CONTACT FLOCCULATION FILTRATION USING NATURAL COAGULANTS FOR DEVELOPING COUNTRIES

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ABSTRACT

Contact flocculation filtration using natural coagulants for developing countries.

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Contact flocculation filtration using natural cationic polyelectrolytes extracted from seeds of the tree *M.oleifera* were found to be effective in the treatment of low turbidity waters. The coagulant was dosed immediately prior to the filter inlet, with subsequent flocculation and deposition occurring in the filter bed. This single stage treatment option was considered appropriate for developing countries, due to observed treatment performance, robustness of operation and reduced treatment costs. This work extends and complements previously successful studies on the treatment of medium to high turbidity raw waters using *M.oleifera* seed.

Laboratory studies using twin 100 mm diameter filter columns, were undertaken with the following variables: turbidities of 5-75 NTU; filtration rates of 5-20 m/h; filter depths of 70 and 120 cm; dual and single media beds, and media sizes of 0.50-1.00 mm and 0.85-1.70 mm. Deeper beds and smaller media were found to considerably reduce filtrate turbidity when using *M.oleifera* seed. The consequent headloss increase was only significant with the higher turbidity waters; dual media beds were most effective on such waters. Turbidity removal was reduced at higher filtration rates (10m/h), due to lower retention times in the bed, and increased detachment of retained particles causing early turbidity breakthrough. Higher filtration rates with another natural coagulant, chitosan, increased turbidity removal and prolonged the time to breakthrough, due to the reduction in surface removal in the filter. At lower loading rates (5 NTU at 5 m/h), removal with *M.oleifera* seed was comparable with chitosan and aluminium sulphate, with the additional advantage of a lower headloss. Optimisation of the hydraulic variables for a specific coagulant was considered necessary to ensure maximum output and filtrate quality.

Field trials on a low turbidity natural raw water indicated that *M.oleifera* reduced the turbidity by >95% and bacterial numbers by 100% at the optimum dose. Despite the rise in organic matter in the final water, trihalomethane levels were not excessive.

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CHAPTER 1

INTRODUCTION

1.0 Introduction

1.3 billion people in the developing world lack safe drinking water (IRC, 1994) and 80% of disease in developing countries is as a result of an inadequate water supply and sanitation (Schertenleib, 1992).
These are facts that cannot be ignored, considering a global population increase of 1.7% (IRC, 1994).

The availability of clean and safe drinking water is a necessity of life, and is generally taken for granted in the industrialised world. Costs are not the most important consideration, and sophisticated treatment processes with expensive chemicals are often utilised. The focus in treatment development terms, has shifted toward removal of chemicals present in minute quantities, which may cause chronic illnesses. Examples include disinfection by-products (DBP) and pesticides.

The situation in many developing countries is very different, with approximately 2,000,000 cases of infant mortality per year due to the consumption of contaminated water (World Development Report, 1992). It is predicted that 80% of the world's population will be living in developing countries by the year 2000, the majority in rural and suburban regions (WHO, 1981). Populations in developing countries are generally more susceptible to water borne diseases than those in developed countries, due to malnutrition induced low immunity. The situation is often exacerbated by natural disasters and political upheaval, which can lead to high numbers of displaced peoples.

The treatment of water for potable purposes in developing countries is fraught with problems. Three distinct reasons have been identified by Schulz & Okun (1992). Firstly, the shortage of skilled labour to operate and maintain water treatment works (WTW), coupled with an abundance of unskilled labour especially in rural areas. Secondly, water utilities are ineffective bodies due to lack of finance and a high turnover of staff. Thirdly, and most importantly, there is a lack of finance for the operation of WTW. It is estimated that the ability of consumers in the developing world to pay for water treatment is between a fifth and one-twenty fifth of that in the Western World (Wagner, 1982a). This not only affects the technologies that can be used, but also the chemicals that are purchased. Often WTW in developing countries have been designed and constructed by western engineers, using technologies appropriate for

industrialised countries. Furthermore the high costs of importation of many of the chemicals used are prohibitively expensive.

It is now regarded as axiomatic by many development bodies that WTW and treatment processes should be as simple, robust, affordable and most importantly reliable (Williams, 1985; Ellis, 1990). This does not necessarily mean the development of new technologies, but rather the innovative application of proven ones. According to Arboleda (1976) and Wagner (1982a) this involves, amongst other factors, the use of indigenous material for construction and operation of the plant. One example is the use of locally available, natural chemicals to act as coagulants in the treatment process.

Natural coagulants are extracts of plant or animal origin, some of which have been in use for many centuries, while others have a more recent history. The natural coagulant used in this study is extracted from the seeds of the tropical tree *Moringa oleifera* Lam. (*M.oleifera*) (syn. *M.pterygosperma* Gaertn). The seeds are reported to have been used to clarify water at household level in Sudan at the beginning of this century (Jahn & Hamid, 1979; Jahn, 1981). The many advantages to developing countries of using natural coagulants, and especially *M.oleifera*, are discussed fully in Chapter 2. Briefly, these are a reduction in costs due to the use of indigenous materials, and the bolstering of the local economy leading to sustainable development. Environmental concerns are of low priority for many developing countries, as a consequence of the immediate pressures of population growth and economic survival. This matter was considered at the UN's 'Earth Summit' 1992 in Rio de Janeiro (Tebbutt, 1998). It was concluded that sustainable practices need to be established now, in order to avoid the problems presently faced by the West, caused by mismanagement of their resources during their economic development.

Surface water is often the only available source for drinking water in many developing countries, since exploitation of ground water sources can incur high costs. Surface waters are characterised by a high particulate load and a correspondingly high concentration of pathogenic organisms in the rainy season. In the dry season, however, the particulate load is often low but the water may still be microbially contaminated. Conventional treatment systems incorporating coagulation, flocculation, solid-liquid separation and disinfection are effective on waters with a high solid load. Treatment generally includes the addition of a chemical coagulant to promote agglomeration of the particulate matter, followed by subsequent removal

in sedimentation tanks and filter beds (discussed more fully in Section 2.2). Using the same treatment on waters with a low particulate content is inherently inefficient, involving excessive use of coagulants to provide effective treatment. Thus alternative treatment processes are sought for waters with low particulate loads and turbidity. Turbidity is a measure of the quantity of suspended material in the water such as clay, silt, colloidal particles and microscopic organisms. It is defined as "an optical property of the sample causing light to be scattered and absorbed not transmitted" (15th Edition of Standard Methods, 1980). It is also dependent upon the size, shape and refractive index of the particles and is generally measured in Nephelometric Turbidity Units (NTU). The significance of turbidity on water quality is confirmed by the high correlation between turbidity, colour, odour, taste problems, and bacterial content. It is the most commonly used parameter in determining effectiveness of treatment, as measurement is simple and rapid.

Alternative treatments of low turbidity waters consist of slow sand filters (SSF) and direct filtration (DF), consisting of coagulation, flocculation, filtration and disinfection. The former has the disadvantage of using a large area of land due to the low flow rates required for effective treatment (0.1-0.2 m/h). Furthermore only very low turbidity waters can be treated by this process; 10 NTU is generally regarded as the maximum (Huisman & Wood, 1974). The latter has the disadvantage of requiring a flocculation stage with supplementary chemicals and coagulants, incurring additional construction and maintenance costs. A further alternative is contact flocculation filtration (CFF). This is a process whereby the coagulant is dosed immediately prior to the raw water entering the filter bed. Subsequent flocculation and deposition occurs when the particles are in 'contact' with the filter media. The main advantage of this is that individual flocculation and sedimentation stages are not required, thus reducing construction and operational costs. The requirement for chemical coagulants to ensure optimal operation of the filters is a disadvantage of the process compared to SSF. Coagulants are not always readily available and also can add significantly to the operational costs. Consequently the use of the natural coagulant extracted from the seeds of tree *M.oleifera* was investigated in conjunction with the CFF process for low turbidity water in developing countries.

1.1 Aims & objectives of the research

Previous work at Leicester University has been undertaken into the effectiveness of *M.oleifera* seed on high turbidity waters, using conventional treatment processes. It was therefore necessary to conduct tests on low turbidity waters, in order to assess its full potential for use in water treatment. As discussed, the use of conventional treatment is inefficient for use on low turbidity waters, therefore the feasibility of using CFF was considered as an alternative treatment.

The first phase of this study was laboratory based, using a pilot rig in the laboratory at Leicester. Preliminary investigations had been undertaken into the use of *M.oleifera* seed in the CFF process using a small filter column operated as a batch process. The initial results were most encouraging and consequently the rig was developed further to allow a fuller investigation to be undertaken. Chapter 3 details the modifications undertaken on the filter rig as well as the development of the methods and protocols used in the study. The physical parameters investigated included varying media size and type, bed depth and configuration, and also flow rate. The effects of changing these parameters and conditions on residual turbidity and headloss development are considered in Chapter 4. The water used in the laboratory consisted of a kaolin clay suspension diluted in deionised water. This was used since it would not have been practical to obtain a natural low turbidity water in sufficient quantities. Furthermore it was essential to maintain a consistent raw water quality throughout the study. The use of synthetic turbid suspensions is well established; it has been found to have similar characteristics to river water samples as a result of the presence of kaolin clay in weathered tropical soils (Weglin, 1986).

The raw water turbidities considered in the laboratory study were between 5-75 NTU at filtration rates of 5-20 m/h. The bed depths used were 70 and 120 cm. These parameters were chosen as they represent typical values encountered in the operation of the majority of deep bed filters. The turbidity values were chosen to represent waters that may typically be treated using CFF. Waters with turbidities of less than 5 NTU, would only require disinfection. Upper limits of raw water turbidities which can be treated by DF vary from 25-200 NTU (Schulz & Okun, 1992; Culp, 1977; McCormick and King, 1982). Excessively higher raw water turbidities would require a separate sedimentation stage before filtration. Most rapid flow

filters (RFF) operate at flow rates of between 5-10 m/h, although Montgomery (1985) states that filter runs of 30 m/h are possible if adequate pretreatment is provided. For this research, flow rates of 5-20 m/h were considered, with the emphasis on filtration rates of 5 and 10 m/h. The results described in Chapter 4 indicated that the combination of CFF with *M.oleifera* seed was effective. In order to put this into context, *M.oleifera* seed application was compared with two other coagulants, namely aluminium sulphate (alum) and chitosan, an extract of crustacean shells. This comparison is detailed and discussed in Chapter 5.

The second phase of the work consisted of a field study using a natural raw water. The kaolin suspension used in the laboratory study is not entirely representative of natural waters, due to the lack of organic content amongst other factors. It had been anticipated that the pilot study would be conducted at Thyolo WTW in southern Malawi during the dry season. The previous study conducted by Leicester University on the treatment of high turbidity water was undertaken at this Works. But this additional visit to Malawi was not possible for various reasons, including economic instability in Malawi, which may have lead to the study becoming very expensive. Much of the equipment used previously at the Works had been damaged or lost and would therefore have needed replacing. Many of the roads in the area were damaged and had become impassable during the previous rainy season, making transport to the Works potentially very difficult. During the course of the laboratory study, the opportunity arose to use an established pilot facility at the Felindre WTW in South Wales, under the auspices of Dwr Cymru Welsh Water Plc. Situated within the main works, the location offered the opportunity for contact and dialogue with various skilled members of staff. The plant also had the advantage of consisting of 4 pilot columns, that could be operated concurrently, increasing the amount of data obtainable from a single run. It was also possible to measure parameters other than turbidity, such as bacteria removal, organic matter and the formation of THM precursor. Details of this study are given in Chapter 6.

1.1.1 Specific objectives

The specific objectives of the pilot study conducted in the laboratory at Leicester University were as follows:

- To establish the range of raw water turbidities that can be treated using the CFF and the natural coagulant *M.oleifera*.

- To establish the maximum filtration rate at which the pilot plant can be operated and maintain effective treatment.

- To determine the potential benefits that may be obtained by increasing the filter bed depth and filter media size.

- To determine the potential benefits of utilising dual media beds of sand and anthracite as compared to single media sand beds.

- To understand more fully the mechanism of floc formation and removal induced by the coagulant extracted from *M.oleifera* seed. Spatially and temporally distributed turbidity and headloss measurements were used for this purpose.

- To optimise the coagulant dose for a range of initial turbidities and flow rates.

- To compare the effectiveness of *M.oleifera* seed with another natural cationic polyelectrolyte - chitosan. Effectiveness was determined in terms of the turbidity of the filtrate, the headloss development rate and the volume of water treated before reaching one of the predetermined termination criteria. The effectiveness of backwashing the filter bed was also considered.

- To compare the effectiveness of *M.oleifera* seed with the proprietary coagulant - alum. This widely used coagulant was compared in terms of the minimal filtrate turbidity achievable, the headloss development rate and the volume of the water treated before reaching one of the predetermined termination criteria. This was evaluated at two raw water turbidities and two filtration rates.

The primary objective of the second phase of work at the pilot plant facility was to complement the research conducted in the laboratory by using a natural water. A low turbidity, natural raw water was used. Specific objectives of the field study included:

- To compare the effectiveness of *M.oleifera* seed with ferric aluminium sulphate, the coagulant used in the main works at Felindre WTW. Effectiveness was determined in terms

of filtrate turbidity and rate of headloss development in the filter bed. Other parameters measured included the degree of bacteria and colour removal. The residual organic carbon present in the final water and hence the potential reduction of THM precursors were also measured.

- To compare the effectiveness of DF and CFF as treatment processes for both *M.oleifera* seed and ferric aluminum sulphate (the coagulant used at Felindre WTW). The effect of including a slow mix (flocculation) stage in the treatment process was therefore investigated.

CHAPTER 2

LITERATURE REVIEW

2.0 Literature Review

2.1 Requirement for water treatment

Raw waters often contain high concentrations of pathogens, but disinfection alone is not always sufficient to render the water suitable for potable purposes. This is particularly so when the water has a high turbidity. The pathogenic organisms are often closely associated with particulate matter that can act as a physical shield against oxidation by the disinfectant. Organic particulate matter present in the water may be oxidised preferentially to the microorganisms, resulting in the need for a large dose of the disinfectant to be used in order to ensure deactivation of the pathogens. Oxidation of the organic matter in the water is itself undesirable. It may lead to the formation of the potentially carcinogenic DBP or organochlorines of which trihalomethanes (THMs) are the most common (Rook, 1974; Bellar, 1974). Maximum removal of suspended matter is therefore an essential requirement of the treatment process before disinfection can occur. Particulate removal in the majority of treatment works is the most costly and lengthy stage of the treatment process.

Figure 2.1 illustrates the treatment train most commonly found at treatment works and is referred to as conventional treatment (AWWA, 1990). The water entering the mixing stages generally contains particles with diameters < 10 μm, and chemical coagulants are added to aid their removal. Immediately after the addition, the water is rapidly mixed in the coagulation stage. The purpose of this relatively short step, ranging from a few seconds to a few minutes, is to destabilise suspended particles such that aggregation can occur. The next step is the flocculation stage, where gentle agitation of the destabilised particles allows the formation of particle agglomerates or floc (from the Latin word 'flocculus' meaning 'tufts of wool' (Purchas, 1981)).

The flocculated water then enters the solid-liquid separation stage. The first step here is clarification, where the majority of the solid load is removed, by settling (sedimentation) or flotation of the particulates. The remaining solids are then removed in the filtration step. The purpose of the clarification step is to reduce the load entering the filter bed, which is relatively expensive to clean in terms of the washwater requirement.

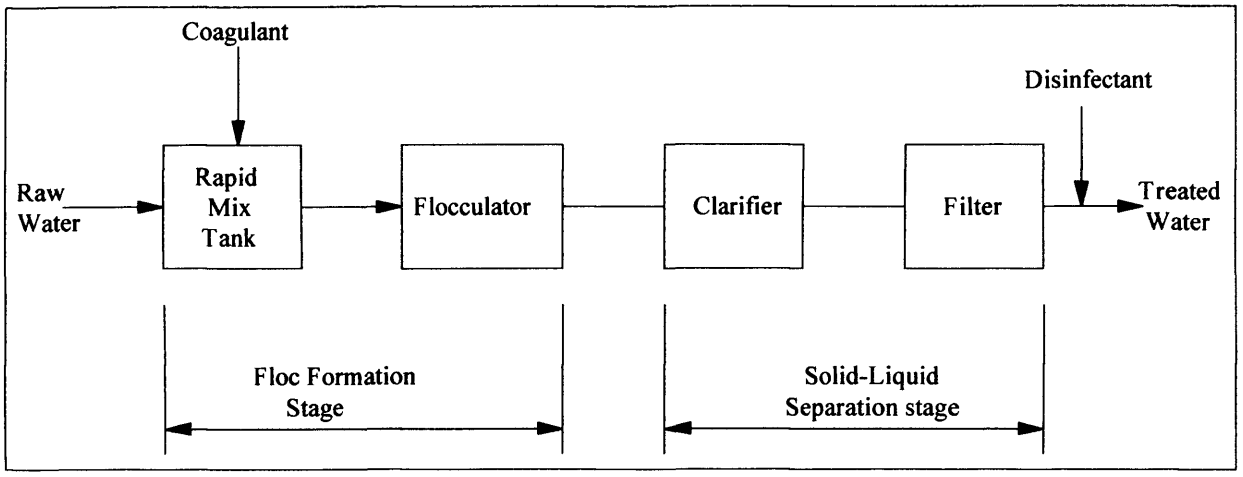

Figure 2.1 Basic schematic of a conventional water treatment works.

(Source: adapted from Edzwald, 1986).

Disinfection is the final and most important stage in rendering the water potable. It generally consists of chlorination, although other forms of disinfection are often used, e.g. ozone or ultra violet radiation.

2.1.1 Characteristics of the particulates found in water

The particulates found in raw waters can generally be divided into two groups based on size: suspended solids which are particles > 2.5 μm, and colloidal particles which range from 5 nm to 2.5 μm. Particles smaller than 5 nm are considered to be in true solution. Particles larger than about 1 μm can be removed relatively simply by sedimentation and filtration. Smaller particles will not settle or be filtered out, but will remain in suspension indefinitely unless aggregation is utilised to facilitate their removal (Montgomery, 1985). Table 2.1 illustrates the expected time intervals for various sized particles to settle. Clearly in order to produce high quality water within a reasonable time span, aggregation of the particles into larger agglomerates must be promoted. The majority of solids encountered in water have a net negative charge on their surface, with the consequence that repulsion occurs between particles. These are termed stable particles, as they will not aggregate unless this surface charge is reduced. Chemicals are added to the water in order to cause the colloidal particles to aggregate together, and allow their removal.

The particles in natural waters can be further classified as organic and inorganic particles. The former consists of colloidal particles such as proteins, humic and fulvic acids, viruses and larger suspended biological matter and microorganisms. They are therefore of considerable concern in water treatment. Organic particles are usually hydrophilic in nature, i.e. they have a high affinity for water molecules and lack a clear boundary at the interface with the aqueous phase. The water molecules bound to the surface of the particle, act as a barrier to collisions with other particles. This makes these particles relatively stable and consequently difficult to remove from the water.

Order of Size	Diameter of Particle (mm)	Total Surface Area[1]	Time Required to Settle[2]
Gravel	10	3.14 cm^2	0.3 seconds
Coarse sand	1	31.4 cm^2	3 seconds
Fine sand	0.1	314 cm^2	38 seconds
Silt	0.01	0.314 m^2	33 minutes
Bacteria	0.001	3.14 m^2	55 hours
Colloidal particles	0.0001	314 m^2	230 days
Colloidal particles	0.00001	0.283 ha	6.3 years
Colloidal particles	0.000001	2.83 ha	63 years minimum

Table 2.1 Effects of decreasing the size of a sphere on settling rates.

(Source: Adapted from AWWA, 1971).

[1] Area for particles of indicated size produced from a particle 10 mm in diameter with a specific gravity of 2.65.
[2] Calculations based on sphere with a specific gravity of 2.65 to settle 30 cm.
[3] Colloids: from the Greek word *'kola'* meaning glue, are stable insoluble substances. They are so minute that the random motion of the water molecules are sufficient to keep them from settling under gravity.

Inorganic particles such as clays and silts are mostly hydrophobic (water repelling) in nature. These particles have a clear boundary with the aqueous phase and are therefore relatively simple to separate from the water. Some inorganic particles, however, are hydrophilic as water molecules will bind to their surfaces (Stumm & Morgan, 1981).

In practice there are not always clear classification divisions. Organic biocolloids such as bacteria, algae or humic substances adsorb onto the surfaces of inorganic particles and the product exhibits heterogeneous surface properties. Hydrophilic colloids often envelop hydrophobic colloids, since the latter actively repel water molecules and attach to other

colloids. Such agglomerations are then very difficult to destabilise and remove from the aqueous phase. This is explained in the next section.

2.1.2 Particle stability

The stability of hydrophobic particles is mainly a consequence of electrostatic repulsion between particles. Anions or cations can accumulate at the surface of particles and electrostatically repel particles with similar charges. Hydrophilic particles remain stable partly as a consequence of the electrical charges on their surfaces from the disassociation of inorganic groups, but mainly due to their high affinity for water molecules.

Charged particles in solution are counter-balanced by clouds of ions of opposite charge as a result of the need for electro-neutrality. Thus charged particles have counter-ions surrounding them forming what is known as an 'electrical double layer', which extends into the solution and its concentration is in proportion to the primary charge of the particle (Kruyt, 1952). The ions are held in place by weak electrostatic forces. These are easily broken as the particle moves through the solution as a result of thermal agitation. Figure 2.2 illustrates the double layer surrounding a negatively charged particle. It consists of a fixed layer of cations adjacent to the surface of the particle, known as the 'Stern' layer. Surrounding this is a layer of moveable diffuse ions which extends out into the bulk solution until electro-neutrality is reached, forming the Gouy-Chapman layer (Gouy, 1910; Stern, 1924). From Figure 2.2 it can be seen that there is a rapid drop in potential at the plane of shear between the particle surface and the diffuse layer, and a more gradual drop between the diffuse layer and the point of electroneutrality. The overall potential between the surface of the particle and the bulk solution is the Nerst potential and the smaller potential between the Stern layer and the Gouy-Chapman layer is the zeta potential. When two particles with the same primary charge are brought together, the electrical double layers begin to interact. If the zeta potential exceeds the attractive van der Waals' forces, then repulsion ensues and the particles are said to be stable. Van der Waals' forces are very weak and are possessed by all molecules in the universe, regardless of charge or composition. They arise when two molecules approach each other and slight charges are temporarily induced due to small movements on the electrons surrounding each molecule. The force only comes into effect when the particles are within relative close proximity to each other and is inversely proportional to the distance between particles. Its magnitude is determined by the density and composition of the particle and not the aqueous phase in which it is suspended.

If the electrostatic repulsive forces can be overcome, then the van der Waals' forces will bring the particles together and the particles will make contact and form floc.

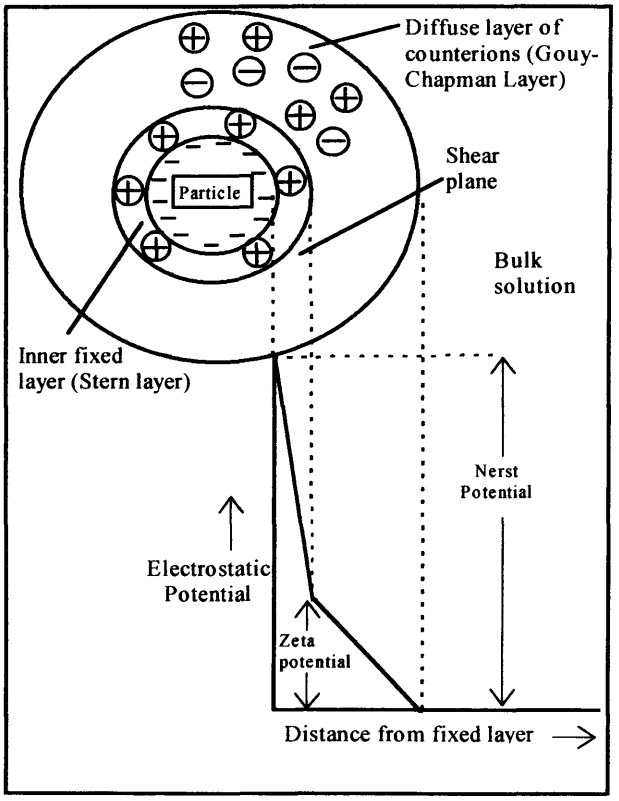

Figure 2.2 Schematic showing the potentials which exist at the solid-liquid interface.

(Source: AWWA, 1992).

One mechanism of reducing the repulsive forces is by using coagulants. These can act by allowing counter-ions to specifically adsorb onto the particle surface, hence reducing or changing the surface charge, or by increasing the ionic strength of the solution in order to reduce the zeta potential of the particle. These mechanisms will be discussed more fully in Section 2.2.2.

2.2 Coagulation and flocculation in water treatment

The terms 'coagulation' and 'flocculation' are often used interchangeably, to describe the processes of destabilisation and particle aggregation. The definitions used in this research are based on those devised by Packham (1963); such that coagulation is the term used to describe the process of reducing the zeta potential of stable particles in order to cause their destabilisation. Whereas flocculation is the term used to describe the physical processes that aggregate these electrostatically destabilised particles to form floc.

2.2.1 Coagulants

As previously noted from Table 2.1, colloidal particles will remain in suspension and not settle out unless destabilised. Removal of colloidal particles is achieved by the addition of chemicals known as coagulants. Coagulants function by reducing the electrical repulsion between particles and bring about their aggregation. The most commonly used coagulants in water treatment are metallic salts and polymers.

The metal salts are inorganic chemicals and consist mainly of hydrolysed aluminium or ferric ions. The structure and behavior of these ions have been researched extensively since the 1960s (Black, 1961; Stumm & O'Melia, 1967; Jiang & Graham, 1996; Srinivasan *et al.*, 1999). Alum is considered to be the most important and widely used coagulating agent worldwide (AWWA, 1992). In common with other metal coagulants it has a proven track record, is cost effective (Herman. 1986), relatively easy to handle and is generally readily available in developed countries (Kawamura, 1991). Furthermore floc formation is rapid and is effective in the treatment of low turbidity waters with a high colour and organic matter concentration. (Logsdon *et al.*, 1993). It is also effective in removal of undesirable chemicals, e.g. fluoride (Williams & Culp, 1986).

The drawbacks with the metal coagulants are that they are pH dependent, the floc can be weak, particularly in cold waters, and large volumes of sludge are produced as a by-product. This causes the dual problem of increased costs associated with disposal of the sludge and also rapid clogging of filter media leading to the requirement for more frequent backwashing. The sludge is also difficult to dewater due to its gel-like properties (Bache *et al.*, 1996).

The specific disadvantage of aluminium based coagulants is the concern over residual aluminium species (both soluble and insoluble) remaining in the treated water. Their occurrence has been linked with various neuro-degenerative diseases such as parkinsonian dementia, amyotrophic lateral sclerosis and Alzheimer's disease (Vogt, 1986; McLachlan *et al.*, 1992). A recent study by the French Institute of Health & Medical Research has found a higher than average incidence of dementia characteristic of Alzheimer's in an area in south west France. The water in this region has a concentration of aluminum >100 μg/L, which although considered to be relatively high, is still below the European standard of 200 μg/L (Peigne-Séraline, 1998). The report has refueled the debate on lowering the maximum allowable concentration (MAC) of aluminum in the drinking water. However, to date there is still no conclusive evidence showing a positive link between aluminium and Alzheimer's disease, and no firm conclusions can be drawn from the numerous studies which have been undertaken (Srinivasan *et al.*, 1999).

The case of the accidental discharge of concentrated alum into the treated water in Camelford in Cornwall, UK in 1988 (Gray, 1994), and the resulting contamination of the water highlighted a further problem. Particular care must be taken in its use and storage to ensure that such incidents do not occur. The adverse publicity arising from reports of the effects of aluminum on human health has led many water companies to seek alternative coagulants (Letterman & Driscoll, 1988). For example, Thames Water has been attempting to phase out its use in all their treatment works over the last few years (Gray, 1994).

Ferric chloride is another commonly used metal coagulant. Ferric salts are considered to be more effective at the removal of natural organic matter (Crozes *et al.*, 1995). But they have the disadvantage of requiring complete removal from the water, since even low residual concentrations can cause taste and colour problems in the supply. Iron can also lead to corrosion problems in the pipework and the salts can be contaminated with other metals, e.g. chromium, nickel, manganese and lead, all of which find their way into the finished water.

Presently much research is being conducted into the production of pre-polymerised aluminium and ferric coagulants (Berrak, 1992; Jiang, 1995; Jiang *et al.*, 1996). These have many advantages over conventional metal salts, such as lower residual aluminium and iron concentrations in the treated water, a wider working pH and better floc separation. However

there are also some disadvantages associated with their use, such as their high costs and a lack of operational experiences in their use.

Polymers are also often used as coagulants in water treatment. These are long chain molecules consisting of between 10^2 to 10^5 repeating molecular units (monomers) bonded co-valently. Examples of polymers are polyacrylamides and polyamines. Polyelectrolytes are charged polymers with water soluble organic or inorganic ionisable groups attached to them, such as carboxyl or ammonia. The overall charge on the polymer is dependent upon the pH and the ionic strength of the solution. Polymers with a preponderance of negative charges are called anionic polyelectrolytes and those with positive sites are cationic polyelectrolytes. Nonionic polymers are those with no, or very small overall charges.

Polymers are not effective in the treatment of all waters. Despite forming strong floc their effectiveness is limited with low turbidity and highly coloured waters (Edzwald *et al.*, 1987). There are also concerns about the long term toxicity and carcinogenicity of using synthetic polymers, as well as the possibility of contamination from the manufacturing process (Letterman & Pero, 1990). Japan has adopted regulations against the use of synthetic polymers in the treatment of water for potable purposes, and certain polymers have been banned in Germany and the Netherlands (Jackson, 1992). Natural coagulants are therefore an attractive alternative to synthetic polymers. There has been an increase in interest in recent times in their use for water treatment; reasons for which are considered below:

- Natural coagulants are often extracts of indigenous materials which are potentially more readily available in some parts of the world than proprietary chemical coagulants. This is of major importance to many developing countries, where imported chemical coagulants are not easily available and can be expensive, involving the use of valuable foreign exchange. The cost of alum, the most commonly used coagulant, is estimated to be as much as seven times more expensive in many African countries than in the USA than, which need to import it from Europe (Schulz & Okun, 1992). The consequence of this is that many treatment works underdose their coagulants, with the result that a low quality filtrate water is produced. Therefore if cheaper coagulants can be found, it would be a welcomed benefit to many developing countries. Using indigenous materials will also help bolster local economy and expand industrial development.

- A reduction in sludge volume, with the use of certain natural coagulants, to an estimated fifth of that produced by alum (Ndabigengesere *et al.*, 1995). Metal coagulants form hydroxide floc which are very voluminous, gelatinous and acidic (AWWA, 1990) compared to that formed by polymers. In addition the sludge formed by natural polymers is biodegradable, making land-based disposal possible.

- Reduction in the requirement for acid and lime for pH adjustment before and after treatment (Ndabigengesere & Narasiah, 1998).

- Natural coagulants tend to have a broader dose response, resulting in a more robust treatment process.

- Use of a renewable resource will slow the depletion of non-renewable mineral resources, thereby reducing the consumption of petrochemical based synthetic polymers.

- A reduction in the use of synthetic chemicals in the treatment of potable water is of benefit to water companies pursuing environmentally sensitive polices.

A potential limitation to the use of natural coagulants is that bacterial concentrations in the treated water may increase due to the presence of organic substrate in the water. This problem can be overcome by disinfecting the treated water, before consumption, if it is to be stored for a length of time. The use of disinfectants such as chlorine, in the presence of organic polymers can also be a problem: the formation of the potentially carcinogenic DBP (Rook, 1974).

Examples of natural coagulants are chitosan, a cationic derivative of chitin extracted from the exoskeletons of arthropods and sodium alginate an anionic extract of kelp (brown seaweed) (Kawamura, 1991a; Murcott & Harlem, 1993). Chitin is the second most abundant natural polymer on earth, and has already been used for water treatment purposes in many countries (Brzeski, 1987; Kawamura, 1991a). Alginate has been used in water treatment for over 30 years in Japan (Kawamura, 1959 & 1991a). The use of fish bones and scales, mainly as a coagulant aid, is being investigated by the National Environmental Engineering Research Institute in India (Okun & Schulz, 1983).

Reports of plant material having the ability to make *"bitter water sweet"* date back many thousands of years (Exodus 15 v.22-25). Seeds which can yield charged molecules that have a use in water treatment include: *Strychnos potatorum* - the Indian "clearing nut" or nirmali seed. It was reportedly used 4,000 years ago (Schulz & Okun, 1992), and is still used today in villages in India (Chaudhri *et al.*, 1989; Vasudevan *et al.*, 1989). *Vicia faba* - the broad bean or the "Egyptian bean" and *Arachis hypogaea* the "Sudanese bean" cultivated in the African Nile delta are used for household water treatment. The seeds are rubbed on the inner surface of earthen vessels, which are then filled with raw water and left to stand over night. In Peru the sap of the cactus *Opuntia fiscus indica* "tuna" and a powder from dry roasted *zea mays* were discovered by sailors to be water clarifiers. Bentonite clay, known as "Rauwaq", is used by the Sudanese as an effective clarifier. Its use then spread throughout the Islamic world by Koranic teaching and is now used in many Arab countries (Jahn, 1981). Other materials also investigated for their coagulating properties are potatoes, lentils, guar plants, red sorella and fenugreek (Bhole & Shrivastava, 1983; Okun & Schulz, 1992) with varying degrees of success.

The coagulant primarily considered in this study is extracted from the seeds of the tropical tree *M.oleifera.* The crushed seed suspensions yield low molecular weight, highly basic, water soluble proteins, which act in a similar manner to synthetic polymers (Gassenschmidt *et al.*, 1995). The seeds are reputed to have been used to clarify water at household level in Sudan at the beginning of this century (Jahn & Hamid, 1979; Jahn, 1981, & 1988; Jahn *et al.*, 1986; Madsen *et al.*, 1987). The potential for its use in water treatment has been realised by many researchers (Jahn, 1988 & 1991; Jahn *et al.*, 1986; Olsen, 1987; Ndabigengesere *et al.*, 1995; Ndabigengesere & Narasiah, 1998). Much of the work has been conducted at jar test level, consisting of rapid and slow mixing of the water after the addition of the coagulant, followed by a period of settling. The supernatant is then decanted off and used. Its use at household level was investigated in Sudan, where testing consisted of a simulation of the treatment processes used in the villages (Jahn, 1986), and the treatment of high and low turbidity waters with *M.oleifera* and bentonite. Effectiveness of treatment was measured in terms of removal of turbidity and the pathogen *Schistosoma mansoni* (Olsen, 1987). Ndabigengesere *et al.* (1995) Ndabigengesere & Narasiah (1998) compared the use of shelled and non-shelled seeds with alum in jar tests on high turbidity. Ndabigengesere & Narasiah (1996) considered the effect of mixing parameters in jar testing on removal rates. Muyibi & Okuofu (1995) conducted jar tests to consider the use of *M.oleifera* as a primary and secondary coagulant in the treatment of low

turbidity surface waters in Nigeria. The team at Leicester University have conducted pilot and full scale trials on the treatment of high turbidity waters in a WTW in southern Malawi (Sutherland *et al.*, 1994). Raw water turbidities of 3-4000 NTU were encountered and conventional treatment was used.

The topic of this research is unique since it consists of pilot scale studies into the feasibility of use and optimisation of CFF in the treatment of low turbidity waters, using the natural coagulant extracted from the seeds of *M.oleifera*.

2.2.2 Mechanisms of destabilisation and aggregation of particles

The process of destabilisation, or coagulation, is achieved by various electrochemical processes, depending on the coagulant used. These are the electrostatic 'patch' mechanism, the 'bridging' mechanism, reduction of interactive energies and the 'sweep' floc mechanism. Some coagulants act primarily by one mechanism, although depending on the raw water and the treatment process, a combination of the processes may be involved. Flocculation is purely a physical process by which the destabilised particles or microflocs, 1-100 μm are induced to collide and aggregate to form larger flocs (Argaman & Kaufman, 1970). The particles can be randomly bombarded by water molecules in a process of perikinetic flocculation, causing the particles to collide and flocculate. This Brownian movement is inherently stochastic and slow in nature. Furthermore, it is temperature dependent, such that the collision rates increase with higher temperatures due to the increase in the particles' thermal energy. The collision rate can be increased by hydraulic or mechanical agitation of the dosed water utilising an outside energy source. In treatment works this is achieved by the dosed water entering flocculation tanks and undergoing a slow mix stage. This is described as orthokinetic flocculation (Kruyt, 1952) and is necessary in order to flocculate particles <1 μm within an acceptable time limit.

