 minutes respectively. Similarly, Krasner et al. (1992) also found that the increase of EBCT led to a higher rate of BDOC and AOC reduction. Carlson and Amy (1998) found that filters at EBCT of around 2-3 minutes could remove up to 90% of DOC. EBCT around 6-12 minutes was found to be the optimal contact time for TOC removal (cited in Clark and Boutin, 2001) while Dussert and Stone (1994) stated that efficient AOC removal rate can be obtained in less than 5 minutes contact time and pesticides about 15-20 minutes (Binnie et al., 2002).

Although higher EBCT can increase the organic removal in biofiltration, the increase of removal rate of organic matters is not proportional to the increase of EBCT (Huck et al., 1994). It was reported that the increase of EBCT in the range less than 25 minutes can enhance the removal of BDOC but at higher contact time, the BDOC reduction was marginal (Krasner et al., 1992). Urfer et al. (1997) suggested that EBCT should be decided based on the purpose of water treatment. For example, longer contact time is more suitable for the purpose of removing chlorination by-product precursors and reducing chlorine demand than removing ozonation by - products and AOC. Carlson and Amy (1998) observed a considerable ozone by-product removal rate with an EBCT as low as 1 minute while DOC removal started steadily at 2 minutes of EBCT in the

anthracite biofilter. In practice, EBCT can be adjusted by changing the filter bed depth or hydraulic loading rate.

In contrast, Hozalski (1996) reported that TOC removal is not a function of EBCT in the range 4-20 minutes of contact time. The above finding was based on the condition of temperature at $22.5^0$C and ozone dosage 2-4 mg/mg TOC. He stated that there were several other factors, which contributed to TOC removal. They included water temperature, ozone dosage, loading rate, range of EBCT, backwashing and type of filter media.

Hydraulic loading rate (HLR) is directly related to EBCT. Carlson et al. (1996) stated that DOC removal was insensitive to the hydraulic loading rate in the pseudo state of the biofilter. The DOC removal rate was equivalent at the low and acclimatized loading rate but less at high HLR ($17.5mhr^{-1}$). A study on slow sand filter by Bernardo and Rivera (1996) showed that higher filtration velocity resulted in lower filter run time. Wang et al. (1996) during their study on the behaviour of NOM in sand filter observed that at a given EBCT, organics removal was not affected by the change of hydraulic loading rate. Carlson and Amy (1998) found that the biomass profile in a biofilter is a function of HLR. The initial biomass concentration in biofilter was greater and penetrated deeper at high HLR. However, they did not record any effect of HLR on BDOC removal rate.

### 2.5.4 Influent characteristics

Influent is the source of nutrients providing the growth of the microbiological community, thus the components and concentrations of feeding influent have direct effect on biofilters. Vahala et al. (1998) investigated the influence of nutrients on biofilters by adding phosphorous and a mixture of inorganic nutrients into feeding influent. Their results showed that there was no statistically significant change on organic removal (TOC and $UV_{254nm}$ absorbance) and the amount of attached biomass onto media (which was analysed by adenosine triphosphate (ATP)). On the other hand, the number of bacteria that were measured by HPC increased in filter effluent due to the increase in nutrients. The researchers suggested that the addition of phosphorous stimulated the growth of bacteria in general, but bacteria could not attach onto the filter

media and then released to effluent. Another study on sulphate reduction in sulphur detrinification processes using biological filter reactors showed that high TOC and nitrogen removal rate can be obtained if the influent ratio of C/N was 2.2 and S/N 0.7 (Ikemoto and Komori, 2003).

In other research, Daifullah et al. (2004) reported that the increase in initial humic acids concentration led to the rise of the humic acids uptake in biofilters. Huck et al. (1994) in their modeling study of biofilter found a proportional relationship between the removal rate of BOM and precursors of THMs disinfection by-products with influent concentration.

In contrast, other authors observed a constant removal rate when the influent concentration changed (Urfer, 1998). Loosdrecht et al. (1990) explained that although concentration of influent increases, biofilm formation depends on the chemical potential of reactants and not on the concentration.

### 2.5.5 Backwash

After a long term operation, biofilters will be clogged because of the accumulation of biological and non biological particles in the pores. This problem can be solved by applying backwash techniques that will detach particles and excess biomass. Backwash can be done by using water and/or air to remove excess particles attached onto the filter media. There is a concern that biomass might be removed during backwash thus microbiological activities in biofilter can be impaired. However, many researches proved that backwash do not have any adverse effect on the performance of biological filter as long as the attached biomass removal during backwashing does not exceed 60% (Chaudhary, 2003, Hozalski et al.,1999; Carlson et al., 1996).

Ahmad et al. (1998) had assessed different backwash strategies including air scour, chlorinated water wash and high rate of water flow. The results showed that filters which were backwashed "at collapse pulsing air scour and then by water at 25% bed expansion" can reach a higher peak in the ripening state in comparison with the filters which were backwashed by water only. A filter that was non air-scoured backwashed also had higher initial head loss than an air-scoured backwashed filter. By measuring the heterotrophic plate count (HPC), Ahmad et al. (1998) also confirmed that backwash

by water was less strong than air scour backwash; thus in the case of backwashing with water alone, biofilm removal will be limited. However, the biological accumulation was found to be similar in both air scour and non air scour backwashed biofilters.

In addition, backwashing the biofilter with chlorinated water was studied to determine the effect of chlorine backwash to biofilter performance. Ahmad et al. (1998) observed a difference in organic removal between chlorine and non-chlorine backwashed biofilters, especially in the initial state, but this effect was not observed in GAC biofilters. In terms of biological activity, microbial count in the filter that was non-chlorine backwashed was considerably higher than the chlorine backwashed filter but the effect of chlorine to biofilter performance was short-term and only happened in the initial stage of filter operation.

In another study, Liu et al. (2001) recorded a longer time to achieve pseudo steady state in chlorine backwash filters than chloramine backwash filters. Backwashing with chloramine at a dosage of 0.25 mg/L did not show any effect on BOM removal in biofilter at any temperature. Liu et al (2001) also concluded that the effect of chlorine backwash to biofilter performance is related to chlorine dosage, temperature, filter media and BOM components. In their study, chlorine backwash had a stronger effect on anthracite biofilter at low temperature while GAC biofilter tolerated chlorine at any temperature. The presence of chlorine also decreased the glyoxal removal rate from 55% to only 11% in the biofilter. Krasner et al. (1992) stated that bed expansion of 30-40% in a GAC biofilter and about 25% in an anthracite biofilter was required to avoid the formation of "mud ball".

### 2.5.6 Temperature

Temperature is a significant factor that impacts the adsorption process as well as the growth of microbial community. Daifullah (2004) from his study concluded that adsorption is an endothermic process in which the adsorption capacity of carbon is better at higher temperature due to acceleration of slow adsorption steps or the creation of new active adsorption sites. Many studies show that not only biofilter acclimation time but also removal efficiency were affected by temperature.

Many authors agreed that higher temperature can result in better performance of biological filtration. Krasner et al. (1993) showed that the time biofilters needed to reach the steady state to remove glyoxal in 20-25$^0$C was shorter than at around 10$^0$C. Similarly, longer time to reach a steady state and a decreased BOM removal from 55% to about 1% were the result of a temperature decrease from above 20$^0$C to 3$^0$C (Hozalski et al., 1999). This conclusion was also supported by Liu et al. (2001). They observed a higher BOM removal in pseudo steady state at 20$^0$C than in 5$^0$C.

Clark and Boutin (2001) found that there was no difference in DOC removal from the biofilter at 20$^0$C and 35$^0$C while the rate of DOC elimination was considerably lower at 5$^0$C in treating river water. Krasner et al. (1993), Huck et al (1994) and Wang et al. (1995) got similar results that AOC removal by biofilter was slightly different at temperatures higher than 15$^0$C but was significant lower at low temperatures. At low temperatures (< 5$^0$C), the accumulation of biomass in the biofilter was severely inhibited that led to poor performance of biological filtration. When non-chlorine backwashing was employed, the difference in BOM removal at 20$^0$C and 5$^0$C was insignificant in pseudo steady state (Liu et al., 2001). The effect of temperature on biofilter performance was only observed clearly under unfavourable conditions such as chlorinated water backwash, anthracite media and raw water with refractory BOM components.

### 2.5.7 Others

During the operation of biofilters, the presence of oxidants in filter influent may cause some effect on the activity of the biofilter. Chlorine can deteriorate the GAC structure in a GAC biofilter even though biofilm can still be formed. Hydrogen peroxide ($H_2O_2$) and free $Cl_2$ do not influence GAC biofilter because they decay rapidly. However, in anthracite-sand filters, ozone can inhibit the development of bacteria (Urfer et al., 1998). Many authors agreed that ozonation following by biofilter results in better organic matter removal rate than biofiltration only (Hozalski, 1996).

With a certain type of filter media and operational conditions, pH may affect the treatment efficiency of biofilter. At different pH, electrical charges of active groups on media surface and particles in filter influent are changed hence adsorption capacity is changed as well (Daifullah et al., 2004).

## 2.6 Microbiological community in biofilter

### 2.6. 1 General

For a long time, it was assumed that biofilm was one dimensional but now biofilms are known as highly heterogeneous structures (Wuertz, 2003). Many biofilms (schmutzdecke) are spatially non-uniform in terms of physical (density, porosity and diffusivity) and chemical properties. By using laser scan scanning microscopy and nuclear magnetic resonance, biofilm properties are revealed with columnar and cluster shapes that allow water flows deep inside the whole biofilm. This structure of biofilm leads to a wide variety of microbiological communities and their distribution (Wuertz, 2003). The surface of biofilms also is not smooth which can affect the hydrodynamics and promote biomass detachment. Wuertz et al. (1996) divided biofilm into external and internal zone in order to study mass transport dynamic in biofilm. External refers to space between biofilm surface and the bulk solution which is characterized by convective and diffusive mass transport. An 'internal' zone is located between substratum and biofilm surface with convective and dispersive mass transport. Biofilms are usually characterized by thickness, density, and their composition. These parameters are crucial in modelling biological fixed film.

### 2.6.2 Biofilm thickness

Biofilm thickness depends on the volume of adhered organisms, ranging widely from 0.07 to 4 mm. However, it does not mean thicker biofilm has better effectiveness in treating water. The whole biofilm thickness does not have the same capacity in consuming substrates and is classified into total and active film thickness. A thick and uncontrolled biofilm does not lead to a higher substrates removal rate than thinner biofilm if its active film thickness is not thicker. Active film thickness was assumed as the result of transport limitations inside the biofilm. If there is a condition of very thin biofilm, high electron donors and acceptors concentration and high rate of transport, active film thickness can extend to the whole biofilm (Grady, 1983). Activities of organisms are proportional to the quantity of fixed biomass; they increase with the

thickness of active depth (Lazarova and Manem, 1995). After the attachment of microorganisms, biofilm will continuously grow until the biomass loss by decay and attrition happens or sloughing occurs but it rarely exceeds 1000μm. The rate of biofilm growth was balanced by the attrition in a low substrates condition but in rich nutrient environment, biofilm grows and accumulates faster than the detachment rate by shear stress and environmental conditions (Grady, 1983). Hydraulic flow rates, bedding materials and treatment designs can influence the thickness of the biofilm. Under conditions of rapid flow rate and smooth surface media, the growth of microorganisms are limited and form a thinner biofilm in comparison to biofilm developed in porous media (Cohen, 2001).

### 2.6.3 Biofilm density

For a long time, it was assumed that biofilm density is constant and independent with thickness but later researches showed that bacteria density increased with depth into the biofilm thickness (Grady, 1983; Lazarova and Manem, 1995). Biofilm density achieves the maximum value in the active depth. It could be the result of the higher accumulation of aerobic bacteria in active thickness and the lysis of organisms in anaerobic condition in the lower layer (Grady, 1983).

### 2.6.4 Biofilm composition

The majority of components of biofilm are microorganisms and exopolymers (or extracellular matrix). Other cell wall components such as peptidoglycan, lipopolysaccharides and lipids are essential elements for biofilm formation. Proteins usually account for about 10 -15% of the total biomass (Allison, 1998; Lazarova and Manen, 1995).

There is a limitation on the study of microbial composition in biofilm. Microorganisms' diversity in biofilm is extremely large with the presence of bacteria, fungi and yeast, protozoa, virus and invertebrates. A study by Ducan (1988) in slow sand filter found that the top of a 10mm surface area contained 0.1kg organic carbon, $10^3$ bacteria and $10^7$ protozoa per square meter area (Rachwal et al., 1996) but the total cell volume

accounts for only a small part in the composition of biofilm. Microbiological population in biological processes can be divided into two main groups. Generally, the most important groups of microbes in biological processes are prokaryotic (single-cellular organisms) groups in which bacteria account the largest proportion. Another group of microorganisms taking part in biological processes is eukaryote (multicellular organism) which includes fungi, protozoa, protifiers and algae, etc. (Tchobanoglous and Burton, 1991) Biological population is usually measured by microbiological methods like counting total, adenosine triphosphate (ATP) determination, electron transport system activity, fluorescence in situ hybridisation (FISH), ribonucleic acid (RNA) and deoxyribonucleic acid (DNA) measurement, etc. For estimating bacterial activities, the most conventional technique was substrates removal rate such as TOC or DOC analysis (Lazarova and Manem, 1995).

When studying microbial growth with GAC, Wilcox et al. (1983) found a wide range of microorganisms in biofilm. They found that the predominant microbes were oxidative, saprophytic chemogranotrophs which include gram negative, aerobic, non-fermentative bacilli (e.g. Pseudomonas, Chromobacterium, etc) and gram positive, aerobic, spore formers (Bacillus spp). Nitrogen fixing bacteria, actinomycetes were also found in the GAC biofilter. Wuertz et al. (2003) stated that among microorganisms of biofilm in the appropriate environment may have communication to others to maximize nutrient transport within biofilm as well as to control biofilm formation and detachment.

The exopolymers or extracellular polysaccharides (EPS), which are defined as materials removed from microorganisms without disrupting the cells, play a crucial role in biofilm formation. EPS are responsible for biofilm morphology, architecture, coherence, physicochemical properties and biochemical activities (cited in Wuertz et al., 2003). The EPS in the biofilm structure can be up to 50- 90% of enveloping matrix polymers. They comprise carbohydrates, protein, nucleic acids, lipids and humic acids. They also may contain low molecular weigh substances such as acetyl, pyruvyl groups and inorganics (Wuertz et al., 2003).

EPS are important not only in the attachment process of microorganisms to the supported media surface but also in the formation of colonies among bacteria. Also, EPS protects microbes from dehydration and toxic substances (Wuertz et al., 2003). In biofilm structure, EPS in gel-like form acts as "a mechanically stable factor" to

immobilize and accumulate microorganism on the surface in an oligotrophic environment. One of the important functions of EPS is taking part in building the structure of biofilm. It determines a three dimensional biofilm structure by providing polysaccharides cross links. The presence of enzymes such as polysaccharidases, proteases, lipases, esterases, etc. in EPS and cell surface help biofilm convert macromolecules in the environment into low molecular weight nutrients that can be digested by cells. However, these enzymes may degrade polysaccharides and cause biofilm detachment. Another role of EPS is protection of biofilm from non specific and specific host immunity (phagocytosis, antibody response, free radical generation) and biocides (antibiotics and disinfectants). It is assumed that EPS matrix may prevent biocides from reaching the microbial cells inside biofilm by diffusion limitation or chemical interaction with biocides molecules. In addition, EPS can accumulate organic nutrients and adsorb potential toxic xenobiotics (Wuertz et al., 2003).

### 2.6.5 Microbiological growth in biological processes

#### 2.6.5.1 Kinetics of microbiological growth

The microbiological population plays a crucial role in determining the success of biofilter. They convert organic matters and other substances in wastewater into gases and cell tissue. For their proper development, they require an energy source (light or chemical oxidation reaction), carbon source to reproduce new cells and inorganic nutrients such as nitrogen, phosphorous, sulphur, potassium, calcium and magnesium. Among them, organic nutrient is the most important source and is considered as the growth factor for bacterial cells synthesis (Tchobanoglous and Burton, 1991). Besides, environmental conditions also have influence on it such as temperature, pH oxygen, ultraviolet radiation, etc. Grady et al. (1999) summarized the metabolism of microorganisms in the following equation in which they indicated that the source of carbon and nutrients is the most important factors and has a close relation to the growth of organisms:

Carbon source + energy source + electron acceptor + nutrients → biomass + $CO_2$ + reduced acceptor + end products. (Eq. 2.5)

The relation between bacteria growth and nutrients is also expressed by the Monod equation (Eq. 2.6):

$$\mu = \mu_m \frac{S}{K_S + S} \qquad \text{(Eq. 2.6)}$$

where,

$\mu$ is specific growth rate

$\mu_m$ is the maximum growth rate

$S$ is concentration of nutrient in solution

$K_S$ is half saturation constant

Many authors applied this equation in modelling organism growth kinetics even though the Monod equation describes the growth of a single strain of bacteria in single nutrient media and ignores the energy required to maintain cellular machinery when they are not dividing. Figure 2.3 illustrates the relation between bacterial growth and nutrient.

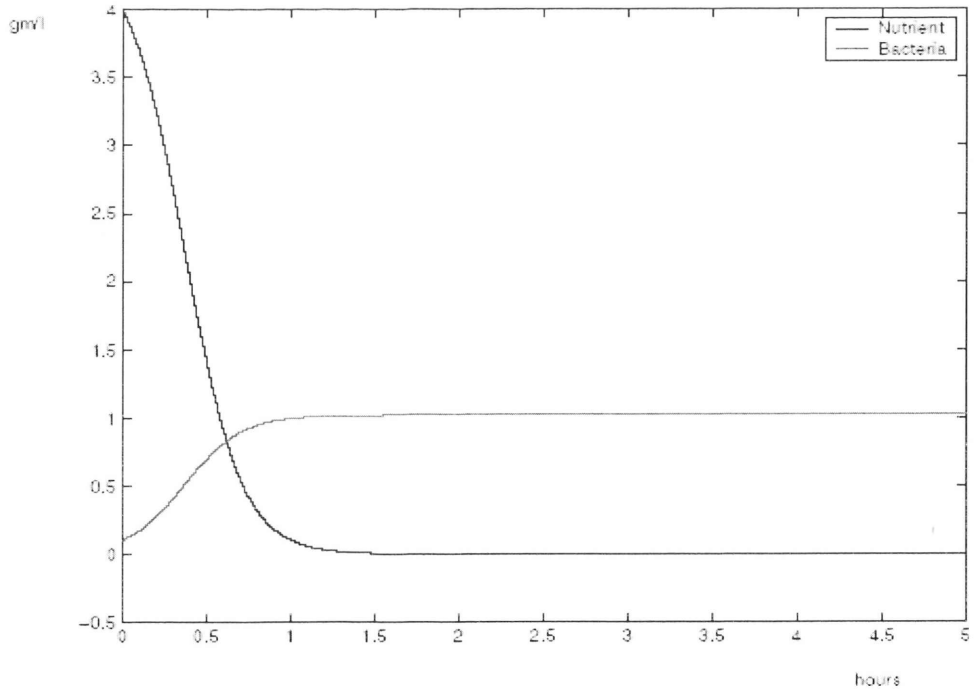


Figure 2.3 A sample of relation of bacteria growth and nutrient concentration in a batch test (Smith, 2005)

Bacteria can reproduce by binary fission in which a cell grows and then is divided into two. Time required for bacteria to complete fission is different, from few minutes to many days, depended on the types of bacteria and environmental conditions. The growth of bacteria in batch condition can be plotted as the logarithm of cell number versus the incubation time. Generally, the growth pattern of bacteria population in a batch culture can be divided into 4 phases as shown in Figure 2.4 (Tchobanoglous and Burton, 1991).

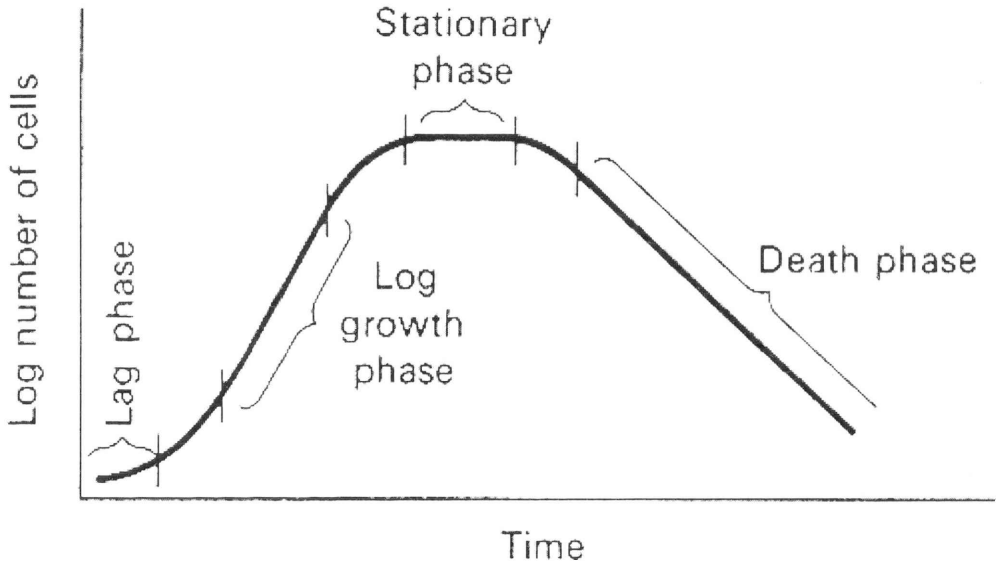


Figure 2.4 Typical bacterial growth pattern (Tchobanoglous and Burton, 1991).

(i) lag phase is the initial stage in which microbes acclimate to their environment and start reproduction;

(ii) log phase (or exponential growth phase) characterized by the highest reproduction rate of organisms due to available of nutrient source;

(iii) stationary phase in which the microbial population is kept constant because of the limited of essential nutrients or oxygen (for aerobic bacteria), and the balance between growth and death rate;

(iv) death phase, happening when the death rate of bacteria exceeds the growth rate due to the lack of nutrients and the presence of toxics released during bacterial metabolism in the environment.

In a biofilter, microorganisms pass through the lag and log phase in the acclimation stage. They are maintained in the stationary phase during biological filtration by receiving nutrients and oxygen in feeding water. Adjusting operational conditions provided the optimum conditions for microbial development that determines the success of the biofilter.

### 2.6.5.2 Biological filtration modeling

There were a few researchers trying to predict the behaviour of biofilter and the breakthrough curve by mathematic model. Both physical and biological parameters are employed in order to model biological filtration. However, those parameters especially biological parameters are difficult to get. Thus it is hard to find well-fitted curves that can describe precisely biological filtration process. Based on the assumption that biofilm was in the steady state condition, Rittmann and McCarty (1980) developed a model of steady-state-biofilm kinetics for single substrates. The assumptions of this model are:

(i) rate of substrate diffusion and rate of substrate utilization were the same;

(ii) minimum bulk substrate concentration ($S_{min}$) was required for maintaining steady state of biofilm.

Fick's second law and the Monod equation were used to describe the mass transfer and microbiological kinetics. In 1987, Rittmann and McCarty developed a steady-state biofilm model of biofilm on activated carbon (BFAC) using parameters of film transfer, biodegradation, adsorption of a substrate and biofilm growth. This model described all mechanisms related to biofilm kinetics in the biofilter and can be applied to different kinds of bioreactors. However, the assumption that biofilm was homogeneous and biofilm density was constant in the whole biofilm did not reflect exactly the real condition.

Hozalski and Bouwer (2001) simulated non-steady state of biodegradable organic matter and biomass in biofilter. The model, called "BIOFILT", described the non-steady state condition of biofilter which was caused by biomass removal or shearing during periodical backwash. It was expressed by the following one-dimensional advection-dispersion equation:

$$\frac{\partial S}{\partial t} = D' \cdot \frac{\partial^2 S}{\partial x^2} - v \cdot \frac{\partial S}{\partial x} - \frac{kSX_{susp}}{K_S + S} - \frac{\alpha}{\varepsilon} J_f \qquad \text{(Eq. 2.6)}$$

where

$S$ is substrate concentration (mg/L)

$D'$ is the dispersion coefficient ($m^2/s$)

$v$ is fluid velocity (m/s),

$t$ is time (s)

$x$ is the distance along the biofilter length (m)

$k$ is the maximum rate of substrate utilization (mg/mg/s)

$K_s$ is the Monod half-velocity coefficient (mg/L)

$X_{susp}$ is the suspended cell concentration (mg/L)

$A$ is the specific surface area of the media

$J_f$ is the flux of substrate into the biofilm ($mg/m^2/s$)

$\varepsilon$ is the bed porosity

$\alpha$ is a constant

The BIOFILT model was quite successful in describing full scale biofilter in drinking water treatment. However, the model still needs to be developed more with knowledge about GAC adsorption mechanisms, kinetics of organic biodegradation, the mass transfer, biomass loss by backwashing and routine operations.