(I) The 'patch' mechanism of charge neutralisation

This mechanism occurs when a charged polymer attaches itself to the surface of a particle by electrostatic forces, forming a patch-like pattern (Kasper, 1971, Gregory, 1973). The majority of suspended matter in natural waters is negatively charged, including organic matter at the pH of most natural waters. Therefore cationic polymers are generally used. A consequence of this

mechanism is that the particles may have an overall negative charge but still be considered to be destabilised, due to the localised region of positive charge. If the positive surface then comes into contact with a negatively charged surface then attraction and aggregation will occur. The molecular weight of the polymer is of no significance, but the charge density is (Ghosh & Leu, 1986). Polymers inducing flocculation by the 'patch' mechanism, produce more compact floc than do polymers inducing flocculation by the 'bridging' mechanism as discussed below (Hughes & Ramsden, 1995). Figure 2.3 illustrates this mechanism schematically.

Forces other than those of coulombic origin can cause attachment of the coagulant to the particle due to specific adsorption. Examples are van der Waals' forces, hydrogen bonds or specific chemical reactions (Gregory, 1977). Hydrogen bonds are electrostatic attractions that arise when hydrogen atoms are co-valently bonded to an electronegative atom. Small charges within the hydrogen molecule occur due to the slight shift of the electrons in the covalent bonds and these cause electrostatic attractions.

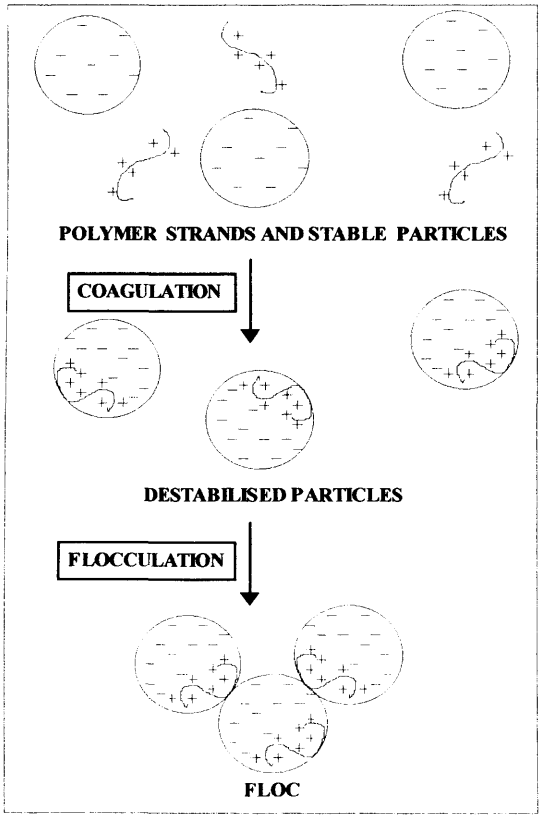

Figure 2.3 Schematic representation of the electrostatic 'patch' mechanism of floc formation.

The effect of these forces is to reduce interparticle repulsion and induce aggregation following particle collision, thus forming floc. The strong electrostatic interactions between cationic polymers and anionic particles, result in adsorption continuing beyond the point of neutrality. Thus if an excess of the polymer is added, charge reversal will occur, causing restabilisation of the particles. The Critical Coagulant Concentration (CCC) occurs when <50% of the particulate surface is covered with the polymer. In this case it is related to the concentration of particles in the water and their surface area. A higher dose would lead to the complete coverage of the particle surface and eventual restabilisation.

(II) The bridging mechanism

Polymeric bridging occurs as segments of a polymer chain adsorb onto the surface of the particles, linking them together and forming floc. The concept of polymeric bridging was first established by Ruethrwein & Ward (1952) and later refined by La Mer & Healy (1963). This method explains how anionic or nonionic polymers were able to destabilise negatively charged particles, since the 'patch' mechanism could not explain this phenomenon.

The sequence of events leading to destabilisation and flocculation by 'bridging' are depicted in Figure 2.4. Step 1 is the addition of the long chain polymer into the water and its contact with the stable colloidal particle. It is essential for the operation of this mechanism that the polymer adsorbs onto the particle in such a way that a large proportion of it extends into the solution to enable attachment to other particles. Gregory (1996) considered linear polymers to be more effective at 'bridge' formation than branched or cross linked polymers. As illustrated in Step 1, attachment and destabilisation occur during the coagulation stage. The polymer can be cationic, anionic or nonionic in nature, since the bonds formed can be due to electrostatic forces, ion exchange, hydrogen bonds or van der Waals' forces (Gregory, 1977). After destabilisation there are two possible courses. In step 2A, it can be seen that the polymer strands attach to particles with their tail-ends extending into the solution. The tails are free to adsorb onto other stable or destabilised particles, 'bridging' them together. Floc are thus formed during this slow mixing stage. Alternately secondary adsorption can occur, as seen in step 2B. This situation occurs if the polymer is not successful in finding an adsorption site on another particle and reabsorbs onto itself due to inefficient mixing of the solution. In step 3 the CCC is exceeded. The CCC for 'bridge' formation has a linear, stoichiometric relationship with the surface area of the particulate material (Stumm & O'Melia, 1967). Therefore optimum

removal will occur when 'half surface coverage' is achieved - a concept devised by La Mer (1964). If each particle has numerous polymers attached to their surfaces, then particle restabilisation occurs, not necessarily by charge reversal, but rather by saturation of all the available sites for chemical bridging. This situation not only leads to inefficient removal of the particulate matter, but also considerable wastage of coagulant. Step 4A & B should also be avoided. Subsequent to step 2A, where floc are formed, high shear forces can rupture previously formed floc, forming smaller fragments, as shown in step 4A. These fragments can be restabilised as the polymer folds back over its surface, as shown in step 4B.

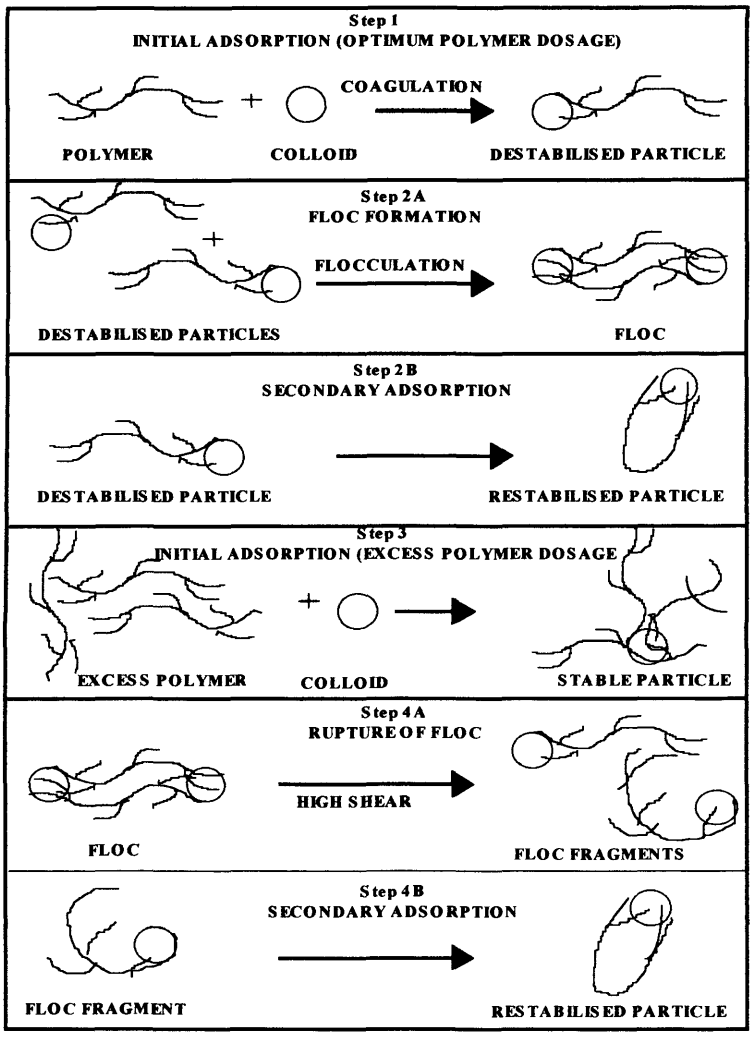

Figure 2.4 Schematic representation of the 'bridging' mechanism.

(Source: O'Melia, 1969).

Polymer bridging produces stronger floc than adsorption flocculation does, since the polymeric chains can provide multiple links with each particle (Gregory, 1988). If shear forces in the flocculation stage become very high, then eventually the floc will begin to break-up. Unlike floc formed by the 'patch' mechanism, disruption of floc formed by the bridging mechanism is irreversible. In fact it has been suggested that a possible method of indicating which mechanism of floc formation is in operation, would be the reversibility or otherwise of floc formation (Ditter *et al.*, 1982; Gregory, 1996).

(III) Reduction of interactive energies

As described in Section 2.2.2, all particles are surrounded by a diffuse layer of counter-ions. In the case of the particle in Figure 2.5 diagram A, the negative particle is surrounded by a layer of positive ions. The thickness of the this layer determines the zeta potential and consequently the stability of the particle. In order to induce particle destabilisation and aggregation, the interactive energy of a particle which causes repulsion must be reduced. This can be achieved in one of two ways:

Firstly, the addition of high valence counter-ions which would specifically adsorb onto stable particles, reducing the magnitude of the zeta potential, as illustrated in diagram B. Once the potential is reduced, particle interaction can take place. Attachment of the counter-ions can be due to electrostatic forces, van der Waals' or hydrogen bonds, or a specific chemical reaction. Thus restabilisation can occur if an excess of these specifically adsorbing counter-ions is added causing charge reversal.

Secondly, increasing the ionic strength of the bulk solution surrounding the particle, as illustrated in diagram C. This has the effect of reducing the extent of the diffuse layer thereby compressing the double layer, and in turn reducing the repulsive forces between the particles. Polyelectrolytes which induce flocculation by this means are termed indifferent electrolytes, since their surface charge is insignificant. Their only effect is to increase the ionic strength of the solution thus reducing the thickness of the double layer and zeta potential.

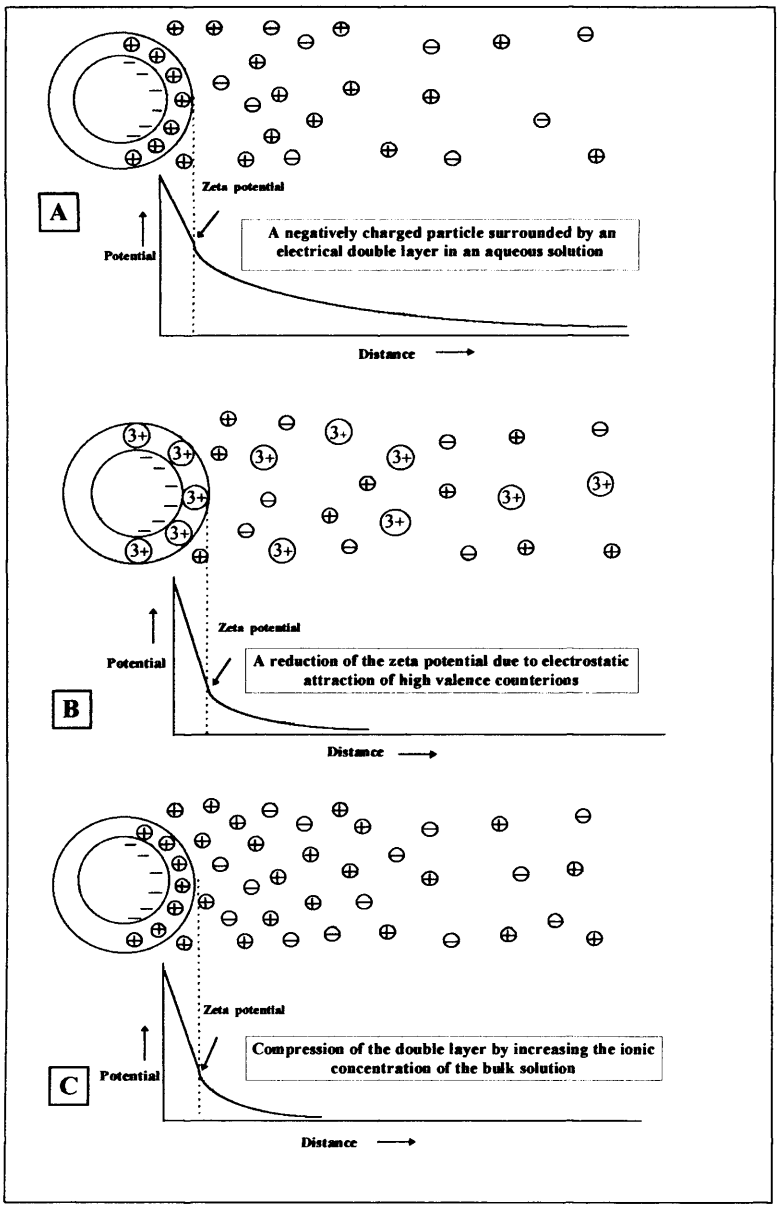

Figure 2.5 Destabilisation by reducing the interactive energies between particles

Aluminium and iron salts were considered to effect destabilisation by the mechanism described in diagram B, since the high valence of the Al^{+3} and Fe^{+3} ions would be very efficient at reducing the charge on the surface of negatively charged ions. This was explained by the Schulz-Hardy Rule, according to which the activities, or coagulating power, of a divalent and trivalent ion are 80 and 640 times greater than a monovalent ion, respectively (Black, 1960). But there are flaws in this theory, since not all monovalent ions have the same extent of activity (O'Melia, 1969). Moreover when the metal salts are added to water, parallel and sequential reactions occur, the rate of which depends on many factors, specifically pH. It is only at low pH values that the particles can be destabilised by adsorption of the metal ions onto their

surfaces (Letterman & Iyer, 1984). Figure 2.6 illustrates how the positively charged trivalent ions adsorb onto the surface of negatively charged particles, thereby lowering the zeta potential and the electrostatic repulsion between particles. Particle aggregation and floc formation can then ensue. The pH of most waters during coagulation is between 6-8, and Al^{+3} and Fe^{+3} ions are only present for a matter of seconds before hydrolysis products are formed. It has thus been concluded that charge neutralisation can not be the only or primary method of floc formation induced by the metal salts, and that enmeshment by a precipitate occurs at higher pH values (Gregory, 1977; Stumm & O'Melia, 1967). This is described in the next section.

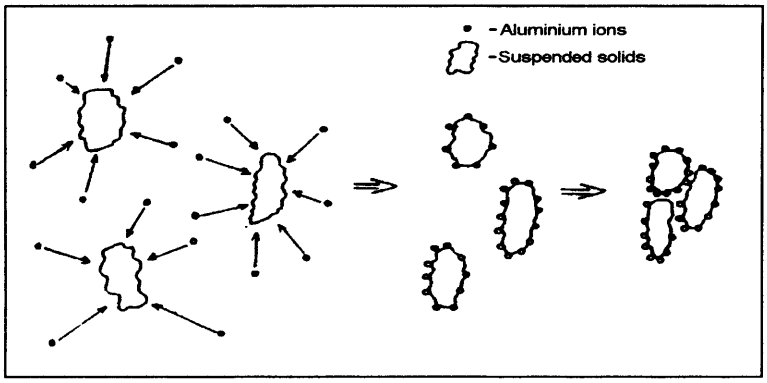

Figure 2.6 Coagulation and flocculation by alum trivalent ions

(Source: Licskó, 1996).

(IV) Enmeshment or 'sweep' flocculation

The following describes floc formation by enmeshment using alum, but it applies to all metal salts. In aqueous solutions the aluminium ion is surrounded by six water molecules. As a consequence of the high positive charge on the Al^{+3} ion, the O-H groups polarises resulting in disassociation of H^{+}.

$$[Al(H_2O)_6]^{3+} \leftrightarrow [Al(H_2O)_5OH]^{2+} + H^{+} \qquad Eq\ 2.1$$

The excess acid liberated by this reaction is neutralised in alkaline waters by the carbonates and hydroxides, and therefore the equilibrium shifts to the right. The unhydrolysed Al^{+3} only exists at a pH value of between 4-5. At pH values of 6-8 the disassociation continues until the uncharged insoluble metal hydroxide is formed, as seen in Equation 2.2.

$$[Al(H_2O)_6]^{+3} \leftrightarrow [l\ (H_2O)_5OH]^{+2} \leftrightarrow [Al\ (H_2O)_4(OH)_2]^{+} \leftrightarrow [Al(H_2O)_3(OH)_3]_{(s)} \quad Eq\ 2.2$$

At even higher pH values the precipitate begins to redissolve and the aluminate ion is formed:

$$[Al(H_2O)_5(OH)_4]^{-1}$$

Each hydrolysis step causes a reduction in the charge on the ion complex. It would therefore be expected that the coagulation efficiency would also be reduced if the mechanism of destabilisation was simply double layer compression. But this is not the case, since a metal complex adsorbs onto a particle surface and effects its destabilisation. In fact, if excess coagulant is present then charge reversal occurs. This would not be possible if the only means of destabilisation was double layer compression.

The sequence of hydrolysis reactions described is highly simplified, since there are many intermediate reactions. Many polynuclear complexes are formed which have been shown to be effective coagulants (Gregory, 1977). In practical terms, however, the main coagulant in operation after the addition of aluminium sulphate is the insoluble metal hydroxide. The process involved is the 'sweep' floc mechanism of particle removal. The particulates are enmeshed into the amorphous structure formed by the hydroxide precipitate, as are dissolved organic molecules and heavy metals (Dempsey *et al.*, 1984). This effect is illustrated in Figure 2.7. These primary particles are said to be 'swept out' of suspension (Gregory, 1989). The bonds involved in this adsorption include van der Waals' forces, hydrogen and ionic bonding (Crozes *et al.*, 1995). Montgomery (1985) suggests that nucleation may occur on the surface of the particulate, and the amorphous precipitate is then able to grow around the particle and further entrap other small particles. The rate of floc formation therefore increases with a rise in the turbidity of the raw water, since there are more nuclei for floc to commence formation. Also at higher turbidities, flocculation can occur with lower doses of coagulant. This form of destabilisation and aggregation is not dependent upon charge neutralisation, so the optimum coagulant dose does not correspond to minimum zeta potential. Optimum performance of alum will depend on the pH of the raw water, and the dose used. Figure 2.8 illustrates how these two factors determine whether it is 'sweep' floc coagulation or charge neutralisation mechanism which cause floc formation. At a pH of between 6.5-8.5 and a dose of about 30-200 mg/L 'sweep' flocculation will dominate, and a pH of 5-6.5 and dose of about 20-200

mg/L adsorption will dominate. But this is not the entire picture, since at specific pH values and doses, restabilisation can occur or both mechanisms can operate simultaneously.

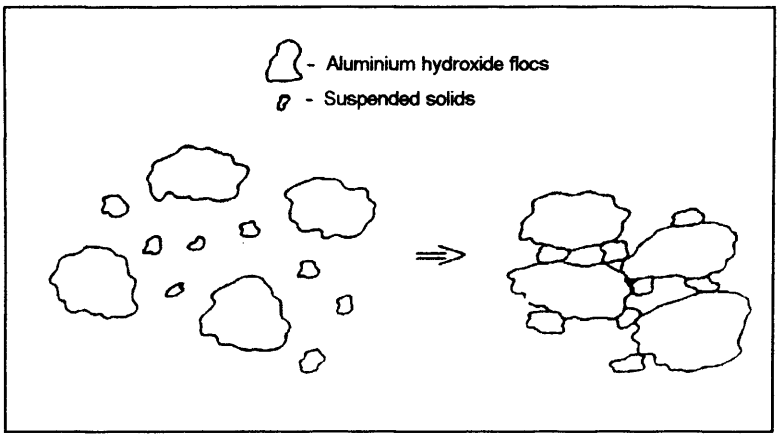

Figure 2.7 Coagulation by voluminous aluminium hydroxide flocs - 'sweep' floc coagulation *(Source: Licskó, 1996).*

'Sweep' floc coagulation has many disadvantages. Firstly, large volumes of wet metal sludge is produced which can be expensive to dewater. Secondly, rapid filter blinding occurs, resulting in a higher backwash requirement, and an increase in operational costs. Thirdly, higher concentrations of coagulant are required to remove particulates by entrapment than would be required for coagulation by reduction of the energy barrier as discussed above. It is estimated that anywhere between 5-40 times more coagulant is required to induce coagulation by entrapment than charge neutralisation (Grutsch & Mallatt, 1977). But the advantage is that coloured organic matter in low turbidity waters can be removed and such waters are generally considered to be very difficult to treat. Reduction of the charge by adsorption or compression of zeta potential are not effective methods of removing low concentrations of particles and organic colloidal matter.

The intensity of mixing in the coagulation and flocculation stages are important. The temperature of the water effects the extent of mixing by firstly, increasing the rate of Brownian Motion and therefore the rate of perikinetic flocculation; and secondly by decreasing the viscosity of water and allowing a greater distribution of the coagulant throughout the water. During coagulation it is essential that a high degree of turbulence is achieved at the point of addition of the coagulant (Elimelech *et al.*, 1995). The design of flocculation tanks is based on

the concept of velocity gradient which describes the degree of mixing at any point in the liquid system. The velocity gradient, G, was first described by Camp & Stein (1943) and is derived from the power input, the volume of water and the viscosity of water. The power input in hydraulic flocculator is based on headloss in the tank. The duration of the mixing stage (t) is also important. The rapid mix stage with metal salts tends to be very brief and with very high shear rates, since there are a sequence of hydrolytic reactions which occur to form the precipitate, generally within 1-7 seconds (Enkel, 1997). The G and t values required by metal salts during coagulation are generally considered to be about 300 s^{-1} and 1 minute respectively (Collins *et al.*, 1987; Ashan *et al.*, 1996). During the flocculation stage the values are about 20-75 s^{-1} and 10-60 minutes receptively (Twort *et al.*, 1994). The mixing intensity and duration for polymers during coagulation are not clearly defined, although the rates are not as high as with metals and generally the duration is longer. The mixing intensity must be sufficiently high to ensure the even distribution of the polymers throughout the water and prevent restabilisation due to readsorption of the polymer chain. Additionally there is evidence that droplets of polymer solution can survive in the dosed water and act as a nucleus for floc formation; this may or may not be desirable (Flynn, 1984). But very high shear rates can cause polymer scission and/or floc break up, which would be detrimental to the process. At lower than optimal G values, the coagulant is not dispersed uniformly, causing local over- and under-dosing to occur.

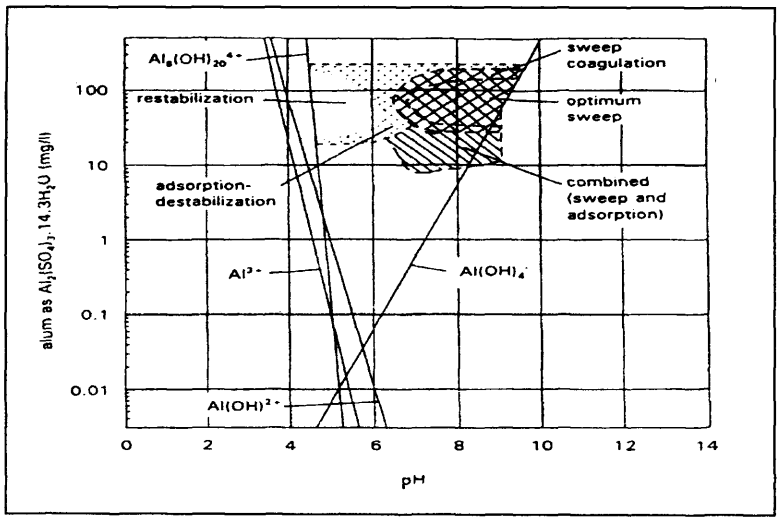

Figure 2.8 Alum coagulation diagram, illustrating the dependence floc formation on pH and the dose of coagulant used.

(Source: Stumm & O'Melia, 1968).

A lower rate of mixing is required during the flocculation stage, in order to induce floc formation. The correct rate is necessary to prevent polymer scission and floc breakup, especially with the large molecular weight, bridge-forming polymers. Another factor is that higher mixing intensities cause polymer chains to uncoil and extend further, making more effective bridge-formers (Horn & Merrill, 1984).

The optimal extent of mixing, for any coagulant, will depend on many factors; such as the turbidity, organic content and pH of the raw water. The type of solid-liquid separation stage employed subsequent to mixing, will determine the size of the optimum floc required. 'Bridge' forming polymers and metal salts tend to be more effective where there is a sedimentation stage prior to filtration, since voluminous 'settleable' floc are created. Whereas small polymers that induce charge neutralisation and the formation of pin-floc, would be more appropriate for use with DF and CFF. Sludge disposal after the treatment process is also a significant factor.

2.3 Filtration

Thousands of years ago it was recognised that filtration was an effective process for improving the clarity of water. Baker (1949) refers to the use of filters in India for water treatment in the year 1000 BC. Since then filters have been used to a lesser or greater extent, with variations in the flow rate, media type, size, depth, etc. In the 1800s, SSF were the main form of treatment for potable water. By the 1900s they had mainly been replaced by RFF (Williams & Culp, 1986).

The term 'filtration' generally implies some form of straining process, whereby the particles removed are larger than the pores in the media through which the fluid is passing (Herzig, 1970). In such cases removal would occur on the surface of the filter and therefore deep beds are not necessary. Filtration in water treatment parlance is different, since it is generally through granular filters with depths of between 0.5-1.5 m, where the pores in the bed are far larger than the impurities to be removed. The particles and colloids generally removed are in the size range of 0.1-10 μm, whereas the smallest filter media (generally sand) would have diameters of 0.50-1.00 mm and therefore pores of about 1.00 mm (Tebbutt, 1998). Consequently, the particles would be passing through pores that are up to 10, 000 times their own diameter and so straining can be eliminated as a major removal mechanism.

SSF consists of relatively shallow beds of about 0.6-0.9 m, the media is ungraded sand ranging in size between 0.25-0.33 mm, and the flow rates range between 0.1-0.2 m/h. Most of the filtration occurs in the top layer, where a slimy layer of retained material develops with an active flora and fauna. This biologically active zone, the *schmutzdecke* layer, results in efficient removal of many pathogens. In RFF bed depths of between 0.5-2.0 m and flow rates of between 5-15 m/h are generally used. The media size is in the range of about 0.50-1.00 mm, and becomes stratified after backwashing, with the lightest grains on the surface and the heaviest towards the base of the bed (Kawamura, 1991b; Schulz & Okun, 1992).

SSF have the advantage of not requiring a floc formation stage prior to the filtration process, nor regular backwashing of the filter bed. They are therefore cheaper to operate and require less skilled labour. SSF are very effective at removal of biologically active material, and also pesticides (Foster *et al.*, 1990). RFF have the advantage however, of being more compact

since the flow rates are much higher. They are also more flexible in terms of being able to cope with surges in turbidity of the raw water.

There are various means by which particles are removed from the flow during the process of filtration. The process is dependent upon physical parameters such as the characteristics of the filter media and flow rate, and also by chemical characteristics of the water, which in turn is affected by the pretreatment process. Removal by straining in rapid filter is prevented by ensuring the particles size does not exceed the pores size, since it is detrimental to the treatment process. If straining dominates then effective use of the filters is prevented and backwashing is required prior to complete bed exhaustion.

For particles to be removed by filtration, they must initially be transported to the media grains and then they can attach to the surface. Removal is therefore a two step process, each step consisting of any one of a number of possible mechanisms (Ives, 1980; 1982; Amirtharajah, 1988; O'Melia & Stumm, 1967). These are discussed further in section 2.3.1 to 2.3.3.

2.3.1 Transport Mechanisms in Filtration

Flow through granular media filter beds is generally laminar, that is to say that the streamlines are ordered and do not cross. In order for the particles to be removed from the flow they must deviate from the fluid streamlines to reach the surface of the media grains, otherwise known as collectors. There are various physico-hydraulic factors that are responsible for transport of the particles to the collectors, the calculation of which assumes that the particles are spherical. These transport mechanism are shown schematically in Figure 2.9 and discussed further below.

2.3.1.1 Gravity (Sedimentation)

Transport by sedimentation occurs when the particles in the flow have a higher density than water. The gravitational force causes the particles to maintain a constant velocity throughout the bed. Thus causing them to cut across the streamline flow paths and settle on the upper part of the collector. The rate of removal by sedimentation can be calculated by dividing the settling velocity (V_S) by the filtration velocity (Agrawal, 1966; Yao, 1968). V_S can be calculated using Stokes' Law:

$$V_s = \frac{g(\rho_1 - \rho)d_p{}^2}{18\mu} \qquad \textit{Eq. 2.3}$$

Removal by sedimentation is calculated by:

$$S = \frac{V_s}{V} = \frac{g(\rho_1 - \rho)d_p{}^2}{18\mu V} \qquad \textit{Eq. 2.4}$$

Where S is the rate of removal by sedimentation, g, ρ_1 is the density of the particle, ρ is the density of water, d_p (μm) is the diameter of the particle, μ is the dynamic viscosity of water (10^{-3} kg/m s) and V is the filtration rate (Ives, 1980; Ives, 1982; Montgomery, 1985). Therefore sedimentation is dependent upon the size of the particle, the ratio of particle to water density, and the filtration velocity. Ives (1982) concluded that particles in the size range 2.5-25 μm are most affected by this transport mechanism.

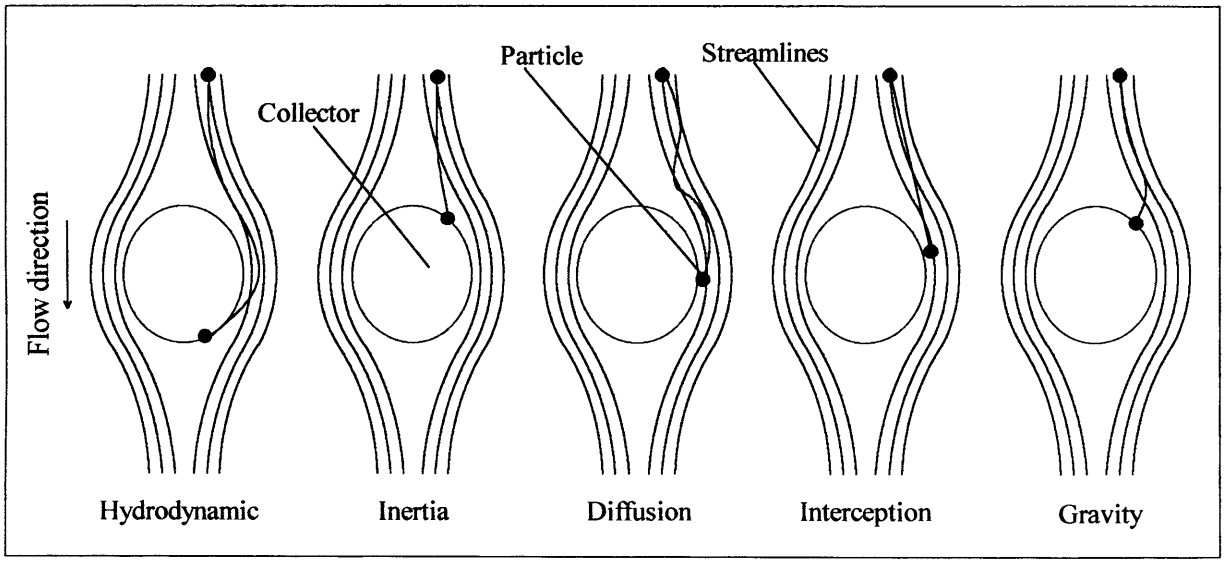

Figure 2.9 Simplified diagrams of particle transport mechanisms

(Source: adapted from Ives, 1980).

2.3.1.2 Diffusion (Brownian motion)

This method of particle transport is brought about by the random bombardment of particles by water molecules, due to their thermal energy. It is only relevant for particles in the colloidal range (5 nm-2.5 μm) as they are closer to the size of water molecules. The efficiency of the collectors with regard to this mechanism can be calculated using the following equation:

$$B = 0.9\left(\frac{KT}{\mu d_p d_m V}\right)^{2/3} \qquad \textit{Eq. 2.5}$$

Where B is the extent of removal by Brownian diffusion, K is Boltzman's constant (energy per degree K = 1.38 x 10^{-23} J deg^{-1} $molecule^{-1}$), T is the absolute temperature (°K) and d_m (cm) is the diameter of the media (Levitch, 1962).

Transport by this mechanism is therefore related to the particle and collector diameters, velocity of flow and the temperature of the water. Higher temperatures will cause an increase in the kinetic energy of the system and consequently higher collision rates.

2.3.1.3 Interception

This describes the contact that occurs as a result of a particle remaining in its flow path around the collector, then coming within half its diameter distance from the collector and being intercepted. This transport mechanism requires laminar flow and a specific size ratio of particle to collector. Traditionally this was viewed as a distinct transport stage, but it can also be considered to be the final mechanism in all cases of contact between particles and collectors. It is characterised by the following equation:

$$I = 1.5\left(\frac{d_p}{d_m}\right)^2 \qquad \textit{Eq. 2.6}$$

Where I is the extent of removal by Interception. As I approaches 1 then straining becomes the dominant mechanism of removal in the filter bed.

2.3.1.4 Inertia (Impaction)

This transport mechanism describes the movement of relatively heavy particles; their inertia when approaching a collector surface would be greater than the hydrodynamic forces that would tend to sweep them past. The higher the flow rate the higher the rate of impaction of the particles onto the surface of the collector. This is an insignificant mechanism in water filtration, due to the relatively low velocity of the particle and the high viscosity of water (Ives, 1980).

2.3.1.5 Hydrodynamic

In laminar flow through filter pores, the water velocity varies from zero at the surface of the collector to a maximum at the pore center. Therefore velocity gradients exist across the pores which cause the particle to experience lateral forces and rotate in the flow field. If the particle is not spherical, as is generally the case, then the centre of gravity will not coincide with the hydrodynamic center, and the forces on the particle will be off-balance. This will cause an increase in particle rotation, increasing the chances of the particle coming into contact with the surface of the collector. The process is unpredictable and very difficult to calculate. There is therefore no quantitative analysis of this mechanism.

2.3.1.6 Combined Effects

The majority of raw waters contain a wide range of particle sizes, especially after coagulation. Different mechanisms of transport can be active at various stages of removal. Interception as an independent transport mechanism and inertia are not considered to be important in water filtration. Diffusion is most effective for particles smaller than 2.5μm, whereas sedimentation and interception are the dominant mechanisms for larger particles; as verified experimentally by numerous investigators (Yao *et al.*, 1971; Fitzpatrick and Spielman, 1973; Ghosh *et al.*, 1975). The contact efficiency for all of the above mechanisms was found to be at a minimum for particles in the size range of 1 μm (Yao *et al.*, 1971). This effect is illustrated in Figure 2.10. The significance of this is that the majority of bacteria and pathogenic microorganisms e.g. *Cryptosporidium* and microsporidia range in size between 1-5 μm, and therefore experience limited removal during filtration if there is no flocculation stage prior to filtration. The problem

is further exacerbated by the increased resistance of such organisms to disinfection by chlorine (Friedman-Huffman & Rose, 1989).

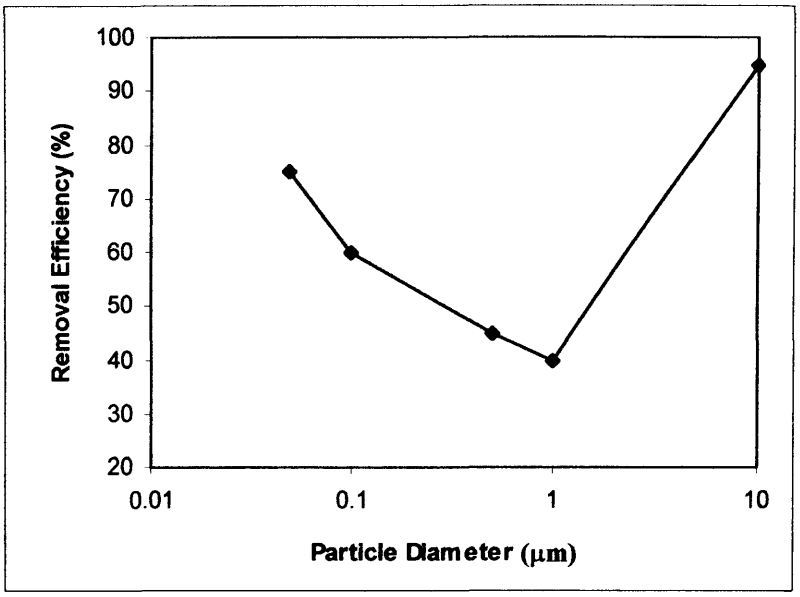

Figure 2.10 Removal efficiency as a function of particle size, showing a minimum for particle with a diameter of 1μm.

(Source: Ives, 1980).

2.3.2 Attachment mechanisms

As the destabilised particles and/or floc approach the surface of the collectors, short range, physicochemical forces will commence affecting particle dynamics. Whether the particle attaches to the collector or not, will depend on these interstitial forces. These consist of electrical forces between the surfaces of the particles and the collectors, and van der Waals' forces. The electrostatic forces can be repulsive or attractive in nature depending on the charges of the two surfaces. They are generally weak, since the charge on the majority of particles is about -20 mV and they are only considered to act when the particle comes within 1μm of the collector (Stein, 1940; Yao, 1968; Kavanaugh, 1974). The negative charge on most of the particles can be reduced or even reversed by the addition of coagulants to the water. These can form a 'patch' of positive charge on the surface of the particle, which can then in turn form an attachment point with the collector surface. Alternatively, they will reduce the negative potential around the particle so that it can come close enough to the collector for

other forces to commence acting. This is analogous to the flocculation of particles described in Section 2.2.2.

Particles may also be removed if they are sedimented out of the flow and onto the surface of the collector once any opposing charges have been reduced. Sedimentation was previously described as a transport mechanism, but it can also be considered an attachment mechanism. The process is dynamic and it is therefore not always clear where one stage ends and the other commences. Each interstitial pore in the filter bed can be considered to effectively act as a small sedimentation 'tank'. The efficiency of the filter is improved by using small grains with irregular surfaces, thus increasing the surface area for attachment and also the number of 'tanks'.

Destabilised particles entering the filter bed will be removed, not only by sedimentation, but also by specific adsorption and 'bridging'. Again these processes are analogous to coagulation and flocculation. As described previously, destabilised particles may have 'patches' of positive charge on their surface, or they may have a long chain polymer attached to their surfaces. These can then form attachment points at the collector surface. In some cases these attachment bonds may be stronger than those formed by sedimentation.

Another force that affects the removal process is the viscous resistance of a film of water. A film of water trapped between a particle and a collector will have to flow outwards from the contact point between the two particles in order for them to make contact. Its effect can be to delay the attachment process to such an extent that eventually the force of the flow of water will move the particle beyond the collector and therefore it will remain in suspension (Spielman, 1978). The only relevant influence on the viscosity of water is temperature, a factor that can not practically be altered.

The existence of a range of removal mechanisms is agreed by most researchers, but the relative effects and sphere of influences are not so clear (Ives & Sholji, 1965). Factors such as size of the media, flow rate and viscosity of the water will have an effect; but the extent of destabilisation of the particles entering the filter is of more significance, since particle capture is more likely to occur when the particle and the collector have an opposite charge.

There will not necessarily be the same dominant mechanism throughout the process. For example as a run commences, interstitial velocities would not be very high, and sedimentation as a transport and attachment mechanism may be dominant. As the run proceeds, interstitial velocities increase due to clogging of the filter beds and the consequent reduction in pore size. Inertia may then become the dominant mechanism of removal. As clogging of the bed increases further, straining may become more prominent, particularly in the surface layers.

2.3.3 Detachment Mechanisms

As the filtration process continues the local flow velocities increase due to clogging of the filter media. This causes the hydrodynamic shear forces in the pores to exceed the attachment forces. Subsequently, previously removed particles become re-entrained into the flow. Detachment can also occur independently of clogging if there is a sudden surge in the flow rate, resulting in deterioration of the quality of the filtrate. Another mechanism of detachment has been termed the 'avalanche' effect. This may occur after many hours of operation of the filter, with an accumulation of deposits on the media grains. As more particles are removed unstable particles or deposits may be 'knocked' back into the flow. This has been observed in pilot perspex filters. Large deposits of particles on media grains in the columns are seen to be 'knocked' off their attachment sites into the flow, and occasionally re-deposited further down the filter (Ives, 1980). The stronger the attachment forces between the particles and the media, the less likely that detachment will occur. This is one of the advantages of using polyelectrolytes as a coagulant, especially long chain, 'bridge' forming polymers, since they form stronger adhesive forces as discussed in Section 2.2.2.

2.4 The three stages of a filter run

The filtration process can be divided into three stages based on the turbidity of the treated water. These can be seen graphically by plotting the turbidity of the treated water against time, as illustrated schematically in Figure 2.11. The initial stage is the 'ripening' or 'working-in' stage, which is characterised by high turbidity that is reduced dramatically over a short period of time (Amirtharajah, 1988, O'Melia & Ali, 1978). Turbidity removal during the ripening period is exponential because previously removed particles act as collection sites for subsequent particles entering the filter, thereby increasing the rate of removal. This stage is

usually short relative to the length of the entire run, and is controlled by the flow rate, media size and the extent of backwashing of the bed prior to the commencement of the run.

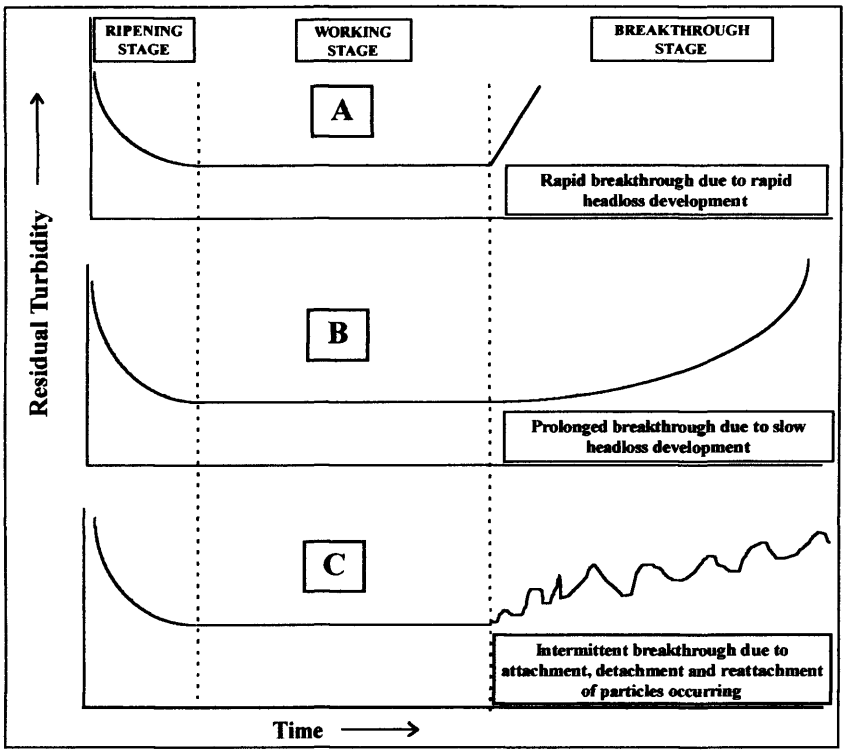

Figure 2.11 The three stages of filtration with variation is the breakthrough stage.

The second stage is the 'working' stage, where the majority of turbidity is removed and the filter is at its most efficient. The length of this run will depend on the operation of the filter. If conditions are optimal in the bed then this stage will be extensive. The final stage is the 'breakthrough' stage, where the residual turbidity begins to increase until a termination point of the filter run is reached (Adin and Rebhun, 1979). The increase in the residual turbidity can be rapid, slow or intermittent, as seen in Figure 2.11. The variation in the rate of breakthrough as seen in A and B can be due to headloss development rate, which in turn can be caused by numerous factors such as the deposition pattern of the particulates. C illustrates intermittent breakthrough, which would occur if attachment bonds between the filter media and the particulates were weak. Previously removed particles would become re-entrained into the flow and cause discrete breakthrough episodes to occur. The effect of this is to clear a section of the bed, thus making collector sites available for the attachment of further influent particulates. This then causes a decrease in residual turbidity for a short time.

Although the three stages are distinct in terms of turbidity removal, they occur simultaneously for different sized particles (Moran *et al.*, 1993). This is a consequence of turbidity not differentiating between different size particles and not directly relating to measurement of the concentration of particles present (AWWA, 1992). The significance of this is that certain particle sizes may reach the breakthrough stage while the turbidity readings indicate that the run is still in the working stage.

2.5 Optimisation of a filter run

It is a well-documented fact in granular filtration that the majority of removal at the initial stages of filtration, occurs in the surface layers of the bed. As the run continues, successive layers become more active in the removal process. An active, or 'clogging', front proceeds down through the bed gradually saturating or exhausting each layer. The front is the region of the bed where maximum removal is occurring. Above the front is a region of very low removal rate as the bed will be saturated and the interstitial velocities are high. Below the front, there is a region of relatively clean filter media that has not been active in the removal process. In a well-operated filter bed the clogging front will reach the bottom of the bed, indicating that the entire depth of the filter is utilised. The front will advance more rapidly where the particulate attachment forces are weak (Adin & Rajagopolan, 1989).

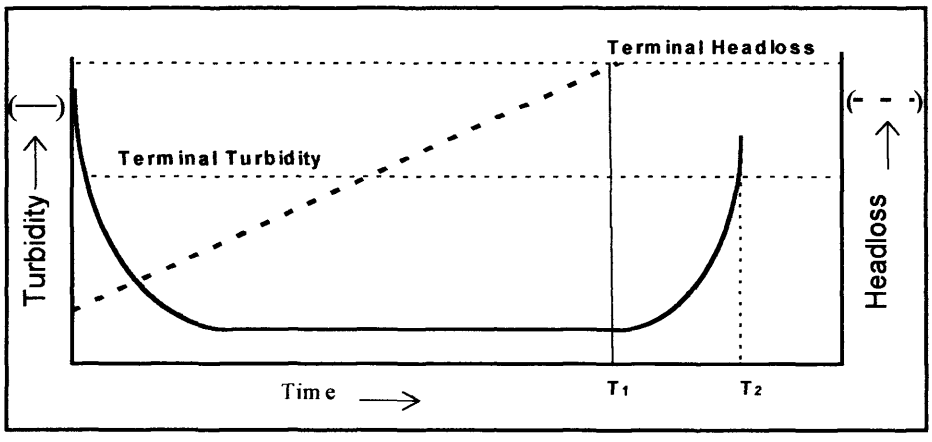

Figure 2.12 Schematic description of optimum filtration times.

(Source: Adapted from Montgomery, 1985).

Mints (1966) concluded that optimal conditions in the filter bed are attained if terminal headloss coincides with turbidity breakthrough. Figure 2.12 is a schematic of the pattern of headloss development and turbidity breakthrough in a typical filter run. Therefore optimal conditions would be achieved when $T_1 = T_2$ (Mints, 1966; Ives, 1969; Ives, 1982; Montgomery, 1985); this would ensure that the filter bed was fully utilised before backwashing was required.

2.6 Contact flocculation filtration (CFF) and direct filtration (DF)

In conventional treatment, rapid filtration follows coagulation, flocculation and sedimentation, as shown in Figure 2.1. This treatment train is necessary for high turbidity waters, but has the disadvantage of high construction costs. In the treatment of relatively low turbidity waters, certain stages can be eliminated, thereby reducing construction, maintenance and operational costs. DF and CFF are examples of such treatment process. In DF a sedimentation tank is not necessary, and in CFF separate sedimentation and flocculation units are not required, as illustrated in Figures 2.13 & 2.14 respectively. In CFF, flocculation occurs when particle and coagulants are in contact with the filter media. With CFF deeper penetration of the particles into the filter bed can occur leading to firmer attachment to the media. Ideally the floc formed in contact with the filter media is small and dense (pin floc), as compared to the large floc formed in a flocculation tank (Treweek, 1974). Calculation of the G and t values in the filter bed is complex. One possible equation which can be used to calculate G is that derived by Gregory *et al.*, (1981):

$$G = \frac{8Q}{3\pi R^3} \qquad \textit{Eq. 2.7}$$

Where Q is the volumetric flow rate and R is the inner radius of the mixing tube. In the case of the filter bed, each pore can be considered to be a tube. Therefore using the porosity of the filter media, the G value for each filter pore can be calculated and multiplied by the number of pores present in the bed to obtain the total G value for that particular filter. However, this was considered to be unrealistic, as particle deposition in the filter bed results in a dynamic porosity. Consequently, the G value is also dynamic and its calculation of little significance. Calculation of t for the filter bed is also difficult because it is unclear when flocculation ends

and deposition occurs. For these reasons no attempt was made to calculate the mixing parameters in the filter bed.

Another benefit of using CFF is that a reduction in coagulant requirement, since small 'filterable' floc are required and not large 'settleable' floc. Consequently, the volume of sludge produced is smaller and more compacted, making disposal cheaper (Datta, 1985). Schulz & Okun (1992) report on a treatment works in Jordan reducing its chemical costs by an estimated 80% by switching operation from conventional treatment to CFF. Construction and maintenance costs are generally also less. The AWWA & ASCE (1990) estimate a 30% reduction in cost through the elimination of the sedimentation stage. A further advantage is the increase in flow rate through the treatment works (Adin & Rebhun, 1974; Larson, 1987), thus reducing the land requirement for the treatment works.

Drawbacks of using CFF includes shorter runs leading to a higher washwater requirement, which can be in excess of 6% of the treated water. In comparison, conventional treatment requires between 3-4% (Schulz & Okun, 1992). Furthermore, without the buffering capacity of a sedimentation tank, the range of raw water turbidities which can be treated is limited. Thus, more careful monitoring of the treatment process is required with CFF than with conventional treatment.

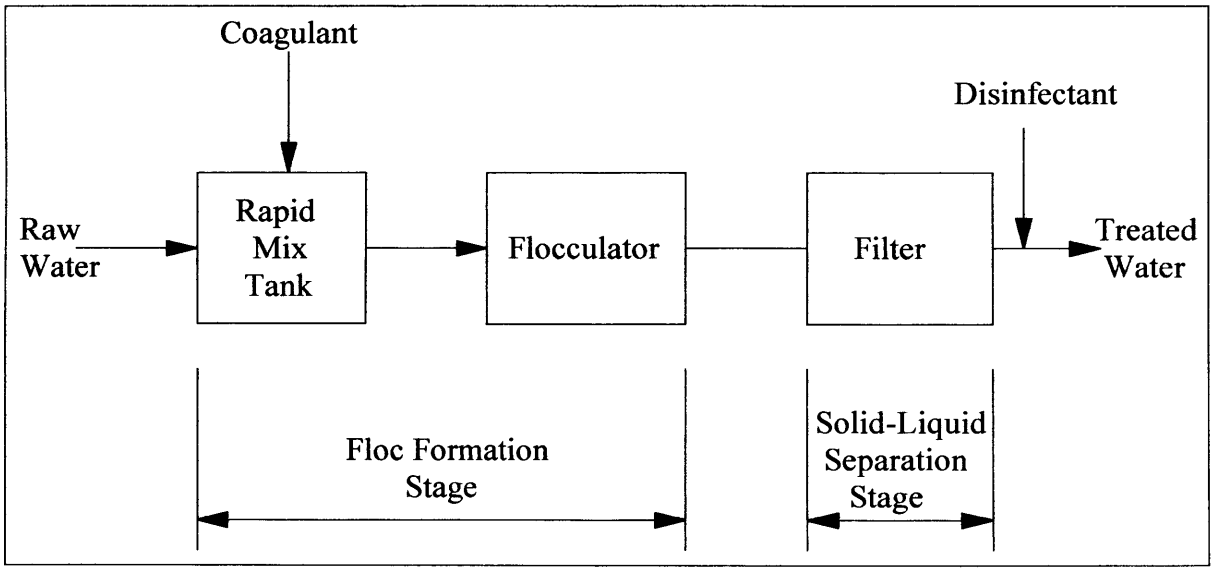

Figure 2.13 Schematic of the Direct Filtration process.

(Source: adapted from Edzwald, 1986).

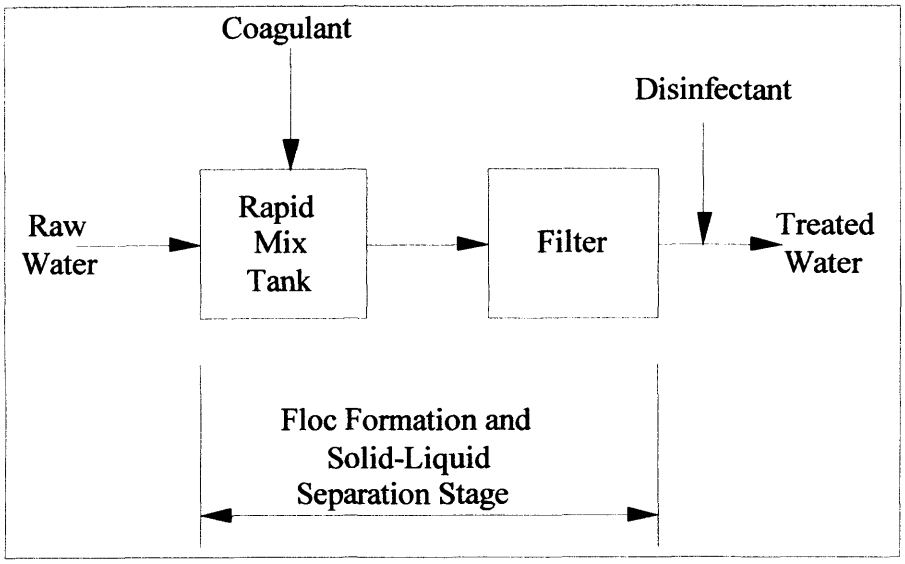

Figure 2.14 Schematic of the Contact Flocculation Filtration process.

(Source: adapted from Edzwald, 1986).

In some situations it may be possible to operate a full conventional treatment works in a DF or CFF mode for a certain period during the year. During the rainy season in tropical countries, surface water turbidities can be as be as high as 3-4,000 NTU, as a result of surface run-off from fields and the churning up of river silt. But during the dry season, flow rates in the rivers are low, turbidities can be below 20 NTU. In such situations it is not necessary to have a separate sedimentation stage to ensure sufficient removal. The advantage of this is not only a more rapid treatment works process, but also a reduction in coagulant demand.

The concept of DF was explored back in the 1900s, but was not successful because filter runs were too short due to rapid clogging and early breakthrough of turbidity. DF and CFF have become more popular in recent times with the introduction of dual and multimedia filter beds. The layer of large media at the surface prevents rapid clogging of the bed and allows deeper penetration of the particulates before removal occurs. The layer of sand acts as a final stage to improve the quality of the filtrate. Larger media permits a wider range of raw water turbidities to be treated.

DF and CFF have roles to play in various situations. Many small treatment works in developed countries, especially in the US with low turbidity waters, have traditionally used disinfection as the sole form of treatment (Public Works, 1992). In more recent years, concerns have arisen

over the presence of certain pathogenic protozoa such as Giardia, *Cryptosporidia* and microsporidia, and also DBP in the final. Consequently filtration has become necessary; DF and CFF are attractive solutions due to relatively low costs and suitability for treatment of low turbidity waters. DF is presently an effective and common form of treatment for upland coloured waters of low turbidity in Scotland and northern England (Graham *et al.*, 1992).

DF and CFF have a role to play in the treatment of water supplies in towns and small cities in developing countries. For such locations there is a need for effective and appropriate water treatment: not necessarily simple enough that unskilled labour can be used, as is the case with very small communities, nor as mechanised and complex as would be found in large developed cities. DF and CFF are seen as falling into this category, since construction and chemical costs are reduced (Schulz & Okun, 1992).

2.7 The natural coagulant - *Moringa oleifera*

M.oleifera (syn. *Moringa pterygosperma* Gaertn) is a member of the monogeneric family *Moringaceae* (Plate 1). There are 14 other species, the seeds of which all exhibit coagulating properties (Jahn, 1988). The tree is indigenous to northwest India, from the river Chenab eastwards and in the Tarari tract of Uttar Pradesh, and in Northern Pakistan and Afghanistan (Ramachandran, 1980; Morton, 1991). The name "*Moringa*" is thought to have originated from the south Indian Dravidian word "Morunga", since the tree originated from the Western Himalayas and Eastern Punjab (Dastur, 1951). The Latin word "*oleifera*" means 'oil bearing'. More common names for the tree are 'drumstick', 'neverdie', 'Ben tree' or 'horseradish'.

Although native to the Asian sub-continent, *M.oleifera* is now found throughout the tropical and subtropical belts (Ramachandran *et al.*, 1980), in particular among Indian communities. It was planted by the British in Africa, reputedly to provide shade along the shores of the River Nile. The pods are a popular food amongst many Indian settlers, and the trees are commonly grown in their gardens. Thus it is suggested that the migration of many Indians into East Africa also had the effect of introducing the tree into Africa (FRIM, 1997; KEFRI, 1999). The spread of the tree to Jamaica, Bermuda, Cuba, Haiti, Barbados and Trinidad is credited to an Englishman in 1784 (Fawcett & Rendle, 1814). *Moringa* is known as the "*Clarifier tree*" (Shagara al rauwaq) by the inhabitants of the shores of the River Nile, "*White pepper*" (svetta maricha) in India, "*The magical bean tree*" (Kelor) in Indonesia, "*Angel Plant*" and "*Tree From Paradise*" in South America, "*Tree of The King*" in Mali and "*Tree from Mecca*" in Niger (Morton, 1991). Thus indicating the wide spread distribution of the tree and that its potential has long been realised (Jahn, 1981).

Reports of the use of *M.oleifera* as a natural coagulant in water treatment date back to the beginning of this century, when Arab-Sudanese women used the strong coagulating properties of the seeds of the tree to treat water for drinking purposes at household level. The treatment process involved placing the seeds of *M.oleifera* into a cloth bag with a thread attached, and swirling it around a bucket of dirty water for about 20 minutes. The water was then allowed to stand for a number of hours and the supernatant decanted off for drinking purposes. This has been described by Jahn (1981) as the 'tea bag principle'.

2.7.1 The various uses of the *M.oleifera* tree

The main interest in the tree for the purposes of this research is the water clarification properties of the seeds. As is indicated by the tree's numerous vernacular names, many other products and uses have been realised. The most popular of which are listed below.

- Many parts of the tree are edible, as is evident from the numerous recipes for dishes involving the use of the pods, leaves, seeds, flowers (ECHO, 1997; EERG & Inter Care, 1998, Fuglie, 1999). The leaves are rich in protein, vitamins A and C, and are considered an ideal supplement for rice-rich diets. The Church World Service (CWS) in Senegal has been actively promoting the use of dry leaf powder in West Africa as a sustainable solution to malnutrition. The advantage of using *M.oleifera* leaves, is that the tree bears leaves well into the dry season and also, once dried, the leaf powder can be stored for long periods of time, and can be a valuable source of many vitamins. The pods contain many of the essential amino acids and vitamins and as such is a highly nutritious vegetable. The immature seeds, also rich in protein and vitamins, and are eaten like ground nuts or cooked in stews. The flowers are rich in vitamins A and C, potassium and calcium and are served up raw in salads or soups and sauces (Busson, 1965; Subadra *et al.*, 1997; Mathur, 1997; ECHO, 1998; Fuglie, 1999). Plate 2 shows the pods, leaves and flowers on a *M.oleifera* tree. Plate 3 shows some of the dishes prepared from the various parts of the tree.

- The leaves and pods are a valuable animal fodder (Jahn *et al.*, 1986). Successful trials have been undertaken using the leaves as pig fodder in Haiti, where it was found to be nutritious and able to withstand severe pruning (Price & Meitzner, 1996). The leaves and the pods have also been used as pig feed in Brazil, where it was found that the animals thrived on the new feed. Furthermore *M.oleifera* products were found to be more economically viable than buying in commercial feed (Medcraft, 1999).

- The oil expelled from the seeds is a high quality cooking oil, with an oleic acid content of about 73% (EERG, 1996). It is pale yellow in colour with a mild nutty flavour. The oil content of the seed is dependent upon variety and cultivation techniques. Reported concentrations include: 22% (Morton, 1991); 28-29% from seeds harvested from Malawi, Zimbabwe and Mozambique (Machell, 1997); 39% from seeds harvested in Malawi (Ibid.); 30% from seeds harvested from Burkina Faso (CWS, 1998); 37% from seeds harvested

from Ghana (Donker, 1992). It is an effective lubricant used by watchmakers in India (Nautiyal & Venkataraman, 1987). It is valued by perfume manufacturers for its ability to absorb and retain scents, and used for illumination since it is a smokeless fuel (Irvine, 1961; Mayer & Stelz, 1993). Organisations in various countries are currently investigating the potential for cultivation of *M.oleifera* trees for oil extraction, for human consumption, as a cash crop and for use in the cosmetic and aromatherapy markets. Examples of these are: Technology Consultancy Centre in Ghana (Donker, 1992); Harmony Foods Ltd., in Zimbabwe (Machell, 1997); CWS in West Africa (CWS, 1998); Optima of Africa Ltd. in Tanzania, who are distributing seeds and seedlings free of charge to farmers, and then guaranteeing the purchase all the seeds for an agreed fixed price for 5 years (Optima of Africa, 1999).

- The presscake produced after expelling oil from the seeds, contained the coagulant used in water treatment (EERG, 1994). Optima of Africa Ltd., are investigating the use of the presscake in water treatment for commercial purposes. The oil, presscake and whole seed can be seen in Plate 4.

- Many parts of the tree are considered by many communities to have special powers of healing for: rheumatism; ascites; venomous bites and as a cardiac stimulant. The leaves are used as an emetic or made into a paste to heal external wounds (Morton, 1991). They also have a folkloric reputation as being an affective hypotensive agent (Siddiqui & Khan, 1968), which has been shown to be due to the presence of isothiocyanate and thiocarbamates compounds (Faizi *et al.*, 1994). The leaves are used as a treatment against scurvy and catarrh, as a consequence of their vitamin content (Nautiyal & Venkataraman, 1987) and also ringworm (Cáceres *et al.*, 1991). The gum produced by the stem is also claimed to have some medicinal properties (Nautiyal & Venkataraman, 1987). The seeds and the roots contain 4-(α-L-rhamnosyl) benzyl isothiocyanate, an antibiotic component with bactericide, fungicide and cercaricidal properties (Olsen, 1985). Seed preparations are also effective against skin infections caused by *Staphylococcus aureus* and *Pseudomonas aeruginosa* (Price, 1992; Eilert *et al.*, 1981), gastrointestinal disorders and diarrhoea (Jahn, 1979). The pods have been used as a herbal treatment for diabetes in the Middle East for many years (Sharma & Raina, 1982).

- The woody stems are ideal for the use in the manufacture of pulp and paper according to the Forest Research Institute of India (Kantharajah & Dodd, 1991), and also for rayon (Nautiyal & Venkataraman, 1987) and cordage (Ramachandran *et al.*, 1980).

- As a consequence of its rapid growth rate, *M.oleifera* is often used for live fencing, fire wood, windbreaks, intercropping with staple food crops and as a 'nurse' tree (Mayer & Stelz, 1993; Folkard *et al.*, 1995).

- The tree has been reported to withstand flooding and high levels of salinity as well as growing well on marginal lands and therefore useful in areas that are considered too hostile for the growth of many other species (Nautiyal & Venkataraman, 1987).

- The flowers of the tree are an attractive source of nectar for honey bees (Kapadia, 1993).

- An antibiotic substance is released when the leaves are dug into the soil, and they are therefore used to prevent damping-off disease of seedlings.

- Laboratory based studies have indicated that *M.oleifera* seed could be effective as a water softening agent (Muyibi & Evison, 1995) and as a sludge conditioner (Ademiluyi, 1988).

- Seed husks used to form high quality microporous activated carbon for use as low cost adsorbents in water treatment (Pollard *et al.*, 1995; McConnachie *et al.*, 1996).

2.7.2 Cultivation of *M.oleifera*

In the natural environment, the tree will withstand temperatures of up to 48°C and a minimum of -1°C, the ideal temperature is between 22-25°C (FRIM, 1997). It thrives in areas with average rainfall of 750-2,000 mm. The geographic location of the tree is limited to tropical and subtropical regions, since it requires strong light. It will grow in all soil types except stiff clays, and prefers alluvial sandy soils (Nautiyal & Venkataraman, 1987). It appears to grow best at altitudes of 100-700 m, but is still found at altitudes of 1000 m above sea level (FRIM, 1995). The author has personal experience of trees flowering and bearing pods at height of 2,000 m in Harare, Zimbabwe.

Estimates of growth rates range from 1 m per year (Nautiyal & Venkatraman, 1987) to 4-5 m per year (Jahn, 1988; FRIM, 1997; CWS, 1998). Initial flowering has been noted after only 6 months and seed harvesting after 12 months, with flowering and fruiting continuing throughout the year, but declining during the dry season. Estimates of yields vary from 1.5-4.5 kg of seed/tree/year (Jahn, 1981; Nautiyal & Venkatraman, 1987; CWS, 1998).

Propagation can be achieved by using cuttings (Ramachandran *et al.*, 1980; Nautiyal & Venkataraman, 1987), or tissue culture (Kantharajah & Dodd, 1991), or from seeds (FRIM, 1995).

A problem associated with using *M.oleifera* seed as a coagulant is obtaining a sufficient quantity for a treatment works to operate effectively. One possible solution is to cultivate fast growing trees, which have more harvests per year with larger pods containing seeds. Currently the Indian short stem variety of *M.oleifera*, Periyakulam 1 (PKM1), is being investigated by the Kenyan Forestry Research Institute (KEFRI) for this purpose. In India, the Jaffna variety had been cultivated to produces pods 120 cm long (Ramachandran *et al.*, 1980). A further variety is cultivated as an annual crop which fruits within 165 days, bears fruit continuously for one year, and then becomes exhausted (Morton, 1991). *M.stenopetala* (Bak.f.) Cuf., a related species, has also to be investigated with a view to hybridisation with *M.oleifera* due to its high survival and growth rates (EERG, 1996). In many countries, *M.oleifera* is already well known where plantations have been established to exploit the nutritional benefits of the tree. Such schemes have been established by: Trees for Life in Orissa, India (Mathur, 1997); CWS in West Africa (Fuglie, 1999); Acäo Evangéliça in Patos, north eastern Brazil (Medcraft, 1999); missionaries in Jeremie, Haiti (Price & Meitzner, 1996); Binga Tree Project in Matabeleland, Zimbabwe (EERG, 1997); Arusha Consumer Services Cooperative Society Ltd., Tanzania (EERG, 1997). Therefore establishing similar plantations in order to obtain the seeds for water treatment purposes would not be impractical

Pests that affect the tree include, *Diplodia* (root-rot), termites (Martin and Ruberté, 1975) and certain root and stem borers (Krantz *et al.*, 1977). Certain green caterpillars cause defoliation during the rainy season, but it was found that the trees recovered during the following dry season (Mayer & Stelz, 1993). This relatively small number of diseases affecting the tree may in part explain the popularity of *M.oleifera* throughout so many countries.

2.7.3 *M.oleifera* products for water treatment

For water treatment purposes, the pods are left to dry on the tree before harvesting. The seeds are removed from the pods and shelled. The soft kernels are then crushed into a fine powder. The powder is then mixed with clean water to make up 1-3% dosing solutions. This is added to the raw water in the same manner as any other coagulant. The preparation of the suspension is relatively simple, due to the water solubility of the active agent. No pH adjustment of the water is required since the polymer has an isoelectric point at pH 10-11 (Tauscher, 1994; Ndabigengesere *et al.*, 1995). The isoelectric point is the pH at which electroneutrality is reached. The polyelectrolyte therefore has an overall positive charge in most natural waters, but some negatively charged sites on the polymer may exist. This is a very important feature of *M.oleifera*, since controlling the influent pH on a treatment works in developing countries can be very difficult, due to potential shortages of chemicals, dosing equipment and skilled labour.

The active constituent in the seed is proteinaceous (Fink, 1984; Gassenschmidt *et al.*, 1991; Ndabigengesere *et al.*, 1995). There are two proteins responsible for the coagulating activity with molecular weights of approximately 6,000-10,000 daltons. The relatively low molecular weight of the protein coupled with its high charge density has led researchers to conclude that the most probable mechanism of destabilisation using *M.oleifera* seeds is charge neutralisation via 'patch' adsorption (Tauscher, 1994; Gassenschmidt *et al.*, 1995). Generally it is only large polymers that induce destabilisation by the 'bridging' mechanism, since the polymer must be larger than the particles which are being destabilised. Moreover, 'bridging' polymers are usually anionic or nonionic, and particulate attachment is not generally electrostatic (La Mer & Healy, 1963). Ndabigengesere *et al.* (1995) undertook a series of jar tests to obtain the optimum dose of *M.oleifera* on a model turbid water. The zeta potential of the treated water samples was also measured. It was found that maximum turbidity removal occurred when the zeta potential was zero. Thus the dominant mechanism of particle destabilisation was considered to be due to charge neutralisation leading to the 'patch' mechanism.

The effectiveness of *M.oleifera* varies considerably depending on the treatment process involved. Turbidity removal rates range from 80-99.5% for highly turbid Nile water (Madsen *et al.*, 1987), 90-99% using kaolin suspension (Folkard *et al.*, 1993), 99% at pilot and full scale at the Thyolo treatment works in Malawi (Sutherland *et al.*, 1994) and 26% using water obtained from the Challawa treatment works in Nigeria (Muyibi & Okuofu, 1995). Bacterial

removal rates range from 90-99.99% with very turbid raw waters (Madsen *et al.*, 1987) to 98-100% for relatively clear water (Jahn, 1988). The dose required to achieve these removal rates varied between 15 and 200 mg/L.

M.oleifera seeds have also been used successfully in the treatment of domestic wastewater. Reductions of 40-50% in the Biochemical Oxygen Demand and Chemical Oxygen Demand, and 70% of the suspended solids and faecal bacteria were achieved by coagulation and flocculation in the primary settlement stages (Travis *et al.*, 1992).

Using *M.oleifera* seed as a coagulant in water treatment produces a relatively small volume of sludge. The sludge produced by aluminium and iron coagulants tend to be voluminous and difficult to dewater. This is considered to be due to the large volume of hydroxide precipitate which needs to be formed to entrap the particulate matter. Therefore, not only is sludge produced by *M.oleifera* biodegradable and therefore valuable as a fertilizer and soil conditioner, but it is also relatively cheap and easy to dewater and dispose of.

As previously noted, the presscake produced after oil extraction from the seeds, can also be used in water treatment. Comparative jar tests carried out in the laboratory at Leicester University on presscake and whole seed sample, found that raw waters of 300 NTU were reduced to 10 NTU by a dose of 20 mg/L of either suspension. Thus the extraction of the oil as the main or by-product, as an extra source of income, would not affect the water treatment capabilities of the seed. It has been estimated that if a water utility established and maintained a plantation of *M.oleifera* trees for oil production and the extraction of the coagulant in the presscake, a net operating profit would be achieved (EERG, 1994). Optima of Africa Ltd., a company based in Tanzania, is investigating the viability of the use of the presscake in water treatment, as they are currently cultivating the tree for the oil extraction purposes.