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# CHAPTER 3

# EXPERIMENTAL INVESTIGATION

## 3.1 Introduction

This chapter gives a detailed description of materials used and procedure followed for various experiments performed to achieve the objectives of the present study as outlined in Chapter 1. The experimental set-up, materials used and analysis methods adopted are discussed separately in this chapter.

## 3.2 Materials

### 3.2.1 Wastewaters and water used in the experiments and their characteristics

#### A. Synthetic wastewater

A synthetic wastewater was used to study the performance of the laboratory-scale biofilters. Synthetic wastewater is representative of secondary treated sewage effluent (Seo et al. (1996). Its component comprises many kinds of organic and inorganic compounds. As well as the easily biodegradable matters found in secondary treated sewage effluent, this synthetic mixture also contains some persistent organic compounds (less biodegradable), including humic acid, tannic acid, lignin and polysaccharides. The average total organic carbon (TOC) concentration of the synthetic wastewater was approximately 10 mg/L. The constituents of synthetic wastewater are shown in Table 3.1.

The molecular weight (MW) distribution of the mixed synthetic wastewater ranges from 290 to about 34100 daltons with the highest fraction at 940 – 1200 daltons. The averaged MW of the synthetic wastewater was approximately 29500 daltons.

Table 3.1 Constituents of synthetic wastewater used

| Compounds | Concentration (mg/L) |
|---|---|
| Beef extract | 1.8 |
| Peptone | 2.7 |
| Humic acid | 4.2 |
| Tannic acid | 4.2 |
| Sodium lignin sulfonate | 2.4 |
| Sodium lauryle sulphate | 0.94 |
| Acacia gum powder | 4.7 |
| Arabic acid | 5.0 |
| $(NH_4)_2SO_4$ | 7.1 |
| $K_2HPO_4$ | 7.0 |
| $NH_4HCO_3$ | 18.8 |
| $MgSO_4.3H_2O$ | 0.71 |

**B. River water**

A part of the experiments in this study was conducted with river water taken from Hawkesbury River belonging to Hawkesbury – Nepean Rivers system, Sydney. The river drains approximately 22,000 $m^2$ of the Sydney basin. It provides potable and non-potable water to Sydney's population, wastewater disposal, commercial fishing, recreation, and a habitat for aquatic flora and fauna (EPA-NSW, 2001)

River samples used in this study were taken from the Hawkesbury River in Richmond suburb, Sydney. The characteristics of river water are shown in Table 3.2. Some values of biological parameters were obtained from an investigation by NSW Department of Land and Water Conservation (Sonter et al., 2002).

Table 3. 2 Characteristics of river water used (Hawkesbury River)

| Parameter | Average value |
|---|---|
| Turbidity (NTU) | 4.8 |
| TOC (mg/L) | 6.0 |
| Total viable cells (CFU/mL) | $2.09 \times 10^6$ |
| Feacal coliforms (CFU/100mL) | 25.8 |
| Enterococci (CFU/100mL) | 47 |

### 3.2.2 Filter media

#### A. Granular Activated Carbon

In this study, granular activated carbon (GAC) was used. The physical properties of the GAC are shown in Table 3.3. The most important characteristics of GAC are their extremely large surface area (more than 1000 $m^2$/ g GAC) and high porosity that makes GAC have advantages in adsorbing substances and microorganisms in wastewater. GAC was washed with distilled water then dried at 103°C and desiccated prior to use.

Table 3.3 Physical properties of GAC (Calgon Carbon Corp., USA)

| Specification of the GAC | Estimated Value |
|---|---|
| Iodine number, mg /(g.min) | 800 |
| Maximum Ash content | 5 % |
| Maximum Moisture content | 5 % |
| Bulk density, $kg/m^3$ | 748 |
| BET surface area, $m^2/g$ | 1112 |
| Nominal size, m (80% finer than) | $3 \times 10^{-4}$ |
| Average pore diameter, Å | 26.14 |

**B. Anthracite**

In this study, anthracite used was manufactured from an Australian coal seam by James Cumming & Sons P/L, Australia. The anthracite produced by this manufacturer contains less carbon content than standard anthracite but still preserves all other features. The physical properties of anthracite used are presented in Table 3.4.

Table 3.4 Physical properties of Anthracite

| Specification of the Anthracite | Estimated Value |
|---|---|
| Uniformity Coefficient | 1.30 |
| Acid Solubility | 1% |
| Alkali Solubility | 1.5% |
| Hardness | |
| • Hardgrove Grindability Index | 50 |
| • Friability | 20% max for 15 minutes |
| Durability | Attrition loss < 0.35% per year |
| Specific Gravity | 1.45 |
| Bulk Density | 660 to 720 kg/m$^3$ |

**C. Sponge**

Synthetic sponge is a new material used in water and wastewater treatment as a filter media for biological filtration. The advantages of sponge are its light weight, high porous and simple in usage. Sponge used in this study was in the form of reticulate foam produced by Joyce manufacturer, Australia. Its properties are shown in Table 3.5.

Table 3.5 Properties of sponge (polyurethane foam)

| Specification | Value |
|---|---|
| Chemical composition | polyester-polyurethane |
| Density ($kg/m^3$) | 28 |
| Number of cells ($cells/inch^2$) | 80 |
| Tear resistance (N/m min) | 740 |
| Tensile strength (kPa/min) | 150 |

**D. Floating medium**

The media used in floating media system was polypropylene bead. Its diameter was 3.8mm with the density about $0.87 g/cm^3$.

## 3.3 Experimental set-up

In this study, biological filtration experiments were conducted using transparent acrylic filter columns. These columns have an internal diameter of 2 cm and length of 80-150cm with outlet pipes along and in the bottom of the column. Prepared filter media (GAC or others) was packed into the column up to the required depth. For sponge and floating media, a stainless steel mesh was placed on the top of the media bed to keep the media in the column. The experimental set-up is shown in Figures 3.1, 3.2 and 3.3. The columns were operated in down flow mode. Feeding water was pumped from a feeding water tank to the top of the columns and then passed through the filter bed. An overflow outlet was placed above the filter bed to keep a constant head. Effluent samples were collected at the bottom of the column or from the middle pipes along the column for analysis.

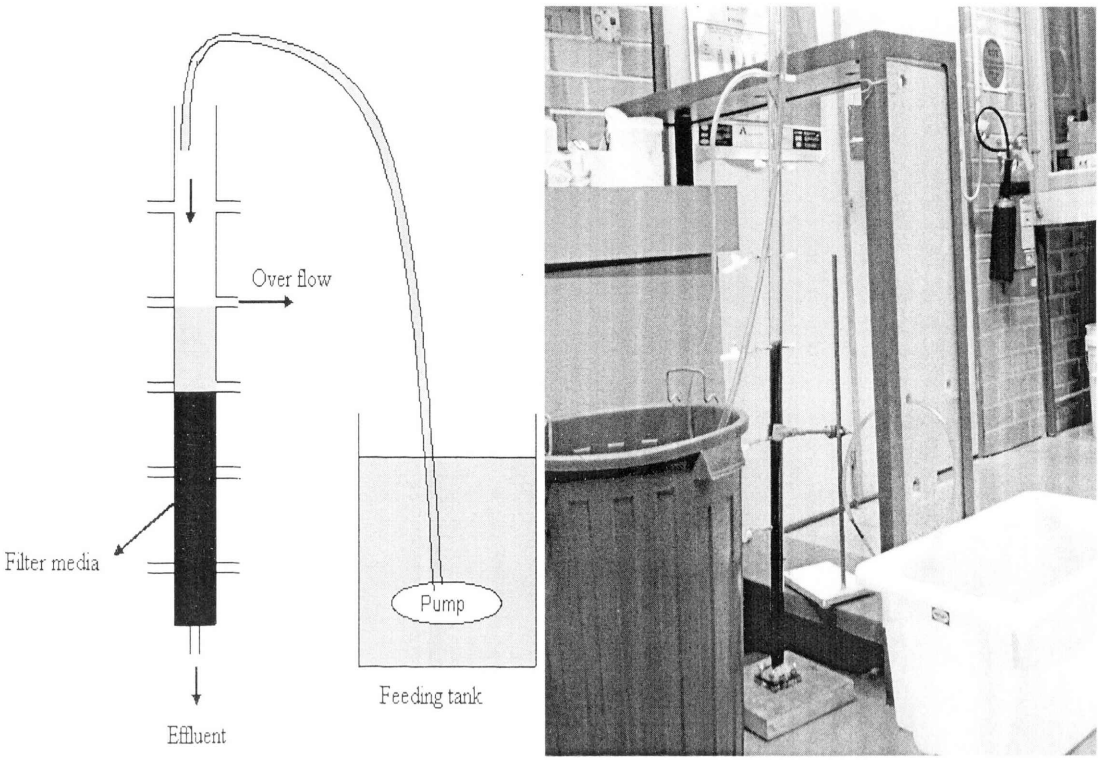


Figure 3.1 Schematic diagram of lab-scale GAC/anthracite biofilter experimental set up

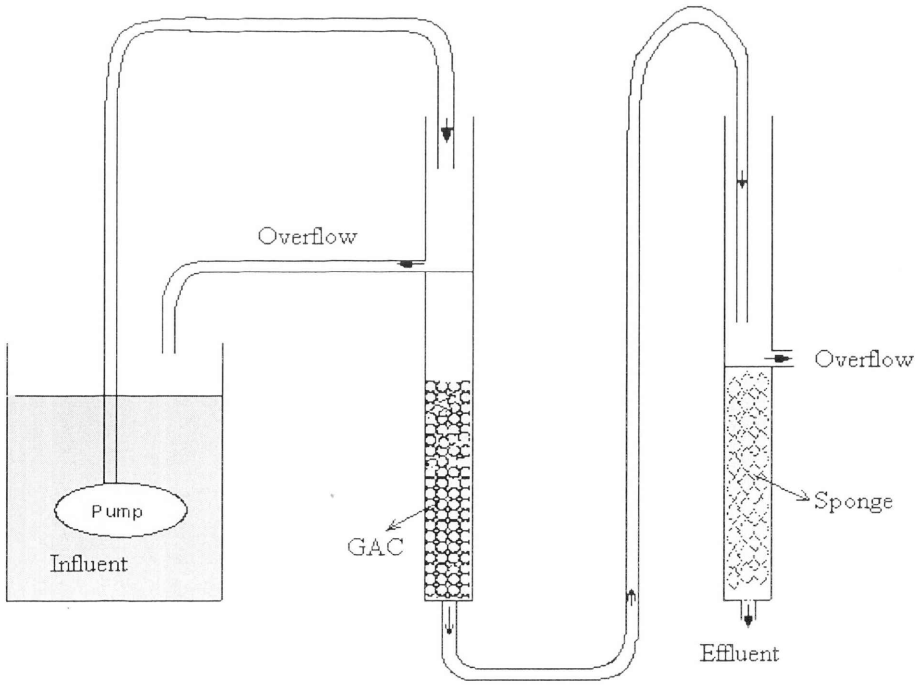


Figure 3.2 Schematic diagram of GAC-sponge biofilter experimental set up

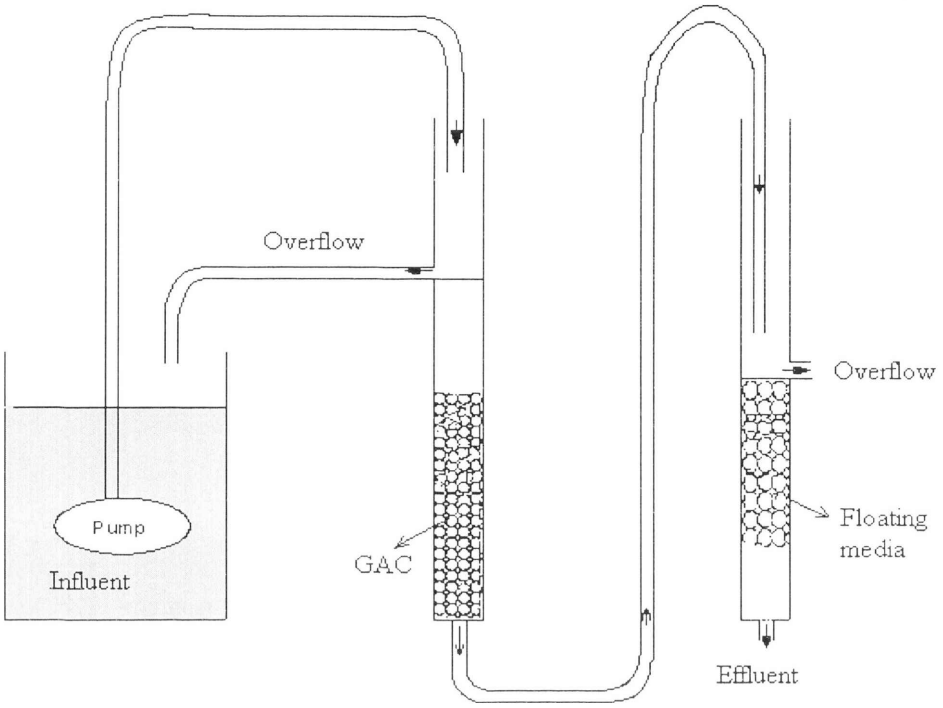


Figure 3.3 Experimental set-up of GAC-floating medium biofilter

During the biofilter operation, daily backwashing was applied to remove particles and excess biomass that can cause biofilter clogging. For GAC and anthracite filters, backwashing was conducted using tap water in the up-flow direction from the bottom of the column. During backwashing, the GAC/anthracite bed was expanded up to 60% for 2 minutes. Excess mass was removed from the column through overflow pipe and can be collected for analysis. For floating medium column, backwashing was conducted daily using up-flow air followed by down-flow water wash.

## 3.4 Analytical methods

### 3.4.1 Total organic carbon measurement

Total organic carbon (TOC) concentration of water was measured by using Multi N/C 2000 analyzer (Analytik Jena AG). The sample was oxidized into end products in a combustion tube at high temperatures (700 – 950$^0$C) by the following equations:

$$R + O_2 \longrightarrow CO_2 + H_2O \qquad \text{(Eq. 3.1)}$$

$$R\text{-}N + O_2 \longrightarrow NO + CO_2 + H_2O \qquad \text{(Eq. 3.2)}$$

$$R\text{-}Cl + O_2 \longrightarrow HCl + CO_2 + H_2O \qquad \text{(Eq. 3.3)}$$

where *R* is substance containing carbon

The amount of $CO_2$ is quantified by non-dispersive infrared gas reactor (NDIR) and calculated with a calibration function as total carbon (TC). Inorganic carbon is determined by reactions between sample and acid in a total inorganic carbon (TIC) reactor. TOC value (mg/L) is determined by Equation 3.4.

$$TOC = TC - TIC \qquad \text{(Eq. 3.4)}$$

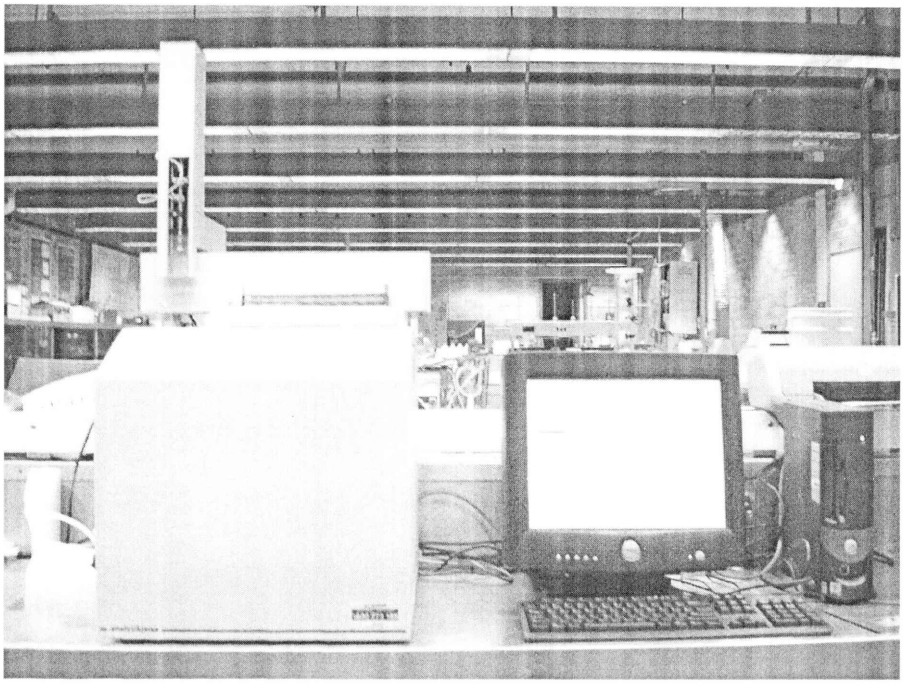

Figure 3.4 Multi N/C 2000 analyzer (Analytik Jena AG)

### 3.4.2 Adsorption analysis

In this study, the GAC adsorption capacity with synthetic wastewater (presented for secondary treated sewage effluent) was evaluated by equilibrium, kinetic and fixed bed adsorption. Equilibrium adsorption is described and compared with the Freundlich, Sips and Talu models. The linear driving force approximation (LDFA) model was used to evaluate kinetic and fixed bed adsorption of GAC.

#### A. Equilibrium adsorption experiments

Equilibrium adsorption experiment of GAC was conducted at room temperature ($25^0$C). Different doses of dry GAC ranging from 0.15 -3g/L were distributed into 250mL flasks containing 100mL of synthetic wastewater. The component of synthetic wastewater was demonstrated in Section 3.2.1. A flask of synthetic wastewater was also added as the control sample. All flasks were shaken continuously for 72 hours on a shaking table at speed 130 rpm. After 72h of shaking, samples of synthetic wastewater were taken from all flasks and filtered through 0.45 μm filter before analyzing TOC.

The adsorption capacity of GAC was determined as the following formation:

$$q = \frac{(C_0 - C_e)V}{M} \qquad \text{(Eq. 3.5)}$$

where

$C_0$ is initial TOC concentration of synthetic wastewater

$C_e$ is the final concentration of the solution (mg/L)

$V$ is the volume of solution (mL)

$M$ is the weight of used GAC (g).

**B. Kinetics adsorption experiments**

In the experiments on kinetics adsorption of GAC, clean and dry GAC was distributed into 2L beakers with synthetic wastewater at four concentrations (0.5, 1, 2, and 3 g GAC/L). The solution with GAC was mixed well using a mechanical stirrer at 130 rpm for 6 hours (Figure 3.5). During the experiments, samples of synthetic wastewater were taken from those beakers at different periods of time for TOC concentration.

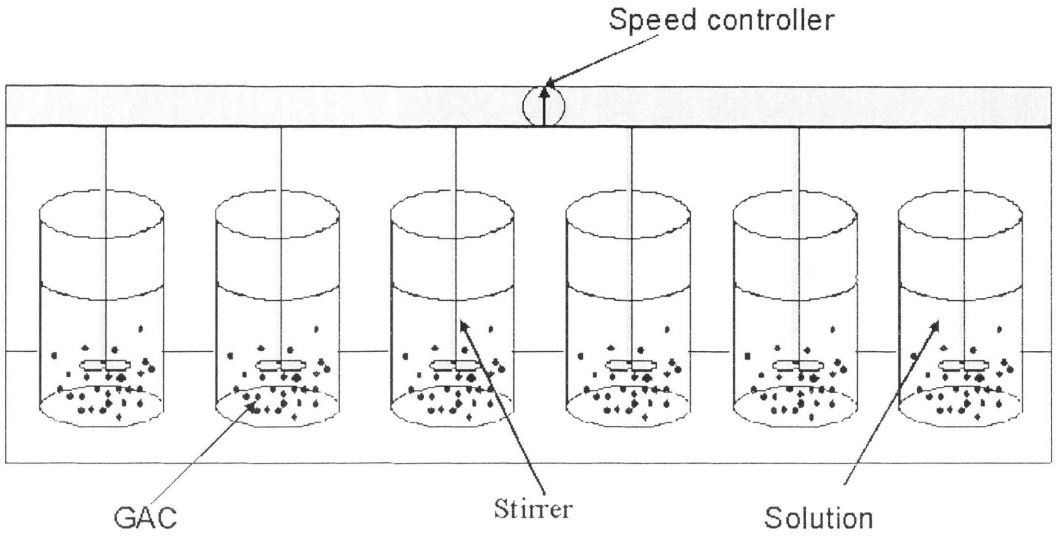


Figure 3.5 Schematic of the batch experiment using Jar test apparatus

**C. Fixed bed adsorption experiments**

In the experiment, 2cm - diameter columns were employed to study the adsorption of GAC used in a fixed bed column. Similar to GAC column set up described in Section 3.3, prepared GAC was packed into the columns at 5, 10, 15 and 30 cm bed depths with the weights of 7.5, 15.0, 22.5 and 45.0 g respectively. All columns were fed by synthetic wastewater in the down-flow direction at the filtration rate of 2 m/h. Effluent samples were collected at the bottom of the columns at different periods of time during 6 hours for TOC analysis.

The linear driving force approximation (LDFA) model was used to evaluate kinetic and fixed bed adsorption of GAC biofilter.

### 3.4.3 Biological measurement

#### A. Dry mass measurement

The quantity of biomass which was attached and grown on the surface of supported filter media during biological filtration processes was measured in terms of dry mass. Dry mass attached on GAC was measured in both batch and filtration (column) mode. In batch mode, the same amount of GAC (250mg) was placed into different beakers with synthetic wastewater. Wastewater in those beakers was replaced every two days to provide enough nutrients to facilitate biofilm growth onto the GAC surface. After a certain time of operation, the whole amount of GAC was taken out from a beaker for dry mass measurement. The GAC was dried in the oven at $103^0$C for 24h and desiccated to eliminate moisture prior weighing. Total adsorbed biomass was calculated as the different mass of filter media before and after contacting with synthetic wastewater.

Similarly, a series of GAC columns was set up under identical operational conditions such as GAC weight, bed depth, filtration rate and backwashing. The whole amount of GAC was also taken out of the column after operation for dry mass measurement as described above.

This method was modified from volatile suspended solids (VSS) measurement. To measure VSS, the sample is first filtered through a weighed glass-fiber filter paper. Filter paper with retained residue is dried and weighed at $103^0$C for at least 1 hour (m1). Sample is then burnt at $550^0$C for 20 minutes and weighed (m2). VSS is calculated as the following equation:

$$\text{VSS (mg/L)} = \frac{(m1 - m2)x1000}{V} \qquad \text{(Eq. 3.6)}$$

where $V$ is volume of sample (mL).

In this study, this method could not be directly applied because GAC was burnt when heated at $550^0$C. Therefore, dry mass can only be measured after being dried at $103^0$C. The dry mass data obtained by this measurement may not give very accurate information about biomass growth because there may also be other materials adhered onto the surface of the filter media in addition to the biomass. However, this method is simple, practical and it can give a reasonable estimation on the biomass growth in the biofilters.

**B. Biological oxygen consumption measurement**

Biological oxygen consumption rate (or oxygen uptake rate) gives an estimation of the active rate of aerobic microorganisms in a biological system. In this study, a biological oxygen monitor (Yellow Springs Instrument, YSI probe model 5300) was used to measure the dissolved oxygen consumption rate by microorganisms (Figure 3.6).

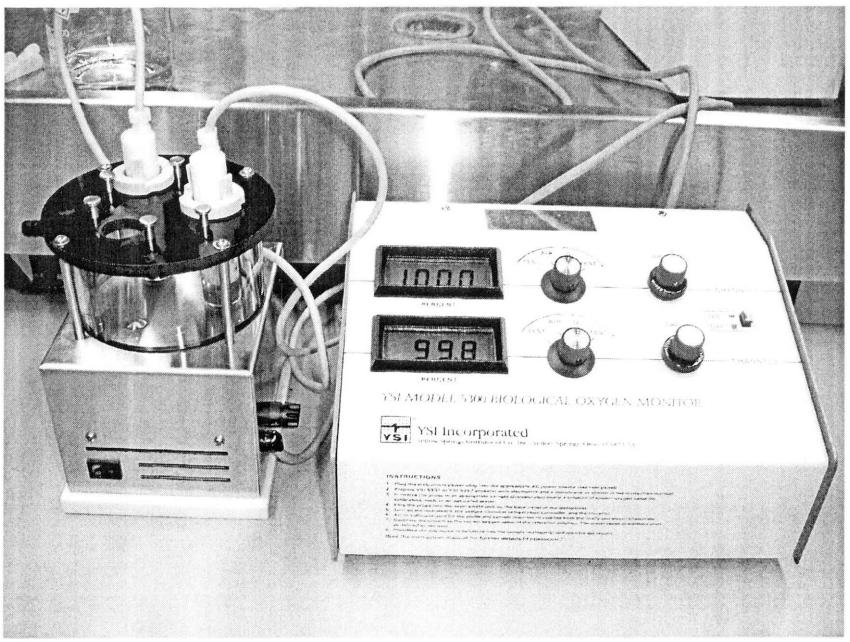


Figure 3.6 Biological oxygen monitor (Yellow Springs Instrument, YSI probe Model 5300)

Filter media with biomass were taken from the top layer of the biofilter for DO consumption measurement. About 5mg of GAC with attached biomass was taken from the biofilter and placed in closed chambers together with 3mL of biofilter feeding water. It is important that no air bubbles existed in the chambers and oxygen from air could not come in. The solution was mixed by magnetic stirrers during the measurement to ensure that dissolved oxygen was evenly distributed in the whole chamber and diffused into the electrode. Oxygen concentration in the sample was measured by a micro oxygen probe which is a polarographic system comprising of a platinum cathode, silver anode, KCl solution and a membrane. DO measurement was conducted in a constant temperature ($26^0$C) because temperature has a strong effect on membrane permeability and oxygen solubility in water.

The rate of oxygen uptake rate (%) was recorded every minute for 30 minutes. The monitor was calibrated prior to each measurement with air saturated solution in which dissolved oxygen content was set at 100%. Dissolved oxygen consumption was plotted as a function of time.

### 3.4.4 Microbiological analysis

#### A. Viable counts of bacteria

The amount of bacteria present in water samples and on GAC surface was analyzed by using the spread plate technique. This measurement was conducted in the microbiology PC2 laboratory, University of Western Sydney, Sydney, Hawkesbury campus. The steps in the measurement procedure follow the sequence below:

1. prepare and dry required number of nutrient agar plates;
2. design dilution series of samples as directed below:

    a) based on the initial load of influent samples, design appropriate dilutions.

    b) in doing so it is important to remember that:

i) a count of 25-250 colonies is needed on at least one spread plate;

ii) in order to be sure of a count, at least one dilution on each side of that expected to yield the appropriate colony count should be plated. A minimum of 3 dilutions should be plated for each dilution series.

3. prepare all dilutions in 0.1% bacteriological peptone water to maintain the viability of bacteria in the sample;

4. aseptically transfer 0.1mL of each selected dilution to Nutrient agar plate and disperse it uniformly throughout the plate using a sterile glass spreader;

5. incubate the plates at $37^{0}$ C for 24hrs;

6. examine the plates and count number of colony forming units of original sample using following formula:

   **Number of Colony forming Units (CFU)/mL or Gm = No of colonies on plate x dilution factor x 10**

7. incubate a control plate with no culture.

**B. Total Coliforms**

A total coliforms count was conducted using the spread plate technique. The procedure for total coliforms test is similar to the viable counts test presented in section 3.4.4.A. The difference between the two tests is the nutrient medium. MacConkey medium is used for the enumeration of total coliforms.

**C. Fecal coliforms**

Fecal coliforms content in water samples was detected by using the membrane filtration technique. The enumeration of the fecal process was conducted by the following steps:

1. place a sterile type HA white filter (grid side facing up) aseptically in sterile filter holder;
2. pour 50mL of sample to be filtered into a funnel of filtration unit;
3. apply the vacuum to start filtration;
4. rinse the funnel walls with at least 50mL of sterile phosphate buffer; if subsequent samples of water are to be tested this rinse will provide sufficient cleaning of the funnel between tests; this step will also improve the accuracy of the method by collecting microbes which adhere to the walls of the funnel;
5. a fresh filter paper will be required for each test;
6. remove the top section of the funnel and aseptically transfer the membrane filter to a sterile Petri dish containing M-FC medium; the membrane must be applied to the medium by placing the edge vertically on one side of the medium and then lowering it by a rolling action; this will avoid air being trapped between the medium and the membrane;
7. invert dish and incubate at $44.5^{o}$ C for 24hrs;
8. count number of colony forming units and calculate number of organisms per 100 mL sample (noting that a 50 mL sample was used);
9. make appropriate dilutions in 0.1% bacteriological peptone water if the colony count is out of 25-250 range.

**D. Microbial identification using BIOLOG system**

Microbes in the biofilter were identified by using a BIOLOG system (Biolog Inc. California, USA, 1993). The BIOLOG principle is based on the bacterial response to 95 different carbon sources. A positive result is indicated by a purple colour caused by the increase of bacterial respiration and reduction of tetrazolium dye. Bacterial strain is recognized by its typical 'breathe print" pattern. An automated plate reader will read this pattern and match it to patterns of species library to give the bacterial identification. (Dlamini, 1997)

To prepare for the Biolog test, bacteria were cultured in the Biolog Universal Growth Medium with 5% of sheep blood at $35^0$C for 12 hours. Cells were picked from agar plates and suspended in saline to reach the turbidity which allowed 55% transmittance. Suspended cells were then transferred to a micro plate and inoculated at $35^0$C for 4 hours. Microlog 3 software was used to interpret the visual reading form the plates. *Providencia stuartii* was used as a quality control strain of the test (Dlamini, 1997).

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# CHAPTER 4

# RESULTS AND DISCUSSION

## 4.1 GAC adsorption

The adsorption capacity of GAC affects not only substrates adsorption during the adsorption phase but also the accumulation of microorganisms for biological phase of biofilter. Therefore, estimation of adsorption capacity of GAC is necessary for the biofilter design and operation. In this study, adsorption equilibrium, kinetics and fixed bed were used for evaluating GAC adsorption. Mathematical modeling was also used for predicting GAC adsorption behavior.

### 4.1.1 Adsorption equilibrium

Adsorption equilibrium experiment of GAC with synthetic wastewater was conducted at room temperature ($25^0$C) for 72 hours. With the use of synthetic wastewater, the equilibrium adsorption of GAC was evaluated in a multi-component system. It was described and predicted by Freundlich, Sips and Talu isotherms. Equilibrium parameters obtained from the simulation are presented in Table 4.1. It can be seen that the Freundlich and Sips models gave the same value of constant n of 1.71 and residual of 1.57, and resulted in almost same equilibrium adsorption curves (Figure 4.1). This finding confirmed the conclusion of Kumar et al. (2004) that Sips (or Freundlich- Langmuir) model prediction follows the Freundlich curve when initial solute concentrations were not very high.

**Table 4.1 Equilibrium adsorption isotherm parameters**

| **Isotherm models** | | | | | |
|---|---|---|---|---|---|
| **Talu** | | **Sips** | | **Freundlich** | |
| Parameter | Value | Parameter | Value | Parameter | Value |
| Qm | $5.19 \times 10^{13}$ | qm | $3.85 \times 10^{9}$ | | |
| K | 0.27 | b | $9.16 \times 10^{-11}$ | k | 0.3529 |
| H | 2.71 | n | 1.71 | n | 1.71 |
| Residual | 1.04 | Residual | 1.57 | Residual | 1.57 |

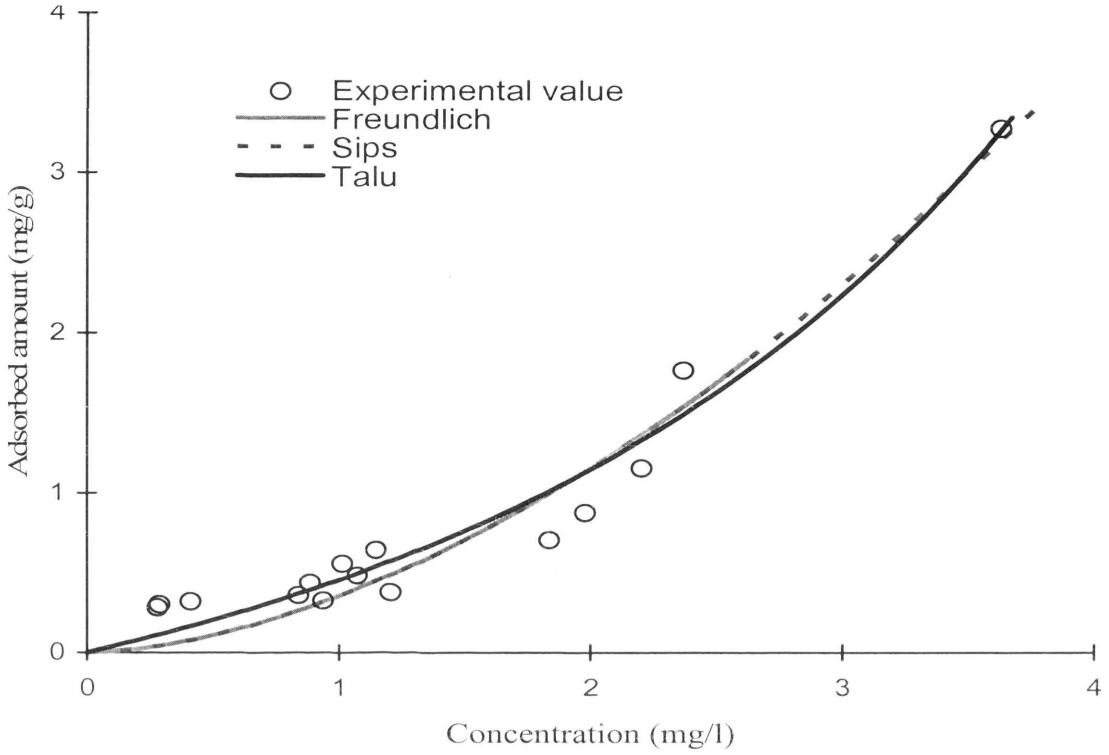



Figure 4.1 Adsorption equilibrium of synthetic wastewater by GAC with different isotherm models (contact time =72 hours, mixing rate = 130 rpm, temperature = $25^0$C)

Among the three simulation models, association theory (by Talu and Meunier) fitted well with experimental data with a residual of 1.04. It agrees with results reported by Chaudhary et al. (2003) that Freundlich cannot fully describe the adsorption in a multi-component system. The Talu association theory model was a well fitted model in describing GAC equilibrium adsorption. Chaudhary (2003) also found that the adsorption equilibrium depends on characteristics of wastewater such as initial concentration and inorganic content.

### 4.1.2 Adsorption kinetics

The saturation of GAC adsorption capacity with time was investigated in adsorption kinetics experiments. The adsorption kinetics of GAC were evaluated at four different concentrations (0.5, 1, 2 and 3g/L). The results of GAC adsorption kinetics are presented in Figure 4.2. As expected, higher GAC loading rate resulted in better organic adsorption due to the availability of adsorption sites on GAC surface. For all the four concentrations studied, organic matter in the synthetic wastewater was quickly adsorbed within the first 2 hours and then the organic adsorption rate by GAC remained constant. In other words, during the first two hours, the available sites on the GAC surface for adsorption was abundant which led to high rate of organic adsorption (more than 40% with all four concentration of GAC. When all the adsorption sites were occupied and the rate of adsorption and desorption was balanced, organic removal by adsorption was constant.

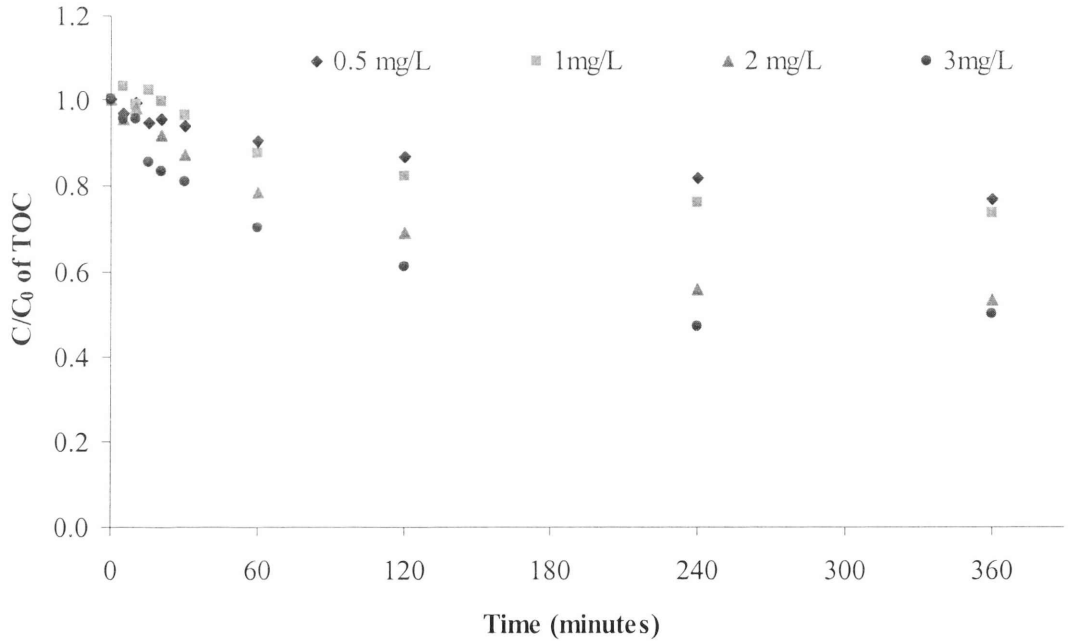



Figure 4.2 Adsorption kinetics of GAC with organic matter in synthetic wastewater (average TOC of synthetic wastewater = 10 mg/L, mixing rate = 130 rpm)

Kinetic adsorption of GAC with organic matter in multi-components systems can be described by the linear driving force approximation (LDFA) model (Tien, 1994). LDFA model is a simple model using lumped index parameter (TOC) but is adequate in describing the mass transfer rate (Chaudhary, 2003). It was assumed that the uptake rate of substances to adsorbents is linearly proportional to the driving force caused by the concentration difference between adsorbent surfaces and the solution. In adsorption kinetics, mass transfer and diffusion of adsorbate particles from the bulk liquid phase to the adsorbent surface determine the rate of adsorption. Values of film mass transfer coefficient ($K_f$, m/s) and diffusion coefficient ($D_s$, $m^2/s$) obtained from the LDFA model are presented in Table 4.2.

Table 4.2 Kinetics adsorption parameters of LDFA model for GAC adsorption

| TOC of influent concentration (mg/l) | GAC weight (g) | Film mass transfer coefficient $K_f$ (m/s) | Diffusion coefficient $D_s$ ($m^2/s$) |
|---|---|---|---|
| 10.73 | 0.5 | $2.99 \times 10^{-5}$ | $3.34 \times 10^{-13}$ |
| 9.51 | 1.0 | $0.80 \times 10^{-5}$ | $5.72 \times 10^{-13}$ |
| 9.51 | 2.0 | $1.01 \times 10^{-5}$ | $7.00 \times 10^{-13}$ |
| 9.51 | 3.0 | $1.40 \times 10^{-5}$ | $3.18 \times 10^{-13}$ |

For the same solute concentration, mixing rate, the value of $K_f$ was found to increase with the increase in GAC loading rate. This change led to a higher adsorption rate of GAC particle at its higher loading. This result agrees with the observation of Chaudhary et al. (2003) in which the film mass transfer coefficient also increased with an increase of GAC loading. They also found that the Freundlich constant was not significantly affected by the change of GAC loading rate in the range of less than 5g GAC/L.

With the $K_f$ and $D_s$ given in Table 4.2, the kinetic adsorption data of GAC with synthetic wastewater was well fitted to the LDFA model (Figure 4.3).

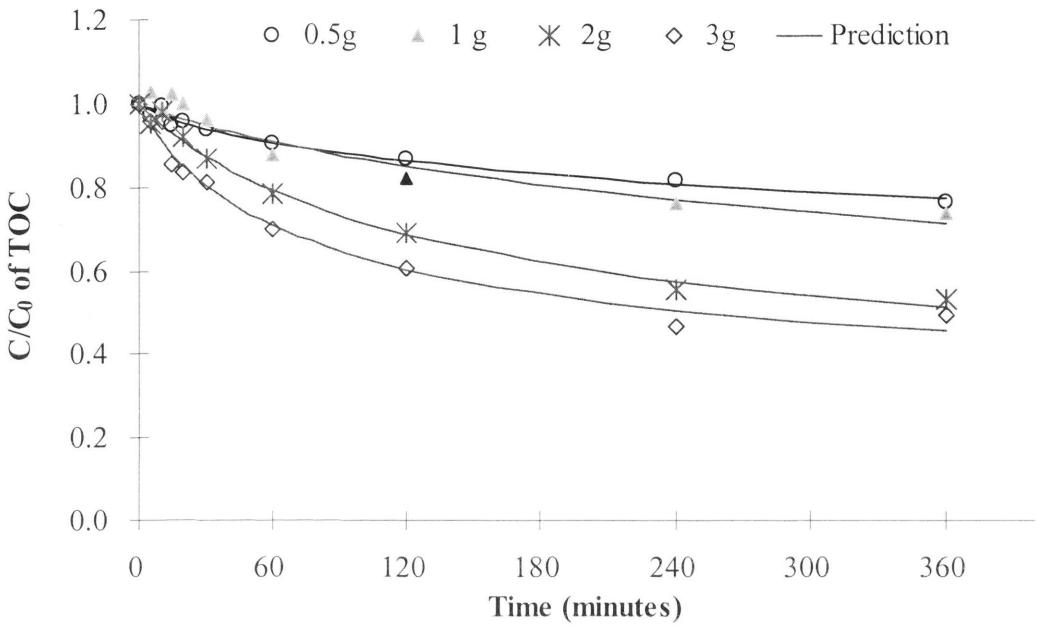



Figure 4.3 Prediction of adsorption kinetics of GAC in batch system with LDFA model (average TOC of synthetic wastewater = 10 mg/L, mixing rate = 130rpm)

### 4.1.3 Fixed bed adsorption column

The fixed bed adsorption reflects the behavior of adsorbents in conditions which are close to practice. Thus, it is strongly affected by design and operating conditions (Shim et al., 2004). In an adsorption of fixed bed column with predetermined values of filter media, bed depth and influent concentration, the external diffusion controls the mass transfer from bulk solution to the surface of adsorbent.

Physical parameters such as film mass transfer coefficient ($K_f$), diffusion coefficient ($D_s$) and axial dispersion coefficient ($D_L$) are essential to describe and predict adsorption behaviors of GAC in fixed bed adsorption. $K_f$ was estimated as a function of fluid velocity and adsorbent size. The $K_f$ value was obtained by minimizing the mean deviation between experimental and calculated values of the concentration decay curve. The diffusion coefficient was determined from the relation of fluid concentration and temperature; and $D_L$ was a function of molecular diffusion or turbulent mixing. Among these parameters, the

mass transfer coefficient has the greatest impact on the exit concentration and temperature breakthrough curve. It was found to depend on adsorbent characteristics and hydrodynamic (Shim et al., 2004). The value of $K_f$, $D_s$ and $D_L$ calculated from the fitting of experimental and predicted data are presented in Table 4.3

Table 4.3 Experimental conditions and physical parameters calculated for of GAC fixed bed adsorption.

| **Bed depth (cm)** / **Parameter (unit)** | 5 | 10 | 15 |
|---|---|---|---|
| Initial TOC concentration (mg/L) | 9.92 | 9.92 | 9.92 |
| Velocity (m/h) | 2 | 2 | 2 |
| Solid phase mass transfer ($K_S$ x$10^{-5}$) | 1.65 | 1.03 | 0.9 |
| Axial dispersion coefficient ($D_L$ x $10^{-7}$) | 4.03 | 0.97 | 1.08 |
| Film mass transfer coefficient ($K_f$ x $10^{-6}$) | 4.92 | 2.85 | 2.29 |
| Diffusion coefficient ($D_s$ x $10^{-13}$) | 6.19 | 3.88 | 3.40 |

Similar to adsorption kinetics, mass transfer rate controls the rate of adsorption. In this experiment, since no mixing device was used, this led to a lower value of the film mass transfer coefficient. The prediction curves for different bed depths are shown together with experimental data in Figure 4.4.

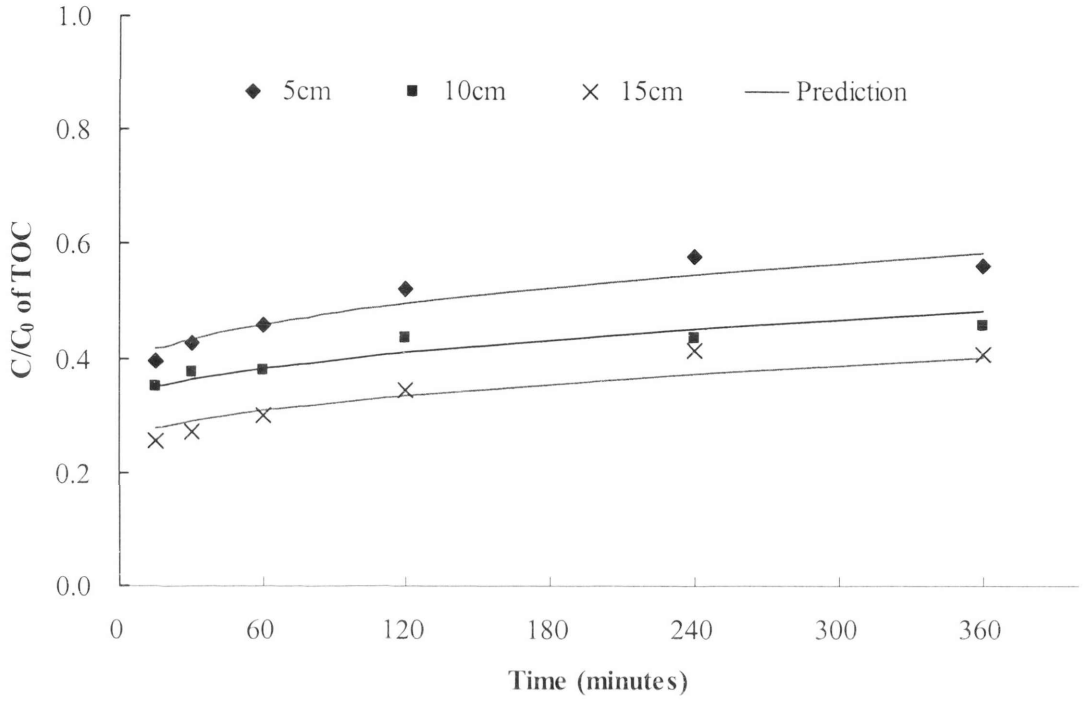



Figure 4.4 Adsorption kinetics results with GAC fixed bed adsorption column. (filtration rate = 2m/h, average TOC of influent = 9.92mg/L)

As can be seen in Figure 4.4, organic removal by GAC adsorption increased with the increase of the bed depth as expected. The adsorption decreased slightly with time during these short term experiments. The experimental data of the GAC fixed bed adsorption fitted well with the prediction curve of the LDFA model. The fixed bed adsorption of GAC in multi component system was also studied by Chaudhary (2003). The study showed that initial concentration of influent, bed depth and filtration rate can affect the GAC adsorption rate. Biodegradation of organic matter should be taken into account in the adsorption modeling.

In general, GAC has high adsorptive capacity with synthetic wastewater. A large proportion of organic matter was removed from synthetic wastewater by adsorption of GAC. In terms of mathematical modeling, GAC adsorption equilibrium predicted by the association theory model fitted best with the experimental data than Freundlich and Sips simulation. The

LDFA was successful in describing and predicting GAC adsorption behaviours in the multi-component system of the batch and fixed bed adsorption experiment with synthetic wastewater.

## 4.2 Long term performance of GAC biofilter

### 4.2.1 Long term performance of GAC biofilter with synthetic wastewater

In this study, a GAC biofilter was operated in the laboratory in order to evaluate its effectiveness in removing organic matter for a long period of operation (more than 6 weeks). Four acrylic columns were employed for GAC packed to different bed depths (5, 10, 15 and 30 cm). All four columns were fed with synthetic wastewater and operated at a low filtration rate of 2m/h and backwashed daily. The filtration rate was kept low as the GAC bed depth was shallow (less than 30cm) to shorten the acclimation time of biofilter.

The efficiency of GAC biofilters was evaluated in terms of TOC removal. As can be seen in Figure 4.5, the GAC biofilters led to consistent TOC removal for a long time of operation without regeneration of the activated carbon. Even after 42 days of continuous running, those biofilters could maintain an organic removal efficiency of 40-50% with 15cm bed depth and 60 -70% with a 30cm GAC bed depth. Even with only a 5cm bed depth of GAC, a TOC removal rate of 25% can be maintained for more than 40 days.