M.oleifera seeds contain compounds exhibiting strong bactericidal properties. Eilert & Nahrstdet (1981) isolated an antimicrobial agent called 4-(α-L-rhamnosyl) benzyl isothiocyanate (a glycosidic mustard oil (Barth *et al.*, 1982)). There are therefore potential health implications involved with the use of seeds in potable water treatment. Other studies have indicated that there are mutagenic, genotoxic and diuretic compounds present in the roasted *M.oleifera* seeds (Villasenor *et al.*, 1989a & b; Càceres *et al.*, 1991 & 1992). It is unclear if these compounds are found in the non roasted seeds. Toxicity tests conducted on

guppies, protozoa, bacteria and coliphages indicated that the permeability of cytoplasmic membranes were affected by the seed suspension (Grabow *et al.*, 1985). The toxic effect was almost instantaneous on the epithelial cells of the gills of the fish and the oxygen uptake mechanism of the cell membrane of the protozoa. This effect was also seen in the cells of the gastrointestinal tract in the liver of rats (Barth *et al.*, 1982). The lack of toxic effects on some bacteria and phages further indicated that the site of action of the toxicant is the cytoplasmic membrane. Since cell walls present in some bacteria can protect the membrane, and viruses do not possess a membrane as higher organisms do. This is not likely to be significant to human health since concentrations of between 200-1000 mg/L were used in these tests and doses of 200 mg/L is the highest concentration used for coagulation purposes. Also the potentially harmful effects of the toxin are eliminated in the intestinal tract due to interference by organic matter (Grabow *et al.*, 1985). It is unlikely that the seeds have carcinogenic properties, indicated by the negative results of the Ames *Salmonella* mutagenicity assays on various seed preparations (Grabow *et al.*, 1985). The *Salmonella* reverse-mutation assay is the most widely used short term in vitro tests for carcinogenic compounds. It consists of testing for the ability of a chemical to induce histidine-requiring strain of Salmonella to revert to a non-histidine-requiring wild strain (Calabrese & Canada, 1986).

Grabow *et al.* (1985) concluded their study with the following statement: "There is no evidence that the toxin, which occurs in the cotyledon of *M.oleifera* seeds..... may have short term toxic, long term chronic or carcinogenic effects on humans under conventional conditions of utilisation of the seeds for.... water treatment purposes.... All indications are that potentially harmful effects of the toxin in coagulated water are eliminated in the gastrointestinal tract as a result of interference by organic matter and instability of the toxin."

Berger *et al.* (1984) summarised their study with two important conclusions. Firstly, since the coagulant is used to form floc that are removed from the water, it is safe to presume that the majority of the coagulant will be trapped in the sludge. Secondly, since the use of various parts of the tree as a food source is so extensive, especially the pods and the seeds, it would appear unlikely that the seeds would be toxic to humans (Jahn, 1979).

An essential component of an analysis of a new coagulant is an economic consideration of its production. It is, however, very difficult to estimate the cost of *M.oleifera* seed. There are variations in labour, land and material costs in each region. As well as variations in fertility of

the land, which will affect the spacing between trees and potential yields. KEFRI found yields to vary from 2-480 kg/ha at Marigat, on one of their research stations in a very dry region of western Kenya. But it was anticipated that this yield would increase to 2 ton/ha through a process of selection (Chagwony & Odee, 1998). Estimates of yields from West Africa are 4.9 ton/ha, which is the equivalent of about 39-44x10^6 seeds/ha (CWS, 1998). The latest estimates of cost of establishment, management and maintenance of *M.oleifera* plantations from planting to harvest by KEFRI are KES 111,494.03/ha (= £1,114.94/ha) (Odee, 1999). This estimation, however, would only be accurate for the station based at Marigat in eastern Kenya. Plantations in other countries would vary considerably. Despite these difficulties, an economic assessment of replacing the use of alum with *M.oleifera* seed in the treatment work in Thyolo in Malawi was made in 1990. It was estimated that under optimum conditions, *M.oleifera* would cost 18% that of alum (Folkard *et al.*, 1993b).

Currently the major problem associated with using *M.oleifera* seed for water treatment is an insufficient supply of seeds. In areas where the tree is already cultivated, the seeds and/or pods are an important food source, and there is therefore a potential conflict of interests in the use of the seeds. The supply of the seeds would need to be increased, and this could be achieved in a number of ways:

- Cultivation of the tree extensively as a cash crop in areas where it will thrive, in the same manner that tea and coffee are grown in restricted regions and then sold globally.

- Establishment of plantations of the tree in conjunction with specific treatment works to ensure a consistent and reliable supply.

- Encouraging local farmers to grow the tree by ensuring a market for the seed and possibly the oil produced.

- Extraction of the proteins active in water treatment from the presscake, thus not competing with oil production from the seeds. Research conducted by the EERG, Campden & Chorleywood Food Association, UK and Optima of Africa Ltd., has lead to the production of 'Phytofloc', which is an extract of the presscake containing the proteins active in water

treatment. Further details of this process can not be given for reasons of confidentiality (Optima of Africa, 1999).

- Synthesis of the active protein by cloning the nucleotide sequence using plastids in *Escherichia coli* (Tauscher, 1994). This high tech solution would mainly be appropriate for the supply of the coagulant to treatment works in developed countries.

There are a number of operational considerations in using *M.oleifera* seeds in water treatment. Firstly, the increase in organic matter in the treated water. The potential exists for regrowth of the bacteria in treated water which is allowed to stand for extended periods of time. Madsen *et al.* (1987) reported a regrowth of certain pathogens in the treated water over a 24 hour period, following a 90-99.99% reduction within the first 2 hours. *Salmonella typhimurium* and *Shigella sonnei* consistently regrew and *E. coli* was occasionally found. This secondary increase in pathogen numbers is a greater problem with warmer tropical waters, which present ideal conditions for the regrowth of certain faecal bacteria. Another problem with the increase in organic matter is that it may combine with chlorine, acting as a precursor for the formation of THM that are potentially carcinogenic (USEPA, 1991). This point will be discussed further in Chapter 6. A method of controlling the organic content of the final water would be to use an extract of the seed rather than the whole seed as the coagulant. Work undertaken in the laboratory at Leicester University has shown that concentrating the active proteins by freeze drying the water soluble fraction of the seed outperformed the whole seed on a weight for weight basis (Folkard & Grant, 1989).

Secondly, the effectiveness of the coagulant suspension is diminished over time. Once the seed suspension has been prepared then it must be used within 8-12 hours, depending on the temperature. The active proteins begin to breakdown and are thus no longer effective as a coagulant, possibly due to the action of proteases produced by bacteria present in the seed or the water. The use of *M.oleifera* seed in a full scale treatment works would therefore entail the preparation of seed suspensions at regular interval to avoid the need for storage of the suspension.

Thirdly, seed powder and shelled seeds need to be stored in airtight containers to ensure the coagulant remains active. Seed powder stored in the laboratory at Leicester University in airtight containers for 2 years has remained active as a coagulant. The majority of coagulants

whether of natural or synthetic origin require storage in airtight conditions to prevent degradation, and therefore *M.oleifera* seed powder is not unique in requiring such attention.

Despite the fact that using this natural coagulant in developing countries clearly offers some significant advantages, *M.oleifera* seeds have not been utilised as a coagulant to any great extent. It has been used in traditional water treatment practices in Sudan, on a household level (Jahn, 1981) and by the Njemps people from Lake Baringo in Kenya since 1973 (Muluvi, 1998). A full scale trial and a number of pilot studies have been undertaken at Thyolo treatment works in southern Malawi (Sutherland *et al.*, 1994). The lack of uptake of the use of *M.oleifera* seed in water treatment may be due to the fact that it is viewed as being low technology and therefore in some ways inferior to the synthetic chemicals. Ndabigengesere & Narasaih (1998) consider the solution to this to be to emphasise the advantages of natural coagulants while utilising modern technology to supply the water, thereby reducing the cost of treatment but not the quality of the water. A further solution would be to make the use of *M.oleifera* seed a viable commercial proposition; as has been be undertaken by Optima of Africa Ltd., who are encouraging the growth of the trees by guaranteeing the purchase of the seeds. With the purchase of sufficient seed, the company is then able to produce the oil and the coagulant on a commercially viable scale.

CHAPTER 3

MATERIALS, EQUIPMENT & METHODS

3.0 Materials, Equipment & Methods

This chapter identifies the materials, equipment and methods used in the first phase of the research, consisting of the laboratory study conducted in the Department of Engineering at Leicester University.

3.1 Materials

3.1.1 The model turbid raw water

The raw water used in the experimental filter runs was made by the addition of varying amounts of stock kaolin clay solution to deionised water and then mixing thoroughly. The procedure for the make-up of the stock kaolin solution is described in Appendix 1. The use of kaolin clay to simulate natural waters is an accepted procedure (McCooke & West, 1978). The justification for its use is that it adds particulate matter to the water, and it remains stable and will not self-flocculate, which can happen with some natural raw waters. For the majority of tests conducted, the raw water consisted of kaolin stock solution diluted in deionised water, to ensure chemical consistency. In practice, natural raw waters generally provide enhanced treatment performance compared to kaolin in deionised water, due to the ionic content and the increased particle size range. Therefore using kaolin suspended in deionised water is a more problematic water to treat. Tap water was used in the make-up of the raw water in the tests conducted with alum as the coagulant. This was necessary since alum requires a certain amount of alkalinity in the water in order to effectively form floc, as discussed in Section 2.2.2. Deionised water does not contain alkalinity and therefore alum would not be able to form the hydroxide precipitate that is responsible for entrapping and subsequently removing particles.

3.1.2 Preparation of *M.oleifera* seed powder and suspension

Preparation of the suspension to be used in all the experimentation consisted of firstly reducing the seeds to a fine powder and then using the powder to prepare the suspension. The initial filter runs conducted in the laboratory used seeds harvested by the Forestry Research Institute of Malawi (FRIM) in the Nsanje region in Southern Malawi in 1995. This initial seed stock was eventually exhausted and later tests were conducted using seeds harvested by KEFRI in January and February 1997. All the seed stocks were stored in airtight containers in the

laboratory at Leicester University until use. Comparative tests of the effectiveness of the two seed batches were undertaken. This consisted of jar testing in the laboratory. Jar tests are the most commonly used method for evaluating and optimising the coagulation- flocculation processes (Bratby, 1980; Kawamura, 1991a). The jar stirring equipment used was manufactured by Aztec Environmental Control Ltd. The test consists of placing six 1-litre beakers containing the water to be treated under six mixing paddles. A specific coagulant dose is added into each beaker, and a set mixing regime is started. In this case it consisted of fast mix at 300 rpm for 2 minutes, and then a slow mix at 30 rpm for 15 minutes. Each of the 1-litre samples was then filtered through 2 sheets of Whatman No. 1 filter paper, and the filtrate turbidity was then measured. This procedure was repeated three times for the two batches of seed powder. The results obtained are shown in Figure 3.1. Since there was no significant difference in performance of the two seed batches, it was considered acceptable to change seed supplies during the course of the study.

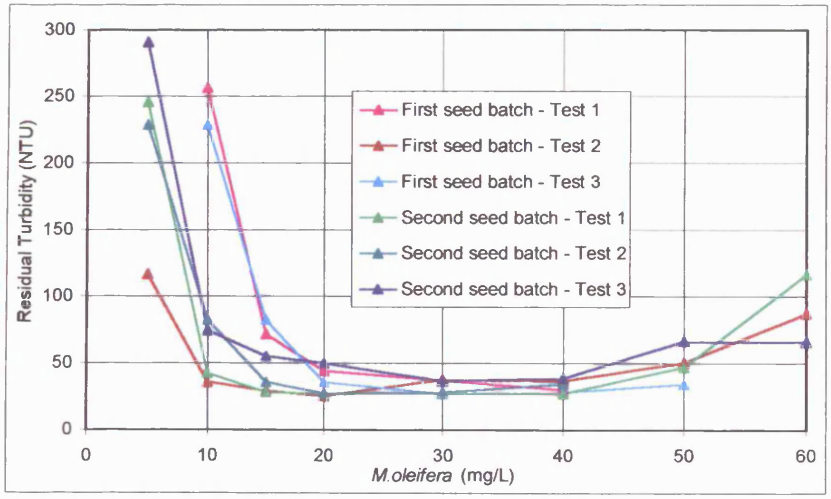

Figure 3.1 A comparison of the consistency of effectiveness of the two seed supplies used in the laboratory tests.

(Seeds shelled by hand and ground using a coffee grinder. Raw water turbidity 300 NTU. Jar testing consisted of 2 minutes @ 300 rpm, 15 minutes at 30 rpm and 30 minutes settling)

3.1.2.1 Preparation of seed powder

The preliminary step seed powder preparation consisted of shelling the seeds. The first seed batch was manually shelled. The second much larger batch was shelled using a stone milling machine. The shelled seeds (kernels) were then reduced in size to a fine powder. This increases

the surface area and hence the rate of dissolution of the coagulant when water was added. Previously it has been found that inadequate grinding of the kernels to produce a powder, resulted in inefficient floc formation and turbidity removal (Travis *et al.*, 1992). Size reduction is achieved by cutting the kernels into very fine pieces in a process of comminution. With the first seed batch this was achieved by cutting the seeds using a domestic coffee grinder. Recognising that the pilot rig testing would require a large amount of seed material, it was decided to establish an industrial scale up procedure that could be consistently adopted. For the second batch a full scale industrial comminuting machine was used. A range of comminution equipment manufactured by Urschel International Ltd. was tested. These operate by rotating the product at very high speeds inside a stationary reduction head. The resulting centrifugal force pushes the product outwards and against the inner periphery of a cutting edge using an impeller. Since the product passes through the head at very high speed, there is no random particle movement and therefore a consistent particle size can be assumed. This is necessary to ensure that a consistent amount of the active protein is dissolved in each batch of seed suspension prepared from the powder.

The criteria used to find the optimum comminuting machine was two-fold. Firstly a comparison of the effectiveness of the seed powder at reducing turbidity, compared to that produced by the coffee grinder. Secondly the amount of heat produced during the comminution process. This was important because excessive heat could denature the active proteins. Furthermore, excessive heat would cause oil to exude from the kernels, which may effect blinding of the fine knife-columns in the cutting head on the machine. Hence, the temperature of the seed powder was measured as it was produced. For the former criteria, a series of jar tests was undertaken using the various batches of seed powder. The results of these comparative tests are shown in Figure 3.2. It can be seen that the Comitrol Processor model 3600 (diocut impeller), was as effective as the seed powder produced by the coffee grinder. It was assumed that the proteins were not denatured by this process, as there was no significant rise in temperature of the powder and no oil was evident in the cutting head.

Further checks were made to asses the range of particle sizes produced by the two methods of comminution. Table 3.1 indicates the percentage by weight of each particle size range present in the sample. It can be seen that for both samples the particles were mainly between 0.500-1.000 mm, and the largest group ranged in size between 0.500-0.855 mm. Although there are some differences in the size profiles of the two samples, effectiveness in terms of turbidity

removal during jar tests were very similar, as seen in Figure 3.2. Consequently, it was considered acceptable to change the method of producing the seed power from the coffee grinder to the Comitrol 3600.

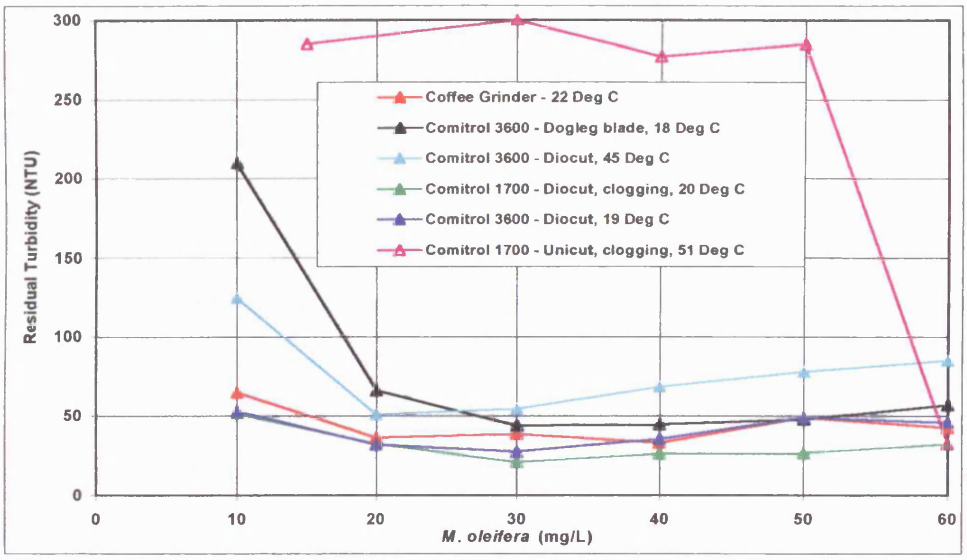

Figure 3.2 A comparison of jar test results obtained using seed powders produced by various comminutors and the coffee grinder, showing the type of blade used, the extent of clogging and the temperature of the resultant powder .

(Raw water turbidity - 300 NTU. Jar testing consisted of 2 minutes @ 300 rpm, 15 minutes at 30 rpm and 30 minutes settling)

Sieve Aperture (mm)	% by weight (seed powder produced by the coffee grinder)	% by weight (seed powder produced by the Comitrol 3600)
> 1.000	17.2	21.7
0.850-1.000	9.9	14
0.500-0.850	35.2	29.7
0.500-0.425	16.8	18.1
0.425-0.315	12.6	8.5
0.315- 0.250	6.5	4.1
< 0.250	1.8	3.9

Table 3.1 Results of sieve analyses on the seed powders produced by the two methods of comminution.

3.1.2.2 Preparation of seed suspension

In order to prepare a litre of 1% seed suspension, the following materials were used:

- 10 g of seed powder

- 1 litre of deionised water
- 1.7 ml of sodium bicarbonate stock solution. (Stock solution consisted of 65 g of sodium bicarbonate per litre of deionised water.)

The seed powder and a small amount of the water, containing the sodium bicarbonate solution, were mixed into a paste using a pestle and mortar. Gradually the remaining water was added to the paste and mixed thoroughly. The resulting suspension was strained through a muslin cloth. The suspension was then allowed to stand for 5 minutes and the supernatant decanted off and used as the coagulant solution. It was necessary to prepare fresh seed suspension for each run, as has been suggested by previous workers (Jahn, 1981 & 1986) and by experimentation undertaken by the author. This is considered to be due to the potential bacterial degradation of the active proteins. The reduction in activity of the seed suspension was often accompanied by the smell of sulphur, which was an indication of the breakdown of sulphur bonds in the protein (Freemantle, 1990).

The dose of *M.oleifera* seed in this research is given in mg/L, but it should be noted that this refers to the whole seed. The dose of the active material used is considerably less. One method of estimating this dose was to measure the weight of soluble matter in the seed, since this fraction contains the coagulant (Folkard *et al.*, 1993). Laboratory testing showed that approximately 30% by weight of the seed is water soluble. This agrees with other researchers' estimations of 25% (Ndabigengesere & Narasiah, 1998). Therefore, the dose quoted in this research is a minimum of 3 to 4 times greater than the dose of the active component due to the extraneous seed material present. This is important when comparing doses of *M.oleifera* seed with other coagulants.

Initially the seed suspensions were prepared using tap water, but significant differences in the in effectiveness of the treatment of various batches of suspension led to the use of deionised water. This lack of consistency was considered to be due to the variation in manganese and calcium content of the tap water in the Leicester area, resulting from the use of more than one source water. Sodium bicarbonate solution was added to the deionised water during the preparation of the seed suspension for two reasons. Firstly deionised water is unrepresentative of natural waters, since it lacks an ionic background. Also, it has previously been found that protein extraction is more effective in waters with an ionic concentration (Moen, 1995). This 'salting-in' effect was explained by Okuda *et al.* (1998) to be due to the increase in protein-

protein dissociation as the ionic strength of the solution increases. The increase in protein bond breakage causes an increase in its solubility and therefore coagulation efficiency. Secondly, it ensures repeatability, since small variations in the ionic concentration of the deionised water would be masked by the addition of a relatively large concentration of the sodium bicarbonate solution.

3.1.3 Preparation of the chitosan solution

The chitosan solution was prepared by mixing 1 g of dry chitosan into 100 ml of 1% acetic acid solution on a heated magnetic stirrer for about 12 hours. The chitosan was supplied by Rigest Trading Ltd., UK., and was manufactured entirety from shrimp shells; the molecular weight was $1x10^6$-$2x10^6$ daltons. The acetic acid was prepared by the addition of 1 ml of 99.8% acetic acid (Sigma-Aldrich Chemicals) into 100 ml of deionised water. The 1% chitosan solution was subsequently diluted down to a 0.01% dosing solution using 1% acetic acid. The dosing solution was stored in a glass stoppered flask and used within 2 weeks.

3.1.4 Preparation of the alum solution

The solution was prepared by the addition of 10 g of Al_2 $(SO_4)_3.14H_2O$ (BDH Chemicals, General Purpose Reagent) into 1 litre of tap water. The solution was thoroughly mixed and stored in a glass stoppered flask for up to 1 month, based on work conducted by Schulz and Okun (1992).

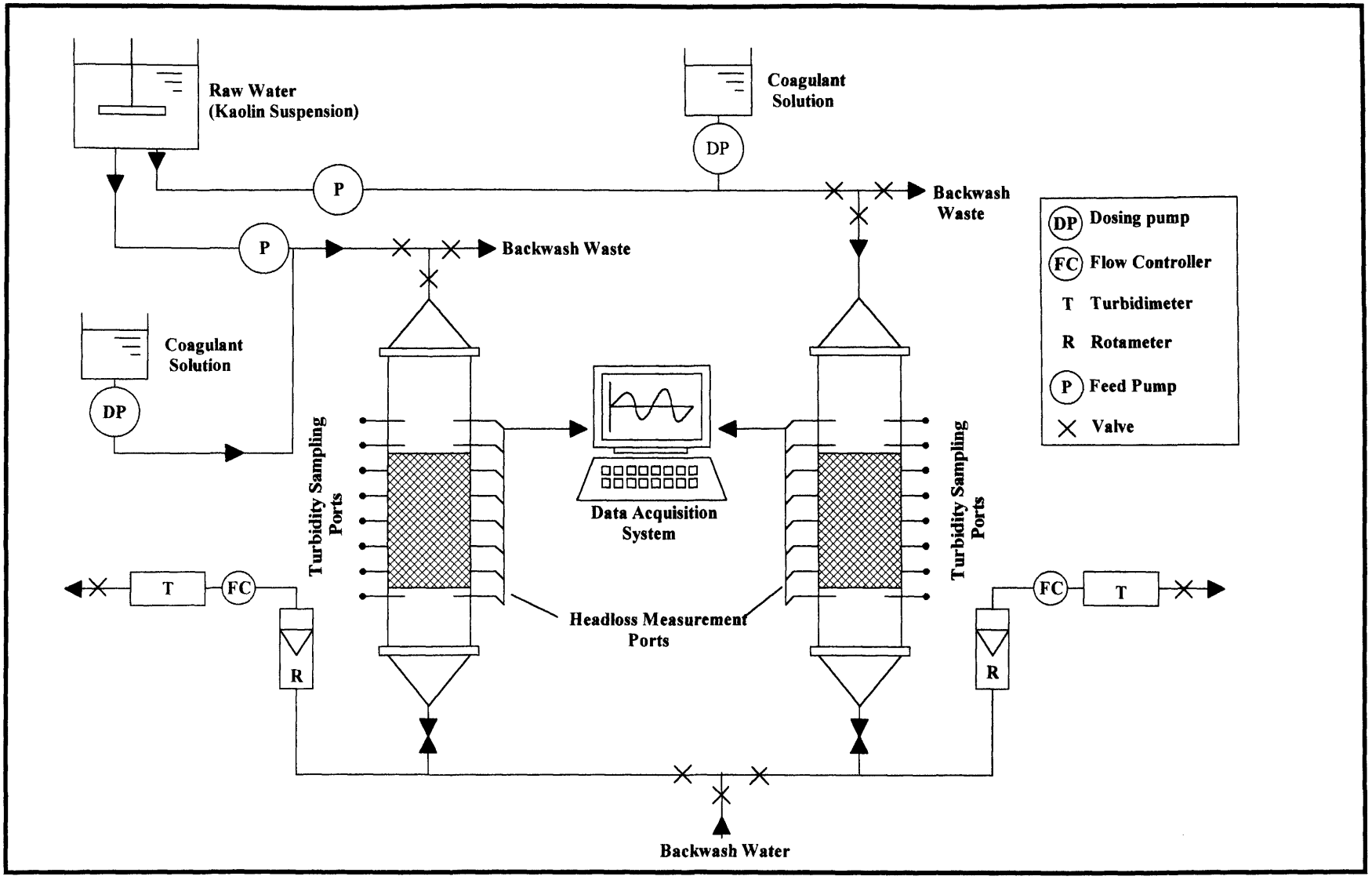

Figure 3.3 Schematic of the laboratory pilot rig.

3.2 Equipment

The laboratory pilot rig is illustrated schematically in Figure 3.3. Plate 5 shows the two perspex columns, of internal diameter 100 mm, with the header tanks above. Plate 6 shows the headloss and turbidity sampling ports along the walls of the two columns.

3.2.1 Rig development

As discussed in Chapter 1, preliminary investigations into the use of *M.oleifera* seed in conjunction with the CFF were undertaken on a laboratory rig at Leicester University. The same rig was used but substantially modified and reconstructed for the present study. The principle modifications were as follows:

- The old rig consisted of 1 column. The modified rig consists of two columns, which can be operated in parallel in upflow or downflow mode, or alternatively in series. For example it can be used as an upflow-downflow filtration system. Figure 3.3 illustrates the rig in the downflow mode. The pipework has push fit joints to allow easy conversion into upflow mode.
- Headloss data on the original rig was measured manually using simple piezometric tubes. On the modified rig, headloss was measured and logged continuously using a pressure transducer, linked to a computer via a data logger.
- The original filtration system was gravity fed. The water in the new rig was pumped around the system. Flow controllers were added and placed after the rotameters in each column. This ensured that the flow through the beds remained constant, despite minor flow disturbances in the flow through the pumps and increases in back pressure in the bed due to clogging of the filter media.

3.2.2 Filter media

The three types of filter media used in this study were:

- Sand classified as British Standard (BS) sieve size 16-30 (particle size range 0.50-1.00 mm) and a uniformity coefficient (UC) of approximately 1.4.

- Sand classified BS sieve size 10-18 (particle size range 0.85-1.70 mm) and a UC of approximately 1.4. Both sands were supplied by United Mineral Suppliers Ltd.
- Anthracite classified as BS sieve size 7-14, grade 2 (particle size range 1.2-2.5 mm), with a hardness of between 2.75-3.25 on the Moh Hardgrove Index, a UC of < 1.5 and a specific gravity of between 1.35-1.45. The anthracite was supplied by Waterforce UK.

There is a maximum size of media grain that should be used in a pilot column, due to the column wall effect on the porosity of the filter bed. According to the AWWA (1992), the ratio of the average media grain diameter to the column diameter should be no less than 1:50. As the column diameter was 150 mm, the media was considered to be appropriate for use in the experimental pilot columns.

3.2.3 Headloss measurement

Headloss development in the filter bed was monitored at regular intervals throughout the depth of the filter media via a number of ports, the position of which can be seen in Figure 3.4 and Plate 5. This was achieved by 8 brass tubes of diameter of 3 mm, extending 50 mm into the media bed. The tip of each tube was covered by a fine mesh that excluded sand from entering the tube. The other end of the tube was connected to a solenoid valve. Plastic tubing then connected all 8 valves to a pressure transducer. The use of a computer program (via a selector box) allowed valves to be opened and closed at set time intervals to sample and record the transducer response signals. These signals were then converted into pressure readings, with reference to the top measurement port, based on a calibration obtained using a mercury manometer, which was undertaken prior to every run. This data was then stored in a database on the computer to facilitate data acquisition, display and analysis. The significance of measuring headloss at various depths in the bed is that it indicates the extent of clogging of the media and the distribution of the particulate matter deposited in the bed.

3.2.4 Turbidity measurement

Turbidity was measured using a HACH turbidimeter (Ratio/XR), as seen in Plate 7. This instrument was maintained and calibrated regularly by the equipment supplier Camlab Ltd. using Formazin standards. This calibration was then checked at weekly intervals in the

laboratory using Gelex Secondary Standards. The final filtrate turbidity was measured in a flow-through cell in the turbidimeter. Turbidity of the water was also measured at intervals along the column. This was achieved by the use of turbidity sampling ports at various depths in the media, their position are diametrically opposed to the headloss measurement ports and can be seen in Figure 3.4 and Plate 5. The ports consist of stainless steel tubes extending 5 mm into the media, with their other end connected to sample draw-off tubes. Roller clamps at the end of these tubes allowed for sample draw-off at low flow rates, avoiding disturbance to the flow regime in the bed. Samples of 30 ml were drawn off, with the first 20 ml used being discarded to ensure the sample measured was obtained from the bed and not a remnant of the previous sample present in the tube.

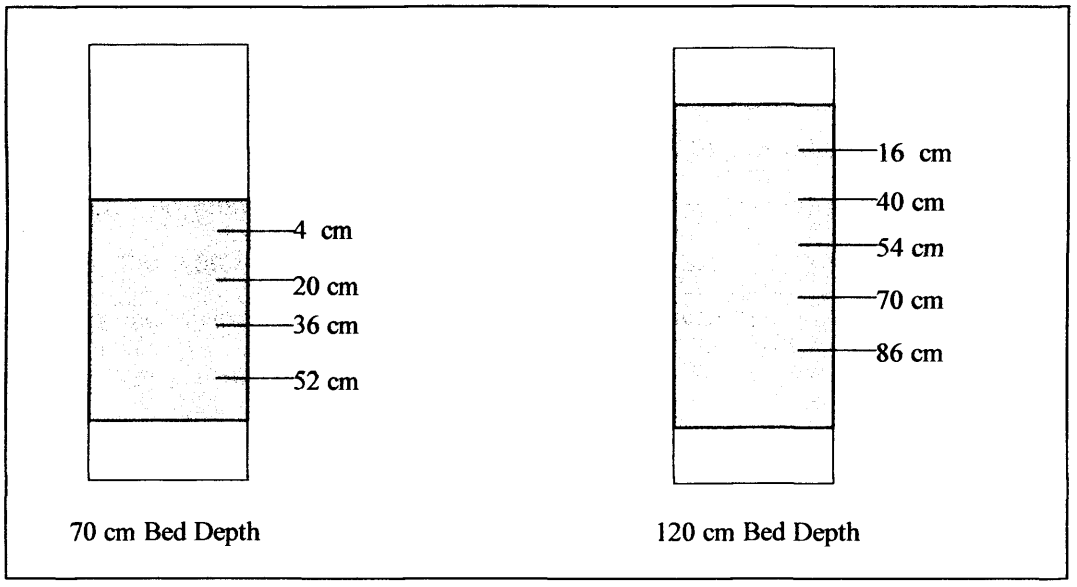

Figure 3.4 Schematic of the positions of the headloss and turbidity measurement ports in the two bed depths considered.

3.3 Methods

3.3.1 Operation of the pilot plant

The raw water was prepared in the header tanks and was stirred continuously after the addition of the kaolin suspension to ensure consistency of the concentration of the suspension. The pH, conductivity and temperature of the water were measured and recorded. The water level in the tank was kept constant by two feeder tanks. The water was pumped from the tank, dosed with the coagulant, then entered the column via a plastic tube with a diameter of 5 mm and a length of 2.73 m. Dosing was achieved by a peristaltic pump powered by a separate power supply, as seen in Plate 8. The power input into the pump was then varied according to the required dose rate and checked at regular intervals throughout each run. Rapid mixing occurred in the narrow tube before entering the column. Turbulence in the flow was also induced by the change in velocity of flow from a 12 mm diameter pipe to a 5 mm tube. The passage of the flow through two elbow joints before entering the column further enhanced mixing. Once in the column, the water entered the filter media, where headloss and turbidity removal were monitored at the various set depths in the bed. The flow was then measured by a rotameter and kept constant by a flow control valve. This automatically adjusted for variations in the flow by changing the diameter of the orifice through which the water entered. Finally the turbidity of the filtrate was measured in a turbidimeter via a flow through cell. The turbidity reading was measured continuously and registered on a chart recorder. Each experimental run was repeated a minimum of 3 times in order to ensure repeatability of results in terms of filtrate quality, headloss development throughout the bed, and volume of filtrate produced. Some variation in the results were found, which were generally attributed to ineffective deionisation of the water and the consequent contamination of the seed suspension or the raw water. Variations in the headloss data were generally caused by the presence of small pockets of air trapped in the filter bed. This problem was eliminated by firstly backwashing the bed thoroughly using a high flow rate, ensuring complete fluidisation of the filter media; and secondly by tapping the column wall before the start of each run to ensure all the bubbles attached to the media surfaces were dislodged.

The filter runs were continued until specified termination points were reached, as discussed below. After each run the filter bed was backwashed using tap water. The washwater entered

the base of the column, travelled up through the filter media and out of the top of the column to waste. The velocity of flow of the backwash water was sufficient to create a 50% volumetric expansion of the media. The washing process was continued until the washwater had a turbidity < 2 NTU.

In this study the filter runs were terminated when the residual turbidity exceeded 5 NTU or the headloss increased to over 240 cm, which ever was reached first. The residual turbidity limit is based on the WHO Guideline value for potable waters in developing countries (WHO, 1996). The headloss development limit is based on a practical maximum value (Adin & Rebhun, 1974; O'Melia & Stumm, 1967).

3.3.2 Rig commissioning

Initial tests consisted of running the new rig and comparing the results with those obtained from the old rig. Similar results were obtained with the two rigs using raw waters with turbidities of 35 and 50 NTU, at filtration rates of 10 m/h, and a bed depth of 70 cm. This indicated that the results found in this study were not merely a function of the pilot column.

Experimental runs were then conducted to compare the treatment performances in the two columns in the new rig. These ensured that any variation in effectiveness of the treatment process was due to the parameter being investigated and not the configuration of the column. Typical comparative runs can be seen in Figures 3.5 and 3.6. It can be seen that turbidity removal rates and headloss development were virtually identical in the two columns.

The effect of raw water temperature was also evaluated. The temperature of the water in the header tanks ranged from 5-20°C, which was considered potentially influential on treatment effectiveness for two reasons. Firstly, flocculation rates are higher at higher temperatures. Secondly, transport mechanisms in the filter bed are directly affected by temperature, and indirectly affected by viscosity, as seen in Equations 2.4 and 2.5. Consequently filter runs with the same hydraulic conditions conducted at different times of the year with varying raw water temperatures, were compared. It was found that the temperature variation of the water had no significant effected on treatment performance.

A further investigation was undertaken into the effect of flow in the 1 m space above the media bed in the filter column, termed the free water zone (FWZ). It was considered possible that slow mixing - orthokinetic flocculation, was occurring in this zone due to the decrease in flow velocity. If this was the case then the consequences were two-fold. Firstly, the treatment being investigated would be more analogous to DF than CFF. Since the objective of this research was to investigate the process of CFF, floc formation and deposition must occur in the filter bed. Secondly, investigation into the effects of media bed depth would be influenced by the extent of mixing before filtration and not just variation in bed depth, since the increase in depth would involve reducing the FWZ. Potentially orthokinetic flocculation may be occurring in the FWZ, such that particle collision and floc formation were occurring due to the fluid motion caused by velocity gradients. The velocity gradient, or G values, can be calculated based on the headloss generated by the flow through the FWZ. At a filtration rate of 10 m/h, the G value in the FWZ was calculated to be 0.16 s^{-1} (details of calculations are given in Appendix 2). For effective flocculation to occur, G values of between 30 and 100 s^{-1} would be required (Montgomery, 1985). Thus by calculation, the flocculation potential in the FWZ was considered to be insignificant to the treatment process and confirmation was then sought experimentally.

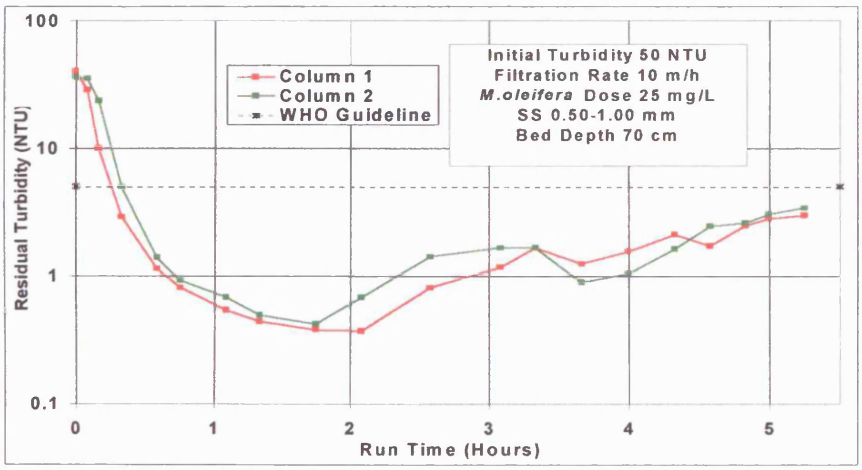

Figure 3.5 Turbidity removal in the two columns of the new rig.