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 may have been lost during biofilter backwashing of the filter but the loss of biomass created possibly more available sites on the GAC surface for adsorption of micro-organisms and organics, and thus the impairment was balanced (Chaudhary, 2003).

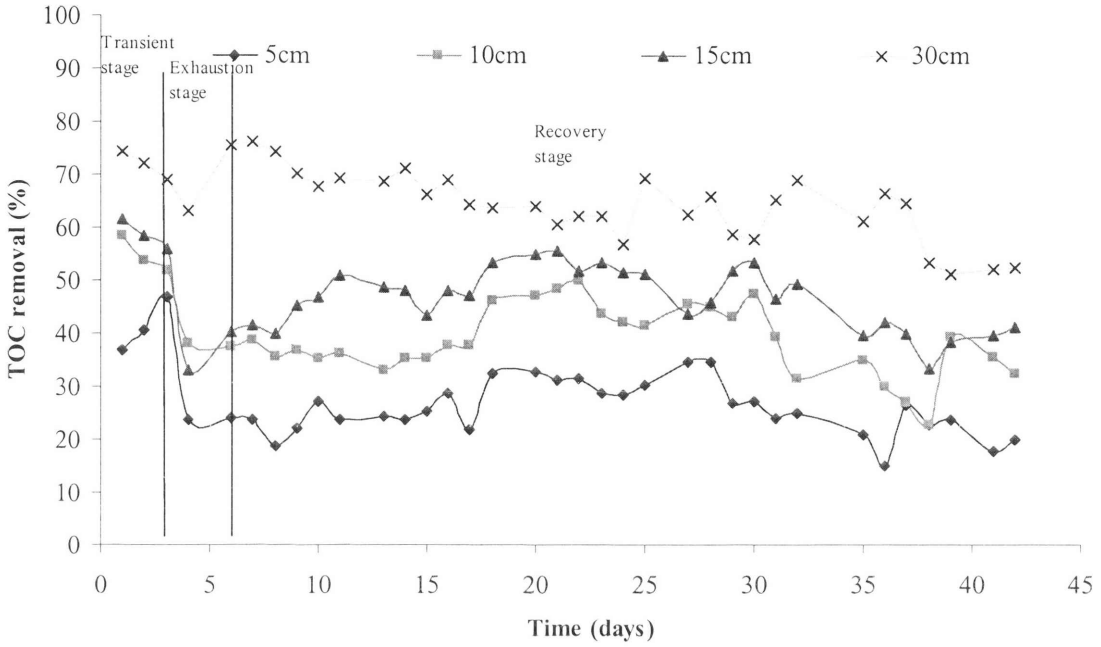



Figure 4.5 TOC removal efficiency of the GAC biofilter with different filter bed depths (filtration rate =2 m/h, column diameter = 2 cm, average influent TOC = 10mg/L)

As can be seen in Figure 4.5, TOC removal efficiency of biofilters varied with the change in GAC bed depths. As expected, the column with higher bed depth resulted in a better organic removal than the shallow bed depth. There was well over 60% of TOC removal with the 30cm - GAC bed depth, compared to only about 30% of TOC removal with the 5cm bed depth GAC column. It is because the deeper the GAC bed depth, the higher the surface area for adsorption of organic matter and attachment of microorganisms.

Additionally, in the same operational conditions of the biofilter, higher bed depth had longer empty bed contact time (EBCT) and resulted in better organic removal rate. In the longer EBCT column, substances in influent had more opportunities to be adsorbed and biodegraded by microorganisms grown in filter bed. This effect of filter bed depth on organic removal was exhibited not only during the biological phase of the biofilter but also in the adsorption phase (Chaudhary, 2003). Previous studies showed that biofilter can

achieve better organic, DBPs and pesticide removal at higher EBCT (Huck et al., 1994; Servais et al., 1989; Wang et al, 1996; LeChevallier, 1992).

The long term performance of biological filtration varied at different periods. It can be seen in Figure 4.5 that the biological filtration process comprised of three different stages. At the transient stage, TOC removal rate of biofilter was significantly high due to the high adsorption capacity of GAC. GAC adsorption can remove 40-75% organic matter from synthetic wastewater during this stage with 5-30 cm GAC bed depths. While GAC adsorption was gradually exhausted, microorganisms started to be retained on the media and grew up. This stage was observed through a reduction of TOC removal efficiency. When the microbiological community in the biofilter reached its equilibrium, organic removal efficiency increased again and then remained nearly constant for a long period of time. This pattern was observed more significantly in the GAC biofilter with shallow bed depth. In this short filter depth, the small amount of GAC reached its maximum capacity faster than other columns. Due to GAC adsorption was exhausted in a short time, microbial population has not fully developed to take the role of adsorption in removing organic matter from wastewater. This led to a clear difference between each stage of biofilter which is indicated by the difference at TOC removal rate.

### 4.2.2 Long term performance of GAC biofilter with river water

A long term performance of GAC biofilter was also conducted with river water. River water was taken from Hawkesbury River for this experiment. Characteristics of the river water were described in Section 3.2 B. A GAC biofilter of 2 cm - diameter and 15 cm - bed depth was used in this experiment. It was operated at a velocity of 2 m/h and daily backwashing. The results of this experiment are presented in Figure 4.6.

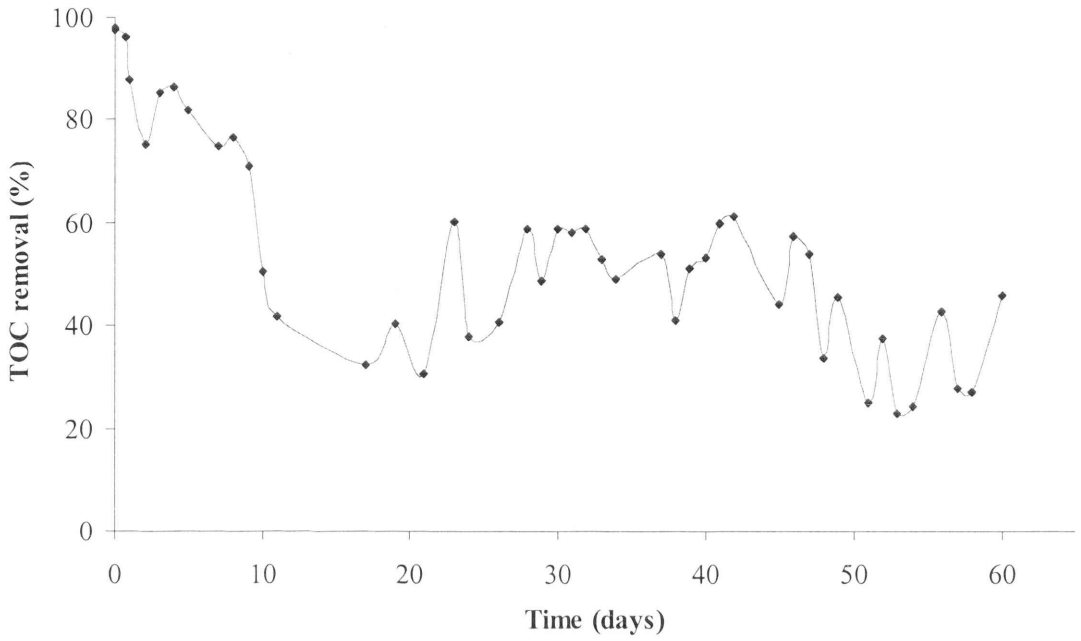



Figure 4.6 TOC removal by GAC biofilter with Hawkesbury river water (filtration rate= 2 m/h, bed depth =15 cm, average TOC of river water= 6 mg/L)

In the GAC biofilter for river water, the TOC removal pattern was similar to the biofilter performance with synthetic wastewater with three different stages. However, there were differences in each phase of biological adsorption processes between the GAC column fed by river water and the synthetic wastewater in the same experimental conditions (filtration rate = 2m/h, bed depth =15cm and daily backwashing). During the adsorption phase, a significantly high TOC removal rate over a rather long time (10days) was recorded in the column fed by river water (e.g. up to 80-95% in comparison to 60% TOC removal in synthetic wastewater feeding column). The acclimation time of the filter for river water was longer and took about 25 days. In addition, the drop of TOC value at the end of the adsorption stage for river water feeding column was less sharp than the synthetic wastewater feeding filter. The slightly lower organic content in river water (average TOC = 6 mg/L) was considered the reason for these differences. The low TOC value of river water also led to higher organic matter removal rate in the latter stage of GAC biofilter by biodegradation. It was suspected that the river water might contain more easily biodegradable substances than the synthetic wastewater. This GAC biofilter maintained a

consistent removal of TOC of about 55% up to 50 days. A marginal decrease of organic removal was recorded after 50 days running this column.

In general, GAC biofilter can be used as an effective treatment in removing organic matters from water and wastewater. The advantages of a GAC biofilter are consistent of TOC removal efficiency, long operational life, and simplicity in operation. In addition, no activated regeneration is required during biofilter operation.

## 4.3 Mathematical modeling of GAC biofilter

Biological filtration is a highly complex system which involves many physical and biological processes. Therefore, only few researchers succeed in describing and predicting biofilter by mathematic models. For GAC biofilter, adsorption and biodegradation are the two main phenomena occurring biofilter. Thus, biofilter modeling was built based on fundamentals of both adsorption and biodegradation mechanisms. Modeling parameters, especially biological figures were obtained from previous studies (Hozalski and Bouwer, 2001; Alonso et al., 1998) and experimental data. For estimating biological parameters, this model was based on the following assumptions (Shim et al., 2003):

- the adsorbent particles are spherical and uniform;
- biological activities only happened in the surface of adsorbent particles;
- adsorption is irreversible;
- the biofilm is thin, homogeneous in terms of thickness, porosity, composition and density; the biofilm can be modeled as a flat plate;
- the curvature effect of the GAC surface is not accounted in modeling;
- the increase of biofilm thickness is only due to the biomass growth;
- substrates are limiting factor of biological activities that was described by the Monod equation;
- intra-pellet diffusion can be described by the Glueckauf approximation;
- biofilm growth led to the change of specific surface area and bed porosity

Parameters estimated in this model were based on many previous studies and are summarized in Table 4.4 (Shim et al, 2003).

Table 4.4 Parameters and equations for modeling biological filtration process.

| Parameter | Equation | Initial and boundary conditions | Index |
|---|---|---|---|
| Transportation substrate in bulk liquid | $\frac{\partial C}{\partial t}=D_{ax}\cdot\frac{\partial^2 C}{\partial z^2}-u\cdot\frac{\partial C}{\partial z}-\gamma_{BIO}-\gamma_{ADS}$<br>$\gamma_{BIO}=k_{\max}\cdot\frac{C\cdot X_S}{K_S+C}$<br>$\gamma_{ADS}=(1-\varepsilon_b)\cdot\frac{3N}{4\pi a_p^3}$ | Initial condition:<br>$C=C_0$<br>Boundary condition:<br>z=0,<br>$D_{ax}\cdot\frac{dC}{dz}=-\nu\cdot\left(C\big\|_{z=0^-}-C\big\|_{z=0^+}\right)$<br>z=L,<br>$\frac{dC}{dz}=0$ | C: the liquid phase concentration.<br>$D_{ax}$ : the axial dispersion coefficient<br>*u*: the interstitial velocity<br>k*max*: the maximum rate of substrate utilization<br>$X_S$ : the suspended cell concentration<br>$K_S$ : the Monod half velocity coefficient<br>S: the concentration of the substrate in the biofilm<br>$\varepsilon_b$ : the bed porosity<br>N: the adsorbate uptake rate per pellet<br>$R_P$ : the pellet radius |
| Biomass suspended in the bulk liquid | $\frac{\partial X_S}{\partial t}=\left(Y\cdot\frac{k_{\max}\cdot C}{K_S+C}-K_d-\frac{\beta}{\theta\cdot\varepsilon_b}\right)\cdot X_S+\frac{1-\varepsilon_b}{\varepsilon_b}\cdot a_f\cdot X_f\cdot\sigma$ | Initial condition<br>$X_S=X_{S0}$<br>Boundary conditions<br>z=0<br>$X_S=X_{S0}$ | Y: the yield coefficient<br>$K_d$ : the decay constant<br>β: the filtration efficiency<br>θ: the empty bed contact time<br>$X_f$ : the cell density of biofilm<br>σ: the biofilm shear loss coefficient.<br>$a_f$: specific surface area |

| | | | |
|---|---|---|---|
| Biofilm diffusion and biodegradation (Andrews and Tien, 1981) | $\frac{\partial S}{\partial t} = D_f \cdot \frac{\partial^2 S}{\partial x^2} - X_f \cdot \frac{k_{\max} \cdot S}{K_S + S}$ | Initial condition: $S=S_0$<br>Boundary conditions<br>x=0,<br>$D_f \cdot \frac{\partial S}{\partial x} = \left(\frac{R_p}{3}\right).\rho_s.k_s.(q_s - \bar{q})$<br>$x=L_f$,<br>$D_f \cdot \frac{\partial S}{\partial x} = k_f.(C-S)$ | $D_f$: the molecular diffusivity within biofilm |
| Biofilm growth and decay | $\frac{dL_f}{dt} = \int_0^{L_f} \left( \frac{Y \cdot k_{\max} \cdot S}{K_S + S} - b_{tot} \right) \cdot dr$ | Initial condition<br>t=0, $L_f=L_{fo}$ | $L_f$: the biofilm thickness<br>$b_{tot}$ : the total biofilm loss coefficient. |
| Support-phase substrate balance | $\frac{\partial \bar{q}}{\partial t} = k_P \cdot (q_S - \bar{q})$ | | $\bar{q}$: the adsorbed-phase concentration<br>$q_S$ :the value of q at pellet surface<br>$K_P$ :the particle phase mass transfer coefficient |
| Bed Porosity and Specific Surface Area (Alonso et al. 1998) | Bed Porosity:<br>$\varepsilon_b = 1-(1-\varepsilon_{b0}).\left[\left(1+\frac{L_f}{R_p}\right)^3 - \frac{P_n}{4}.\left(\frac{L_f}{R_p}\right)^2.\left(2.\frac{L_f}{R_p}+3\right)\right]$<br>Specific surface area:<br>$a_f = \frac{3.(1-\varepsilon_{b0})}{2.R_p}.\left(1+\frac{L_f}{R_p}\right).\left[(2-P_n).\frac{L_f}{R_p}+2\right]$ | | $P_n$: the number of characteristic packing spheres. |

Tables 4.5 and 4.6 present the estimated value of physical and biological parameters, which were then used in the modeling GAC biofilter. The diffusion coefficient, film mass transfer coefficient and axial dispersion coefficient were obtained from the fitting of predicted data with the experimental values. Some of biological parameters (biofilm thickness, maximum growth rate, suspended cell concentration etc.) were obtained from previous studies of Hozalski and Bouwer (2001), and Alonso et al (1998). Not only biofilm formation process but also biofilm detachment was considered in describing biofilter. Parameters were taken into account in this model including the effect of the change of bed porosity and specific surface area due to the increase of biofilm thickness, biofilm decay and shear loss, biofilm thickness, mass transfer and backwashing. Chaudhary (2003) found that the model simulation was sensitive to biofilm thickness, yield, decay, shear loss coefficient, substrate utilization and molecular diffusivity.

Table 4.5 Physical parameters used for theoretical predictions in GAC biofilter

| Parameter | Value | |
|---|---|---|
| Bed depth (cm) | 5 | 10 |
| Velocity (m/h) | 2 | 2 |
| TOC of influent (mg/L) | 11.6 | 11.6 |
| Diffusion coefficient $D_S$ ($x10^{-13}$) | 5.212 | 5.212 |
| Film mass transfer coefficient $K_f$ ($x10^{-6}$) | 4.919 | 2.852 |
| Axial dispersion coefficient $D_L$ ($x10^{-7}$) | 3.145 | 3.145 |

where

$K_f$ is a function of fluid velocity and particle diameter

$D_L$ is function of molecular diffusion and turbulent mixing

$D_s$ is estimated from batch experiment and concentration

Table 4.6 Biological parameters used in the GAC biofiltration modeling

| Biological Parameter | Value | Unit |
|---|---|---|
| Biomass density ($X_f$) | $4.5x10^{3}$ | mg/L |
| Yield coefficient (Y) | 0.34 | mg/mg |
| Decay coefficient ($K_d$) | $7.9x10^{-07}$ | $s^{-1}$ |
| Total biofilm loss efficient ($b_{tot}$) | $1.9x10^{-06}$ | $s^{-1}$ |
| Biofilm thickness ($L\_f_0$) | $1.0x10^{-06}$ | m |
| Suspended cell concentration ($X\_s_0$) | $1.0x10^{-08}$ | mg/L |
| Shear Loss ($\sigma$) | $1.16x10^{-06}$ | $s^{-1}$ |
| Maximum rate of substrate utilization ($K_{max}$) | $1.59x10^{-04}$ | |
| Monod half velocity coefficient ($K_S$) | 0.24 | |

Among different parameters, mass transfer rate, maximum rate of substrate utilization, and biomass yield coefficient were the most important parameters in biofilter modeling (Shim et al., 2004). With the estimated values of physical and biological parameters in Tables 4.5 and 4.6, mathematical prediction of the performance of GAC biofilter in terms of organic removal was made. The results are presented in Figure 4.7.

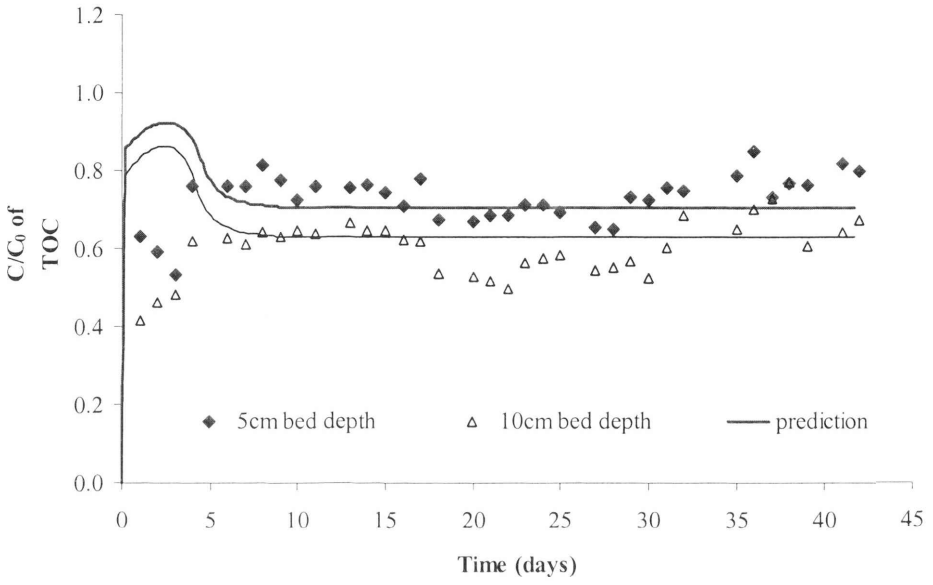



Figure 4.7 Experimental and simulated TOC values (synthetic wastewater, influent TOC = 11.6 mg/L, filtration rate = 2m/h, daily backwashing)

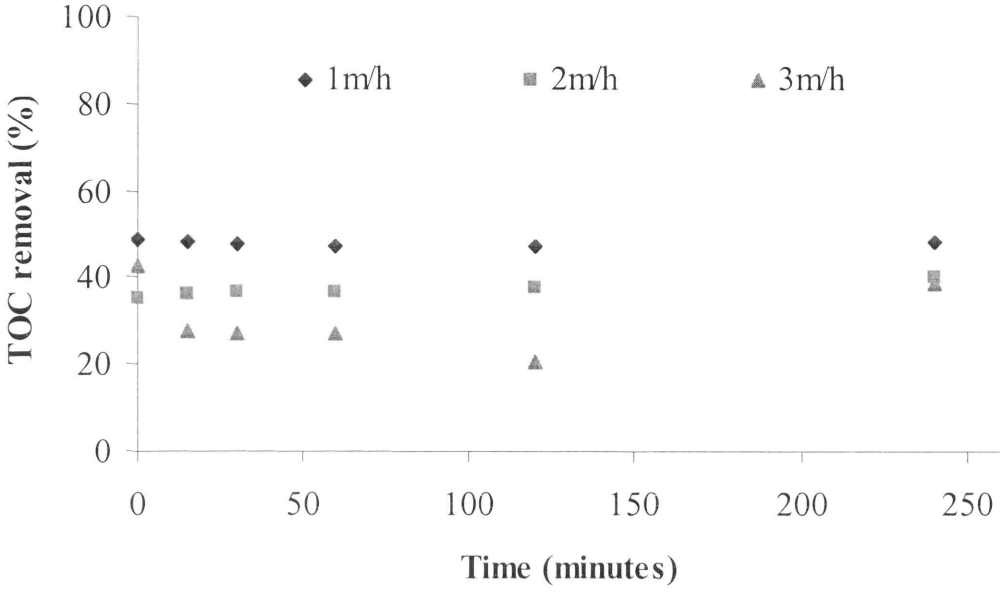
In this simulation model, GAC biofilter performance was described in two stages: adsorption during the initial stage and biodegradation in the latter stage. The prediction curve was fitted with experimental data in steady state of the biofilter but it was not successful in predicting the GAC biofilter in acclimation time when adsorption capacity was exhausting and biological activities were increasing. In practice, the prediction of the steady state of a biofilter is more important as the initial stage (adsorption phase) is only for only short time in the beginning (the first few days). The duration of steady state can be for few months or even a year. Even though there were many biological parameters adopted from other studies, the performance of the biofilter fitted rather well with experimental data in the biological phase in biofilters. However, this simple model cannot describe the biofilter in detail. It could be the lack of experimental data and simplicity in assumption. The parameters obtained from previous studies could not fit well to this model. The input of more parameters is needed to get better results. This model paves the way for modification and evaluation of physical and biological parameters in a more precise way.

## 4.4 Effect of operational conditions on the performance of a GAC biofilter

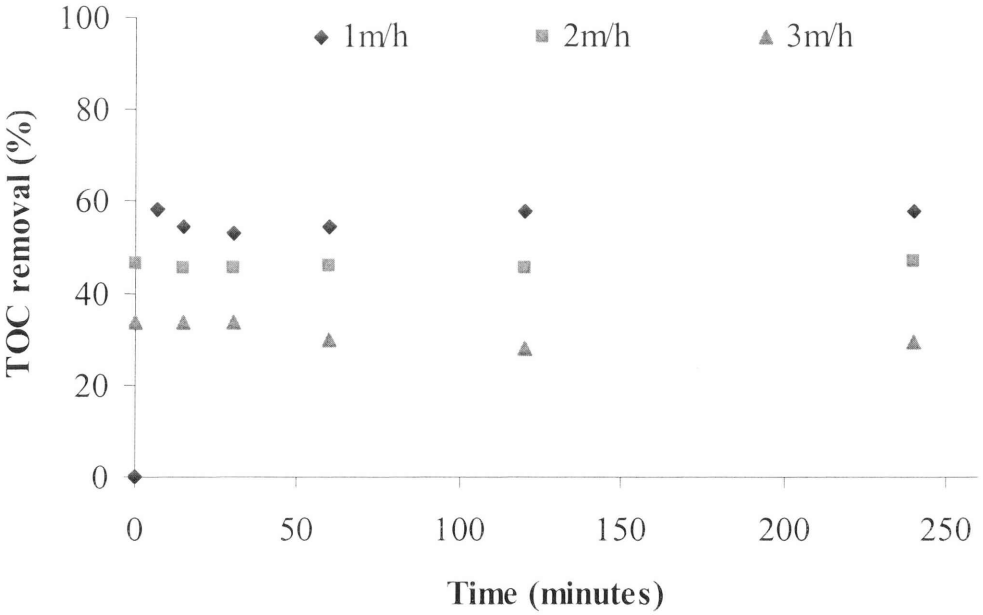
The success of operating a biofilter depends on several factors such as filter media, influent characteristics, operational conditions, etc. During operation and maintenance processes of a biofilter, operational conditions can be varied and consequently affect the efficiency of substances removal. In this study, the effects of filtration velocity and initial organic concentration of influent on GAC biofilter were evaluated during its biodegradation in the steady state. The Effects of filter media and filter bed depth on biofilter performance are discussed in Sections 4.2 and 4.5 of this study.

### 4.4.1 Effect of filtration rate

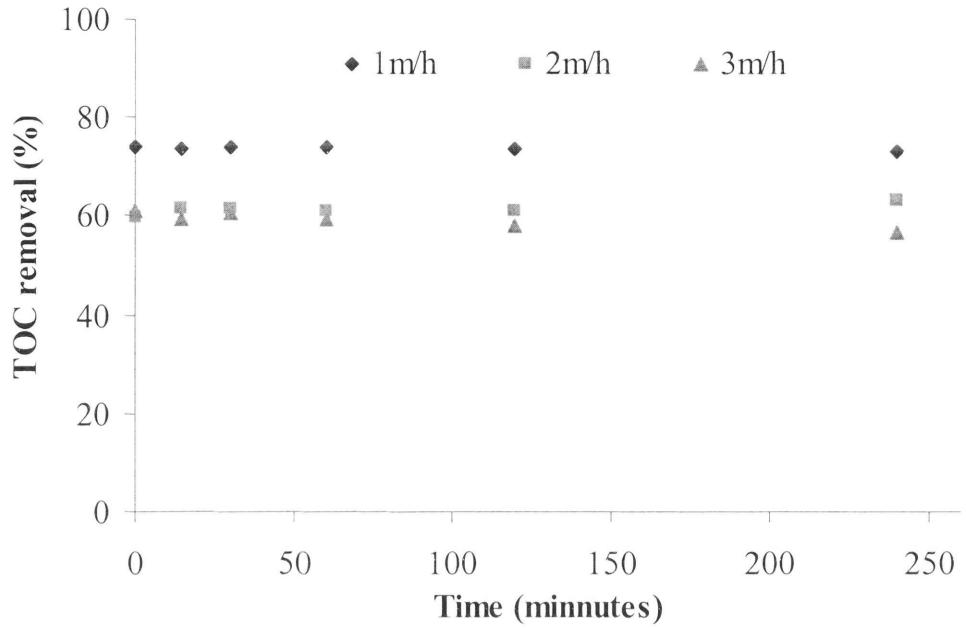
The effects of filtration rate on the organic removal efficiency of GAC biofilter were experimentally investigated. Three GAC columns with different bed depths (10, 15 and 30 cm) were operated at different filtration rates of 1, 2 and 3 m/h with synthetic wastewater. This experiment was conducted with GAC biofilters which were acclimatized at 2 m/h filtration rate with synthetic wastewater for about 40 days until the biofilters were in a pseudo steady stage. As can be seen from Figure 4.8, with increased filtration rates, the effluent quality became inferior to that with lower filtration rate and the organic removal pattern remained unchanged with time. This change occurred in all three GAC columns when filtration rate varied.




(a) GAC bed depth =10cm




(b) GAC bed depth =15cm




(c) GAC bed depth =30cm

Figure 4.8 Effect of filtration rate on the performance of GAC biofilter at different bed depths (filter acclimatization at 2m/h for 42 days prior to this experiment; average TOC = 10 mg/L, column diameter = 2 cm).

In all the acclimatized GAC biofilters, the efficiency in organic removal was decreased with the increase in the filtration rate. The TOC removal efficiency increased from less than 30 % to around 50% when the filtration rate decreased from 3 to 1 m/h in the 10 cm bed depth GAC biofilter. Similarly, an increase in organic removal was observed in the 15 cm and 30 cm GAC bed depth columns with the same change of filtration rate. This effect was observed more clearly in the shallow bed column with a significant drop in the TOC removal rate when filtration velocity increased. The same change of filtration velocity resulted in more than 20 % difference of TOC removal in the 10 and 15 cm bed depth column, but only about 10 % in the 30 cm GAC bed depth column. It seems that the effect was only observed within some range of filter bed depth or filtration velocity. This needs more detailed investigation to have the overall conclusion.

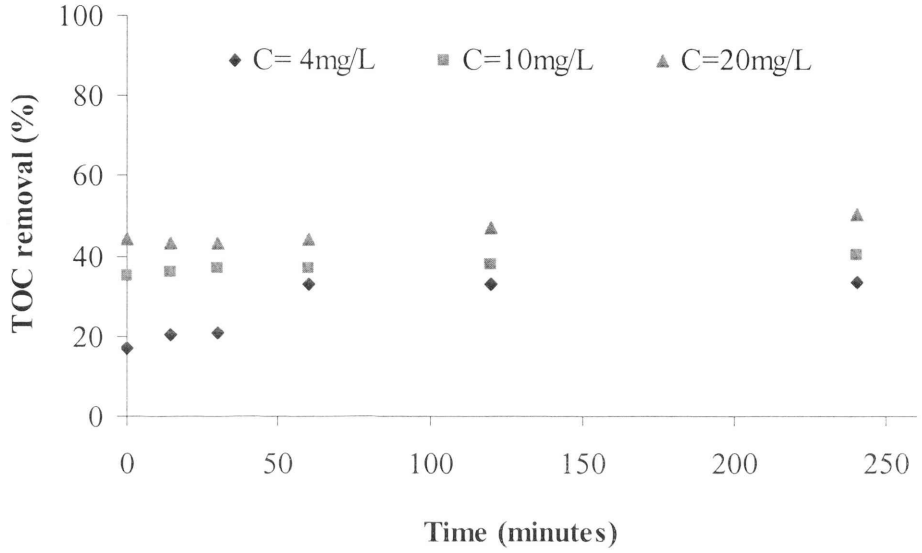
In other words, the change of filtration rate led to the change of EBCT that caused the differences in TOC removal efficiency. In 15 cm - GAC bed depth column, when filtration increased from 1 to 2 and 3m/h, EBCT dropped from 9 minutes to only 4.5 and 3 minutes while in 30cm - GAC bed depth, the same change of filtration rate decreased the EBCT from 18 minutes to 9 and 6 minutes. The longer contact time between filter media and influent provided more time for biodegradation of organic matters so that they could result in better organic matter removal.

This result agreed with many previous studies about the influence of EBCT on the quality of treated water. They all observed that the increase of EBCT led to better organic removal efficiency in the biofilter. LeChevallier et al. (1992) claimed that TOC removal could increase from 29 to 51.2 % with the increase of EBCT from 5 to 20 minutes. In another study by Chaudhary et al. (2003), the decrease of EBCT by increasing velocity also resulted in lower organic removal rate. EBCT is usually suggested as the most important factor in designing biofilter. Wang et al. (1996) found that in a given EBCT, organic removal efficiency was independent of hydraulic loading rate.

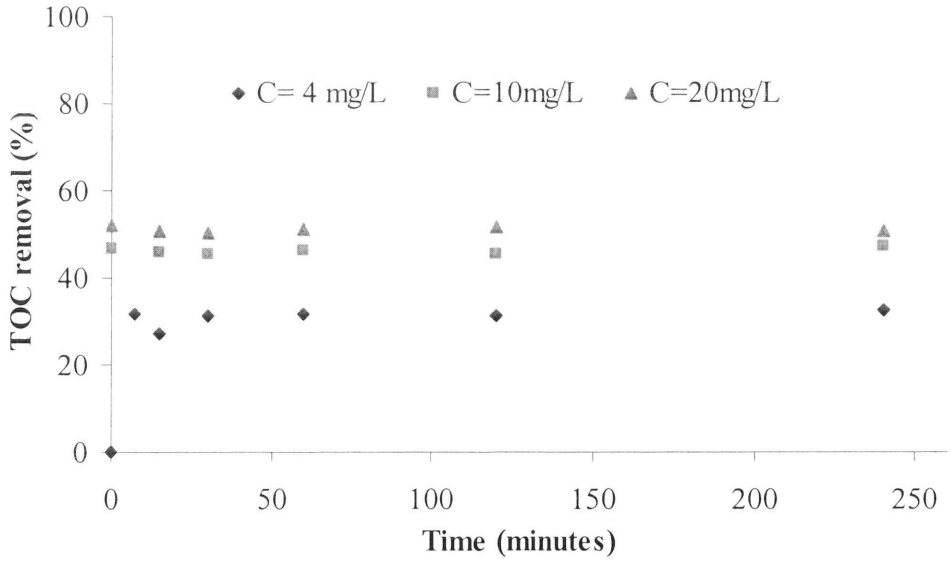
### 4.4.2 Effect of influent concentration

Influent characteristics are one of factors that can directly affect the quality of treated effluent in the treatment process. Previous studies (Carlson and Amy, 1998, Chaudhary, 2003) reported that the influent concentration can affect the quality of treated water. Characteristics of the influent, especially organic content, are more important in systems based on biological activity as it is the source of energy and nutrients for the maintenance and development of a microbiological community.

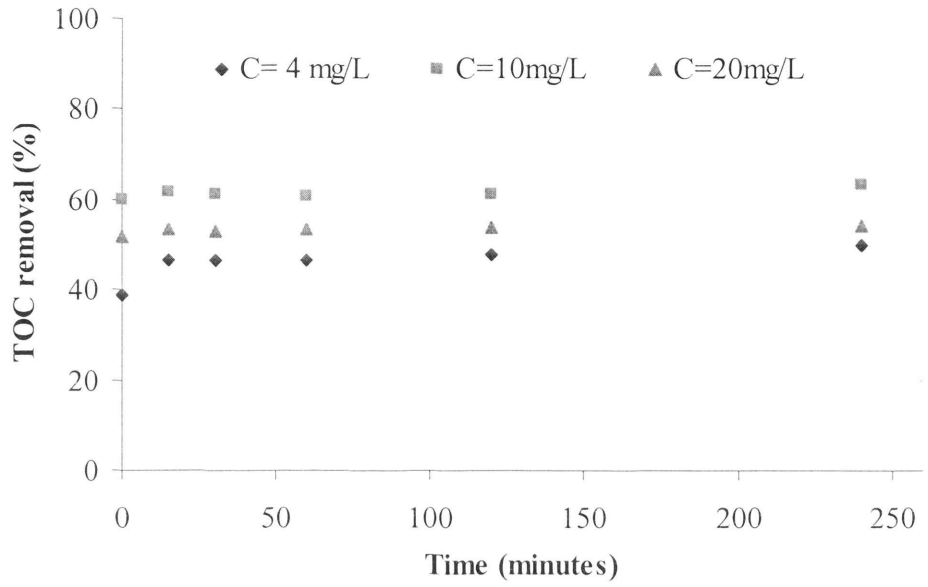
In this experiment, the GAC biofilters were acclimated at a filtration rate of 2m/h with synthetic wastewater (TOC concentration = 10 mg/L). In order to study the effect of influent on biofilter, synthetic wastewater at different TOC concentrations (4, 10 and 20mg/L) were used to feed those columns. The result of this experiment with GAC columns (bed depth = 10, 15 and 30 cm) is shown in Figure 4.9.




(a) GAC bed depth =10cm




(b) GAC bed depth =15cm




(c) GAC bed depth =30cm

Figure 4.9 Effect of influent concentration on the performance of GAC biofilter with different bed depths (Filter acclimatization at 2m/h, average TOC influent = 10 mg/L, column diameter = 2 cm)

In all the three GAC columns used in this experiment, the TOC removal efficiency was increased when the column was fed with higher concentrations of influent. This effect remained consistently over time even after the change of influent concentrations. In a 15 cm - bed depth column, there was about only 30 % TOC removal at 4mg/L TOC influent whereas 55 % of TOC was removed when TOC of influent was increased to 20 mg/L. A similar trend was also observed in a 30 cm GAC bed depth column with an increase from 45 % and 65 % TOC removal with a change of influent concentration from 4 to 20 mg/L TOC, respectively. This could be the result of rising micro-organisms metabolisms in biofilter when more nutrients in feeding water were available. Thus, it led to a higher organic removal rate at a high concentration of influent. This result agrees with the findings of Huck et al. (1994) in which higher influent concentration resulted in better removal rate of BOM and DBP precursors.

However, the increase of TOC removal did not seem proportional to the rise in influent concentration. There was a significant increase in organic removal (>10 %) when TOC concentration of influent increased from 4 to 10 mg/L in comparison to a little increase of organic removal (less than 5%) with an increase of influent concentration from 10 to 20mg/L in all columns. With the condition of this experiment, it might be expected that GAC biofilter can enhance its TOC removal in only some ranges of influent concentration. It might be because all available sites in the supported media surface were occupied by biofilm. When the biofilm reaches its maximum, the continuous increase in nutrients supply no longer promotes microbial development. The independence of removal rate and influent was also reported by Loosdrecht et al. (1990) who explained this phenomenon as a result of potential of reaction among reactants (microorganisms and surface media).

To sum up, the change of filtration velocity and initial concentration of feeding water during operation affected biofilter efficiency of organic removal. The GAC biofilter performed a better TOC removal rate when it was operated at lower filtration rates (longer EBCT) and higher initial organic concentration of influent.

## 4.5 Biomass in GAC biofilter

### 4.5.1 Biomass accumulation

The accumulation of microorganisms onto filter media plays an important role in determining the effectiveness of a biofilter. The speed of deposition and formation of biofilm decides the length of acclimatized time of the biofilter which can affect time and cost of the biofilter set-up. During the pseudo steady state of biofilter, the amount and activity of biofilm directly influence the biodegradation process in removing pollutants.

In this study, in order to estimate the growth of biomass on GAC, batch test and column experiments were conducted with synthetic wastewater and river water (taken from Hawkesbury River, Sydney). Biomass growth was evaluated by measuring dry mass and viable cell number attached on GAC particles.

**A. Dry mass accumulation**

**(i) Dry mass accumulation in batch test**

In this study, the growth and attachment of biomass on GAC was investigated first in the batch mode. A known amount of GAC was mixed in synthetic wastewater. Synthetic wastewater water was replaced daily to make sure that microorganisms always received adequate amount of nutrients for their natural growth.

Figure 4.10 presents the amount of dry mass attached on GAC during the batch test with synthetic wastewater. After about 4 weeks of gradual increase, the dry mass attached on GAC was then stable. The amount of biomass reached the highest value of about 44 mg/g of GAC after 49 days.

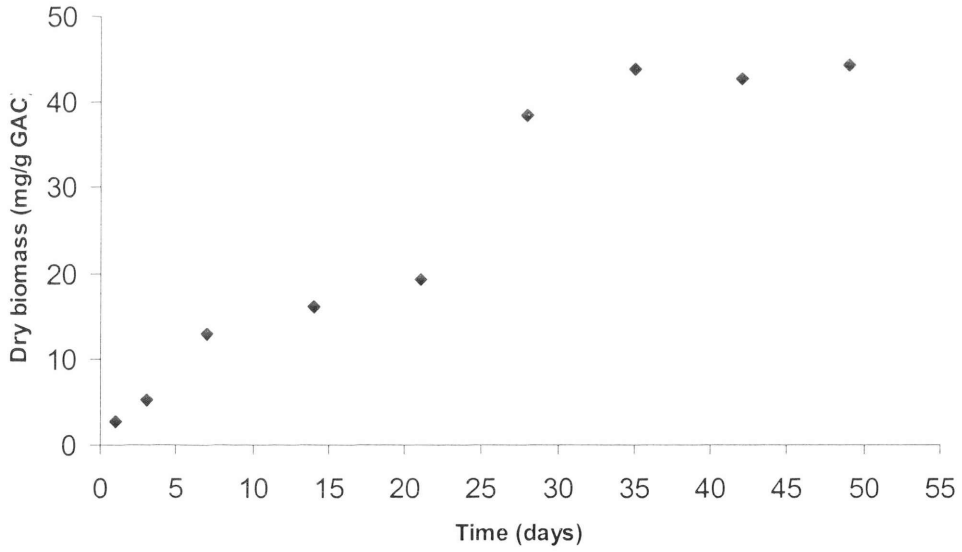



Figure 4.10 Biomass accumulation in GAC with synthetic wastewater in batch mode (GAC = 1g/L mg, average influent TOC = 10 mg/L)

The curve of biomass accumulation in Figure 4.10 can be correlated to microbial growth pattern (Figure 2.5). The initial period with a gradual increase of amount of dry mass corresponded to lag phase of microbial growth when microorganisms start to get used to the new environment. A rapid jump of dry mass was recorded from day 22 to day 30. This

corresponded to the exponential microbial growth phase. According to Deront et al. (1998), the length of this period depended on the organic loading rate. The log phase was followed by the stationary phase in which the amount of biomass nearly constant. The equilibrium between the biofilm growth and detachment is an important feature of this phase. Even though synthetic wastewater was frequently replaced to provide nutrient, the biomass growth on GAC was quite slow due to the lack of oxygen source.

**(ii) Dry mass accumulation in GAC biofilter**

The attachment of biomass on the GAC filter (fixed bed process) was also investigated in this experiment. A series of GAC columns of 5 cm bed depth was operated with synthetic wastewater at a filtration rate of 2m/h and daily backwashing. All GAC with biomass retained on it was taken out of each column and dried for biomass measurement at different periods of time. The results of the amount of dry mass retained on GAC in biofilter over time are presented in Figure 4.11.

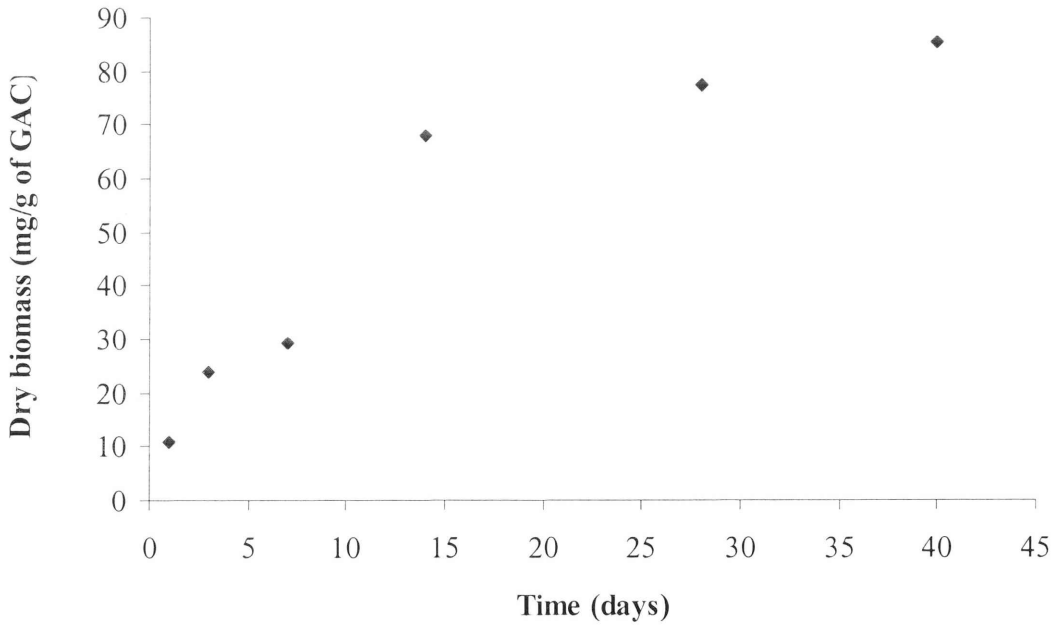



Figure 4.11 Dry mass accumulation in GAC biofilter (GAC bed depth = 5 cm, filtration rate = 2m/h, average influent TOC = 10 mg/L)

The accumulation of biomass on the GAC particles in columns followed the same pattern as in batch mode. Similar to the batch test, the mass retained on GAC increased steadily during the initial stage (lag and log phase in microbial growth curve or acclimation stage in biofilter process).The amount of biomass then remained constant during the stationary phase. However, the dry mass accumulation rate on GAC in the column experiment was faster than that in the batch test. The amount of dry mass was also higher in the column (Figure 4.12). While it took about 30 days for accumulating 44 mg dry mass/ g GAC in batch test, biofilter only needed 15 days to reach the pseudo steady state with double amount of dry mass retained on GAC (85 mg dry mass/g GAC).

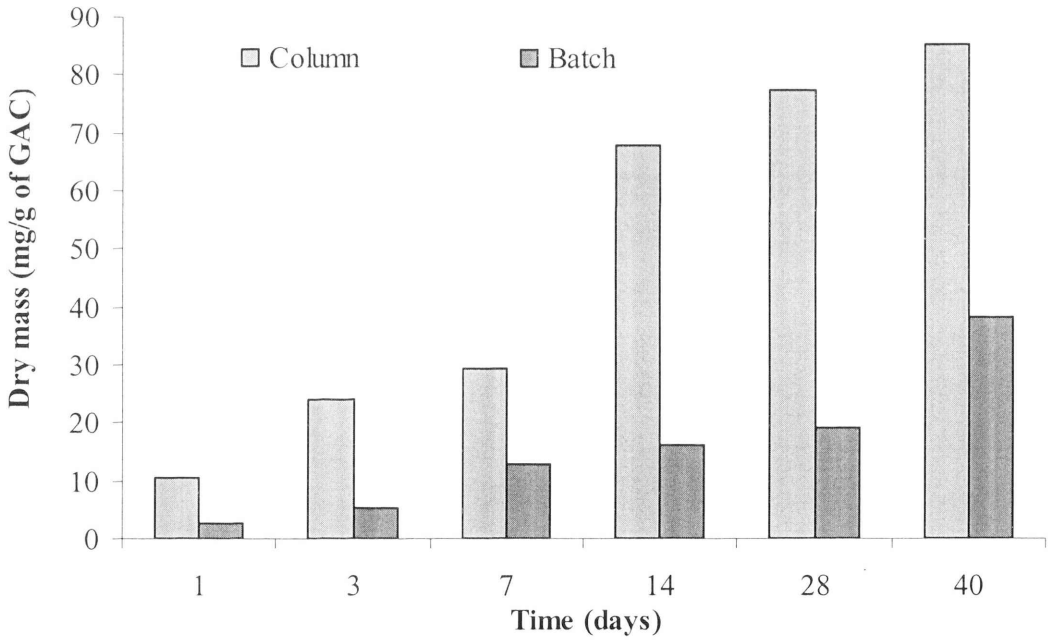



Figure 4.12 Comparison of biomass accumulation onto GAC in column and batch experiment

The difference of biomass accumulation onto GAC was the result of the difference of experimental conditions. Microorganisms in the column experiment were in the favorable conditions with the continuous supply of nutrients at a constant rate. Further, the dissolved oxygen was higher throughout the GAC depth. Therefore, the amount and rate of biomass attached on the GAC in the filter was higher than in the batch. According to Carlson and

Amy (1998), Wang et al. (1995) and Chaudhary et al. (2003), the biomass concentration profile with time was found to depend both on hydraulic and organic loading rates in biofilter.

**B. Viable cell count on GAC**

**(i) Viable cell count in batch test**

Microbiological growth on GAC was investigated in the batch test by measuring the number of viable cells with time. Different amounts of GAC (0.5, 1, 2, 5 and 10 g/L) were added in the same volume of synthetic wastewater solution. Synthetic wastewater in the batch test was replaced on a daily basis in order to supply nutrients for microorganisms. Oxygen was also provided by shaking the solution at a speed of 50 rpm. The same amount of GAC sample was taken out of the solution once every few days for viable cells measurement. The development of microbes on GAC in batch test with synthetic wastewater and river water is presented in Figure 4.13. The same set of experiments was performed with water from the Hawkesbury River. The results with river water are presented in Figure 4.14.