Figure 3.6 Headloss removal on the two columns of the new rig.

The effect of removing the FWZ on the treatment process was therefore also investigated. This consisted of increasing the depth of media in the first column (column 1), thus removing the FWZ. The second column (column 2) was used as a control and consisted of a media depth of 70 cm. The turbidity measurement for column 1 was taken from a depth of 70 cm, thus enabling a direct comparison of the removal of the FWZ. The results obtained from running the two columns are shown in Figure 3.7. It can be seen that with an equivalent bed depth, but reducing the FWZ in column 1, the filtrate quality was very similar to column 2. The effect of the FWZ on subsequent test runs was thus considered to be negligible.

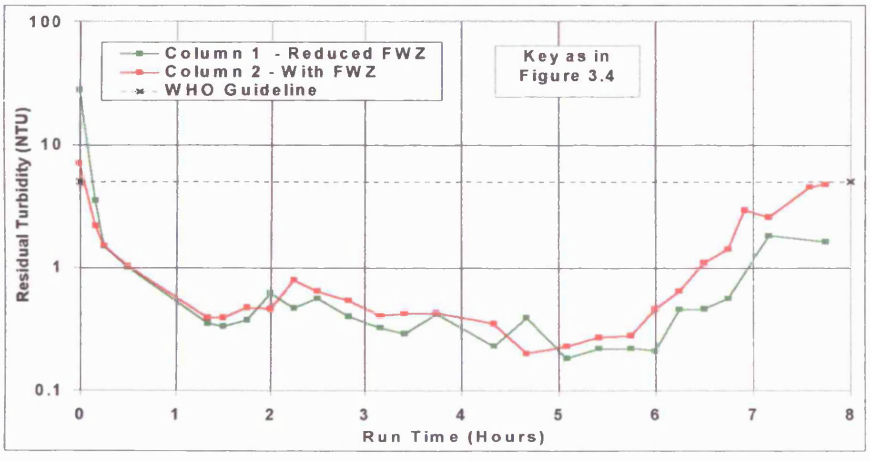

Figure 3.7 Turbidity removal resulting from a reduction of the free water zone

3.3.3 Determination of the optimum dose

The optimum dose for all test conditions was initially found by jar testing, as described in Section 3.1.2. Five doses were selected and added to the jars, the sixth jar being used as a control with no coagulant dose. These were then fast mixed at 300 rpm for 2 minutes, then slow mixed at 30 rpm for 15 minutes. Each of the 1 litre samples was then filtered through 2 sheets of Whatman No. 1 filter paper, and the filtrate turbidity was then measured. A period of slow mixing was used in the jar tests despite the fact that there is no separate flocculation period in CFF. This was because using filter paper to simulate the filtration stage does not allow for 'contact' flocculation to occur in the same manner that would occur in a filter bed. A range of optimal doses was then identified, which was further confirmed using the pilot plant, operating with 2 or 3 different doses. Examples of such curves can be seen in Figure 4.1- 4.5. The optimum dose was found from such curves and based on four criteria:

- The rate of ripening of the filter bed.
- The minimal turbidity reached during the working stage.
- The volume of filtrate produced before either of the termination points was reached.
- Headloss development rate.

CHAPTER 4

LABORATORY PILOT RIG RESULTS AND OPTIMISATION OF THE HYDRAULIC VARIABLES

4.0 Laboratory pilot rig results and optimisation of the hydraulic variables

4.1 Introduction

Deep bed filtration (filtration in deep porous beds) is one of the most commonly used unit operations for particulate removal in water treatment. Consequently a considerable amount of research has been conducted into its optimisation. Attempts to develop theories that quantitatively predict solid removal in a filter bed have met with little success, and as yet no universal model of filtration has been developed. This is due to the heterogeneous nature of the filter media and the particulates in the raw water, coupled with a poor understanding of the hydrodynamics of flow through a clogging bed. Essentially filtration is a dynamic process, which never truly achieves steady state, and its optimisation is often considered to be as much an art form as a science (Kawamura, 1975). A study conducted by O'Melia & Stumm (1967) on the subject of water filtration theory concluded with the following two quotes:

> "*It is apparent that any attempt to work out an exact mathematical description, with theoretical constants, of the filtration process to hold for any conditions of the filter operation is bound to fail. Obviously it will always be necessary to determine the parameters of the process experimentally*" (Mints, 1966).

> "*Pilot studies ....are recommended prior to the design of water treatment plants, as a means of selecting the coagulating chemicals, coagulants and filter aids and filter media; and of determining filter rates, wash rates and size, number and dimensions of units*" (Camp, 1964).

Hence optimal conditions of operation in most filter beds are found by the empirical process of a pilot study. These are generally undertaken prior to the design stage of a treatment works in order to obtain critical information on the effect of the basic hydraulic parameters. Operation at optimal conditions is essential to reduce running costs and produce a high quality filtrate. It is considered that optimal conditions are reached if terminal headloss coincides with turbidity breakthrough (Mintz, 1966; 1969; Ives, 1969), as discussed in Section 2.5.

This laboratory study was conducted in order to investigate the effect of media size, bed depth, filtration rates and media configuration on treatment effectiveness using CFF in conjunction with *M.oleifera* seed. The two sand sizes used in the study had average sizes of 0.75 and 1.28 mm. The former was considered to be within the normal size range used in the majority of treatment works (D. Moran *et al.*, 1993; UMS, 1997). The latter was larger than the media normally used for drinking water treatment, and is generally used in wastewater treatment (UMS, 1997). It was chosen for use in this study to contrast with the smaller media. It was expected that an increase in media size would cause a reduction in the headloss development rate and thereby reduce the rate of turbidity breakthrough and increase the total output of filtrate for higher turbidity raw waters.

Filter bed depths, in RFF, generally range from 0.6-0.8 m (Kawamura, 1991a) or 0.6-1.0 m (Gray, 1994); a 70 cm bed depth was chosen for this study. In order to compare the available collector surface area in the filter bed for the two media sizes considered, a depth of 120 cm was also used. This is explained fully later.

Filtration rates generally used in RFF are in the region of 5-10 m/h (D. Moran *et al.*, 1993 & Kawamura, 1991). But filtration rates up to 25 m/h are used in high rate CFF systems, although with such hydraulic conditions, deeper beds and dual media beds would normally be utilised. Rates of 5, 10, 20 m/h were investigated in dual and single media bed configurations, thus covering the full range of filtration conditions. The filter media used in this study were silica sand and anthracite, the two most commonly used filtration media (Kawamura, 1991b).

The turbidity of the raw waters investigated in this study ranged from 5-75 NTU. The field study, discussed in Chapter 6, was conducted on a raw water with a turbidity of about 5 NTU. This value was also chosen as the minimum turbidity to be considered in the laboratory study, to enable a direct comparison. The maximum turbidity considered appropriate for treatment by CFF range from 25-200 NTU (Culp, 1977), although other parameters also influence optimisation such as colour and organic content of the raw water. Higher turbidities were also investigated in this study because it is possible for a low turbidity water source, which is suitable for treatment by CFF, to have temporary spikes of high turbidity, for example after a storm. Also many surface waters experience seasonal variations in turbidity. Therefore it was considered necessary to investigate the effectiveness of the treatment process in dealing with medium turbidity raw waters.

Many of the above parameters have been investigated for CFF within specific studies (Adin & Rebhun, 1974; Darby *et al.*, 1991; Clark *et al.*, 1992; D. Moran *et al.*, 1993, M. Moran *et al.*, 1993 & Kau & Lawler, 1995). But this research is unique in that the coagulant used was the natural polymer extracted from *M.oleifera* seed, which has not previously been investigated in conjunction with CFF.

The results obtained from studying the effect of these hydraulic variables produced a large quantity of data, the display of which was complex. Outline details of all the runs conducted are shown in Table 4.1. A more detailed display of the spatial and temporal dynamics of turbidity removal and headloss development rates for individual runs are shown in the figures at the end of this chapter.

The effectiveness of the filtration process was investigated in terms of quality of filtrate produced, quantity of filtrate produced before termination and the rate of headloss development across the filter bed. The residual turbidity, i.e. the turbidity of the filtrate, was plotted as a log scale since the variation in turbidity in the working stage was very small. The volume of filtrate produced was measured in m^3/m^2 (the equivalent output for a filter of surface area $1m^2$). This allows a direct comparison to be made between the results obtained from other pilot rigs with different diameter columns.

As the filtration rate through the filter bed was kept constant throughout each run, it may be assumed that any increase in headloss was as a result of particle removal and attachment to the filter media, causing an increase in fluid drag and loss of permeability. Thus this parameter was used in conjunction with the turbidity of the filtrate to indicate the efficiency of removal.

A more informative method of demonstrating the headloss development data is by plotting the hydraulic gradient against time. This is a measure of the headloss development per unit bed depth, in this case cm headloss development per cm bed depth for each section of the bed. It is thus a normalisation of the headloss development data since the sections between measurement ports were of unequal length. This measurement assumes a uniform gradient over the length of the sections between ports. A typical example of this can be seen in Figures 4.15 and 4.16, showing comparative hydraulic gradients with the two media sizes (further examples of this can be seen in Figures 4.43, 4.44, 4.53 and 4.54).

The initial headloss across the larger media was found to be consistently lower than with the smaller media, and with lower filtration rates. Therefore, to make a fairer comparison between headloss development rates, the additional headloss was calculated and plotted as a function of time. The additional headloss is defined as the increase in headloss above the initial clean bed value, and is a commonly used parameter (O'Melia & Ali, 1978; Darby & Lawler, 1990; D. Moran *et al.*, 1993). This data can be seen in the insets in Figures 4.6, 4.30, 4.40 and 4.46. It is a useful parameter when conducting comparative studies, but is of less significance in a treatment works, where the absolute headloss is the terminating factor. A further method of displaying the data can be seen in Figures 4.17 and 4.18, where the spatial increase in headloss is plotted as a function of time

Run No.	Initial Turbidity (NTU)	*M.oleifera* mg/L	Filtration Rate (m/h)	Media Specification (mm)	Bed Depth (cm)	Minimum Turbidity (NTU)	Turbidity on Termination (NTU)	Headloss on Termination (cm)	Total Output (m^3/m^2) [1]	Length of Run (Hours)
A1	75	50	10	S.[2] 0.50-1.00	S. 70	0.8	5.0	182	20.0	2.0
A2	75	25	5	S. 0.50-1.00	S. 70	0.2	5.0	194	75.0	15.0
A3	75	50	10	S. 0.85-1.70	S. 70	2.8	5.0	128	10.0	1.0
A4	75	25	5	S. 0.85-1.70	S. 70	0.6	5.0	80	70.0	14.0
A5	50	35	20	S. 0.50-1.00	S. 70	3.3	5.0	240	16.0	0.8
A6	50	25	10	S. 0.50-1.00	S. 70	0.3	5.0	170	54.0	5.4
A7	50	25	5	S. 0.50-1.00	S. 70	0.6	5.0	105	55.0	11.0
A8	50	25	20	S. 0.85-1.70	S. 70	2.3	5.0	105	26.0	1.3
A9	50	25	10	S. 0.85-1.70	S. 70	2.0	5.0	50.5	33.0	3.3
A10	50	25	5	S. 0.85-1.70	S. 70	0.4	5.0	65	45.0	9.0
A11	35	25	20	S. 0.50-1.00	S. 70	1.7	5.0	N/A	56.0	2.8
A12	35	25	10	S. 0.50-1.00	S. 70	1.0	5.0	170	78.0	8.0
A13	35	15	5	S. 0.50-1.00	S. 70	0.1	3.4	240	150.0	30.0
A14	35	25	20	S. 0.85-1.70	S. 70	2.1	5.0	95	40.0	2.0
A15	35	15	10	S. 0.85-1.70	S. 70	2.0	5.0	53	50.0	5.0
A16	35	10	5	S. 0.85-1.70	S. 70	0.4	5.0	210	154.0	30.8
A17	20	25	20	S. 0.50-1.00	S. 70	1.7	3.0	240	86.0	4.3
A18	20	25	10	S. 0.50-1.00	S. 70	0.4	2.9	240	133.0	13.3
A19	20	10	5	S. 0.50-1.00	S. 70	<0.1	1.0	240	146.0	29.2
A20	20	20	20	S. 0.85-1.70	S. 70	2.8	5.0	240	146.0	7.3
A21	20	20	10	S. 0.85-1.70	S. 70	0.8	1.3	240	135.0	13.5
A22	20	20	5	S. 0.85-1.70	S. 70	0.2	1.0	240	220.0	44.0
A23	10	10	20	S. 0.50-1.00	S. 70	0.9	0.9	240	116.0	5.8
A24	10	10	10	S. 0.50-1.00	S. 70	0.3	0.9	240	223.5	22.5
A25	10	15	5	S. 0.50-1.00	S. 70	<0.1	0.1	240	241.5	48.3
A26	10	15	5	S. 0.85-1.70	S. 70	0.1	0.4	240	333.5	66.7
A27	5	15	5	S. 0.50-1.00	S. 70	0.1	-	-	-	-
A28	5	15	5	S. 0.85-1.70	S. 70	0.2	-	-	-	-

Run No.	Initial Turbidity (NTU)	*M.oleifera* mg/L	Filtration Rate (m/h)	Media Specification (mm)	Bed Depth (cm)	Minimum Turbidity (NTU)	Turbidity on Termination (NTU)	Headloss on Termination (cm)	Total Output (m^3/m^2)[1]	Length of Run (Hours)
B1	75	35	10	S. 0.50-1.00	S. 120	0.8	5.0	240	64.0	6.4
B2	75	25	5	S. 0.50-1.00	S. 120	0.2	3.7	240	77.5	15.5
B3	75	35	10	S. 0.85-1.70	S. 120	3.0	5.0	70	28.0	2.8
B4	75	25	5	S. 0.85-1.70	S. 120	0.7	5.0	80	72.1	14.4
B5	50	25	10	S. 0.50-1.00	S. 120	0.2	0.5	240	65.0	6.5
B6	50	25	5	S. 0.50-1.00	S. 120	<0.1	0.2	240	90.0	18.0
B7	50	25	10	S. 0.85-1.70	S. 120	1.6	5.0	90	65.0	6.5
B8	50	25	5	S. 0.85-1.70	S. 120	0.2	5.0	100	85.0	17.0
B9	35	25	10	S. 0.50-1.00	S. 120	0.2	4.0	240	140.0	14.0
B10	35	25	10	S. 0.85-1.70	S. 120	1.1	5.0	90	85.0	8.5
B11	20	15	10	S. 0.50-1.00	S. 120	<0.1	0.4	240	130.0	13.0
B12	20	15	10	S. 0.85-1.70	S. 120	0.4	5.0	140	190.0	19.0
B13	5	15	5	S. 0.50-1.00	S. 120	<0.1	-	-	-	-
B14	5	15	5	S. 0.85-1.70	S. 120	0.2	-	-	-	-
C1	75	10	25	A.[3] 1.2-2.5 & S. 0.50-1.00	A. 50 & S. 70	0.8	5.0	100	45.0	4.5
C2	50	10	25	A. 1.2-2.5 & S. 0.50-1.00	A. 50 & S. 70	0.6	5.0	140	90.0	9.0
C3	5	5	15	A. 1.2-2.5 & S. 0.50-1.00	A. 50 & S. 70	<0.1	-	-	-	-

Table 4.1 Summary of the main results obtained from the pilot rig to investigate the hydraulic variables.

Sand size 0.50-1.00 mm is classified as BS Mesh Size 16/30, and has a UC of 1.4.

Sand size 0.85-1.70 mm is classified as BS Mesh Size 10/18, and has a UC of 1.4.

When residual turbidity leveled off above 5 NTU, the run was terminated.

A1-A28 = filter media of 70 cm sand, B1-B14 = filter media of 120 cm, C1-C3 = filter media of 50 cm anthracite & 70 cm sand.

[1] Total output for an equivalent filter of surface area $1m^2$, [2] S = sand , [3] A = anthracite

4.1.1 Dose optimisation

The optimum dose for each raw water and hydraulic variable investigated was determined by carrying out a series of dose response runs. The results shown in Table 4.1 were obtained from runs conducted at optimum doses. Figures 4.1-4.4, show several runs which were operated until the working stage. The optimum dose was then found based on the lowest dose to produce the lowest residual turbidity. Two distinct dose responses can be observed in these graphs. Firstly, in all cases increasing the dose from the lowest one used improved the quality of the filtrate. Secondly, a further increase in the dose from the optimum caused a deterioration in the quality of the filtrate with significantly shorter filter runs. It is considered that the addition of insufficient coagulant would cause only partial destabilisation of the suspended particles, as shown by lower turbidity removal rates at low doses of coagulant in all the figures. As explained in Section 2.2.2, a specific minimum number of particles would need to have a 'patch' of positive charge on their surface through polyelectrolyte attachment to induce effective floc formation. Hence, an increase in the dose improved removal rates in all cases, by providing optimal coverage of the charged particles. When the dose was increased to greater than the optimum, an overdose effect was observed. This was caused by the reversal of the charge of the suspended particles, coupled with the adsorption of the excess 'free' protein on the surface of the filter media causing particle-particle and particle-media repulsion. Essentially, the effect of overdosing is a reduction in performance in terms of turbidity removal. A beneficial feature of *M.oleifera* seed coagulant is that increasing the dose above the optimum still effected good turbidity removal. In Figure 4.1, for example, increasing the dose from the optimum of 10 mg/l to 50 mg/L, a five-fold increase, still reduced the turbidity; although the corresponding increase in headloss development would have resulted in a lower total output before termination of the run. The same can also be seen in Figures 4.3 and 4.4, where the optimum dose was found to be 25 mg/L, although equal removal occurred with a dose of 35 mg/L. Increasing the dose further to 100 mg/L still reduced the turbidity, although to a lesser extent than the lower doses. Optimal dosage is expected to occur when there is 50% coverage of the particle with the 'patch' forming polymer, as discussed in Section 2.2.2. At this point a maximum number of collisions result in successful aggregations.

The effective dose range with *M.oleifera* seed is much greater than that found using other coagulants. For example, the range of doses of ferric aluminum sulphate (Ferral) used in the

field study (described in Chapter 6) was between 15-22.5 mg/L; a considerably smaller range than with *M.oleifera*, as found in these results and by other workers (Folkard *et al.*, 1992; McConnachie, 1993). The wide dose range may be due to the use of the whole seed to make-up the dosing suspension. The dose of the active coagulant which is used is be less than 30% of the whole seed suspension dose. Therefore what appears to be a wide dose range is actually a small variation in the active coagulant. The wide dose response is an advantage of using *M.oleifera* seed, since it allows for a more robust treatment regime; an important consideration for WTW in developing countries where there are often shortages in skilled labour and reliable dosing equipment.

4.2 The effect of media size on filtration efficiency.

The effect of this parameter was investigated because of its importance on the treatment process; the size of the media dictates the size of the interstitial pores. The pore size influences the storage capacity for the removed floc, the interstitial velocities, the mechanism of particle transport and attachment to the filter media and the extent of in-pore flocculation. The storage capacity is an important factor when using CFF, since there is no solid-liquid separation stage prior to filtration. The rate of filter pore clogging affects the rate of headloss development across the bed and turbidity breakthrough. Turbidity breakthrough can be caused by either the direct passage of influent particles through the filter, or by the break-off of previously removed particles. Thus, removal rates and distribution patterns throughout the bed are effected. The pore size influences the rate of in-pore flocculation, since for the same number of particles in the water, the smaller the pores the higher the collision efficiency. In addition, for the same bed depth, the smaller the media the greater the available collector surface area. Thus, the extent of in-pore flocculation and potential particulate removal is greater with smaller media (Graham, 1988). It was anticipated that larger media would effect a lower rate of headloss development (due to the decrease in resistance to flow) and a lower turbidity removal rate.

A comparison of the two media sizes was obtained by operating the two columns of the pilot rig in parallel, containing different sand sizes, while maintaining all the other hydraulic variables constant. The results obtained are summarised in Table 4.1. Runs A1-A28 were comparative runs of the two media sizes using a 70 cm bed of sand. Runs B1-B14 were further comparative test of the two media sizes using a 120 cm bed of sand. A selection of these runs are presented graphically in Figures 4.5-4.67 at the end of this chapter.

The principal effects of media size on the filter runs were as follows:

- Larger media resulted in a reduction in turbidity removal compared to the smaller media. The effect of this was that where turbidity breakthrough was the terminating factor, smaller media resulted in the production of a greater volume of filtrate before termination of the run. This was the case for the highest turbidity raw waters considered (35, 50 and 75 NTU), in the 70 cm bed, as illustrated in Figures 4.29, 4.39 and 4.45.

- For lower turbidity waters (10 and 20 NTU), the reduction in headloss development rates with the larger media, extended the operation of the run. The delay in headloss accumulation rate gave a larger volume of filtrate, at the expense of a small reduction in filtrate quality (Figures 4.11 and 4.21).

- The pattern of particulate deposition in the bed was also affected by the media size. This is reflected in the data obtained on turbidity removal and headloss development measurements taken at various depths in the filter bed. A large proportion of the particulates was removed above the highest port with both sand sizes and bed depths. This is illustrated in Figures 4.17 and 4.18; the largest gradient of the headloss curves occurred above 4 cm. The gradient was steeper with the smaller media indicating greater removal. In the 120 cm bed (Figures 4.7, 4.23, 4.41, 4.47 and 4.51) turbidity removal in the surface layer (top 16 cm) was greater with the smaller media than the large. This phenomenon was also seen in the top 4 cm of the 70 cm bed (Figures 4.9, 4.13, 4.31, 4.33 and 4.49). This is further illustrated by plotting the hydraulic gradient as a function of bed depth, as seen in Figures 4.15 and 4.16. The main feature of these graphs is the very rapid increase in the hydraulic gradient above the top port, which was considerably greater with the smaller media. A further trend that can be seen in all the figures noted, is that greater removal occurred deeper into the bed with the larger than the small media. The reduced surface removal and deeper penetration of the particulate matter was caused by the more rapid advancement of the clogging front with the larger media.

4.2.1 Discussion

The difference in the hydraulic conditions with the two media sizes is considered to have caused the reduction in turbidity removal and headloss development rates. As a filter run proceeds, increased deposition in that filter bed causes constriction of the interstitial pores and an increase in fluid drag and interstitial velocities. If the filtration rate is kept constant the increase in the shear forces will cause particle detachment. Furthermore, when the detachment rate exceeds the attachment rate, the result is deeper penetration of the particulate matter, a lower headloss development rate and early breakthrough of turbidity. It is evident from all the filter runs that lower attachment and/or higher detachment of the particles occurred with the larger media. As a consequence of these factors the working layer advanced through the bed at

a higher rate, leading to a more even distribution of the deposited particles, and more effective utilisation of the voids in the filter bed. Thus preventing excessive headloss accumulation in the surface of the bed.

The higher capture and removal rate in the bed of smaller media can be explained by considering the Isolated Single Sphere Model (Yao *et al.*, 1971; Rajagopolan & Tien, 1976; O'Melia & Ali, 1978). Although this is not the only model used to estimate the capture efficiency of a collector in the filter bed, it is the simplest. Moreover, it has been shown to qualitatively agree with more complex models (Tien *et al.*, 1979; Montgomery, 1985). The model is used to estimate the efficiency of collection of a single collector (η), where:

$$\eta = \frac{\text{The number of successful collisions for all particulates in the cross-sectional area perpendicular to the collector}}{\text{The total number of possible collisions in the cross-sectional area perpendicular to the collector}} \qquad \textit{Eq. 4.1}$$

(The total number of possible collisions is the product of the cross sectional area, flow rate and concentration of solids in the water).

For beds of equal depth, smaller media provide a greater collector surface area and therefore greater opportunity for particle collisions and capture, than larger media.

Filter beds of smaller media also have smaller interstitial pores and therefore higher interstitial velocity, than beds of larger media under the same approach velocity (filtration rate through the filter). A consequence of higher interstitial velocities is an increase in headloss development rates, as stated by Darcy's Law that established the basic hydraulics of filtration:

$$\frac{h}{L} = \frac{v}{k} \qquad \textit{Eq. 4.2}$$

where h is the headloss development across a filter bed with a depth L, and a face velocity of v, and k is the coefficient of permeability in laminar flow (Ives, 1980; Tebbutt, 1998).

The rate of headloss development and turbidity removal is not only affected by media size, but also by raw water turbidity. The runs conducted on the higher turbidity waters (35-75 NTU) were terminated due to turbidity breakthrough, whereas those conducted on lower turbidity waters (10-20 NTU) were terminated due to excessive headloss accumulation. It has been found by previous workers that higher turbidity waters effect rapid particle aggregation due to the high number of particle-particle and particle-collector collisions. This can also be seen from Equation 4.1, which states that the number of possible collisions is related to the concentration of particles in the water. With low turbidity raw waters, the collision rate is lower, allowing some particles to pass out of the bed without having the opportunity of forming floc, or attaching to a collector surface (Graham, 1988; Clark *et al.*, 1992). Thus it is concluded that larger floc tend to form with higher turbidity waters, compared to low turbidity waters, where low collision rates result in small floc attaching to the collector surfaces. This is important because for the same volume of material, smaller floc have a greater surface area than larger floc. Higher headloss development rates ensue due to the effect of fluid drag. Other workers have found similar results (Edzwald, 1986; Darby & Lawler, 1990; Tobiason *et al.* (1993).

Concentration of removed particles in the surface layer occurred to some extent with both media sizes as can be seen by plotting the hydraulic gradient as a function of depth. This is evident in Figures 4.15 and 4.16 with a raw water of 10 NTU, Figure 4.43 and 4.44 with a raw water of 50 NTU and Figure 4.53 and 4.54 with a raw water of 75 NTU. There were progressively smaller increases in the hydraulic gradient in each subsequent layer. This effect is also seen by plotting headloss development as a function of time (Figures 4.17 and 4.18). The rate of removal was very similar between the two media sizes until saturation level was reached in the bed of smaller media, and a large increase in headloss in the top section of the bed occurred. The increase in headloss rates in the subsequent sections of the bed was smaller in all cases.

The significance of this is that regardless of the media size, raw water turbidity or filtration rate, the majority of headloss development occurred in the surface layers, indicating the importance of the surface filtration phenomenon. Thus to achieve true deep bed filtration (where maximum bed use is attained), an understanding of why it occurs is required. The surface chemistry of the particles which enter the filter bed would not have been uniform. Accordingly, the particles which were fully destabilised, and had a higher collision frequency, were captured and removed preferentially in the upper layers of the bed. The particles which

then entered the lower sections of the bed were more stable, with a lower collision frequency and less likelihood of capture and removal. Particle capture and ripening are autocatalytic: the process is self-perpetuating, such that captured particles commence acting as collectors them selves and further enhance removal (Clark *et al.*, 1992; Kau & Lawler, 1995). As a consequence of this preferential removal of particles in the surface layers continues. Thus, in order to reduce surface filtration, destabilisation of some of the particles should occur when in contact with the filter media, allowing deeper penetration of the particles before capture and removal.

One major difference between the results obtained here and those found by other workers researching the effect of hydraulic variables such as media size, was the variation in removal efficiencies and headloss development rates between the two media sizes. Adin & Rebhun (1974), using media sizes of 0.6 and 1.2 mm (comparable with the media sizes used in this work), found the smaller media impractical for use with a high molecular weight polyelectrolyte, due to the rapid rise in headloss. Whereas in this study, the headloss development rate was not always prohibitive. It was anticipated that the extent of surface removal with the smaller media may have been a limiting factor, but this was not the case. The percentage of removal in the surface layers of the beds of both media sizes was very similar. This is illustrated in Figures 4.17 and 4.18, where headloss development is plotted as a function of time at various depths. These graphs indicate that headloss development was very similar with the two media sizes up to 60 hours into the run. At the termination of the majority of the runs, the headloss which had accumulated in the surface layer, as a percentage of headloss across the whole bed was about 50-60% for both media sizes. The turbidity removed at the surface, as a percentage of the turbidity removed over the entire depth at the point of maximum removal in this layer, was about 40-50% for both media sizes.

The significance of this is that using larger media in conjunction with *M.oleifera* seed had limited benefits compared with other coagulants. It is known that the *M.oleifera* coagulant forms bonds by the 'patch' mechanism of charge neutralisation (Tauscher, 1994; Ndabigengesere *et al.*, 1995; Gassenschmidt *et al.*, 1995), as discussed in Section 2.7.3. A patch of positive charge on the surface of the particle, forms an attachment point with the collector surface. Alternately, the negative potential around the particle is reduced allowing the particle to approach the collector whence other forces come into play. This type of attachment is relatively weak compared to that initiated by 'bridge' forming polymers, where multiple links

can form with individual particles (Gregory, 1988). Also, 'bridge' forming polymers tend to protrude out into the flow and capture passing particulates. 'Patch' forming polymers are unable to do this and therefore capture efficiency is reduced. Consequently deeper penetration of the destabilised particles occurs prior to capture, with the lower molecular weight polymers, regardless of media size. Surface clogging is reduced and better utilisation of the filter bed can be achieved.

A further variation in the results obtained in this study to those of other researchers, is the comparative rates of filter bed ripening of the two media sizes. Clark *et al.* (1992) found that bed ripening was faster in the filter bed of smaller media than the larger media. This was considered to be due to the relationship between the ripening rate and the specific deposit or fractional voidage (the fraction of pore space taken up by the deposit (Kau & Lawler, 1995)) in each pore. Since ripening is the process of reducing the pore size, having a bed with smaller pores leads to a more rapid rate of ripening. The results obtained in this research, however, indicated that the ripening rates with the two media sizes were of similar duration. This is illustrated in Figures 4.5, where the ripening stage continued for about 5 hours with both media size and bed depths. This effect can also be seen in Figure 4.11, 4.21, 4.29, 4.39, 4.45. As described above, this may be related to the method of bond formation induced by *M.oleifera* seed, allowing deep penetration of the destabilised particles with both media sizes. Whereas, in the research by Clark *et al.* (1992) a greater amount of particle capture and attachment occurred in the surface layer. This was considered to be a function of the coagulant used: Cat Floc-T - a 'bridge' forming polymer, which brought about strong attachment bonds between the particulates and the collector surfaces. Therefore the rate of pore constriction would be significantly enhanced leading to more rapid filter bed ripening

The activation of sequential layers in the filter bed, as seen in Figure 4.13 describing runs A25 and A26, indicates the progress of the working layers. The rate of removal in the top 4 cm of the bed of smaller media began to decrease after 15 hours, indicating that the clogging front had progressed down to the next layer. The section of bed between 4 and 20 cm then became the active layer until 25 hours in to the run. Finally the layer between 20 and 36 cm became active and continued until termination of the run. In contrast, with the larger media the front progressed from the top 4 cm of the bed after only 10 hours. It advanced to the next section after about 20 hours, then into the section between 36 and 52 cm after 30 hours, and finally the bottom layer of the bed after 35 hours.

The progress of the front through the bed was not always visible in such depth profiles. For example runs B13 and B14 illustrated in Figure 4.7, indicates that the active layer for both filter beds was mainly above 16 cm throughout the 13 hours of the run. This was caused by the low load entering the filter bed and therefore saturation of the surface layer only occurred after a long period of operation. The progress of the working front through the bed is therefore very slow. The converse can be seen in runs B1 and B3, Figure 4.51, where the particulate load entering the filter bed was relatively high. With a raw water turbidity of 75 NTU and a filtration rate of 10 m/h, minimal turbidity occurred after 1.5 hours with both media sizes. Breakthrough was almost simultaneous in all the ports of both beds. This is due to the dynamic nature of particle capture and deposition, resulting in removal occurring in all sections of the bed to a greater or lesser extent throughout the run (with the majority of activity taking place in the working layer). But because the load was high, clogging of the filter media and subsequent advancement of the working layer was rapid. This coupled with the large time interval between filtration readings at each depth gave the appearance of breakthrough occurring simultaneously in all layers.

The practical implications of the variation in performance of the two filter media in previous studies are that using larger media results in better utilisation of the filter bed and consequently a lower headloss development rate. This potentially delays filter run termination, where headloss development is rapid, and thereby reduces the backwash water requirement. Furthermore, there is a reduction in energy expenditure, since rapid clogging of the filter bed requires a greater pumping rate to ensure a constant flow rate. In gravity fed filters, clogging results in lower flow rates and a reduction in the total output. Surface filtration, which can occur to a greater extent with smaller media can lead to the development of negative heads in the filter and the formation of air pockets. This situation is undesirable since short circuiting of the flow can result, allowing the passage of the influent water through the filter without treatment. Also compact layers of sand and mud form on the surface of the bed which can lead to the formation of mud balls.

In this study, it was found that the phenomenon of surface filtration was low. With raw water turbidities of 5 NTU, e.g. Figure 4.7 and 4.8, it can be seen that headloss increase was so low with both media sizes, that the reduction in the rate of increase with the larger media was insignificant. The major difference in performance of the two media sizes was the improved filtrate turbidity with the smaller media bed. But with the higher turbidity water of 75 NTU,

Figures 4.47 and 4.48, the more rapid rise in headloss with the smaller media was a more significant factor. Although the reduction in removal rates with the larger media would still be a significant factor in the choice of media size, the advantage of the larger media is more apparent at these higher loading rates.

The physical characteristic of media size cannot be considered in isolation, since larger media is generally associated with deeper beds. Consequently the next section not only considers the effect of increasing bed depths, but also compares deep beds of large media with shallow beds of small media.

4.3 The effect of bed depth on filtration efficiency

The effect of increasing the depth of the filter bed was investigated from two perspectives. Firstly, the range of raw water turbidities which could be effectively treated using CFF in conjunction with *M.oleifera*. Secondly, to study the effect of collector surface area in the filter bed. This was achieved by comparing beds of different depths and media sizes, such that the available collector surface area was maintained. Rather than calculate the available collector surface area in each bed with the different size media, the Isolated Single Sphere Model (described in Equation 4.1), was used. According to this model equal removal would occur when:

$$\eta_1\left(\frac{L_1}{d_{m1}}\right) = \eta_2\left(\frac{L_2}{d_{m2}}\right) \qquad \textit{Eq. 4.3}$$

Where:

η is the single collector contact efficiency.

(1) and *(2)* are the two different filter conditions.

L is the filter media depth.

d_m is the diameter of the media grain.