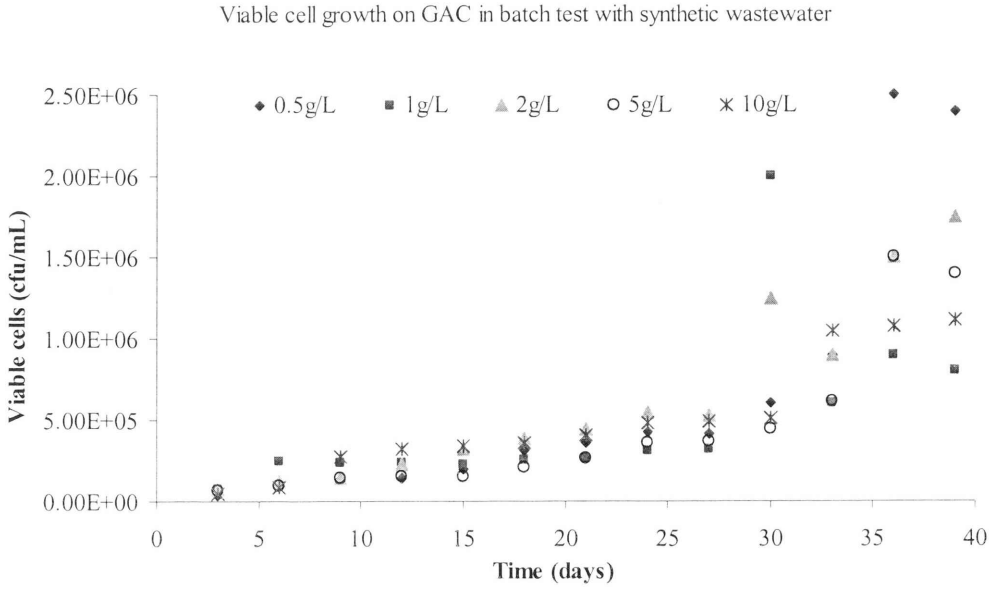



Figure 4.13 Viable cell growth on GAC in batch test with synthetic wastewater

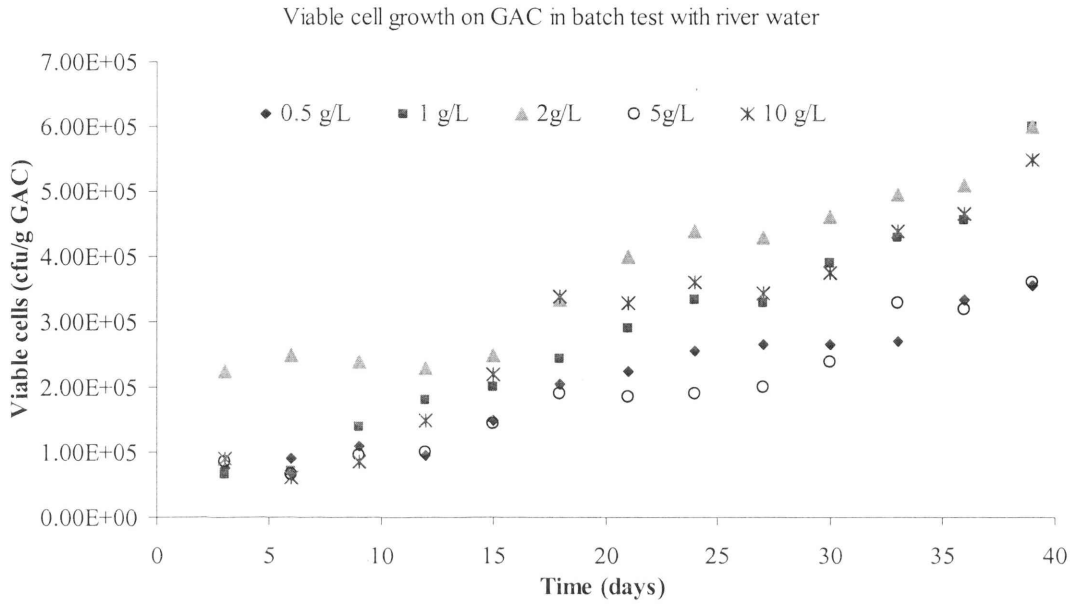



Figure 4.14 Viable cell growth on GAC in batch test with river water

In both sets of this experiment, the growth of biofilm onto GAC gradually increased in a steady manner. A slow increase in the number of bacteria was observed at the beginning of the experiment and then it was followed by a jump in the number of viable cells on GAC. In this batch experiment, the number of viable cells enumerated on GAC grains with synthetic wastewater was significantly higher than with river water. The difference was more substantial after 30 days. The number of bacteria on GAC in the batch with synthetic wastewater was $7.5x10^5 - 1.5x10^6$ CFU/ g of GAC while this number was only $4.0 \times 10^5$ - $6.0 \times 10^5$ CFU/g of GAC in river water.

Similar to the dry mass accumulation in batch conditions, the time required for biofilm on GAC reaching its equilibrium state was quite long. After 40 days of running this experiment, the number of viable cells was found to increase and no equilibrium of microbial community was observed in the batch with synthetic wastewater. In the other batch with river water, viable cells were increasing during the experiment but the increase was slowed down after 25 days.

It can also be seen from Figures 4.13 and 4.14 that the variation of the GAC loading rate led to a different amount of biomass accumulation on GAC. With the GAC dose of 2g/L, a

higher number of viable cells was noticed from the 20th day onwards. However, the difference was not significant.

**(ii) Viable cell count in GAC biofilter**

The deposition of microbes on the GAC in the biofilter was also investigated by using a viable cell count technique. In this experiment, two GAC columns with a bed depth of 15 cm were fed with synthetic wastewater and river water (from Hawkesbury River). Both GAC filters were operated at the filtration rate of 2 m/h. Daily backwash was employed to avoid filter clogging. The microbial population in the influent, effluent and on GAC was estimated by plate count of colony forming units (CFU). With GAC samples taken from filters, microorganisms were detached by agitating GAC in a pre-determined amount of distilled water and then enumerated by the viable cell count analysis process. The results of viable cells number in GAC biofilters are presented in Tables 4.7 and 4.8.

Table 4.7 Viable cell count in GAC column fed with synthetic wastewater (column diameter = 2cm, GAC bed depth = 15cm, filtration rate = 2m/h)

| Time (day) | Influent (cfu/mL) | Effluent (cfu/mL) | GAC granules (cfu/g GAC) |
|---|---|---|---|
| 5 | $2.12 \times 10^6$ | $0.36 \times 10^6$ | $1.50 \times 10^6$ |
| 7 | $1.85 \times 10^6$ | $0.71 \times 10^6$ | $8.50 \times 10^6$ |
| 9 | $1.16 \times 10^6$ | $1.33 \times 10^6$ | $5.40 \times 10^6$ |
| 11 | $13.5 \times 10^6$ | $3.40 \times 10^6$ | $4.20 \times 10^6$ |
| 14 | $2.13 \times 10^6$ | $4.70 \times 10^6$ | $4.80x\ 10^6$ |
| 18 | $1.48 \times 10^6$ | $4.90 \times 10^6$ | $11.00 \times 10^6$ |
| 22 | $2.13 \times 10^6$ | $2.80 \times 10^6$ | $12.50 \times 10^6$ |
| 27 | $2.13 \times 10^6$ | $0.86 \times 10^6$ | $14.80 \times 10^6$ |
| 30 | $1.50 \times 10^6$ | $1.28 \times 10^6$ | $43.00 \times 10^6$ |

Table 4.8 Viable cell count in GAC column fed with Hawkesbury River water (column diameter = 2cm, GAC bed depth = 15cm, filtration rate = 2m/h)

| Time (day) | Influent (cfu/mL) | Effluent (cfu/mL) | GAC granules (cfu/g GAC) |
|---|---|---|---|
| 5 | $0.03 \times 10^6$ | $0.01 \times 10^6$ | $0.1 \times 10^6$ |
| 7 | $3.80 \times 10^6$ | $1.65 \times 10^6$ | $9.2 \times 10^6$ |
| 9 | $4.10 \times 10^6$ | $2.00 \times 10^6$ | $1.9 \times 10^6$ |
| 11 | $2.00 \times 10^6$ | $1.70 \times 10^6$ | $13.2 \times 10^6$ |
| 14 | $1.59 \times 10^6$ | $2.23 \times 10^6$ | $25.0 \times 10^6$ |
| 18 | $1.21 \times 10^6$ | $1.12 \times 10^6$ | $25.0 \times 10^6$ |
| 22 | $0.13 \times 10^6$ | $1.50 \times 10^6$ | $42.0 \times 10^6$ |
| 27 | $0.23 \times 10^6$ | $1.52 \times 10^6$ | $10.1 \times 10^6$ |
| 30 | $1.32 \times 10^6$ | $1.44 \times 10^6$ | $28.0 \times 10^6$ |

In both GAC filters for synthetic waste water and river water, the amount of viable bacteria attached on GAC increased with time. A high number of bacteria was found in the effluent. Vahala (2002) suggested that the presence of microorganisms in effluent was the result of biomass detachment which can be related to the phase of bacterial growth. It means that the number of bacteria in effluent decreases when the colonization on the GAC media reaches the steady state. Servais et al. (1994) reported a similar observation in which bacteria in effluent was highest at the colonization phase with a density of $1.5 \times 10^5$ bacteria/ mL and about $0.5 \times 10^5$ bacteria/mL

In this study, the amount of organisms attached on GAC was underestimated due to the difficulty in detaching all the biofilm from GAC. In addition, this microbiological technique can only measure cells which are capable of reproducing themselves (Lazarova and Manem, 1995).

### 4.5.2 Biomass detachment by backwashing

During the biofilter operation, backwashing was applied to prevent the clogging of the biofilter by removing the excess biomass and suspended particles. The detachment of biomass in the biofilter occurs mostly by backwashing even though biological decay and fluid shear contribute towards the loss of biofilm (Vahala, 2002). Therefore, the effect of

backwashing on the biofilter performance was studied by many authors (Hozalski et al., 1999; Carlson et al., 1996; Ahmad et al., 1998).

In this study, only the biomass loss by backwashing was investigated by determining the dry mass and viable cell count measurement. Backwashing was performed daily by using fresh tap water in an up-flow direction. The bed of the GAC column was expanded about 60% during backwashing for 2 minutes. The blockage in the filter was removed by turbulence and shear stress created in the filter bed during backwashing (Scholz and Martin, 1997). The amount of mass which was removed from the GAC biofilter by daily backwashing is presented in Figure 4.15.

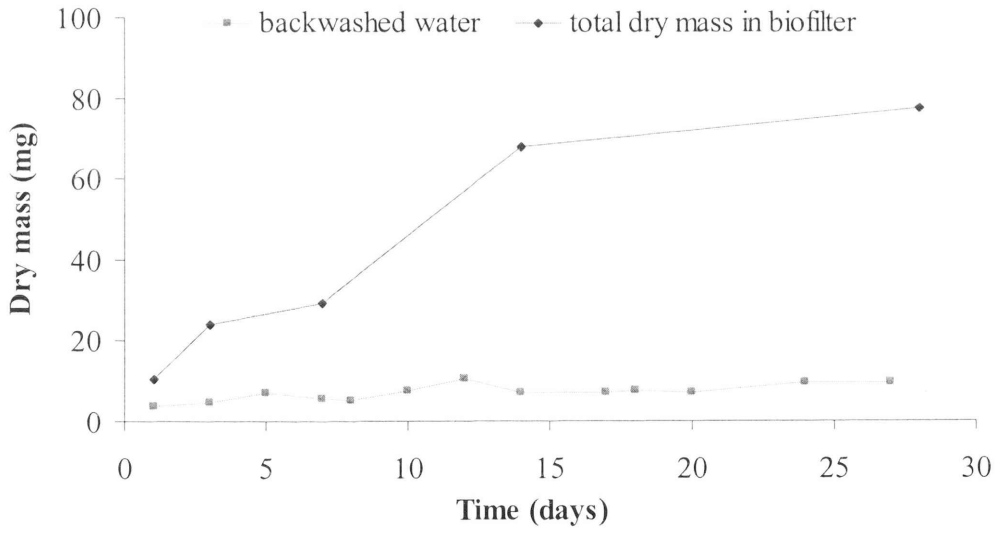



Figure 4.15 A comparison of total dry mass accumulation and the amount of mass detached by backwashing in GAC biofilter (GAC bed depth = 5 cm, filtration rate = 2 m/h, influent = synthetic wastewater, average influent TOC = 10 mg/L, backwashing expansion = 60 %, backwash duration = 2 minutes)

Biomass removal during backwashing was also measured by viable cell count. Table 4.8 shows result of plate count of viable cells of backwashed.

Table 4.9 Viable cell count in backwashed water (column diameter = 2 cm, GAC bed depth = 5 cm, backwashing expansion = 60 %, backwash duration = 2 minutes)

| Time (day) | Synthetic wastewater feeding column (cfu/mL) | River water feeding column (cfu/mL) |
|---|---|---|
| 5 | $4.2x10^3$ | $2.0x10^2$ |
| 10 | $7.8x10^6$ | $3.8x10^6$ |
| 20 | $8.9x10^5$ | $1.4x10^6$ |
| 30 | $4.3x10^5$ | $4.4x10^5$ |

The amount of biomass removed by backwashing increased slightly with the increase of total biomass accumulated in the biofilter. The amount of biomass removed during backwash (in terms of dry mass) was less than 5% of the total accumulated dry mass in the biofilter. According to Hozalski (1999), the biofilter performance was not affected with the rate of biomass removal by backwashing less than 60%. Besides, there was no significant difference in the TOC value of effluent before and after backwashing. It was in agreement with the finding of previous studies that backwashing does not considerably affect the performance of biofilter (Chaudhary, 2003; Hozalski et al., 1999; Carlson et al., 1996).

### 4.5.3 Microbiological activity in biofilter

Microbial community is responsible for biodegradation in the biofilter through its metabolism. Although the quantity of microorganisms was essential for the biological filtration process, the rate of microbial activity determines the efficiency of a biofilter. There are many methods reported to estimate the biofilm activity such as Adenosine triphosphate (ATP), dehydrogenase activity (DHA), DNA, etc. Among them, respiratory measurement is frequently used in water and wastewater treatment to estimate dissolved

oxygen uptake rate (OUR), or oxygen consumption rate or respiratory activity of microorganisms in biological systems (Lazarova and Manem, 1995).

Dissolved oxygen is essential for the metabolism of aerobic microorganism which determines the effectiveness of aerobic biological system. The disadvantages of dissolved oxygen consumption measurement are low sensitivity and reproducibility. In wastewater treatment, oxygen consumption rate represents the amount of oxygen utilized per unit volume per unit time by available microorganism (Ganesh and Ramanujam, 2005). Lazarova and Manen (1995) correlated oxygen uptake rate with the ATP profile in activated sludge. Lindberg and Carlson (1996) also found direct links between respiration rate with biomass growth and substrate removal.

**A. Dissolved oxygen consumption rate in GAC biofilter**

The biological activity of microbes in the biofilter was estimated by measuring their oxygen utilization. Figures 4.16 and 4.17 illustrated the DO consumption of microbes on GAC in biofilter for 30 minutes. In this experiment, a GAC column was operated with synthetic wastewater at a filtration rate of 2 m/h and shallow bed depth of 5 cm to shorten the biofilter acclimation time. Samples of GAC with retained biomass were taken from the top layer of biofilter where the microbial community is most active and well developed during the whole length of the filter (Wang et al., 1995).

During oxygen consumption measurement, oxygen content in the sample steadily decreased with time. The higher rate of oxygen consumption or metabolic rate of microorganisms led to steeper slope of dissolved oxygen content. It can also be seen from Figure 4.16 that the oxygen consumption rate has a linear relationship with time. This indicated a constant rate oxygen of consumption by microorganisms in GAC biofilter.

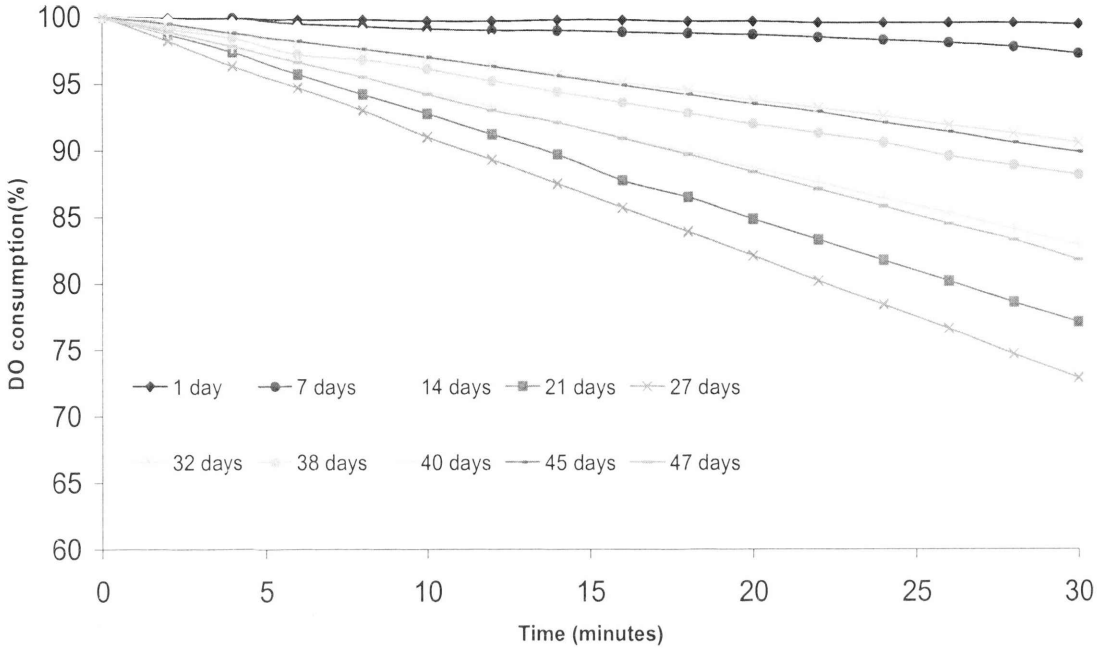



Figure 4.16 The changes in dissolved oxygen content of samples during 30 minutes of respiratory measurement (the days in the Figure caption refers to the number of the day of operation by the biofilter)

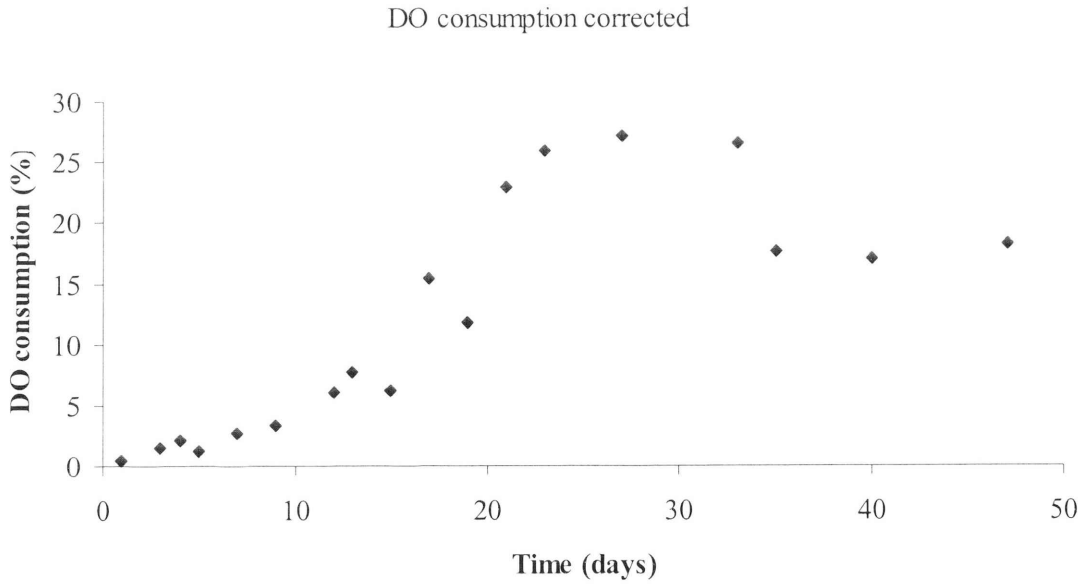



Figure 4.17 DO consumption of microbes on GAC in biofilter (diameter = 2 cm, bed depth =5 cm, filtration rate = 2 m/h, TOC influent = 10 mg/L; DO consumption in 30 minutes)

Figure 4.17 shows the pattern of microbiological activities through the oxygen consumption rate in GAC biofilter. During the first 20 days, the rate of oxygen consumption increased gradually due to the build up of the microorganism community. This period was the acclimation stage of the biofilter or the lag phase of microbial growth. The highest microbial activity was observed from 25-35 days of biofilter operational period. This correspond to the period where the biofilter exhibited the highest TOC removal (Figure 4.18). Except the acclimation time (the first 20 days), the TOC removal rate and respiration rate followed the same pattern. They both reached a peak after about 30 days of filter operation with 35% TOC removal and 30% oxygen consumption in 30 minutes. Servais et al. (1994) observed the same kind of evolution of biological activity but they could not find the explanation for the drop of OUR in the middle of the biofilter steady state.

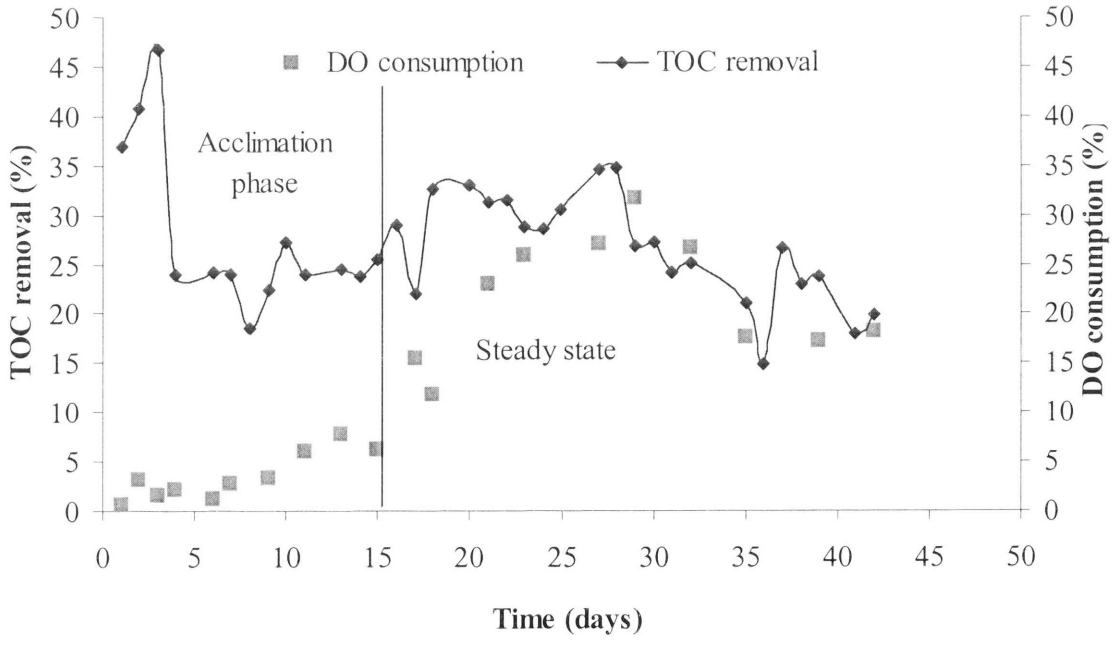



Figure 4.18 Relation between dissolved oxygen consumption rate and TOC removal by GAC biofilter (diameter = 2cm, bed depth =5cm, filtration rate = 2m/h, TOC influent = 10mg/L)

**B. Relationship between oxygen uptake rate and dry mass**

As discussed in previous sections, there is a relation between fixed biofilm on GAC and its respiratory rate in biofilter. The oxygen consumption rate in biofilter is the result of microbial metabolism. Thus, a direct link is expected between the amount of microorganisms and the oxygen uptake rate. It was found that not only microbial growth but also substrates removal correlated with the respiration rate (Ganesh and Ramanujam, 2005).

In this study, it is almost impossible to completely extract all the fixed biofilm from GAC due to its irregular surface. As a result, the amount of biofilm in GAC biofilter is underestimated by the dry mass and the viable cell count measurement. In order to estimate the fixed biofilm on GAC more accurately, a simple mathematical equation between the oxygen uptake rate and dry mass was established. Dry mass was collected from a batch experiment using sponge (polyeuthane foam) as the filter medium with synthetic wastewater. Due the softness of the sponge, the majority of the biomass attached to the sponge can be squeezed out. The same sample of microbes collected from sponge was analysed for both biomass and oxygen uptake rate. Biomass was determined as volatized suspended solids (VSS). The relationship between oxygen consumption rate and biomass is mathematically expressed in Equation 4.1.

$$\text{Oxygen consumption rate (\%)} = 0.1026 \times \text{amount of biomass (mg/L)} \qquad (4.1)$$

$$R^2 = 0.913$$

The linear relationship between the oxygen consumption rate and biomass was obtained during the exponential phase of microbial growth. Figure 4.19 presents the experimental data and mathematically fitted curves for dry mass and dissolved oxygen consumption.

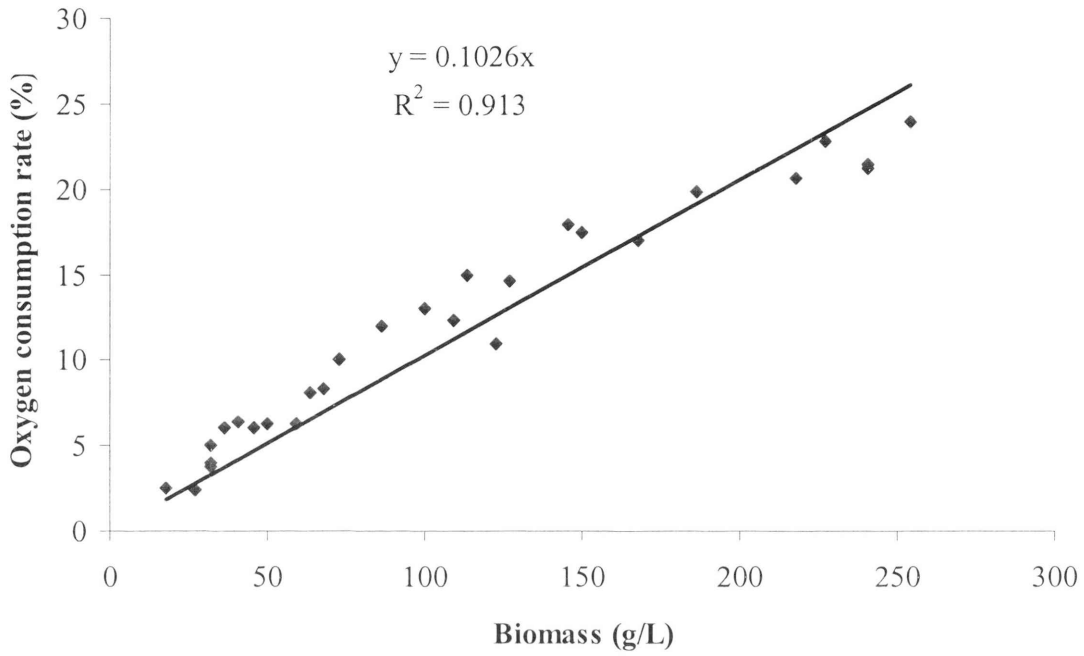



Figure 4.19 Relationship between biomass and oxygen consumption rate of fixed biofilm (oxygen consumption rate was measured in 30 minutes)

The result showed that the oxygen consumption rate could be used as a practical tool to evaluate biological activity in the biofilter. The relationship between biomass and the oxygen consumption rate enables us to estimate the fixed biomass in the biofilter. This is useful for highly porous filter media such as GAC, from which it is impossible to detach the biofilm for measurement.

## 4.6 Microbiological assessment of a GAC biofilter

### 4.6.1 Total coliforms

Microbiological safety is a significant criterion in assessing water quality. In drinking water treatment, biological quality is strictly controlled because it directly affects public health. The reduction of microorganisms in wastewater treatment contributes to better performance in the subsequent steps. It helps to decrease doses of disinfectants and membrane fouling. Among several indicators of biological quality, total coliforms is the most common indicator for evaluating fecal contamination. According to EAP standard (2002), total coliforms do not pose the health risks itself but indicate the presence of other potential harmful bacteria. Bitton (2005) stated that total coliforms is one of the best microbiological indicators of treatment efficiency. The total coliforms group belongs to the family of *Enterobacteriaceae*. They include *Escherichia coli*, *Enterobacter*, *Klebsiella,* and *Citrobacter*. Those bacteria are aerobic and facultative anaerobic, gram-negative, non spore-forming, rod-shaped bacteria (Bitton, 2005).

In this study, total coliforms indicator was used to evaluate the biological quality of water treated by GAC biofilter. The evaluation was conducted when a GAC biofilter was in the steady state. The results of this measurement are shown in Table 4.10.

Table 4.10 Total coliforms of influent and effluent of GAC biofilters for synthetic wastewater and river water.

| Time (days) | Total coliforms (Cfu/mL ) | | | | | |
|---|---|---|---|---|---|---|
| | Synthetic wastewater feeding columns | | | | River water feeding column | |
| | Influent | Effluent 1* | Effluent 2* | Effluent 3* | Influent | Effluent 4* |
| 30 | 620 | 280 | 180 | 220 | 280 | 0 |
| 33 | 620 | 300 | 190 | 200 | 220 | 0 |
| 36 | 630 | 280 | 180 | 220 | 180 | 0 |
| 39 | 530 | 140 | 120 | 110 | 160 | 0 |
| 42 | 580 | 120 | 160 | 150 | 160 | 0 |
| 45 | 640 | 140 | 160 | 150 | 180 | 40 |
| 48 | 700 | 140 | 170 | 210 | 210 | 0 |
| 51 | 630 | 180 | 110 | 100 | 190 | 0 |
| 54 | 770 | 320 | 150 | 120 | 170 | 0 |
| 57 | 480 | 130 | 140 | 80 | 200 | 0 |
| 60 | 480 | 150 | 170 | 140 | 230 | 0 |
| 63 | 550 | 190 | 220 | 170 | 180 | 0 |
| 66 | 490 | 180 | 150 | 120 | 200 | 0 |
| 69 | 480 | 160 | 160 | 150 | 190 | 0 |
| 72 | 520 | 220 | 230 | 200 | 170 | 0 |
| Average TC (cfu/mL ) | **581** | **195** | **166** | **156** | **195** | **3** |
| Average TC removal (%) | | **66.40** | **71.44** | **73.17** | | **98.63** |

* *1, 2 and 3* are the 5, 15 and 30 cm GAC bed depth column for synthetic wastewater

*4* is the 15 cm GAC bed depth column for river water (from Hawkesbury River).

As can be seen in Table 4.10, all the GAC columns in this experiment resulted in a good removal rate of total coliforms. GAC columns eliminated 66-73% of coliforms from synthetic wastewater with the bed depth only 5-30cm. With river water, the GAC biofilter was highly effective in removing coliforms. The total coliforms were not detected in most of the effluent samples. The average total coliforms removal rate of the river water feeding the column was 98.63 %. The results were in accordance with the previous studies on biofilter in removing pathogens. It was found that a biofilter could remove from 20 to more than 90% of bacteria depending on operational conditions (Bitton, 2005).

### 4.6.2 Total fecal coliforms

Besides total coliforms evaluation, fecal coliforms were also monitored. This indicator is more practical in assessing water quality concerning public health. In drinking water evaluation, fecal coliforms detection is the compulsory test for samples which are positive to total coliforms (EPA standards, 2002).

The average numbers of fecal coliforms in influent and effluent of GAC biofilter with synthetic and river water are presented in Table 4.11. This test was carried out for 45 days during the steady state of the GAC biofilters which operated at a filtration rate of 2 m/h and daily backwashing.

Table 4.11 Total fecal coliforms of influent and effluent of GAC biofilters for synthetic wastewater and river water

| Biofilter | Sample | Average total fecal coliforms (cfu/100mL) |
|---|---|---|
| GAC biofilter with river water | River water | 1.47 |
| | Effluent | 0 |
| GAC biofilter with synthetic wastewater | Synthetic wastewater | 13.73 |
| | Effluent 1* | 0 |
| | Effluent 2* | 4.27 |
| | Effluent 3* | 0 |

* *1, 2 and 3* are the 5, 15 and 30 cm GAC bed depth column for synthetic wastewater

It can be seen in Table 4.11 that total fecal coliforms in the influent was present in a small number of less than 20 cfu/100mL for synthetic wastewater and 5cfu/100mL for river water. In effluent samples, no fecal coliforms were detected in the effluent of the filter for river water. Fecal coliforms were detected in only 2 samples of effluent of the filter for wastewater during 45 days of measurement. It is suggested that using GAC biofilter can be a good option for controlling biological quality of water.

### 4.6.3 Microbiological identification in GAC biofilter

After the GAC biofilter reached its steady state, microorganisms were isolated for bacterial identification. Biolog was used for identifying predominant bacteria retained on GAC grain. The results of microbial identification in biofilter fed by synthetic wastewater and river water are shown in Table 4.12.

Table 4.12 Microbial identification on GAC in biofilter

| Column | Predominant microbal genera |
|---|---|
| Synthetic wastewater | • *Pseudomonas Aeruginosa*<br>• *Pseudomonas Alcaligenes*<br>• *Streptococcus oralis* |
| River water | • *Brevibacterium otitidis*<br>• *Enterobacter Cloacae*<br>• *Enterobacter Aerogenes (KLB. Mobilis)*<br>• *Staphylococcus Epidermidis*<br>• *Kocuria Rosea/Erythomyxa** |

* this species only appears in the batch test with river water.

*Pseudomonas* was present on the GAC in the biofilter for synthetic wastewater. These bacteria are Gram-negative, aerobic, non fermentative. Previous studies indicated that *Pseudomonas aeruginosa* was frequently found in water, wastewater, soils and the biofilm of biofilter and drinking water distribution systems (Bitton, 2005). Some researchers used *Pseudomonas Aeruginosa* for developing artificial biofilm (Szewzyk et al., 2000). *Pseudomonas Aeruginosa* and *Pseudomonas Alcaligenes* are among the common opportunistic pathogens of humans and exposed to fecal pollution (Bitton, 2005). Therefore, disinfection is necessary as a post treatment to eliminate those pathogens.

In the biofilter fed by river water, the predominant microorganisms on GAC belonged to three genera, namely, *Enterobacter* (Gram-negative, capsulated bacterium), *Brevibacterium and Staphylococcus* (Gram-positive, non motile bacterium). Enterobacter belongs to coliforms group and opportunistic bacterial pathogens. In the batch test with river water, there was the appearance of *Kocuria Rosea*. It is a gram-positive microorganism belonging to the family Micrococcaceae which can be the opportunistic pathogen in the immuno-compromised patients (Stackebrandt et al., 1996).

It is noted that only predominant bacteria were identified in this study. Thus, this identification does not reflect the whole microbiological ecology in a biofilter. Other studies found that protozoa and rotifers play a significant role in biofilter (Bitton, 2005).

## 4.7 Comparison of the effectiveness of GAC and other filter media in biofilter

The selection of biofilter media is critical in designing biofilter. It directly influences system design parameters, operation and maintenance techniques and expenses. Determining the kinds of filter media is also due to different purposes of water treatment and practical conditions.

In this study, biofilter experiments were conducted with different filter media such as GAC, anthracite, sponge (polyeuthane foam) and floating media (poly propylene) to evaluate the effectiveness of GAC to compare with other filter media for organics removal. GAC, anthracite, floating media and sponge were packed in 2 cm - diameter columns to a bed depth of 30 cm. All the columns were operated at the same condition with synthetic wastewater as a nutrient source. After 6 weeks of continuous operation, all columns reached the steady stage in terms of organic removal.

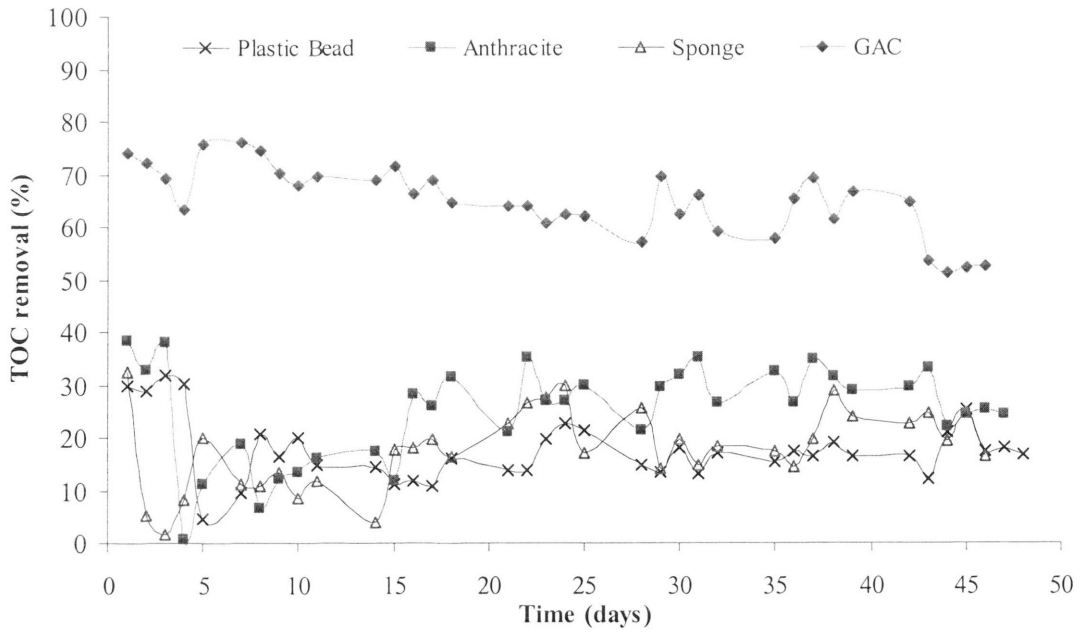



Figure 4.20 Comparison of different filter media for organics removal (average influent TOC = 10 mg/L, bed depth = 30cm, filtration rate = 2m/h)

The results are presented in Figure 4.20 in which GAC was superior to other filter media with higher TOC removal efficiency. While anthracite, floating medium and sponge removed only about 20% of TOC, GAC resulted in a constant TOC removal efficiency of more than 60%. Compared to anthracite, floating media and sponge filter media in terms of eliminating organic matter, GAC provides better removal rate due to its capacity of adsorption of substrates and microorganisms. According to Urfer *et al.* (1997), GAC establishes biofilm more rapidly with better protection against oxidant residual and faster recovery NOM removal capacity after periods out of service.

Except GAC, the other three media exhibited a similar organic matter removal rate of 20%. Even through they did not lead to as high a rate of organic removal as GAC, they all have other advantages. For example, anthracite is a traditional media for biofilter with the advantage of low cost. However it is heavy and requires more energy for backwashing. Sponge is a new material recently employed in water treatment as a biofilter media. It not only removes organic carbon, but also performs nitrification in bioreactors (Machdar et al., 1997). Unlike other medium, floating media can be used as a flocculator which performed a good removal rate of nitrogen and phosphorous (Ngo and Vigneswaran, 1996).

## 4.8 Performance of the combined system of GAC - sponge biofilter

In order to improve water quality by biological process, influent can be pre-treated by biofilter systems. Higher removal of organic matters can be achieved by using a series of GAC filters instead of using anthracite or sponge. However, if considering economical aspects, using many columns of GAC could result in higher expense of set up, operation and maintenance costs. Thus, a combination of GAC with other filter media biofilters can be a good way for treating water and wastewater in both cost and treatment efficiency, especially for small scale of water treatment plants. In addition, the combined system of different filter media biofilter can enhance the pollutants removal from water.

As discussed in Section 4.4, even though anthracite, floating media and sponge do not have a high rate of organic removal, they led to a consistent TOC removal efficiency at lower capital and operational costs. Their efficiency in organic removal remained consistently after a long time of operation. Among anthracite, floating and sponge, sponge has more advantages such as no need of backwashing during operation, easy maintenance, light weight and low cost (Machdar et al., 1997). Machdar et al. (2000) employed sponge in an aerobic biological unit. Their results indicated that sponge exhibited not only an excellent organic removal but also high nitrification efficiency and some extent of denitrification. Therefore, sponge was chosen in this study to investigate the effectiveness of the combination with GAC in organic matter removal. GAC biofilter, which remove the majority of organic matter, was used as pre-treatment for sponge column. Sponge was employed as biofilter media in saturated and unsaturated flow mode column for treating effluent from GAC filters.

### 4.8.1 GAC- unsaturated sponge biofilter

The system comprised two 2cm-diameter columns connected together. The first column was a GAC column with bed depth 30cm. The other column contained 30cm bed of small cubes of sponge (total volume of 94.2 $cm^3$, weight = 2.5 g). Firstly, influent (synthetic wastewater) was pumped into the GAC column, and then effluent from the GAC column was transferred directly into the sponge column. Since the sponge biofilter was operated in unsaturated mode, the influent was passed through sponges and flowed out completely at the outlet of the filter column. The organic removal efficiency of the combined system of GAC - unsaturated sponge biofilter is presented in Figure 4.21.

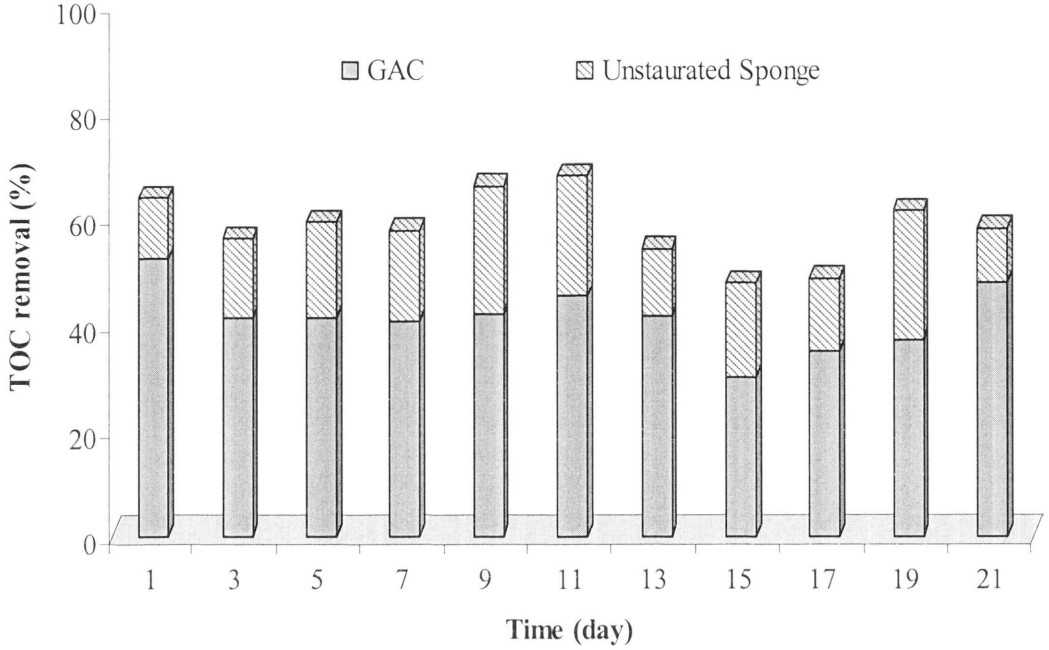



Figure 4.21 TOC removal by GAC-unsaturated sponge biofilter during steady state (filtration rate = 2 m/h, average TOC of feeding water =10 mg/L)

In this experiment, the major part of TOC was removed by the GAC column (about 40%). The unsaturated sponge column following the GAC biofilter continued eliminating further 15-20% of TOC. It is noted that this rate of organic removal was significant with only a small amount and volume of sponge used (total sponge volume = 94.2 $cm^3$, weight = 2.5 g). The addition of the unsaturated sponge column after the GAC biofilter raised the overall organic removal of the whole system up to 60-70%.

Other researchers reported that sponge bioreactor also performed good nitrogen removal (84% of $NO_3$ and $NO_2$ removal with hydraulic retention time less than 1 hour) from wastewater (Machdar et al., 1997). Moreover, there was no maintenance or operation activity, such as backwashing or aeration, related to sponge biofiltration process.

### 4.8.2 GAC- saturated sponge biofilter

The experimental set-up was similar to the experiment of GAC-unsaturated flow sponge biofilter. In saturated operation mode, the sponge was deeply submerged in feed water and

no backwash was applied. Both columns were fed in down flow direction at the same filtration rate 2m/h. During the steady state of both columns, the GAC biofilter demonstrated similar organic removal efficiency (about 40%) to GAC column in combination with the unsaturated sponge column but the saturated sponge achieved a lower TOC removal rate (Figure 4.22).

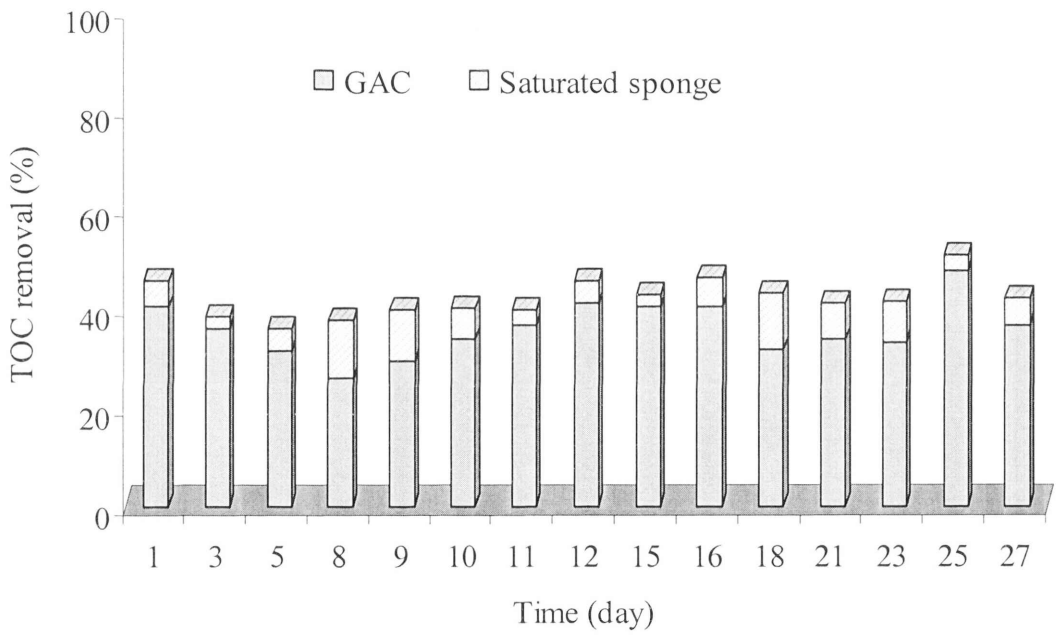



Figure 4.22 TOC removal by GAC-saturated sponge biofilter during steady state (filtration rate = 2 m/h, average TOC of feeding water =10 mg/L)

The lower TOC removal efficiency of the saturated flow sponge biofilter compared the unsaturated column was probably due to the lack of oxygen for microbiological activity. An improvement saturated flow sponge filter performance can be expected if there is an addition of aeration in the system. In addition, shape, size, porosity and arrangement of the sponge can affect the effectiveness of biodegradation process (Agrawal et al., 1997).

## 4.9 Performance of the combined system of GAC – floating medium biofilter

In this study, a GAC biofilter was connected to a column of floating filter medium. Effluent from the GAC column was transferred directly to the floating medium column. The set up of the two columns was as the same as the description in section 3.2

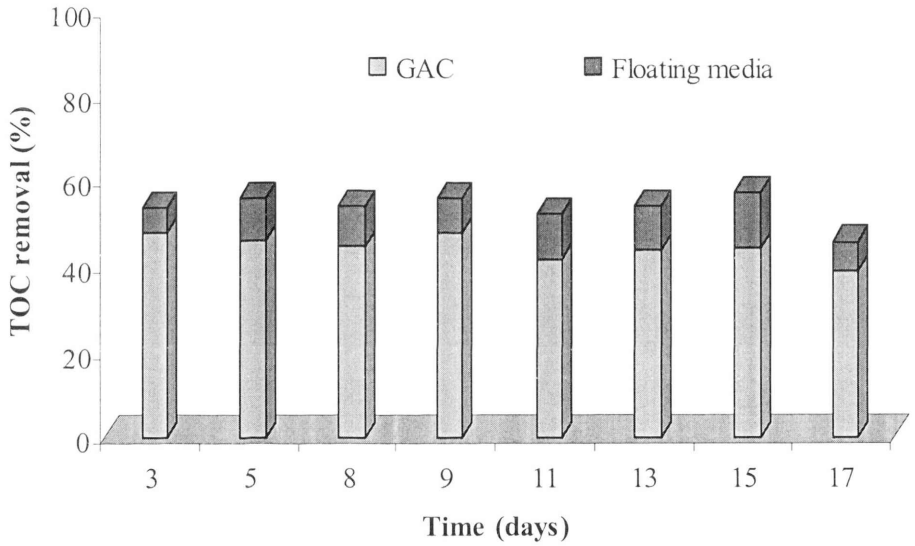



Figure 4.23 TOC removal by GAC-floating media system (filtration rate = 2 m/h, average TOC of feeding water =10 mg/L)

The average TOC removal rate by the GAC biofilter was 40% during steady state. The floating media filter removed an additional 10% of TOC when it was used as a post treatment of GAC biofilter (Figure 4.23). The organic removal rate by floating media was less than the experiment using a single floating media column for synthetic wastewater (Section 4.4). The lower TOC concentration of effluent from GAC column and lack of aeration could be the reason of this result. Some of organics which can be removed by the floating media filter would have already been removed by the GAC biofilter. However, the

floating medium filter was effective in removing phosphorous. A study of down-flow floating medium filter showed that it can remove up to 80-89% phosphorous from wastewater with chemical addition. It is remarkable because phosphorous is the limited nutrient for the growth of bacteria (Ngo and Vigneswaran, 1996). Also in this study, the floating media flocculator was used as pre-treatment of a coarse sand filter with high removal efficiency of $NH_3$-N (87%) and phosphorous (94%). Therefore, the use of floating medium filter after the biofilter can help controlling the biological quality of effluent but it needs further investigation.