If sedimentation is the dominant mechanism of removal in the filter bed, then η of Equation 4.3 can be ignored, because it is the ratio of Stokes' settling velocity to the filtration velocity (as described in Equation 2.4, Section 2.3.1.1), and therefore media size has no effect on contact efficiency and Equation 4.3 can be simplified to:

$$\eta(\text{due to sedimentation})\left(\frac{L_1}{d_{m1}}\right) = \eta(\text{due to sedimentation})\left(\frac{L_2}{d_{m2}}\right)$$

$$\frac{g(\rho_1 - \rho){d_p}^2}{18\mu V}\left(\frac{L_1}{d_{m1}}\right) = \frac{g(\rho_1 - \rho){d_p}^2}{18\mu V}\left(\frac{L_2}{d_{m2}}\right)$$

$$\left(\frac{L_1}{d_{m1}}\right) = \left(\frac{L_2}{d_{m2}}\right) \qquad \textit{Eq. 4.4}$$

However, this simplification would not apply if the particles/floc under consideration were < 2.5 μm, as Brownian motion and not sedimentation would be the dominant mechanism, as described in Section 2.3.2.5. It can be seen from Equation 2.5 that media size does have an effect on η, when Brownian motion dominates. Thus Equation 4.3 for the purposes of this study with average media sizes of 0.75 mm and 1.25 mm and bed depths of 70 cm and 120 cm respectively, would be:

$$\eta \text{ (due to Brownian Motion)} \left(\frac{L_1}{d_{m1}}\right) = \eta \text{(due to Brownian Motion)} \left(\frac{L_2}{d_{m2}}\right)$$

$$0.9\left(\frac{KT}{\mu d_p d_m V}\right)^{2/3}\left(\frac{L_1}{d_{m1}}\right) = 0.9\left(\frac{KT}{\mu d_p d_m V}\right)^{2/3}\left(\frac{L_2}{d_{m2}}\right)$$

$$\left(\frac{1}{0.75}\right)^{2/3}\left(\frac{L_1}{d_{m1}}\right) = \left(\frac{1}{1.25}\right)^{2/3}\left(\frac{L_2}{d_{m2}}\right)$$

$$\left(\frac{L_1}{d_{m1}}\right) = 0.7\left(\frac{L_2}{d_{m2}}\right) \qquad \textit{Eq. 4.5}$$

Calculation of the required bed depths can be made based on Equations 4.4 and 4.5, in order to achieve equal collector surface area with the two media sizes. Considering Equation 4.4, a 70 cm bed of the smaller media would provide the equivalent surface area as a 120 cm bed of the larger media size. But based on Equation 4.5, a 176 cm bed of the larger media would be required. A bed depth of 120 cm was used in this study, since a deeper bed would not have been practical in the pilot columns. Furthermore, a survey conducted by Kawamura (1975) found that the L/d_m ratio generally used in treatment plants was about 950. Based on this ratio,

a bed depth of 121 cm would be recommended for the larger media. Thus beds of 70 and 120 cm were compared in terms of turbidity removal and headloss development. There are three main observations from Figures 4.5-4.54, with regards to bed depth:

- A deeper filter bed, regardless of media size, filtration rate and raw water quality, resulted in higher turbidity removal efficiencies (Figures 4.5, 4.21, 4.29, 4.39 and 4.45) and higher clean bed headloss (Figures 4.30, 4.40 and 4.46).

- Removal rates in the shallow bed of small media and the deeper bed of large media were almost identical (with the same filtration rate and raw water turbidity), as illustrated in Figures 4.5, 4.21, 4.29, 4.39 and 4.45.

- The headloss development rates (both actual and additional), were greater in the shallow bed of small media than in the deeper bed of large media, despite the similarity in removal rates. This can be seen with a filtration of 10 m/h in Figures 4.22, 4.30, 4.40 and 4.46 and 5 m/h in Figures 4.6, 4.40 and 4.46.

4.3.1 Discussion

The higher removal rates with the deeper beds was expected, since deeper beds resulted in higher retention times in the filter bed leading to greater in-pore flocculation. Also the increase in collector surface area, allowed for greater attachment of the particulate matter and therefore a reduction in filtrate turbidity.

The increase in surface area in the deeper beds offered more resistance to flow, and resulted therefore in higher clean bed headlosses. The additional headloss was lower with deeper beds as a consequence of the wider distribution of the removed particulate matter throughout the bed. The higher headloss development in the shallow bed of small media was due to the greater specific deposit. This then leads to greater pore constriction and resistance to flow, resulting in higher headloss development rates.

Removal rates achieved with the two media sizes at bed depths of 70 and 120 cm were compared based on Equation 4.4, and were found to be very similar. This has also been found

by other workers and considered to be due to equal collector surface (D. Moran *et al.*, 1993 (Kau & Lawler, 1995).

There were exceptions to this in the runs conducted on raw waters of 75 NTU, as illustrated in Figure 4.45. In comparing runs A2 (70 cm of small media) and B4 (120 cm bed of large media), it can be seen that the shallow bed of smaller media had a greater removal rate than the deeper bed of larger media. Also runs A1 (70 cm of small media) and B3 (120 cm of large media) had different removal patterns, with a lower turbidity during the working stage and early breakthrough with the smaller media (run A1). Thus with the higher loading rate (turbidity of 75 NTU and filtration rate of 10 m/h), the deeper bed of the larger media did not remove turbidity to the same extent, as the shallow bed of small media. This may be due to rapid particulate penetration occurring with the larger media leading to rapid turbidity breakthrough.

It was found to be advantageous to use relatively deep filter beds when using *M.oleifera* seed coagulant, due to the relatively low attachment rates and high detachment rates. Furthermore, since removal of floc in the filter bed requires in-pore flocculation to occur, increasing the number of particle collisions would enhance the rate of removal. Therefore using small media increases the rate of flocculation. But, the detrimental effect of using smaller media, i.e. the increased surface clogging, is limited since relatively deep penetration tends to occur using *M.oleifera* seed.

The highest quantity and quality of filtrate was produced by the bed of smaller media at a depth of 120 cm, as illustrated in Figures 4.21, 4.29, 4.39 and 4.45. The subsequent rise in headloss in all these runs was not a limiting factor, since terminal headloss and turbidity breakthrough occurred almost simultaneously. When the bed of larger media was used, even though the rate of headloss accumulation was reduced (as seen in Figures 4.22, 4.30, 4.40 and 4.46), the early breakthrough of turbidity resulted in lower total output. Thus, optimal conditions when using *M.oleifera* seed as the coagulant, would be achieved by designing the operational parameters to delay turbidity breakthrough, at the expense of a high headloss development rate.

4.4 The effect of filtration rate on filtration efficiency

The rate of filtration through a filter bed is a very important independent variable in the design and operation of deep bed filters. For economic reasons there is a requirement for high flow through the filters. But excessively high rates can lead to inadequate treatment, resulting in the presence of pathogenic organisms in the filtrate. Hence in the US, water treatment operators need to obtain special permission from regulatory agencies to operate filters at rates above 25 m/h (Kawamura, 1991a), although there are no such restrictions in the UK. There are diminishing advantages of using higher rates of flow, due to the frequency of backwashing and the increase in coagulant demand. The main benefit is the reduction in capital costs with higher loading rates, due to the decrease in filter surface area required to produce a certain total output.

This operating parameter was investigated by obtaining data from filter runs with identical media size, bed depth and initial turbidity, while varying the filtration rates. The results are summarised in Table 4.1 (Runs A1-28 and B1-14), and in the figures at the end of the chapter. The three rates considered were 5, 10 and 20 m/h, as these are typical of rates currently used in the water industry (Kawamura, 1991a; D. Moran *et al.*, 1993; Kau & Lawler, 1995).

The principal effects of increasing the filtration velocity on turbidity removal and accumulation of headloss in the bed were:

- Turbidity removal was consistently lower with the higher filtration rates, as is illustrated clearly with the 10 NTU raw water in Figure 4.19. Figures 4.27, 4.39 and 4.45 also illustrate this with the 20, 50 and 75 NTU waters respectively.

- The corresponding headlosses to the aforementioned figures indicate that clean bed headlosses were consistently greater with the higher filtration rates; additional headlosses were equal to, or less than that with lower filtration rates. This is illustrated in Figures 4.20, 4.28, 4.40 and 4.46 and their insets.

- Surface removal dominated at the lower filtration rates to a greater extent than at the higher rates. This can be seen with the 35 NTU raw water in Figure 4.37 and the 75 NTU raw water in Figure 4.57.

- The distribution of headloss throughout the bed was more homogenous at the higher filtration rates. This is illustrated by the greater spread of headloss gradients at the higher filtration rates shown in Figures 4.56 and 4.58. Also ripening of the bed and turbidity breakthrough occurred earlier at each depth at the higher filtration rates, as indicated by the graphs of turbidity removal as a function of depth (Figures 4.35, 4.37, 4.55 and 4.57).

- The total output of treated water was greater at the lower filtration rates, as can be seen in Figures 4.19, 4.27, 4.39 and 4.45.

4.4.1 Discussion

Increased turbidity removal at lower filtration rates was expected based on filtration theory, and is a consequence of longer retention times and more opportunities for particle capture in the filter bed. Additionally, the mechanisms of particle transport to the collector's surface are affected by filtration rate, as can be seen from Equations 2.4 and 2.5. describing the two principal mechanisms of transport: sedimentation and Brownian diffusion. Moreover, particle detachment increases at higher velocities, due to greater fluid drag near the collector surface. This would induce deeper penetration of particles and floc into the bed and, if the bed is not of sufficient depth, early breakthrough would occur (Rajagopolan & Tien, 1976).

Clean bed headloss was greater with higher filtration rates, as headloss is inversely proportional to the velocity of flow, as stated by Darcy's Law in Equation 4.2 (Section 4.2.1). But plotting additional headloss as the ordinate and the total output as the abscissa, indicates that the rate of headloss increase is lower with higher rates after a period into the run, as seen in Figures 4.56 and 4.58. This may be a function of the reduced removal experienced under such conditions, and not the effect of filtration rate *per se*. But it may also be caused by an increase in detachment of particles and their deeper penetration into the filter bed preventing surface filtration. This can be seen from the data of turbidity removal at various depths. Figures 4.37, 4.55 and 4.57 in each case indicate that turbidity removal at the top port was greater at

the lower filtration rate. The percentage of accumulated headloss in the top section was calculated for both filtration rates at the earliest termination point of both runs. It was found that the increase was greater at the lower filtration rate. With a raw water of 75 NTU and the smaller media (Figure 4.56, runs B1 and B2), the top 16 cm accumulated 68% and 42% of the total headloss at 5 and 10 m/h respectively. With the larger media (runs B3 and B4 shown in Figure 4.58), 48% and 34% of the headloss accumulated in the top 16 cm at 5 and 10 m/h respectively. Furthermore, the headloss development data indicated that there was a more homogenous distribution of the accumulated headloss throughout the bed at the higher filtration rate. The significance of these results is that a rapid increase in headloss in the surface layers at the lower filtration rate is responsible for the rise in headloss. Reducing this effect would thus increase the total output, where rapid headloss accumulation was the cause of termination.

To summarise, at lower filtration rates, particles entering the bed have a greater opportunity of attachment to the collector surface due to the increase in retention time in the filter bed. Contact efficiencies due to sedimentation and Brownian motion of the particles, leading to their capture and removal are also higher at lower filtration rates, hence the enhanced removal rate at lower filtration rates seen in Figures 4.19, 4.27, 4.39 and 4.45. Higher filtration rates also lead to an increase in the inter-pore shear forces and detachment of previously removed particles. The particles thus re-enter the flow and are pushed deeper into the bed, where reattachment or breakthrough may occur. This was evident by the earlier breakthrough of turbidity at the higher filtration rate. These results are in agreement with other research conducted on the effect of filtration velocities (Darby *et al.*, 1991; Adin & Rebhun, 1974).

As a consequence of the reduced removal at higher filtration rates, deeper beds are generally used (Kau & Lawler, 1995). Therefore, a more significant study into the effect of flow velocity would be to compare higher filtration rates in deeper beds with low filtration rates in shallower bed. Based on the Single Sphere Model, in the same manner as Equation 4.3, equal removal would occur with different filtration rates when:

$$\eta_1\left(\frac{L_1}{V_1}\right) = \eta_2\left(\frac{L_2}{V_2}\right) \qquad \textit{Eq. 4.6}$$

To calculate η, the size of the influent particles must be considered. If the particles under consideration were > 2.5 μm, then sedimentation would be the dominant mechanism of particle removal. Based on Equation 2.4, equal removal would occur when:

$$\eta \text{ (due to sedimentation)} \left(\frac{L_1}{V_1}\right) = \eta \text{(due to sedimentation)} \left(\frac{L_2}{V_2}\right)$$

$$\left(\frac{g(\rho_1 - \rho)d_p{}^2}{18\mu V_1}\right)\left(\frac{L_1}{V_1}\right) = \eta\left(\frac{g(\rho_1 - \rho)d_p{}^2}{18\mu V_2}\right)\left(\frac{L_2}{V_2}\right)$$

$$\left(\frac{1}{10}\right)\left(\frac{L_1}{V_1^2}\right) = \left(\frac{1}{5}\right)\left(\frac{L_2}{V_2^2}\right)$$

$$\left(\frac{L_1}{V_1}\right) = 2\left(\frac{L_2}{V_2}\right) \qquad \textit{Eq. 4.7}$$

If the particles were < 2.5 μm, then Brownian Motion would dominate as the mechanism of capture and removal. Hence the influence of filtration velocity on these submicron particles can be calculated from Equation 2.5. Removal rates would thus be equal with the two filtration rates when:

$$\eta \text{ (due to Brownian Motion)} \left(\frac{L_1}{V_1}\right) = \eta \text{(due to Brownian Motion)} \left(\frac{L_2}{V_2}\right)$$

$$0.9\left(\frac{KT}{\mu d_p d_m V_1}\right)^{2/3}\left(\frac{L_1}{V_1}\right) = 0.9\left(\frac{KT}{\mu d_p d_m V_2}\right)^{2/3}\left(\frac{L_2}{V_2}\right)$$

$$\left(\frac{1}{10}\right)^{2/3}\left(\frac{L_1}{V_1}\right) = \left(\frac{1}{5}\right)^{2/3}\left(\frac{L_2}{V_2}\right)$$

$$\left(\frac{L_1}{V_1}\right) = 1.5\left(\frac{L_2}{V_2}\right) \qquad \textit{Eq. 4.8}$$

According to Equations 4.7 and 4.8, equal removal would be achieved in a 120 cm bed operated at 10 m/h, and in beds of 30 and 40 cm respectively at a filtration rate of 5 m/h. But Figures 4.59-61 indicate that a depth of between 52 and 68 cm would in fact be required for equal removal to occur. Thus it would seem that lower filtration rates were less efficient than was predicted by the Single Sphere Model. If however, the calculation of bed depth requirement is based purely on equal retention times in the filter bed, as used by D. Moran (1993) and Kau & Lawler (1995), then η can be ignored and Equation 4.6 becomes:

$$\left(\frac{L_1}{V_1}\right) = \left(\frac{L_2}{V_2}\right) \qquad \textit{Eq. 4.9}$$

Thus the required bed depth at a filtration rate of 5 m/h would be 60 cm, which is in line with the results of this study. In Figure 4.59 it can be seen that similar removal was obtained in the 120 cm bed operated at a filtration rate of 10 m/h as at a depth of 68 cm operated at a filtration rate of 5 m/h. Similarly Figures 4.60 and 4.61 indicate that removal at 10 m/h in the 120 cm bed was close to that at 5 m/h at depths of between 36-68 cm and 52-68 cm respectively. This would suggest that either the contact efficiency did not influence the required bed depth, or that other factors were involved which have not been accounted for. D. Moran *et al.* (1993) and Kau & Lawler (1995) assumed the former situation in their studies, although they offer no explanation for this.

It was considered possible that η did have an effect, but that the reduction in filtration rate also affected the mixing intensity, to the detriment of the coagulation process, by reducing the particle collision frequency. It has however, been shown by previous researchers that the coagulation stage is not critical when using *M.oleifera* seed in conventional treatment. The mixing intensity does not need to be very high, as restabilisation by readsorption is unlikely to occur, as is the case with 'bridge' forming polymers. Flash mixing is not required as is the case with alum. In fact Ndabigengesere & Narasiah (1998) concluded from their study that a period

of rapid mixing was unnecessary prior to flocculation and sedimentation in jar testing. The process of flocculation, however, is considered to be more important with *M.oleifera*. It was noted during jar testing that floc formation with *M.oleifera* was relatively slow compared to other coagulants. Floc would was only visible after 15-20 minutes of slow mixing. The importance of the flocculation stage was also found by McConnachie (1993): the intensity of the slow mix affected the removal rates and the dosage requirement during jar tests. It is therefore concluded that the velocity of flow would not have influenced the destabilisation stage, when using *M.oleifera*, but the rate of in-pore flocculation would have been affected. Generally in-pore flocculation is considered to be insignificant with relatively large polymers. More significant factors are the efficiency of particle-media attachment and the influence of previously removed particles (Graham, 1988). This was concluded because pre-coating the filter media improved removal rates, suggesting that polymer strands attached to the media were able to form bonds with in-coming particles. This is not considered to be the case with *M.oleifera* seed for a number of reasons. Firstly, Tanaka & Pirbazari (1986) found that small linear polymers adsorbed relatively poorly to filter media, possibly because they are unable to 'bridge' the gap between collectors and particles. Secondly, experimentation in the laboratory on pre-coating the filter media with *M.oleifera*, did not increase removal rates. It is therefore suggested that in-pore flocculation was significant with *M.oleifera* seed; the longer the particles remained in the filter pore the greater the opportunity for collision and in-pore flocculation. The culmination of this analysis is that the calculated bed requirement at 5 m/h was not correct, and a depth of between 52-68 cm would in fact be required.

Increasing the filtration rate thus had two effects. The detachment rate increased due the higher shear forces; and the attachment rate decreased due to the shorter retention times. The increased rate of detachment is a major drawback in beds of insufficient depth, since the re-introduction of previously removed particles is an important public health issue, especially where filtration is the last physical barrier for pathogens removal. Where the bed depth is sufficient higher filtration rates can be advantageous as better utilisation of the bed ensues.

The flow velocity also affected the total output prior to termination of the run. Figures 4.19, 4.27, 4.39 and 4.45, indicated that there was a reduction in output volume with the higher velocities of flow through the bed. Other workers (Clark *et al.*, 1992) results indicated the contrary. Higher filtration rates resulted in a higher total output, which was attributed to better utilisation of the filter bed at higher filtration rates. As described previously, higher rates

reduce surface filtration and cause deeper penetration of the particles; so a coagulant that formed strong bonds with the surface media would not be adversely effected by the higher rates. Whereas with *M.oleifera* seed, surface filtration is limited, therefore the necessity for deeper beds was found to be of more significance than increasing the filtration rate.

4.5 A comparison of dual and single media filter beds in terms of filtration efficiency

Sand is the most commonly used filter media, since it is widely available, relatively cheap and performs well as a filter. However, as previously noted, problems can arise when the surface layer of the bed of sand becomes clogged; the resulting pressure build-up in the bed leads to inefficient filter run length. This is a particular problem with high rate CFF, since there is no prior sedimentation stage for floc removal. One solution is to use reverse graded filters, such that the water passes through a layer of large media (with a large holding capacity) before passing through a layer of small media. Although this can be achieved by upflow filtration, the process is fraught with practical problems, such as fluidisation of the bed and bursts of turbidity breakthrough. Another method is to operate a multi-media bed in a down flow mode. Each layer consisting of media of different size and specific gravity (Bratby, 1980; Schulz & Okun, 1992; Kawamura, 1991a, Darby *et al.*, 1991). This results in the larger, less dense media remaining above the smaller more dense media subsequent to backwashing. In dual media filters, anthracite coal is generally used as the coarse surface layer and sand for the bottom layer. Dual media filters are commonly used in industrialised countries to allow the use of higher rates, without reducing the filter runs times (McCormick & King, 1982). There are also advantages for developing countries in using dual media filters. The treatment capacity can be increased by reducing the headloss development rate, at a relatively low cost as no structural alteration is required (Schulz & Okun, 1992). Furthermore, indigenous materials can be used as the coarse, upper layer. Such as crushed coconut shells (Kardile, 1972), fruit stones (Ranade & Agrawal, 1974) and bituminous coal (Paramasivam *et al.*, 1973).

There are disadvantages to using dual media beds, such as the accumulation of sludge at the media interface. Consequently, air scouring during backwashing may be required to breakup these large agglomerates, at additional cost. In order to regrade the filter media subsequent to an air scour, a higher flow of water is required during backwash. This involves the use of larger volumes of water, holding tanks and pipes. Furthermore, the use of higher backwash flow rates combined with less dense media in the upper layer, can result in the loss of media from the filter. There is always an acceptable loss of filter media with any material used; but this tends to be higher with anthracite due to the attachment of air bubbles to pieces of coal

and also the formation of coal-floc aggregates. These have low densities and are thus easily washed out of the filter bed. It is therefore essential that the free water zone - the distance between the top of the filter bed and the top of the filtering unit (referred to in Section 2.3.3) is sufficiently large to allow for bed expansion during backwashing. The brittle nature of anthracite and the constant wearing of the filter material, compared to sand, can result in the formation of fines which are washed out of the bed.

The selection of the correct media size, specific gravity and relative depths of each layer in dual media beds is essential to ensure adequate washing and regrading of the media layers, and prevent loss during backwashing (Kawamura, 1975). Previous researchers have found that the upper grains should be about twice the size of the bottom sand grains, and the top layer about twice the depth of the bottom layer to allow efficient particulate removal and backwashing (Camp, 1964; Kawamura, 1975; Culp, 1977). If the use of an air scour is not possible during backwashing, then the use of larger and heavier media in both layers is recommended. This allows for a large force of water to be applied during backwashing without excessive fluidisation occurring.

The effectiveness of single and dual media beds was compared by considering a monomedia bed of 120 cm sand with a bed of 50 cm of anthracite over a layer of 70 cm of sand. The other characteristics of flow rate, sand size and raw water turbidity were maintained constant for both columns. Effectiveness of treatment was measured in terms of turbidity of filtrate, headloss development rate and distribution of the particulate matter throughout the bed. The results of runs B1, B5 and B13 were compared with runs C1-C3, and the data obtained is summarised in Table 4.1. Figures 4.62-67 illustrate this comparison. The main observations of these results are as follows:

- Filtrate turbidity was marginally lower using the single media than using the dual media bed, for the three raw waters considered. This is illustrated by the filtrate turbidity in both beds in Figures 4.62, 4.64 and 4.66.

- Headloss development at the termination point in the single media bed was double that in the dual media bed. This is illustrated in Figures 4.63, 4.65 and 4.65 showing the headloss development for the runs referred to above.

- Turbidity breakthrough was the cause of termination in the dual media bed with the higher turbidity raw waters (50-75 NTU). Excessive accumulation of headloss was the terminating factor in the single media bed at these influent turbidities.

- Surface removal occurred to a greater extent with the single media than the dual media bed, as shown from both the turbidity removal and headloss development data.

4.5.1 Discussion

The single media bed achieved slightly greater removal than the dual media bed, although the total output of filtrate before termination was lower. This was considered to be due to the reduction in collector surface area in the dual media. As explained in Section 4.2.1, the larger media effects a lower removal and headloss development rate, based on the Isolated Single Sphere Model. Essentially surface removal is reduced in the dual media bed, as shown by the headloss accumulation data. Figure 4.63 illustrates that at the termination point of the run using the relatively low turbidity raw water of 5 NTU (run C3), 22% of the accumulated headloss occurred in the top 40 cm of the dual media bed, compared to 56% in the equivalent depth in the single media bed. Figures 4.65 and 4.67 show a similar trend using raw water turbidities of 50 (run C2) and 75 NTU (run C1). About 12% of the total headloss accumulated in the top 40 cm with the dual media bed, compared to about 65% with the single media for both raw water turbidities. These results were expected, since the section considered consisted entirely of anthracite in the dual media bed, and sand in the single media bed. The reduction in headloss development rate by larger media has previously been explained. The total output was greater with the dual media because headloss development was the reason for termination, and this was lower with the dual media (Figures 4.65 and 4.67). It may be the case that with other raw waters, breakthrough may be the terminating factor, and the single media bed may yield a higher total output. Other workers using a variety of raw waters also found that the total output was greater with dual media beds (Robeck *et al.*, 1964; Sembi & Ives, 1983; Kawamura, 1991a). The total volume of filtrate produced per run will clearly be dependent upon the specific termination criteria, and is therefore not purely a function of the hydraulic parameters.

It is reported in existing literature that the range of raw water turbidities that can be treated is greater with dual rather than single media beds, due to the increased holding capacity (Kawamura, 1991a; Schulz & Okun, 1992). The relatively high turbidities of 50 and 75 NTU were effectively treated by both single (120 cm) and dual media beds. The more rapid rise in headloss in the single media bed, when treating the 75 NTU water compared with the 50 NTU water, indicates that higher turbidity raw waters would effect even greater headloss development rates. The may ultimately be prohibitive and necessitate the use of dual media beds.

Turbidity measurements from the aforementioned runs (Figures 4.64 and 4.66), taken in the sand layer of the dual media bed (depths of 54 and 86 cm), indicate that turbidity increased rapidly at 2.5 and 1 hours into the runs respectively. In the anthracite layer (depths of 16 and 40 cm), the rise in turbidity was more gradual. It is postulated that greater removal may have been achieved if a deeper layer of sand was used at the expense of a shallower layer of anthracite. This was concluded because the majority of the particulate matter is captured and stored in the large voids of the anthracite layer. The sand layer, which acts as a polishing stage, then removes the remaining particles which are inherently difficult to capture. As can be seen from Figures 4.62, 4.64 and 4.64, the bottom layer of sand (between a depth of 86 cm and 120 cm) was very active in the removal process from the beginning of the run. Thus using a deeper layer of the sand would have reduced the rate of particulate penetration, thereby reducing the filtrate turbidity. The main advantage of having a deeper anthracite layer at the expense of the sand layer, is the reduction in headloss development rate. But since headloss was not a terminating factor, it can be assumed that a deeper sand layer would not limit the filtration process in the dual media bed.

4.6 The combined effect of the hydraulic parameters

In order to determine the combined effect of media size, filtration rate and bed depth on removal efficiencies, bed use and headloss development rate, Figures 4.27, 4.39 and 4.45 were considered. Figure 4.27 illustrates the turbidity removal of a raw water of 20 NTU, using a 70 cm bed. Two filtration rates (5 and 10 m/h) and two media sizes were considered. In Figure 4.27 the turbidity removal graphs are paired in terms of filtration rate indicating that media size was of lesser significance. It was found that the hydraulic parameter with the greatest influence was the filtration rate.

Figure 4.39 shows the variation in turbidity removal with a raw water of 50 NTU, considering all three hydraulic parameters. The greatest removal for both media sizes, occurred at a filtration rate of 5 m/h and bed depth of 120 cm (Runs B6 and B8). In order to determine if this rate was a function of depth or filtration rate, the runs are compared with those conducted at 5 m/h using a 70 cm bed (Runs A7 and A10). It can be seen that for both media sizes the removal rates were very similar until the output reached about 30 m^3/m^2, when the shallower bed depth began to show signs of impending breakthrough. If the 5 m/h runs using 120 cm bed depth are compared with those conducted at 10 m/h in a 120 cm bed (runs B5 and B7), it can be seen that there was significantly greater difference between the removal rates from the commencement of the runs, indicating that it was the filtration rate which had the major influence on removal.

A similar trend can be seen in Figure 4.45 with a raw water of 75 NTU. Removal rates in the 70 and 120 cm beds were very similar for both sand sizes at 5m/h (runs A2 and B2, and A4 and B4); but as the run progressed and the media clogged, bed depth became a significant factor, and an insufficient depth resulted in premature breakthrough. Increasing the filtration rate from 5 to 10 m/h, however, produced a lower quality filtrate throughout the run. Indicating that in order to maintain high removal for long periods of time, with a relatively high turbidity raw water and high filtration rate, a deep bed is necessary. Thus, it would seem the main influence on the turbidity removal rate was filtration rate. The synergistic effect of a shallow bed and a large media was to reduce the quality of the filtrate; whereas singularly their effects were limited, particularly at the commencement of the run.

The effect of the hydraulic variables on headloss development (both absolute and additional) can be seen in Figures 4.28, 3.39 and 4.46. In all cases the media size was the major influence on headloss. The effect of deeper beds and higher filtration rates was to increase clean bed headloss, but to reduce the additional headloss development rate. The extent of this effect varied depending on the raw water turbidity. Thus it would appear that the headloss development rate and turbidity removal are affected by different hydraulic variables. It is postulated that this may be as a result of the effect of the hydraulic variables on floc morphology i.e. surface area and volume of the captured particles. A large collector surface area offers a greater resistance to flow due to higher fluid drag and therefore increase of headloss development. Removed particles can also form dendrite like structures which attach to the collector surface. If a particle is retained in the shadow of these structures, then it will have a minimal effect on the headloss development, since it will cause little or no fluid drag on the flow, but the filtrate turbidity will be reduced. Larger particles entering the bed will saturate the attachment sites more rapidly and cause more rapid clogging of the interstitial pores. Thus, the turbidity removal rate is lowered due to a reduction in the number of available attachment sites and/or an increase in the interstitial velocities and shear forces on the previously removed particle. Hence, it is suggested that media size affected the extent of particle aggregation, which in turn altered the surface area and not the volume of the captured material. The effect of media size was therefore greater on the headloss development rate than on the turbidity removal rate. The effect of filtration rate is mainly on the extent of particle penetration into the bed.

Clark *et al.* (1992) found the effect of media size to be more significant than filtration rate on removal rates, bed utilisation and headloss development, whereas Darby *et al.* (1991) found the opposite. Clearly important factors are the range of media sizes, bed depths and flow rates considered, as well as the chemical and physical properties of the raw water. From the point of view of this research the coagulant used is significant. Tanaka and Pirbazari (1986) concluded from their study into the effect of the various physical parameters, that the most significant factor was the choice of coagulant. Specifically, the molecular weight and extent of branching of the polymer. Therefore an explanation for the difference in results obtained by Clark *et al.* (1992) and Darby *et al.* (1991) compared with this research is that the coagulants used initiated particle destabilisation and aggregation in a different manner. The relatively weak bonds formed by *M.oleifera* seed may be influenced to a greater extent by the filtration rate

than are other coagulants. Higher filtration rates cause rapid and deep penetration of the particulate matter throughout the bed, and early breakthrough of turbidity. Accordingly, deeper beds are a necessity with the higher filtration rates, larger media and the use of *M.oleifera* seed as the coagulant.

Headloss development rates are relatively low with *M.oleifera* seed due to the lack of surface filtration as a removal mechanism. Therefore, the increased headloss accumulation resulting from lower filtration rates was not significant. In contrast, if a strong bond forming polymer was used, the rate of headloss accumulation would be significant. Therefore, an increase in filtration rate can be of benefit by decreasing the rate of surface removal and causing deeper penetration of the particulate matter. Premature turbidity breakthrough is unlikely since particle reattachment deeper into the bed will occur with stronger bond forming polymers. An example of this can be seen in Chapter 6, with the use of chitosan - a 'bridge' forming polymer.

In the treatment of the relatively high turbidity waters of 50 and 75 NTU, the lowest turbidity filtrate was obtained in the 120 cm bed of small media operated at 5 m/h, that is to say runs B6 and B2 . Termination of the run was caused by excessive headloss accumulation, in the case of the smaller media. The rate of headloss development was halved with the dual media bed, making the treatment process viable, although with a marginal reduction in filtrate quality. This is illustrated by runs C2 and C1. Optimal treatment conditions for water with a turbidity of 20 and 35 NTU was obtained in the 120 cm bed of small media. Although only a single media bed and a filtration rate of 10 m/h were investigated, conditions were considered to be optimised since turbidity breakthrough and terminal headloss were reached at the same time. The optimal treatment conditions for the 5 NTU water are difficult to predict as the runs were not continued until either of the specified termination points were reached (as described in Section 3.3.1). As expected the highest removal rates were obtained in the 120 cm bed of small media, but although terminal headloss was not reached, after 13 hours into the run the headloss was double that of the dual media bed and the 120 cm deep bed of larger media. It is therefore predicted that using either of the latter bed configurations would increase the total output at the expense of a minimal deterioration in filtrate quality, if the run had been continued until termination.

4.7 Conclusions of the laboratory study into the hydraulic variables

The effect of the various hydraulic variables viz.: media size, bed depth, filtration velocity and media configuration were investigated in terms of optimising the treatment process on the laboratory rig. The following conclusions were drawn from the results obtained:

• It was observed during jar tests using *M.oleifera* seed as the coagulant that floc formed slowly, therefore any parameter which increased the duration of flocculation would bring about better floc formation and removal. As flocculation only occurs in 'contact' with the filter media in the process of CFF, then in-pore flocculation needs to be maximized when using *M.oleifera* seed. In-pore flocculation was considered to be significant in the laboratory study for a number of reasons. Firstly, sedimentation rather than Brownian motion was found to be the dominant mechanism of particle transport leading to capture by the filter media. As sedimentation is only effective with particles > 2.5 μm, then it can be assumed that flocculation occurred to some extent prior to floc/particle capture. Floc would not have formed prior to entering the bed (due to the slow rates of floc formation), therefore in-pore flocculation must have occurred. Secondly, filter runs conducted after pre-coating the filter media did not improve the overall removal rates, indicating that only formed floc are removed from the flow. With high molecular weight 'bridge' forming polymers, it can be envisaged that the polymer chain would attach to the media and then capture particles from the flow. But with low molecular weight polymers (e.g. *M.oleifera* seed), the strands would not be able to protrude out and capture particles. Therefore removal can only occur by 'patches' of positive charge forming on the surface of the negatively charge particles and then for the particles to collide and form floc, which are transported to the collector surface and removed. Accordingly, parameters which reduce retention time in the bed, make conditions unfavorable for floc formation and decrease particulate removal. Deep bed filtration ultimately results in lower headloss development rates. Thus parameters which reduce the headloss development rate are only of real significance at the higher loading rates.

• The wide dose response encountered with the use of *M.oleifera* seed was considered to be a function of the 'patch' mechanism of floc formation and the fact that the whole seed was used in the preparation of the coagulant suspension. The active coagulant was found to make up < 30% of the total seed weight, and therefore >70% of the suspension consisted of extraneous

seed material. This characteristic of *M.oleifera* seed offers a great advantage to treatment works in developing countries, where shortages of skilled labour and accurate dosing equipment may be encountered. The use of *M.oleifera* results in a robust treatment process, that is not dependent upon accurate dosing of the coagulant.

• Treatment of waters with turbidities of 5-75 NTU under various conditions indicated that a combination of a 70 cm depth of filter media and larger media did not bring about effective treatment for long periods of time. Greater removal consistently occurred with the 120 cm bed depth of single and dual media. This can clearly be seen with the treatment of 75 NTU raw water at a filtration rate of 10 m/h in Figures 4.45 and 4.66. With the dual media bed, filtrate turbidity remained below 5 NTU for 3 hours, with the 120 cm bed of small media this period extended to 5 hours, with the 70 cm bed of large media the filtrate turbidity was not reduced below 5 NTU throughout the run. This trend was seen to some extent with all the raw waters considered, since deeper beds and smaller media provided greater collector surface area and retention times in the filter bed and hence enhanced turbidity removal rates.

• The variation in performance of the media sizes can be summarised as greater turbidity removal with the smaller media, and a slower rate of headloss development with the larger media. Thus where conditions were such that turbidity breakthrough was the reason for termination, the bed of small media produced a higher total output, as seen with the 50 NTU raw water in Figures 4.39 and 4.40. Whereas when excessive headloss accumulation was the cause of termination, the larger media produced a higher output (although the quality of filtrate was marginally lower), as seen with the 10 NTU raw water in Figures 4.11 & 4.12. The headloss development rates were higher due to the greater resistance to flow by the increased surface area of the media. Also the reduction in size of the interstitial pores and the consequent increase in interstitial velocities caused a rise in the rate of headloss development. The trade-off between the quality and quantity of the filtrate produced is a matter that needs to be considered at the design stage, since it would not be practical to change the filter media specification during the operation of a plant.

• Deeper beds effected greater turbidity removal due to longer retention times, higher collector surface area and a greater number of interstitial pores leading to an increase in in-pore flocculation. Headloss development rates are also affected by the bed depth: clean bed headloss

was higher due to the greater resistance to flow. The additional headloss was lower due to the greater distribution of material throughout the bed, which in turn reduces the specific deposit in each pore and therefore the extent of pore constriction and resistance to flow.

- In order to assess the effect of filtration rate on the removal process, the influence of bed retention time was removed, by maintaining the ratio of bed depth to filtration rate in two filter beds. Removal rates were found to be very similar in shallow beds at lower filtration rates as deeper beds at higher filtration rates. This indicated that the main influence of filtration rate was due to the retention time in the bed. This was considered to be due to the effect on in-pore flocculation, which was relatively slow with *M.oleifera*. Thus if higher rates are to be utilised, then the reduction in removal efficiency would need to be compensated for by increasing the depth of the filter bed.

- Filtration rate was found to be the most significant hydraulic variable in terms of turbidity removal. This was considered to be due to the effect of filtration velocity on interstitial velocities and therefore the rate of in-pore flocculation and the inter-pore shear forces, which brought about detachment of previously removed particles. Media size was found to be the main influence on headloss development rate. This was considered to be due to the increase in resistance to flow with the smaller media caused by the constriction in pore size.

- A working front can be seen in most of the runs progressing through the bed, in the form of activation of sequential layers of the filter media. This phenomenon is not so obvious when the turbidity loading on the bed is very low or very high. The significance of the progression of the working layer is that for true, deep bed filtration to occur, and for optimal conditions to be reached, the entire bed should be active in floc/particulate removal. If the working front reached the bottom of the bed and the point of terminal headloss were attained simultaneously, then the operational parameters are optimal. This is clearly not only dependent on the raw water quality and the operational variables, but also the termination criteria employed.