In conclusions, the combined systems of GAC biofilter with sponge or floating filter media could improve the effectiveness in removing organic matter, nitrogen and phosphorous.

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Faculty of Engineering

# CHAPTER 5

# CONCLUSIONS

## 5.1 GAC biofilter in water and wastewater treatment

### 5.1.1 GAC bioadsorption mechanisms and models

- GAC exhibited a high organic removal rate by adsorption in both the batch and column experiments. The GAC adsorption equilibrium with synthetic wastewater fitted better with association theory (Talu) than with Freundlich and Sips models. The batch and dynamic (column) adsorption kinetics with organic matter can successfully be predicted by the Linear Driving Force approximation (LDFA) model.

- Long term GAC biofiltration was mathematically modeled using both adsorption and biodegradation mechanisms. The simulation was only successful in describing the biological phase. Details of experimental data on adsorption and biological processes are necessary to obtain specific biological and adsorption coefficients, thus to obtain better result of simulation.

### 5.1.2 Biomass growth on GAC filter

- The rate and amount of biomass which were accumulated onto GAC granules depended on influent concentration and operational conditions such as filtration velocity. The amount of biomass measured as dry mass and numbers of viable cells retained on GAC in the column dynamic experiments with synthetic wastewater were higher than in the batch test and with river water. During the batch test, the attachment of dry mass on GAC reached 44mg/g GAC in 30 days at the equilibrium stage while the amount of dry mass in steady state was 85mg/g GAC after 15 days operation in the column experiment with synthetic wastewater. The colonization time of microorganisms in the batch with synthetic wastewater was also shorter than with river water.

- In the biofilter, the pattern of biomass accumulation followed the lag, log and the stationary phases of the microbial growth curve. After a rapid increase in acclimation time, the biomass in the GAC biofilter remains constant over a long period that makes the biofilter work efficiently for a long filtration time.

It was found that the amount of fixed biomass on filter media was proportionally related to biological dissolved oxygen consumption rate.

- The prominent bacteria isolated from GAC biofilter for synthetic wastewater and river water includes (i) *Pseudomonas Aeruginos, (ii) Pseudomonas Alcaligenes, (iii) Brevibacterium Otitidis, Enterobacter Cloacae, (iv) Enterobacter Aerogenes, (v) Staphylococcus Epidermidis and (vi) Kocuria Rosea*. The full description of microbiological community can be obtained if the identification is extended to protozoa and virus components in biofilter. A more detailed study on microbial population is recommended.

### 5.1.3 Design and operational conditions of GAC biofilter

- Higher GAC bed depth led to better organic removal;

- Decrease in filtration velocity and increase in organic concentrations of influent resulted in higher organic removal. The TOC removal efficiency increased from 25% to 50% as the filtration rate decreased from 3 to 1m/h in a 15 cm GAC bed depth column. The increase of organic concentration of influent from 4-20 mg/L led to a rise of TOC removal from 25%-50%.

- Backwashing did not has any significant effect to TOC removal efficiency

### 5.1.4 Performance of GAC

- The GAC biofilter performed a consistent TOC removal efficiency for a long term of operation. A 2 cm diameter column of 30 cm GAC bed depth at filtration rate of 2 m/h can maintain about 60 % organic removal from synthetic wastewater (the average TOC = 10mg/L) for more than 40 days. With river water, GAC biofilter (bed depth = 15cm, filtration rate = 2m/h) resulted in a TOC removal efficiency of about 55 % for 50 days. It indicated that the GAC biofilter can be used for a long time with reliable efficiency.

- In terms of the biological quality of effluent, the total coliforms removal efficiency of 60 – 98 % was achieved with a GAC biofilter in the steady state.

Especially with river water, almost all fecal coliforms and total coliforms were eliminated by GAC biofilter.

- GAC as filter media was superior to plastic bead, anthracite and sponge in terms of organic removal.

## 5.2 Combined system of GAC and one other filter medium biofilter in water and wastewater treatment

GAC can be combined with other filter media to improve the effectiveness of biofilter. The combination of GAC with sponge or polypropylene not only improved the quality of treated water but also will help in reducing the cost of installation, operation and maintenance. Sponge column in unsaturated filtration mode removed 20% of organic matter from the effluent of GAC biofilter without the need of backwash and maintenance. The saturated sponge column and floating media column can eliminate about 10% TOC from effluent of the GAC biofilter. However, this was just an initial study with the simplest combined systems and experimental conditions. To develop a combined system of GAC and other filter medium biofilter, further study is necessary to modify the combined system and optimize the design and operational conditions. Other parameters such as nitrogen and phosphorous removal should be evaluated. This aspect is recommended in future works.

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# APPENDIX

## Appendix A

## Pilot scale biofilter

Evaluation of a pilot scale of biofilter was conducted in Tamagawa Water Works, Tokyo, Japan.

### A.1 Experimental conditions:

- Raw water: Tamagawa river water
- Filtration velocity: 17m/h
- Filter media: plastic media
- Filter bed depth: 115cm
- Filter diameter: 20cm
- Temperature: 13-20$^0$C

Figure A.1 The biofilter in Tamagawa water works

## A.2 Experimental result

Quality of water treated by the biofilter was assessed in terms of turbidity, DOC, UV absorbance (254nm), chlorine demand and major metal ions (Na, Mg, Al, K, Ca, Mn and Fe). The results of these measurements are presented in Tables A.1 and A.2.

Table A.1. Turbidity, DOC, UV and chlorine demand of influent and effluent of the biofilter.

| Date | Sample | Turbidity | | DOC | | UV254 (1cm cell ) | | Chlorine demand |
|---|---|---|---|---|---|---|---|---|
| | | NTU | Removal % | Conc. mg/L | Removal % | Abs | Removal % | mg/L |
| 11/11/ 2004 | Influent | 10.40 | 63.08 | 1.25 | 7.87 | 0.032 | 3.13 | |
| | Effluent | 3.84 | | 1.15 | | 0.031 | | |
| 15/11 | Influent | 2.70 | 58.15 | 1.29 | 9.07 | 0.039 | 23.08 | 4 |
| | Effluent | 1.13 | | 1.17 | | 0.030 | | 3 |
| 17/11 | Influent | 3.29 | 62.92 | 1.81 | 26.17 | 0.034 | 8.82 | 3 |
| | Effluent | 1.22 | | 1.34 | | 0.031 | | 1 |
| 19/11 | Influent | 3.98 | 72.61 | 1.25 | 18.36 | 0.043 | 11.63 | 2 |
| | Effluent | 1.09 | | 1.02 | | 0.038 | | 1 |
| 22/11 | Influent | 3.84 | 74.48 | 0.98 | 3.30 | 0.045 | 20.00 | 3 |
| | Effluent | 0.98 | | 0.95 | | 0.036 | | 1 |
| 24/11* | Influent | 4.60 | 60.43 | 1.37 | 28.32 | 0.049 | 18.37 | 3 |
| | Effluent | 1.82 | | 0.98 | | 0.040 | | 2 |

* Biofilter was backwashed 1 day before taking sample

The biofilter is effective in decreasing turbidity by about 60-70%. During the short term operation condition, low temperature, and low concentration of DOC in the raw water, the microbial community achieving in the biofilter was not significant to obtain high DOC removal efficiency. The DOC removal rate was less than 30%.

Chlorine demand for the raw water and biofilter effluent ranged from 2-4 mg/L and 1-2 mg/L respectively.

Table A.2. Result of major metal ion of river water and biofilter effluent

| Date | Sample | Metal ion | | | | | | |
|---|---|---|---|---|---|---|---|---|
| | | Na | Mg | Al | K | Ca | Mn | Fe |
| 11/11 | Influent | 16.76 | 4.91 | 0.06 | 3.31 | 25.03 | 4.83 | 0.00 |
| | Effluent | 15.16 | 4.34 | 0.03 | 2.88 | 22.34 | 4.49 | 0.00 |
| | Removal % | **9.58** | **11.73** | **58.92** | **13.13** | **10.73** | **7.02** | |
| 15/11 | Influent | 13.44 | 4.17 | 0.06 | 2.75 | 22.29 | 4.20 | 0.00 |
| | Effluent | 12.96 | 3.56 | 0.03 | 2.67 | 21.76 | 4.08 | 0.00 |
| | Removal % | **3.63** | **14.61** | **49.41** | **3.03** | **2.39** | **2.81** | |
| 17/11 | Influent | 13.21 | 4.13 | 0.04 | 3.18 | 22.16 | 4.18 | 0.00 |
| | Effluent | 13.15 | 3.58 | 0.04 | 2.83 | 22.47 | 4.21 | 0.00 |
| | Removal % | **0.48** | **13.24** | **3.57** | **10.97** | **0.00** | **0.00** | |
| 19/11 | Influent | 15.15 | 4.30 | 0.21 | 2.85 | 22.24 | 4.44 | 0.00 |
| | Effluent | 14.29 | 4.08 | 0.15 | 2.76 | 21.30 | 4.24 | 0.00 |
| | Removal % | **5.67** | **5.03** | **26.25** | **2.99** | **4.24** | **4.56** | |
| 22/11 | Influent | 3.08 | 4.42 | 0.31 | 3.22 | 34.41 | 1.94 | 1.22 |
| | Effluent | 2.90 | 4.04 | 0.31 | 2.97 | 30.06 | 1.94 | 1.30 |
| | Removal % | **5.90** | **8.64** | **0.00** | **7.62** | **12.65** | **0.00** | |
| 24/11 | Influent | 12.87 | 5.24 | 0.32 | 3.86 | 37.20 | 1.42 | 1.17 |
| | Effluent | 2.57 | 3.98 | 0.23 | 3.15 | 25.45 | 1.85 | 1.18 |
| | Removal % | **80.06** | **24.11** | **28.06** | **18.29** | **31.58** | **0.00** | |

Among major metal ions, Sodium and Calcium had the highest concentration in both the influent and effluent.

# Appendix B

# Bacterial identification using BIOLOG system

The positive results of bacterial identification using BIOLOG system are presented below. The BIOLOG results showed the identification of 8 species of bacteria from GAC media in the biofilter. The other closest species are also listed. Samples were taken from biomass on GAC in biofilter after 45 days operation.

**B.1 Brevibacterium otitidis**

Filter: biofilter for river water (Hawkesbury River) after 50 days

Incubation time: 16-24h

Plate type: GP2

Strain type: GP-ROD

Data generation mode: Manual

Number +/b/- reactions: 96/0/0

Database to search: Microlog

| | 1 | 2 | 3 | 4 | 5 | 6 | 7 | 8 | 9 | 10 | 11 | 12 |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| A | + | + | + | + | + | + | + | + | + | + | + | + |
| B | + | + | + | + | + | + | + | + | + | + | + | + |
| C | + | + | + | + | + | + | + | + | + | + | + | + |
| D | + | + | + | + | + | + | + | + | + | + | + | + |
| E | + | + | + | + | + | + | + | + | + | + | + | + |
| F | + | + | + | + | + | + | + | + | + | + | + | + |
| G | + | + | + | + | + | + | + | + | + | + | + | + |
| H | + | + | + | + | + | + | + | + | + | + | + | + |

⇨ Species identification: BREVIBACTERIUM OTITIDIS

Closest species

| | Species | PROB | SIM | DIST | TYPE |
|---|---|---|---|---|---|
| 1 | Brevibacterium otitidis | 100 | 0.800 | 3.00 | GP-ROD CAT+ |
| 2 | Brevibacterium mcbrellneri | 0 | 0.000 | 6.00 | GP-ROD CAT+ |
| 3 | Cellulomonas flavigena | 0 | 0.000 | 7.00 | GP-ROD CAT+ |
| 4 | Corynebacterium nitrolophilus | 0 | 0.000 | 7.00 | GP-ROD CAT+ |
| 5 | Cellulomonas cellulans | 0 | 0.000 | 8.00 | GP-ROD CAT+ |
| 6 | Rhodococcus rhodochrous | 0 | 0.000 | 9.00 | GP-ROD CAT+ |

**B.2 Enterobacter cloacae**

Feeding water of the filter: river water (Hawkesbury River)

Incubation time: 16-24h

Plate type: GN2

Strain type: GN-ENT

Data generation mode: Manual

Number +/b/- reactions: 70/0/26

Database to search: Microlog

| | 1 | 2 | 3 | 4 | 5 | 6 | 7 | 8 | 9 | 10 | 11 | 12 |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| A | - | - | + | + | + | + | + | + | - | + | - | + |
| B | - | + | + | + | + | + | + | + | + | + | + | + |
| C | + | + | + | + | + | + | + | + | + | - | + | + |
| D | + | + | + | + | + | + | + | - | + | - | + | - |
| E | + | - | - | - | - | + | + | - | - | + | - | + |
| F | + | + | + | + | + | + | + | + | + | + | + | + |
| G | + | - | - | + | - | - | + | + | + | + | - | - |
| H | -+ | + | + | + | - | - | - | + | + | + | + | + |

⇨ Species identification: ENTEROBACTER CLOACAE

Closest species

| | Species | PROB | SIM | DIST | TYPE |
|---|---|---|---|---|---|
| 1 | Enterobacter cloacae | 100 | 0.922 | 1.17 | GN-ENT |
| 2 | Klebsiella terrigena | 0 | 0.000 | 6.60 | GN-ENT |
| 3 | Klebsialla pneumoniae ss pneumoniae | 0 | 0.000 | 8.58 | GN-ENT |
| 4 | Enterobacter asburiae | 0 | 0.000 | 8.56 | GN-ENT |
| 5 | Enterobacter aerogenes (KLB. Mobilis) | 0 | 0.000 | 9.28 | GN-ENT |
| 6 | Hafnia alvei | 0 | 0.000 | 9.50 | GN-ENT |

**B.3 Enterobacter aerogenes (KLB. mobilis)**

Feeding water of the filter: river water (Hawkesbury River)

Incubation time: 16-24h

Plate type: GN2

Strain type: GN-ENT

Data generation mode: Manual

Number +/b/- reactions: 73/0/23

Database to search: Microlog

| | 1 | 2 | 3 | 4 | 5 | 6 | 7 | 8 | 9 | 10 | 11 | 12 |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| A | - | - | + | + | + | + | + | + | + | -+ | + | + |
| B | - | + | + | + | + | + | + | + | + | + | + | + |
| C | + | + | + | + | + | + | + | + | + | - | + | + |
| D | + | + | + | + | + | + | + | + | + | + | - | - |
| E | + | - | - | + | - | + | - | - | + | + | - | + |
| F | + | + | + | + | + | + | + | + | + | + | + | + |
| G | -+ | + | - | + | - | + | - | + | + | + | -+ | - |
| H | + | -+ | + | + | - | + | - | + | + | + | + | + |

⇨ Species identification: ENTEROBACTER AEROGENES (KLB. MOBILIS)

Closest species

| | Species | PROB | SIM | DIST | TYPE |
|---|---|---|---|---|---|
| 1 | Enterobacter aerogenes (KLB. mobilis) | 100 | 0.767 | 5.31 | GN-ENT |
| 2 | Klebsialla pneumoniae ss pneumoniae | 0 | 0.000 | 6.47 | GN-ENT |
| 3 | Klebsiella planticola | 0 | 0.000 | 7.94 | GN-ENT |
| 4 | Klebsiella planticola/ornithinolytica | 0 | 0.000 | 8.94 | GN-ENT |
| 5 | Serratia odorifera | 0 | 0.000 | 9.02 | GN-ENT |
| 6 | Enterobacter asburiae | 0 | 0.000 | 10.13 | GN-ENT |

**B.4 Staphylococcus epidermidis**

Feeding water of the filter: river water (Hawkesbury River)

Incubation time: 16-24h

Plate type: GP2

Strain type: GP-COCCUS

Data generation mode: Manual

Number +/b/- reactions: 14/0/82

Database to search: Microlog

| | 1 | 2 | 3 | 4 | 5 | 6 | 7 | 8 | 9 | 10 | 11 | 12 |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| A | - | - | - | + | - | - | - | - | - | - | - | - |
| B | - | - | - | - | + | - | - | - | - | - | + | - |
| C | + | - | + | + | - | + | - | - | - | - | + | - |
| D | - | - | - | + | - | - | + | - | - | - | - | + |
| E | - | - | -+ | - | - | - | - | - | - | - | - | - |
| F | - | - | - | - | - | + | - | - | + | - | - | - |
| G | - | - | - | - | - | - | - | - | - | - | - | + |
| H | - | - | - | - | - | - | - | - | - | - | - | - |

⇨ Species identification: STAPHYLOCOCCUS EPIDERMIDIS

Closest species

| | Species | PROB | SIM | DIST | TYPE |
|---|---|---|---|---|---|
| 1 | Staphylococcus epidermidis | 100 | 0.869 | 1.95 | GP-COC CAT+ |
| 2 | Streptococcus oralis | 0 | 0.000 | 6.13 | GP-ROD CAT+ |
| 3 | Staphylococcus warneri | 0 | 0.000 | 6.34 | GP-ROD CAT+ |
| 4 | Aerococcus viridans | 0 | 0.000 | 6.60 | GP-ROD CAT+ |
| 5 | Staphylococcus hominis ss novobiosepticus | 0 | 0.000 | 6.82 | GP-ROD CAT+ |
| 6 | Staphylococcus pasteuri | 0 | 0.000 | 6.99 | GP-ROD CAT+ |

**B.5 Pseudomonas aeruginosa**

Feeding water of the filter: river synthetic wastewater

Incubation time: 16-24h

Plate type: GN2

Strain type: GNP-NENT OXI+

Data generation mode: Manual

Number +/b/- reactions: 48/3/45

Database to search: Microlog

| | 1 | 2 | 3 | 4 | 5 | 6 | 7 | 8 | 9 | 10 | 11 | 12 |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| A | - | - | - | + | + | + | - | + | - | - | - | - |
| B | - | + | - | - | - | + | - | - | - | - | + | + |
| C | - | - | - | - | - | - | - | - | - | - | + | + |
| D | + | + | + | | - | - | + | - | - | + | + | - |
| E | + | + | - | + | + | + | + | + | + | - | + | + |
| F | + | + | | + | + | + | - | + | + | + | - | - |
| G | + | + | | -+ | - | + | + | - | + | + | + | + |
| H | + | + | - | - | - | + | + | + | + | - | - | - |

⇨ Species identification: PSEUDOMONAS AERUGINOSA

Closest species

| | Species | PROB | SIM | DIST | TYPE |
|---|---|---|---|---|---|
| 1 | Pseudomonas aeruginosa | 100 | 0.867 | 1.98 | GNP-NENT OXI+ |
| 2 | Pseudomonas citronellolis | 0 | 0.000 | 11.26 | GNP-NENT OXI+ |
| 3 | Pseudomonas putida | 0 | 0.000 | 11.58 | GNP-NENT OXI+ |
| 4 | Pseudomonas fulva | 0 | 0.000 | 12.37 | GNP-NENT OXI+ |
| 5 | Pseudomonas putida biotype A | 0 | 0.000 | 12.53 | GNP-NENT OXI+ |
| 6 | Pseudomonas maculicola | 0 | 0.000 | 13.29 | GNP-NENT OXI+ |

**B.6 Pseudomonas alcaligenes**

Feeding water of the filter: river synthetic wastewater

Incubation time: 16-24h

Plate type: GN2

Strain type: GNP-ALL

Data generation mode: Manual

Number +/b/- reactions: 33/0/63

Database to search: Microlog

| | 1 | 2 | 3 | 4 | 5 | 6 | 7 | 8 | 9 | 10 | 11 | 12 |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| A | - | - | - | + | + | + | - | - | - | - | - | - |
| B | - | - | - | - | - | - | - | - | - | - | - | - |
| C | - | - | - | - | - | - | - | - | - | - | + | + |
| D | + | + | + | - | - | - | - | - | - | + | + | + |
| E | - | - | - | -+ | -+ | + | - | + | - | - | - | + |
| F | + | + | | + | + | + | + | + | + | -+ | - | - |
| G | + | + | | + | + | + | - | - | + | + | - | + |
| H | + | - | - | - | - | + | - | - | - | - | - | - |

⇨ Species identification: PSEUDOMONAS ALCALIGENES

Closest species

| | Species | PROB | SIM | DIST | TYPE |
|---|---|---|---|---|---|
| 1 | Pseudomonas alcaligenes | 100 | 0.557 | 6.96 | GNP-NENT |
| 2 | Psychrobacter immobilis | 0 | 0.000 | 14.19 | GNP-NENT OXI+ |
| 3 | Aquaspirillum dispar | 0 | 0.000 | 14.81 | GNP-NENT OXI+ |
| 4 | Aquaspirillum putridiconchylium | 0 | 0.000 | 14.84 | GNP-NENT OXI+ |
| 5 | Acinetobacter calcoaceticus BV ALC | 0 | 0.000 | 15.06 | GNP-NENT OXI+ |
| 6 | Acinetobacter calcoaceticus/genospecies 1 | 0 | 0.000 | 15.55 | GNP-NENT OXI+ |

**B.7 Streptococcus oralis**

Feeding water of the filter: river synthetic wastewater

Incubation time: 16-24h

Plate type: GP2

Strain type: GP-COCCUS CAT-

Data generation mode: Manual

Number +/b/- reactions: 33/0/63

Database to search: Microlog

| | 1 | 2 | 3 | 4 | 5 | 6 | 7 | 8 | 9 | 10 | 11 | 12 |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| A | - | - | - | + | - | - | - | - | - | - | - | - |
| B | - | - | - | - | + | - | - | - | - | - | -+ | - |
| C | + | - | + | + | - | + | - | - | - | - | + | - |
| D | - | - | - | + | - | - | + | - | - | - | - | + |
| E | - | -+ | - | - | - | - | - | - | - | - | - | - |
| F | - | - | -+ | - | - | + | - | - | + | - | - | - |
| G | - | - | - | - | - | - | - | - | - | - | - | + |
| H | - | - | - | - | - | - | - | - | - | - | - | - |

⇨ Species identification: STREPTOCOCCUS ORALIS

Closest species

| | Species | PROB | SIM | DIST | TYPE |
|---|---|---|---|---|---|
| 1 | Streptococcus oralis | 100 | 0.506 | 6.84 | GP-COCCUS CAT- |
| 2 | Aerococcus viridans | 0 | 0.000 | 7.60 | GP-COCCUS CAT- |
| 3 | Streptococcus pneumoniae | 0 | 0.000 | 8.76 | GP-COCCUS CAT- |
| 4 | Streptococcus agalactiae(GPB) | 0 | 0.000 | 9.09 | GP-COCCUS CAT- |
| 5 | Streptococcus sanguinis | 0 | 0.000 | 9.18 | GP-COCCUS CAT- |
| 6 | Streptococcus pyogenes (GPA) | 0 | 0.000 | 9.39 | GP-COCCUS CAT- |

**B.8 Kocuria rosea/ erythromyxa**

Sample: biomass on GAC of the batch with river water (Hawkesbury River)

Incubation time: 16-24h

Plate type: GP2

Strain type: GP-COCCUS Data generation mode: Manual

Number +/b/- reactions: 95/0/1

Database to search: Microlog

| | 1 | 2 | 3 | 4 | 5 | 6 | 7 | 8 | 9 | 10 | 11 | 12 |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| A | - | + | + | + | + | + | + | + | + | + | + | + |
| B | + | + | + | + | + | + | + | + | + | + | + | + |
| C | + | + | + | + | + | + | + | + | + | + | + | + |
| D | + | +- | + | + | + | + | + | + | +- | + | + | + |
| E | + | + | + | + | + | + | + | + | + | +- | + | + |
| F | + | + | + | + | + | + | + | + | + | + | + | + |
| G | + | + | + | + | + | + | + | + | + | +- | + | + |
| H | + | + | + | + | + | + | + | + | + | + | + | + |

⇨ Species identification: KOCURIA ROSEA/ ERYTHROMYXA

Closest species

| | Species | PROB | SIM | DIST | TYPE |
|---|---|---|---|---|---|
| 1 | Kocuria rosea/ erythromyxa | 100 | 0.734 | 4.00 | GP-COCCUS CAT+ |
| 2 | Staphylococcus lentus | 0 | 0.000 | 6.00 | GP-COCCUS CAT+ |
| 3 | Dermacoccus nishinomiyaensis | 0 | 0.000 | 8.00 | GP-COCCUS CAT+ |
| 4 | Staphylococcus sciuri | 0 | 0.000 | 16.00 | GP-COCCUS CAT+ |
| 5 | Staphylococcus gallinarum | 0 | 0.000 | 18.00 | GP-COCCUS CAT+ |
| 6 | Kytococcus sedentarius | 0 | 0.000 | 18.00 | GP-COCCUS CAT+ |