- The reduced filtrate quality and reduction in headloss development in the dual media bed were considered to be due to the lower collector surface area and greater interstitial pore size respectively. With the relatively high turbidity waters (50 and 75 NTU), the reduction in headloss development in the dual media compared to the single media bed of the same depth,

increased the duration of the run. Thus it was considered that for such waters, CFF would only be viable if dual media beds were utilised. The major limitation when using the dual media as compared to the single media bed, was the reduction in filtrate quality. Therefore using a deeper layer of sand at the expense of the anthracite layer would potentially produce higher quality filtrate without increasing the rate of headloss development to any great extent.

- Surface removal occurred to some extent regardless of media size, depth, and configuration, flow rate, water quality or coagulant dose. Optimisation of any treatment process will consist of reducing surface filtration and increasing penetration of the particulate matter before removal occurs. Essentially deep bed filtration is required. An advantage of using *M.oleifera* seed, is that the slow rate of floc formation results in deeper penetration of the particulates prior to capture. Therefore the percentage removal which occurs in the surface layers of the bed as compared to the total removal is relatively low and deep bed filtration is brought about.

- The choice of operational filtration rate and media size and depth depends on the raw water quality and the design of the treatment works, specifically on the amount of available head in the filter bed. There is an economic trade-off between the volume of filtrate produced before termination of the run and the volume of water required for backwashing, coupled with the extra costs of construction and maintenance of deeper beds.

- This work investigated the effect of varying these hydraulic parameters in order to optimise the process and to understand the effect of each one on the removal process. It is recognised, however, that any conclusions reached are specific to the laboratory water in the pilot rig and optimal condition reached in this work would not necessarily apply to other water types. The CFF treatment process is not appropriate for all water types all year round, because even in low turbidity waters, spikes of higher turbidity may lead to the need to use a 'buffer' in the form of pretreatment, to ensure efficient operation of the filter beds.

- The performance achieved with CFF and *M.oleifera* under laboratory conditions indicates that it is a suitable single stage treatment process for low to medium turbidity water (5-75 NTU). Furthermore, it is reported that full scale plants operated in the DF mode perform better than that predicated by pilot studies (Schulz & Okun, 1992). The successful results of this pilot study indicate a great potential for this treatment process at full scale.

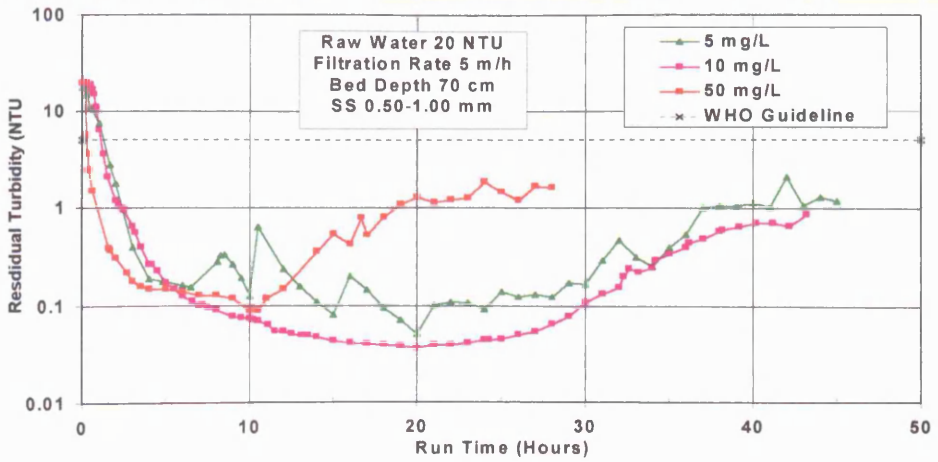

Figure 4.1 Dose response curve for a raw water turbidity of 20 NTU; sand size 0.50-1.00 mm.

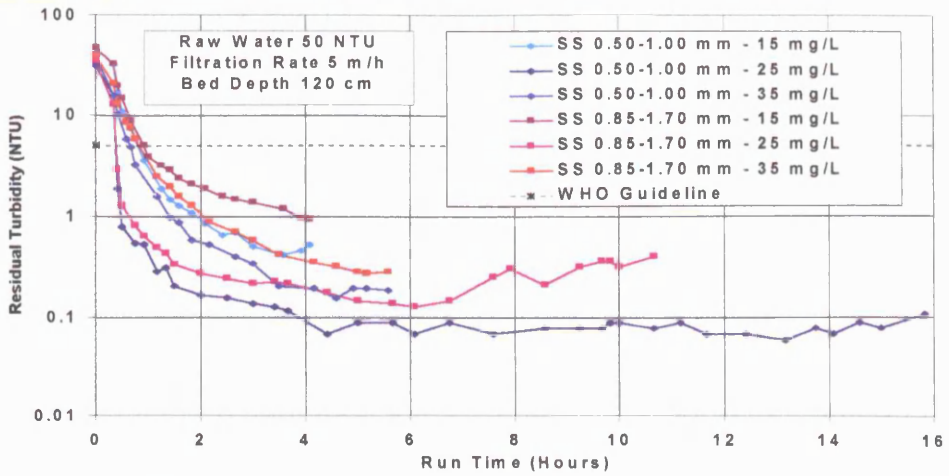

Figure 4.2 Dose response curve for a raw water turbidity of 50 NTU.

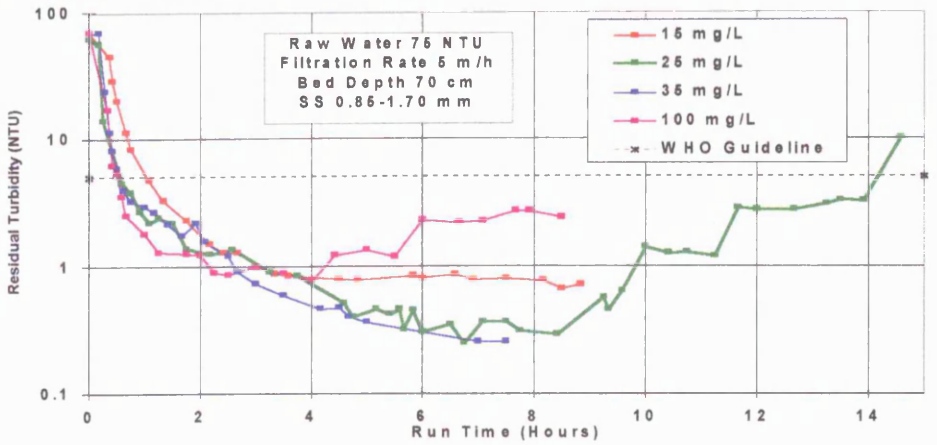

Figure 4.3 Dose response curve for a raw water turbidity of 75 NTU; sand size 0.85-1.70 mm.

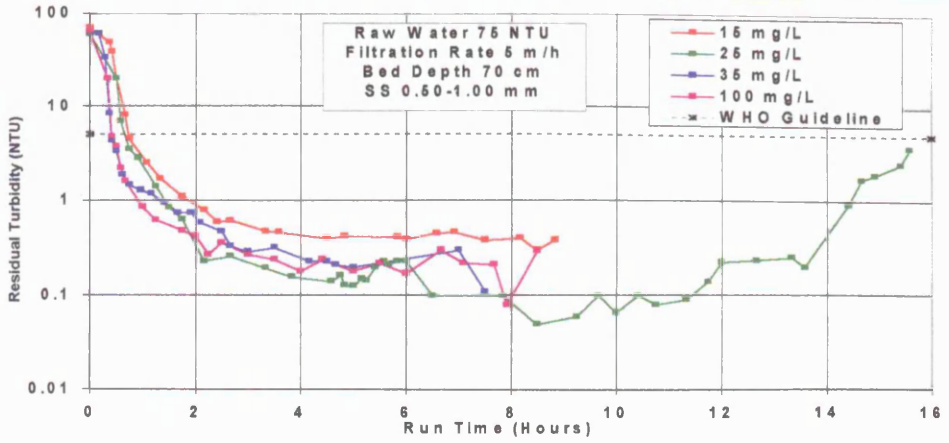

Figure 4.4 Dose response curve for a raw water turbidity of 75 NTU; sand size 0.50-1.00 mm.

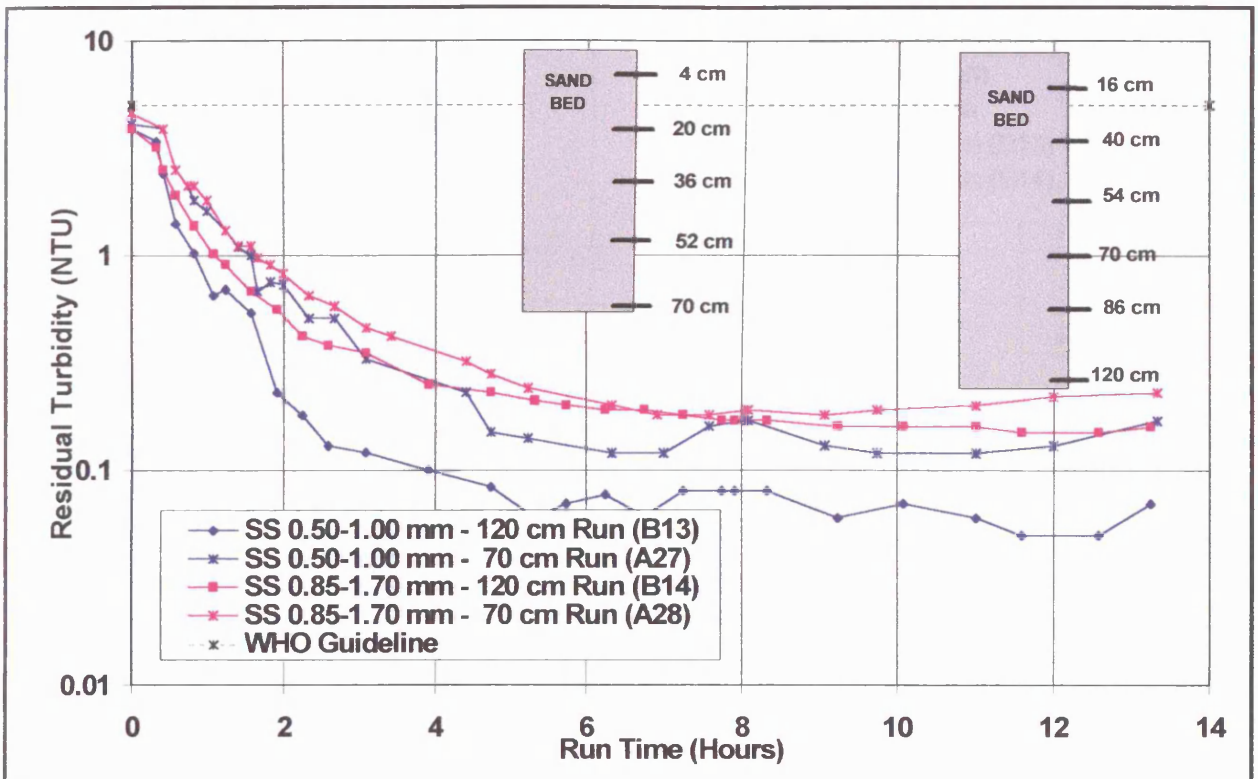

Figure 4.5 Turbidity reduction as a function of bed depth and sand size; raw water turbidity 5 NTU, filtration rate 5 m/h.

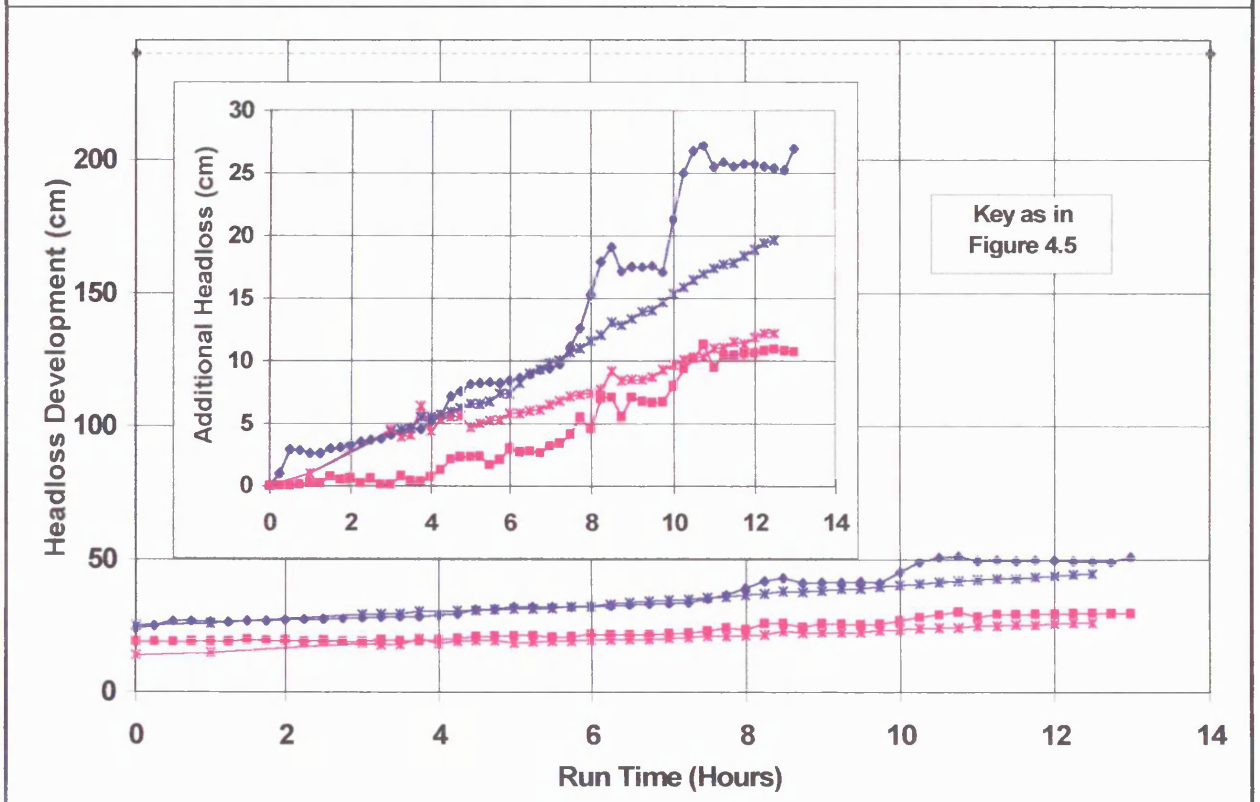

Figure 4.6 Absolute and additional headloss development as a function of bed depth and sand size; raw water turbidity 5 NTU, filtration rate 5 m/h.

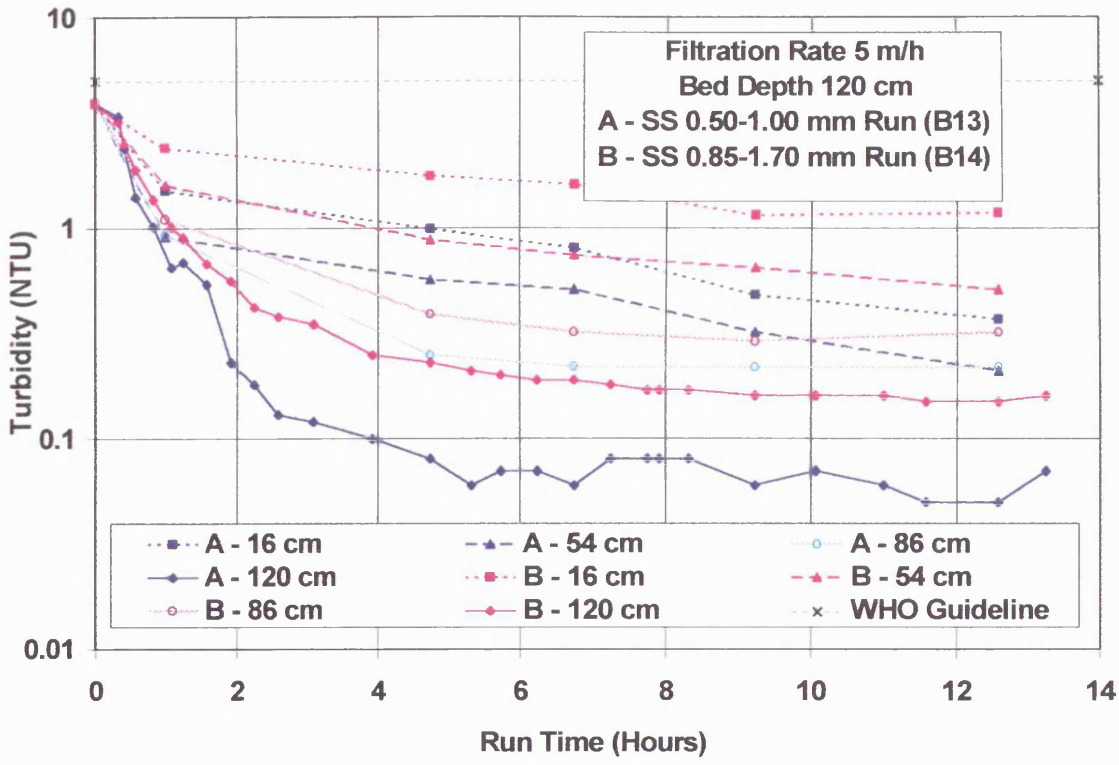

Figure 4.7 Turbidity reduction as a function of depth; raw water turbidity 5 NTU, bed depth 120 cm.

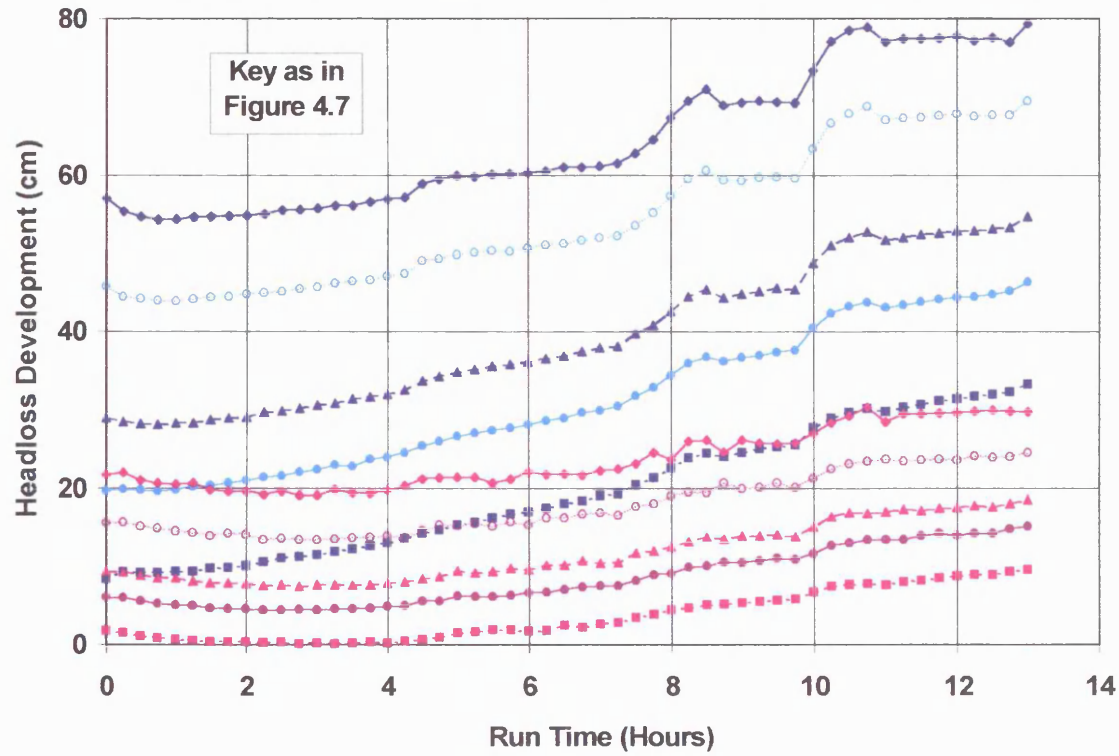

Figure 4.8 Headloss development as a function of depth; raw water turbidity 5 NTU, bed depth 120 cm.

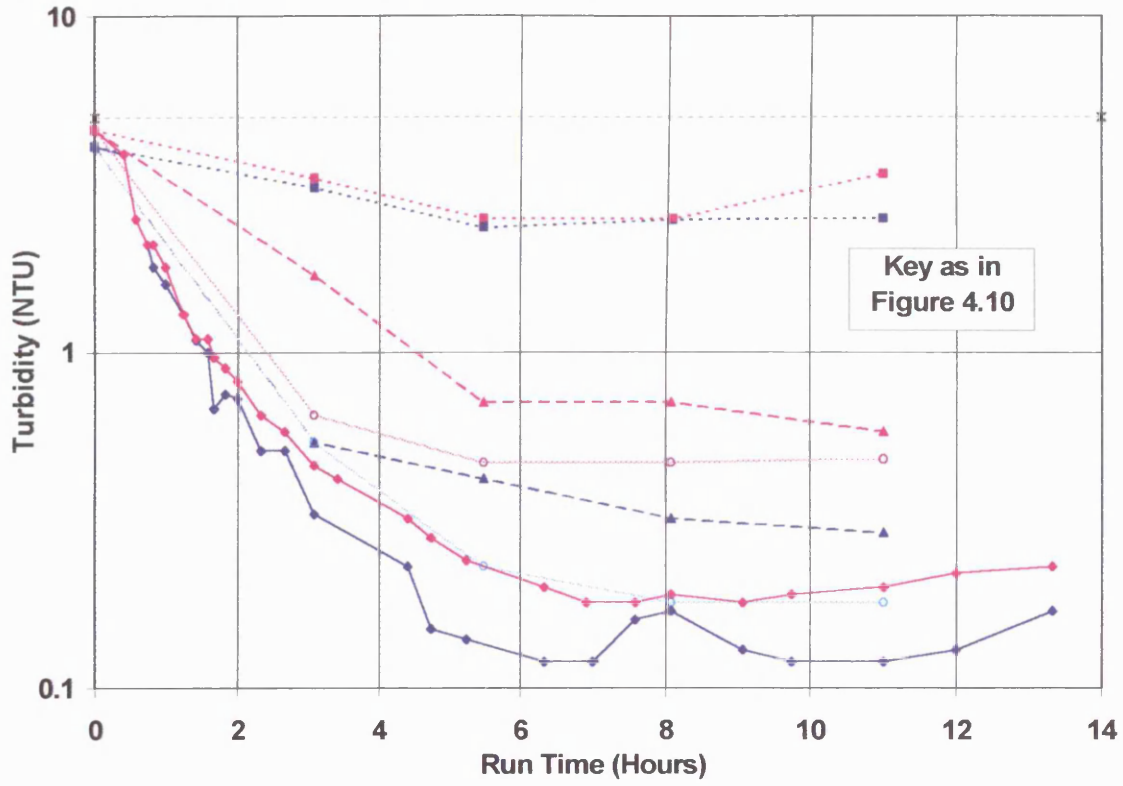

Figure 4.9 Turbidity reduction as a function of depth; raw water turbidity NTU, bed depth 70 cm.

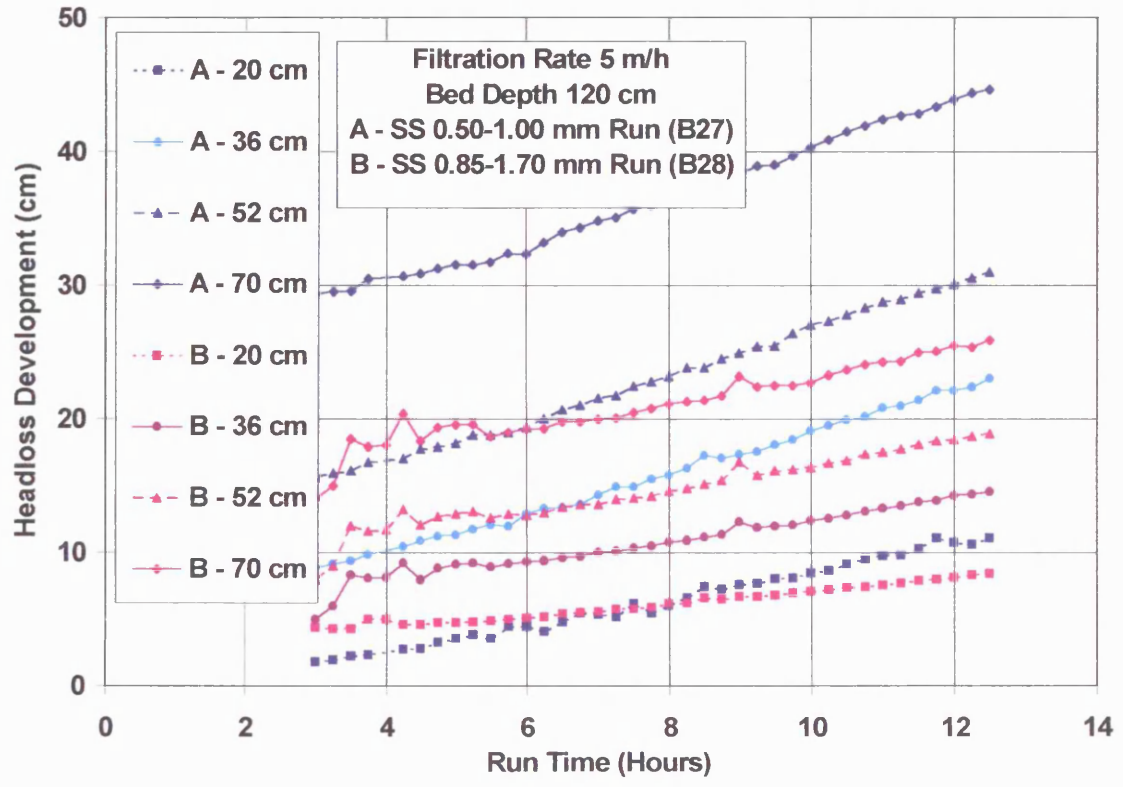

Figure 4.10 Headloss development as a function of depth; raw water turbidity 5 NTU, bed depth 70 cm.

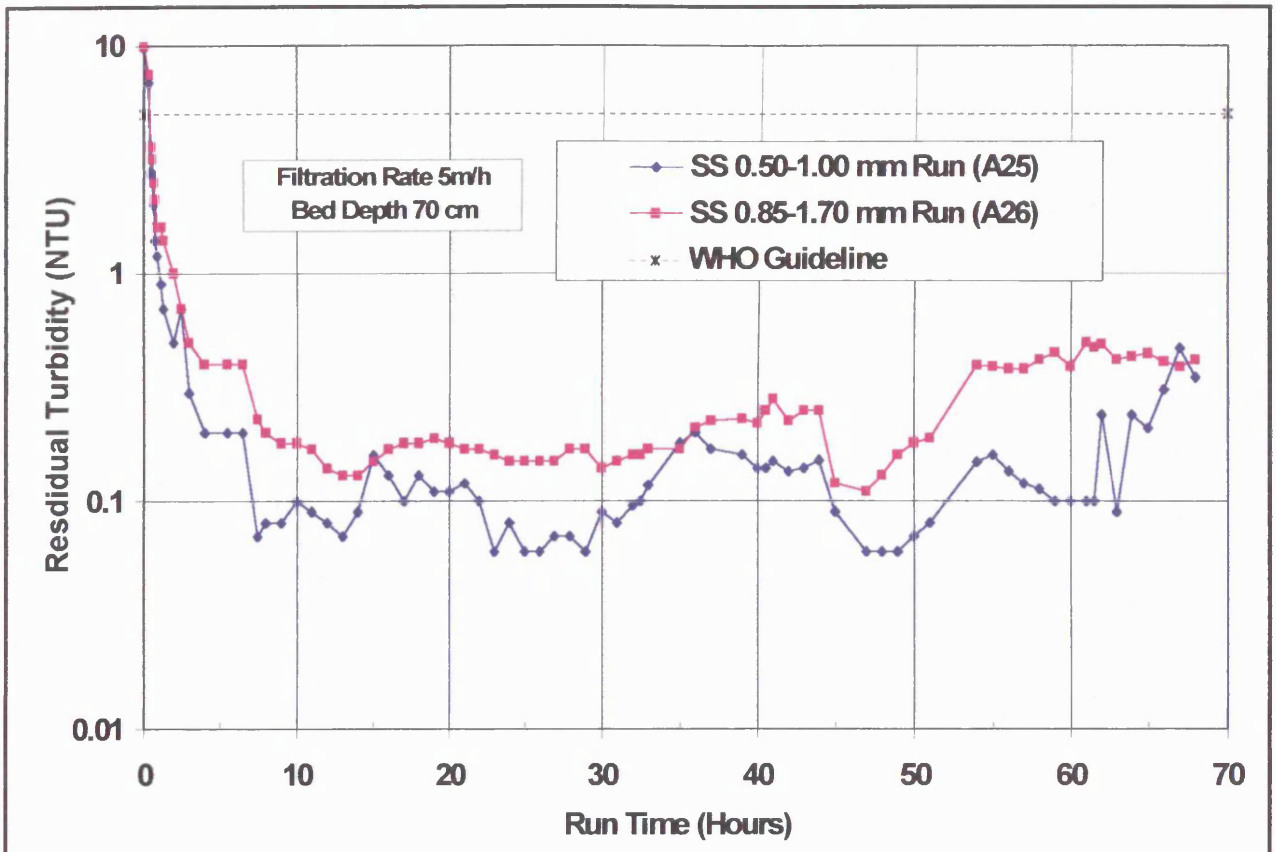

Figure 4.11 Turbidity reduction as a function of media size; raw water turbidity 10 NTU.

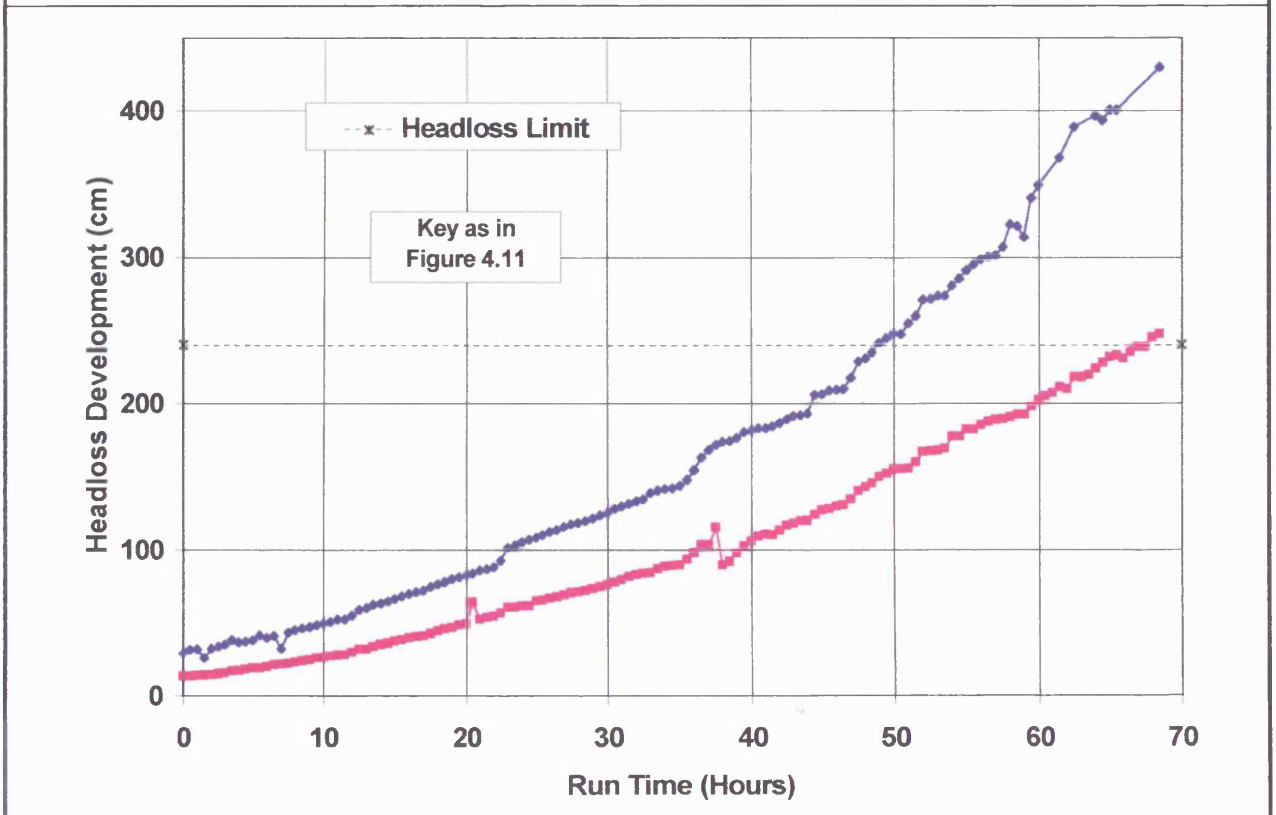

Figure 4.12 Headloss development as a function of media size; raw water turbidity 10 NTU.

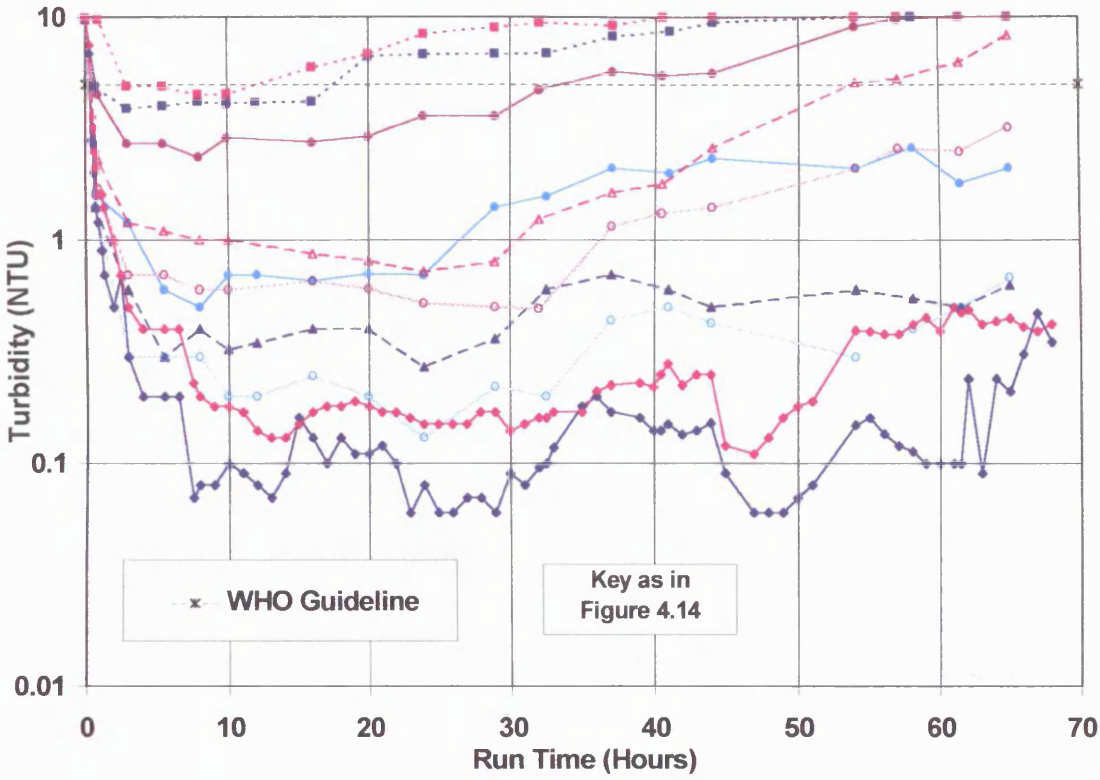

Figure 4.13 Turbidity reduction as a function of depth; raw water turbidity 10 NTU.

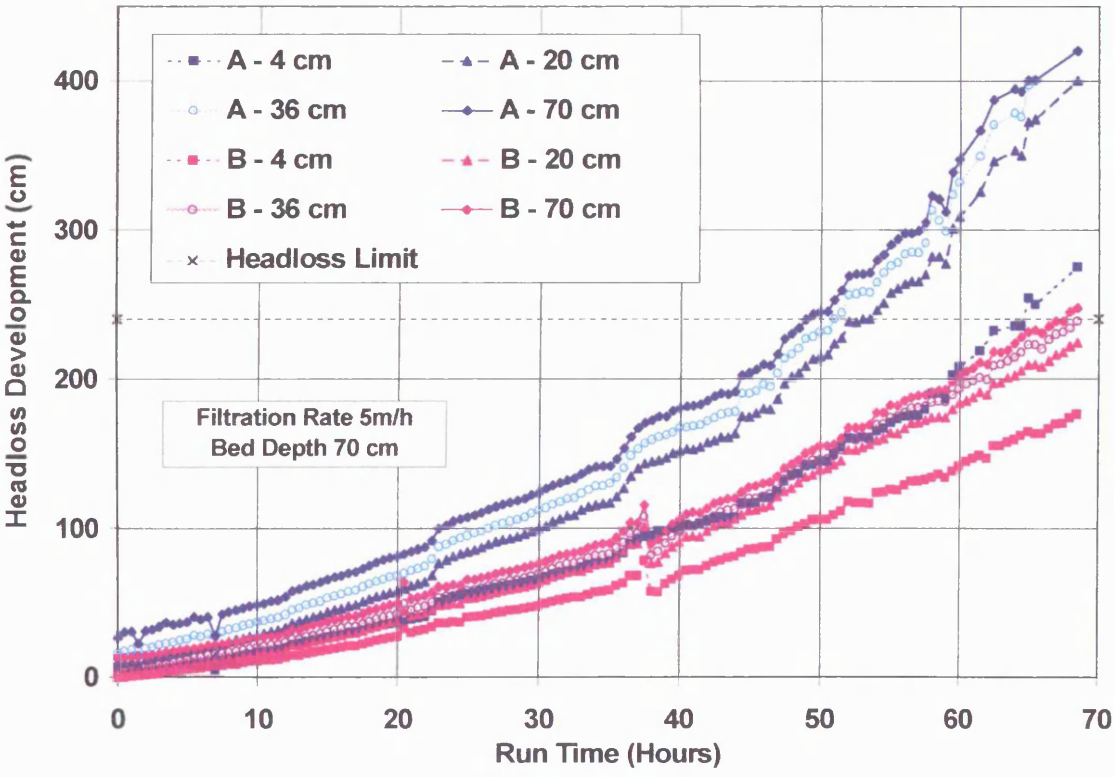

Figure 4.14 Headloss development as a function of depth; raw water turbidity 10 NTU.

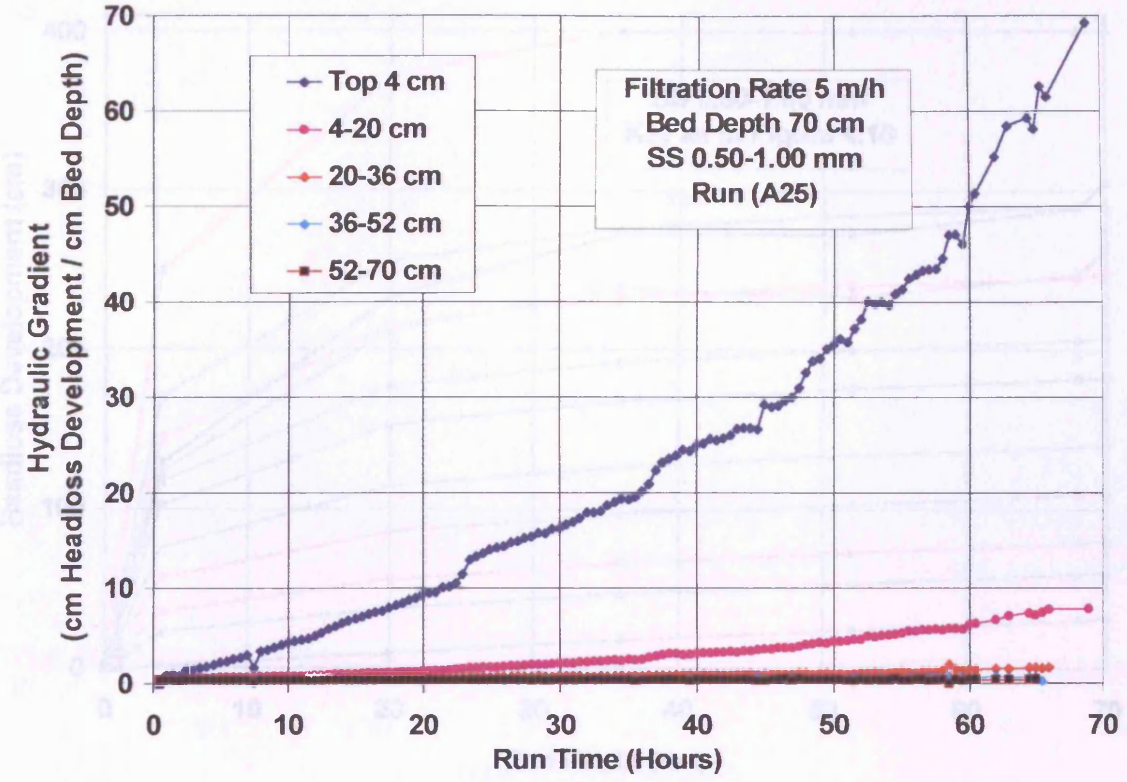

Figure 4.15 Hydraulic gradient as a function of depth; raw water turbidity 10 NTU, sand size 0.50-1.00 mm.

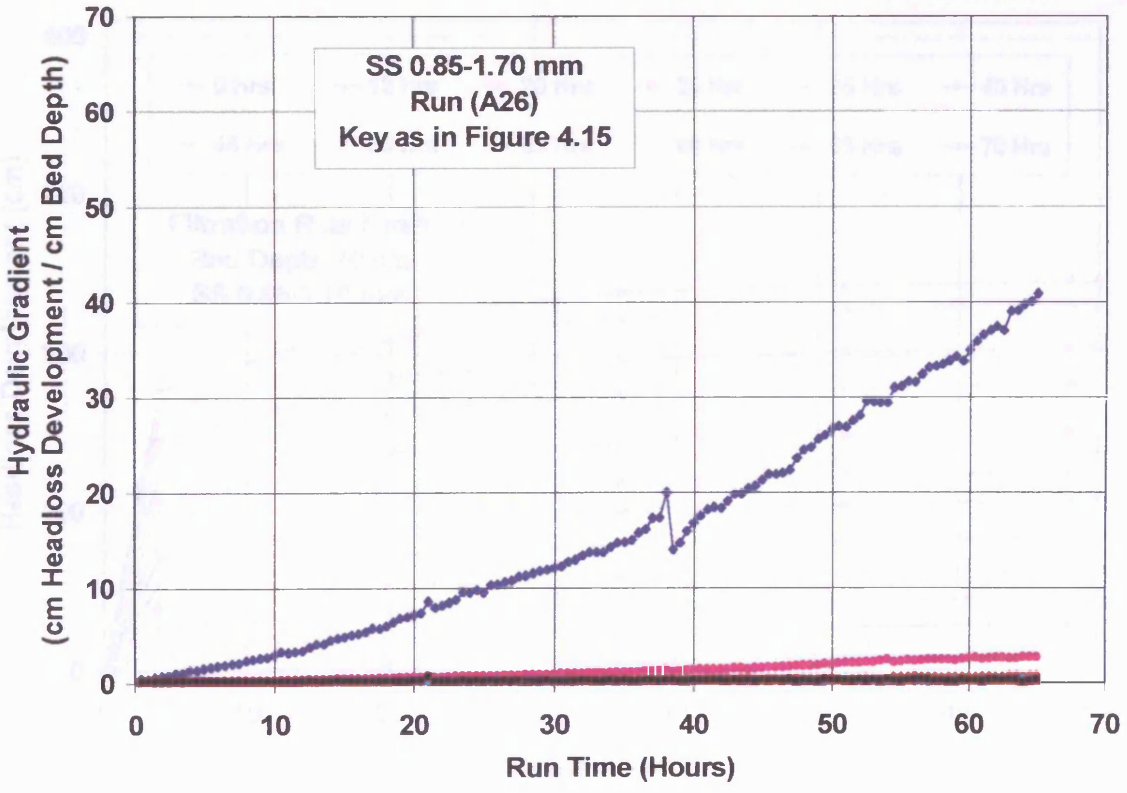

Figure 4.16 Hydraulic gradient as a function of depth; raw water turbidity 10 NTU, sand size 0.85-1.70 mm.

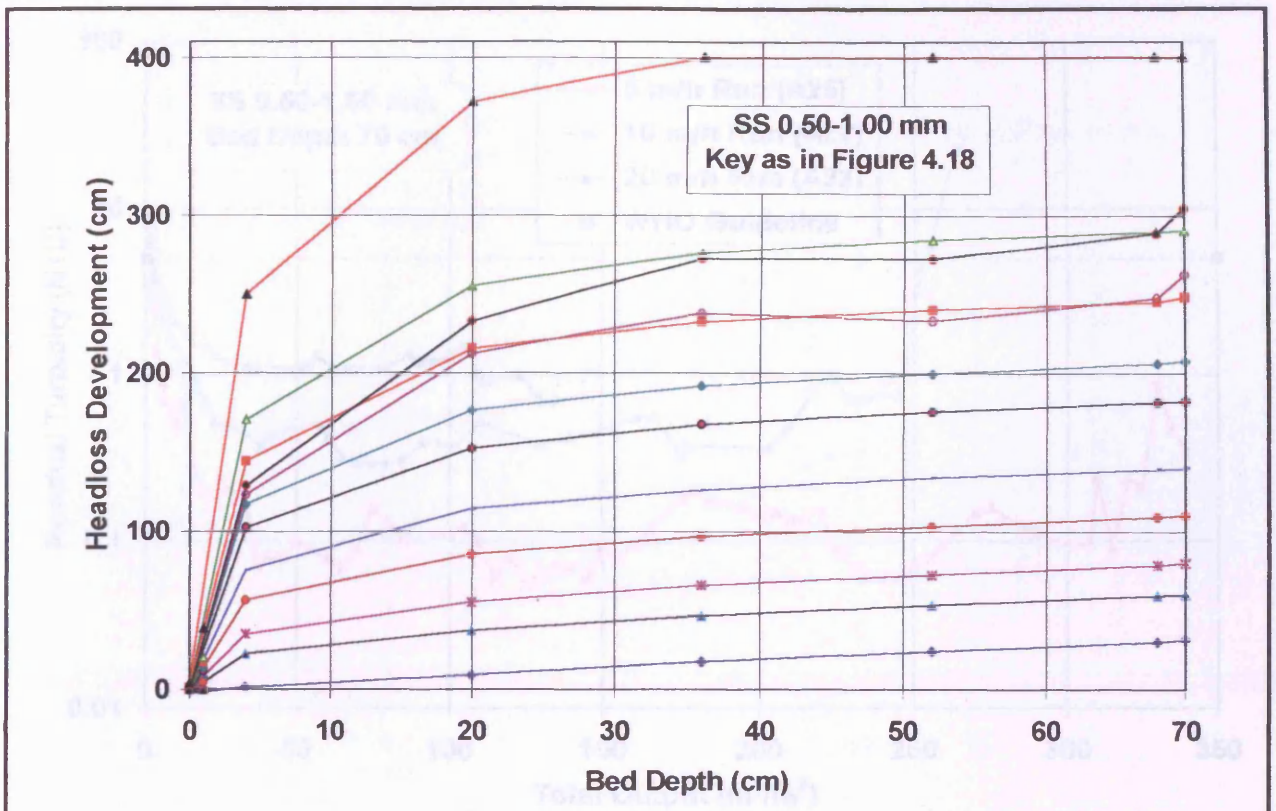

Figure 4.17 Headloss development as a function of time; raw water turbidity 10 NTU, sand size 0.50-1.00 mm.

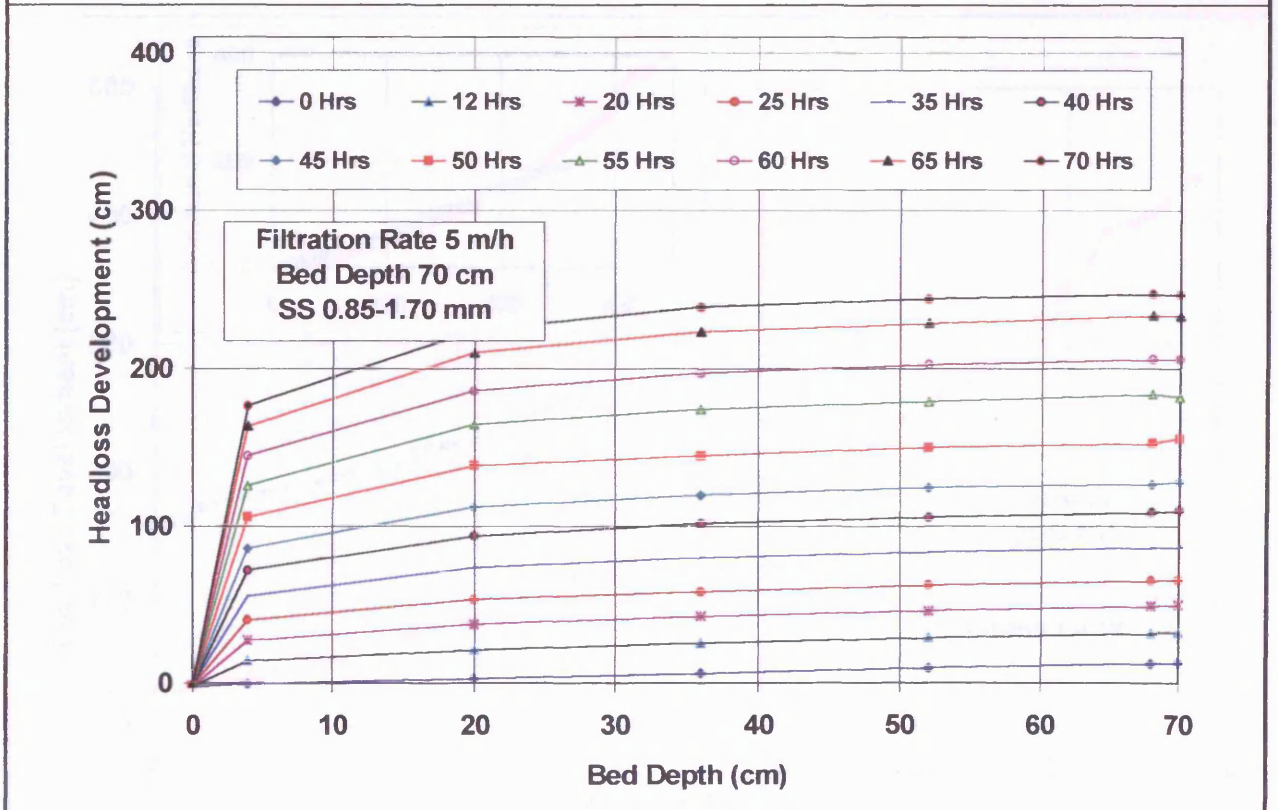

Figure 4.18 Headloss development as a function of time; raw water turbidity 10 NTU, sand size 0.85-1.70 mm.

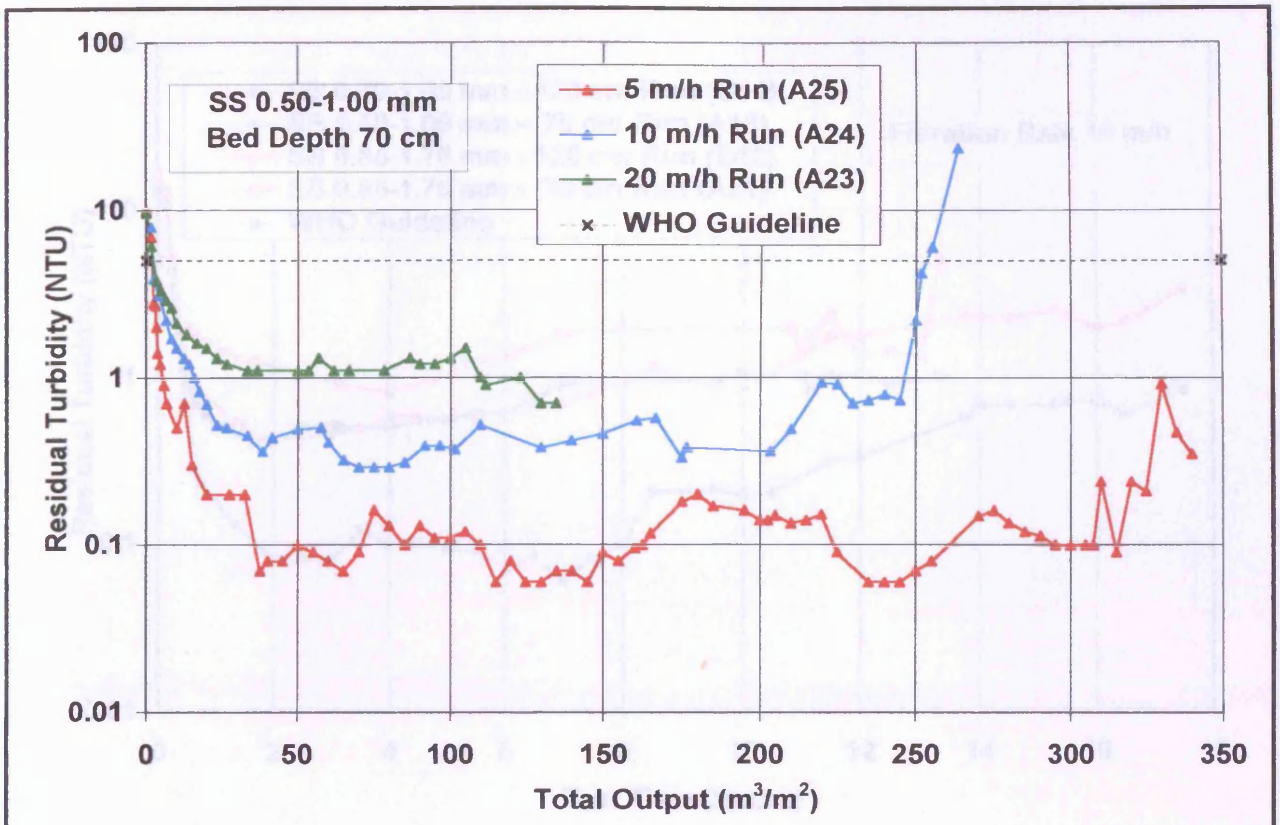

Figure 4.19 Turbidity reduction as a function of filtration rate; raw water turbidity 10 NTU.

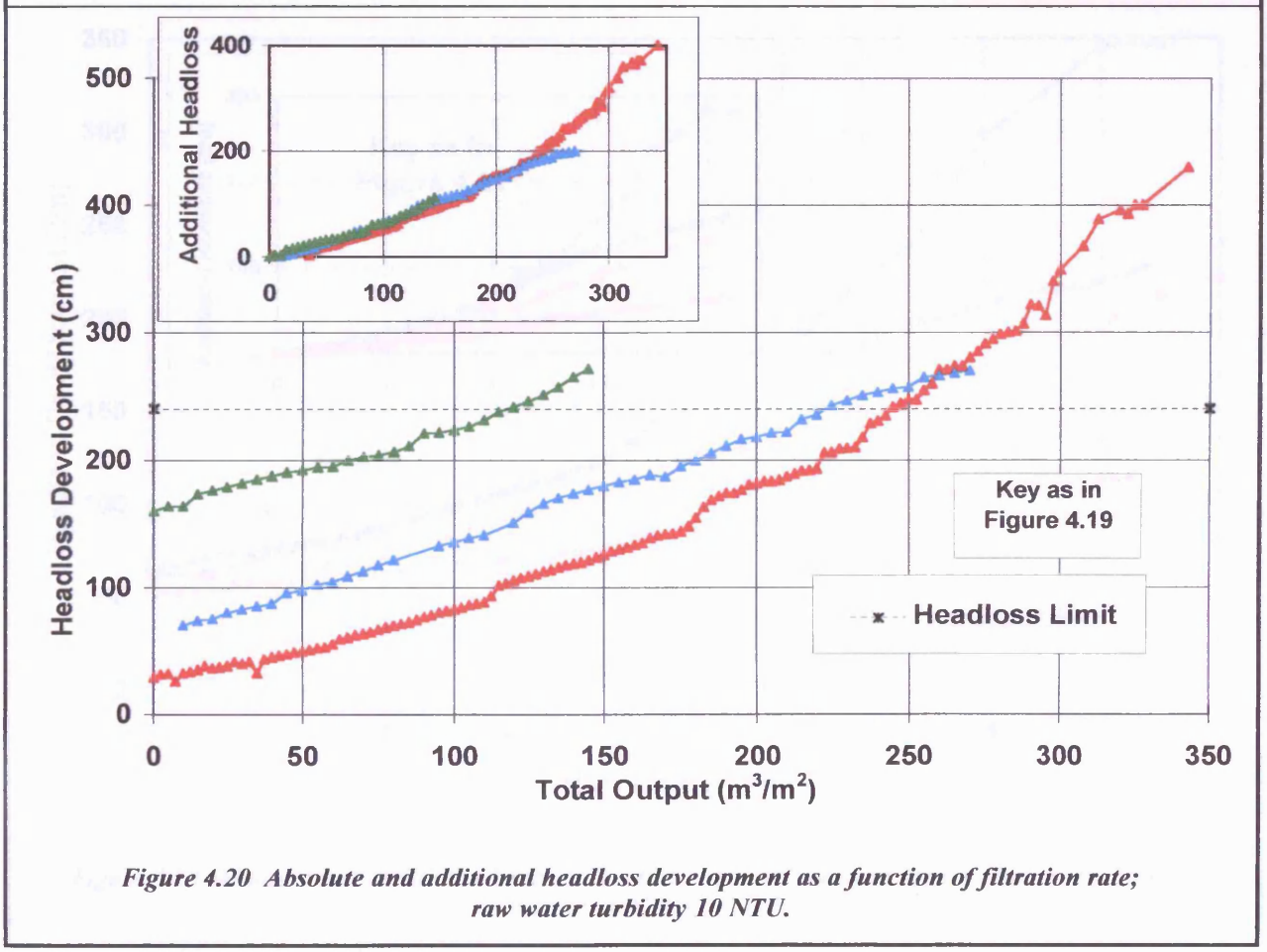

Figure 4.20 Absolute and additional headloss development as a function of filtration rate; raw water turbidity 10 NTU.

Figure 4.21 Turbidity reduction as a function of bed depth & media size; raw water turbidity 20 NTU.

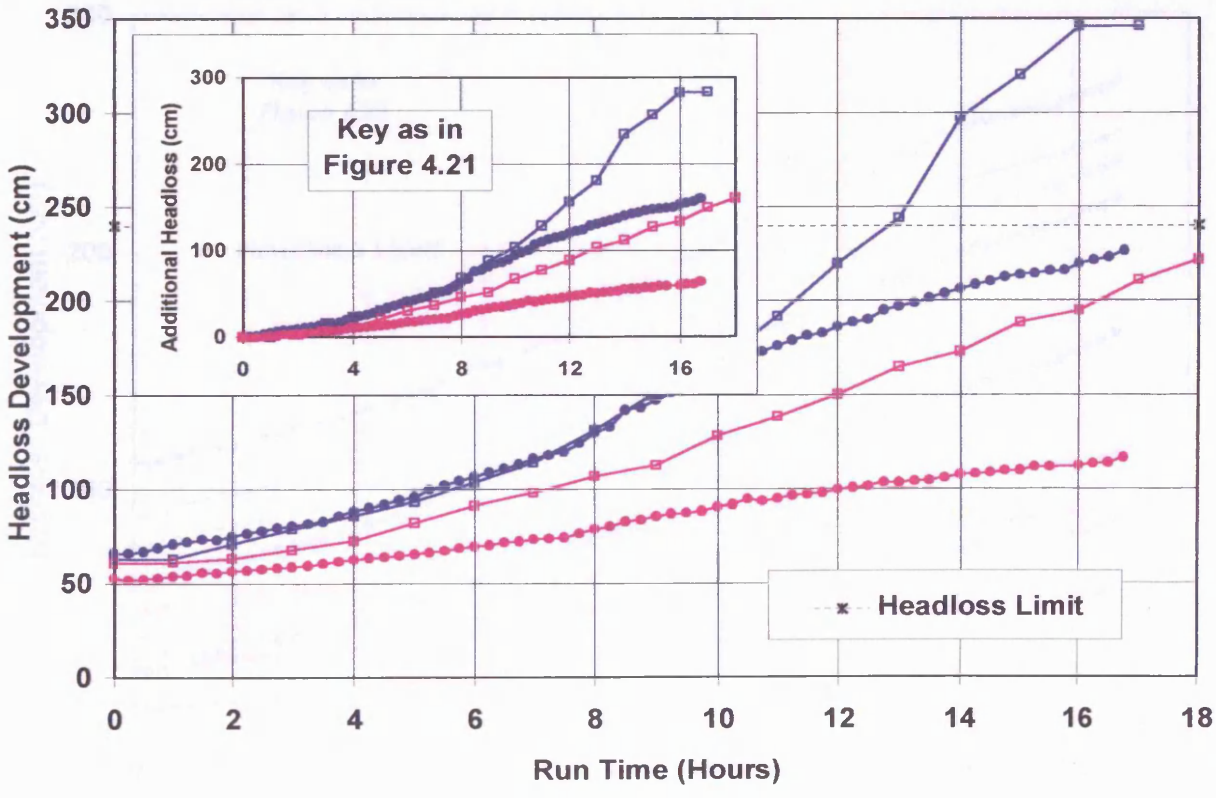

Figure 4.22 Absolute and additional headloss development as a function of bed depth & media size; raw water turbidity 20 NTU.

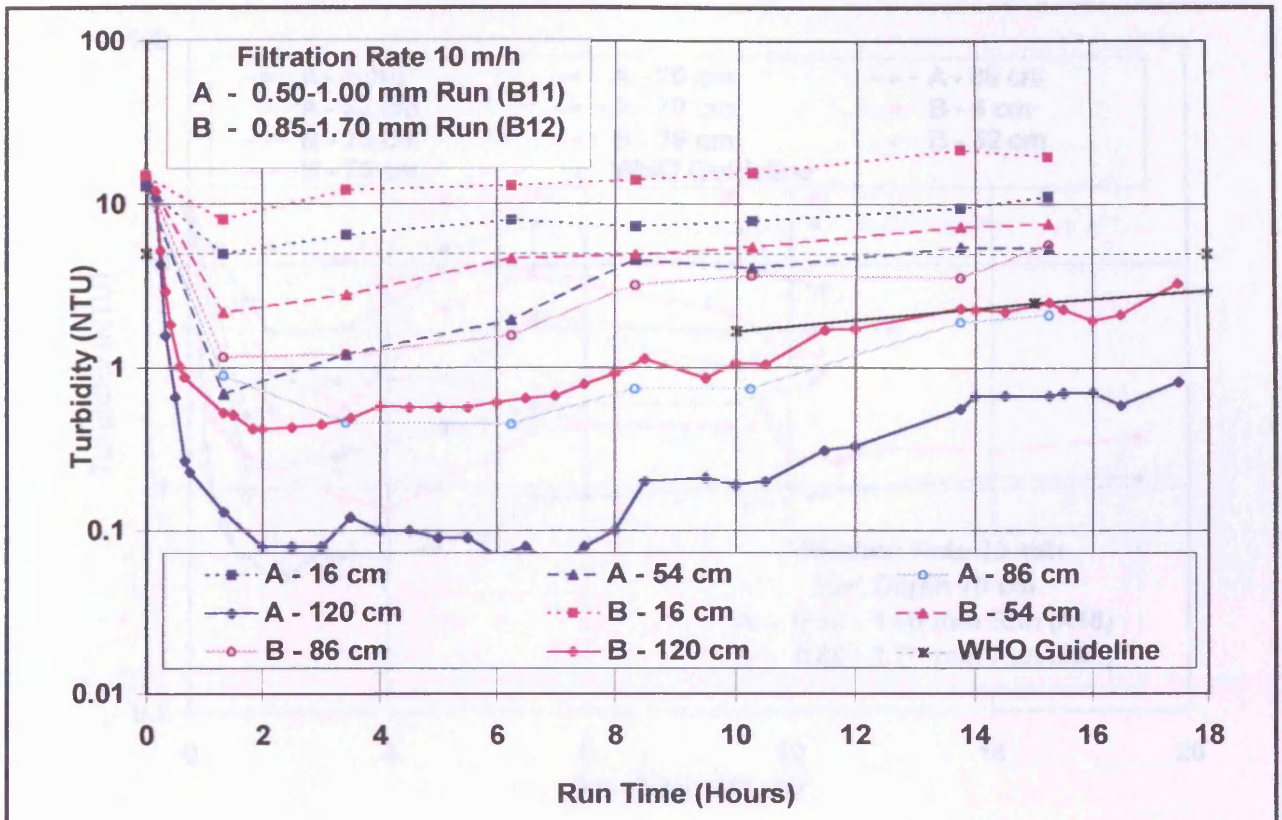

Figure 4.23 Turbidity reduction as a function of bed depth; raw water turbidity 20 NTU, bed depth 120 cm.

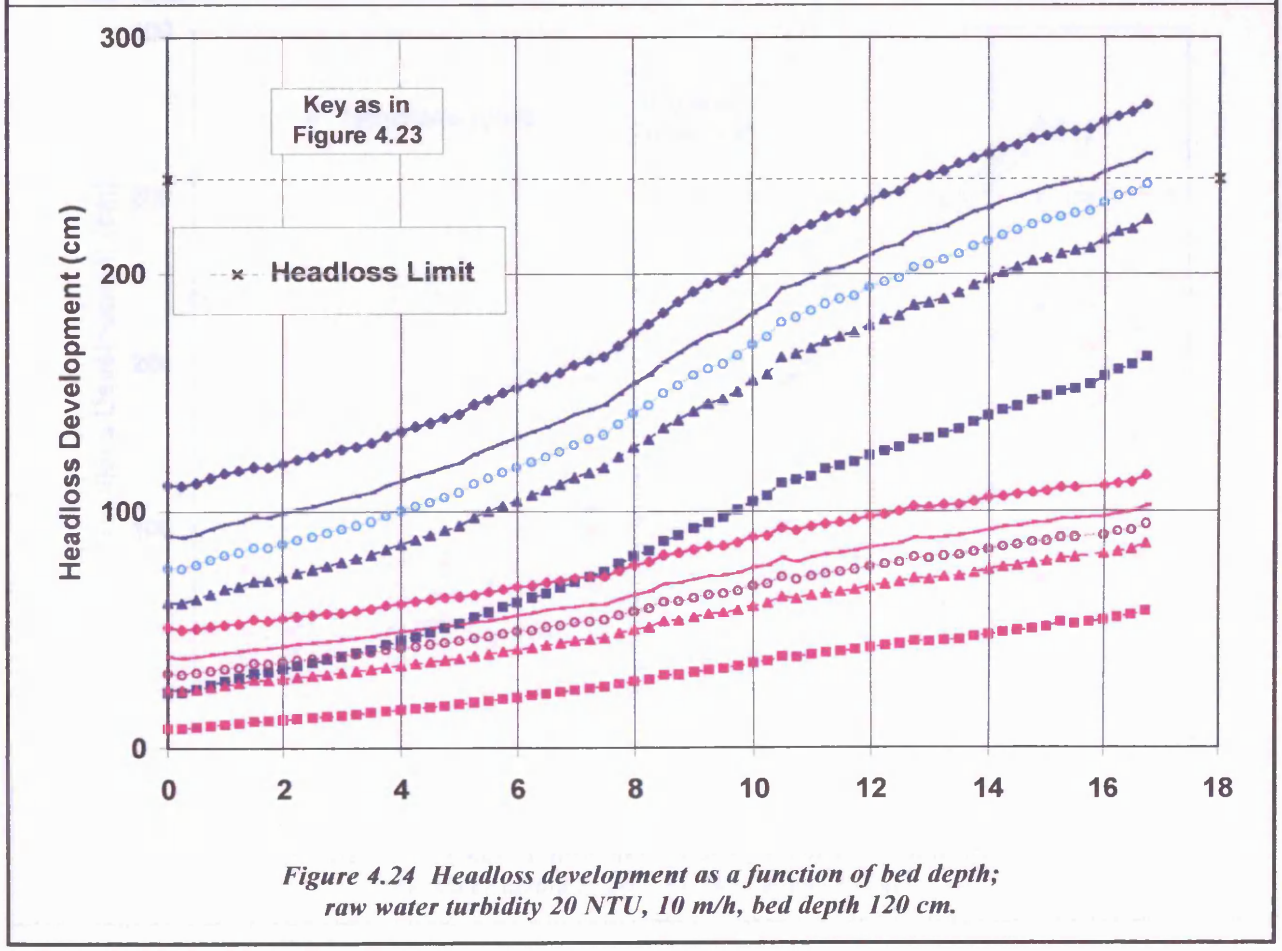

Figure 4.24 Headloss development as a function of bed depth; raw water turbidity 20 NTU, 10 m/h, bed depth 120 cm.

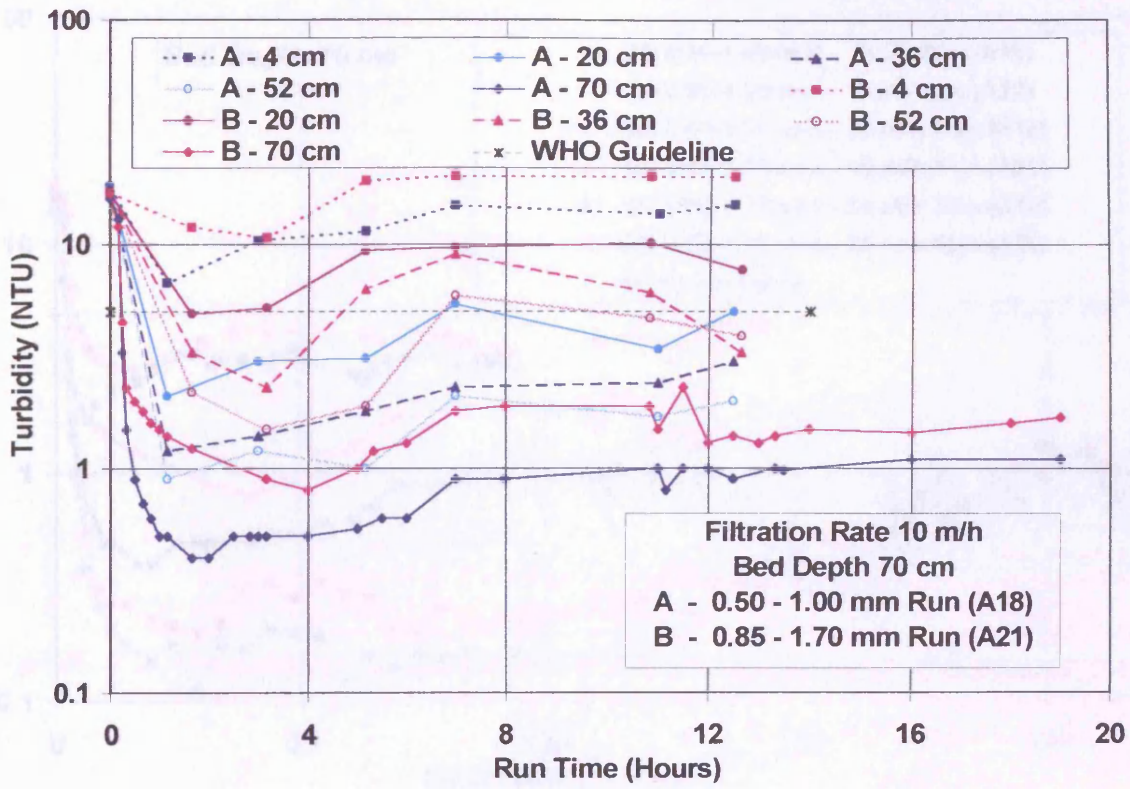

Figure 4.25 Turbidity reduction as a function of bed depth; raw water 20 NTU, bed depth 70 cm

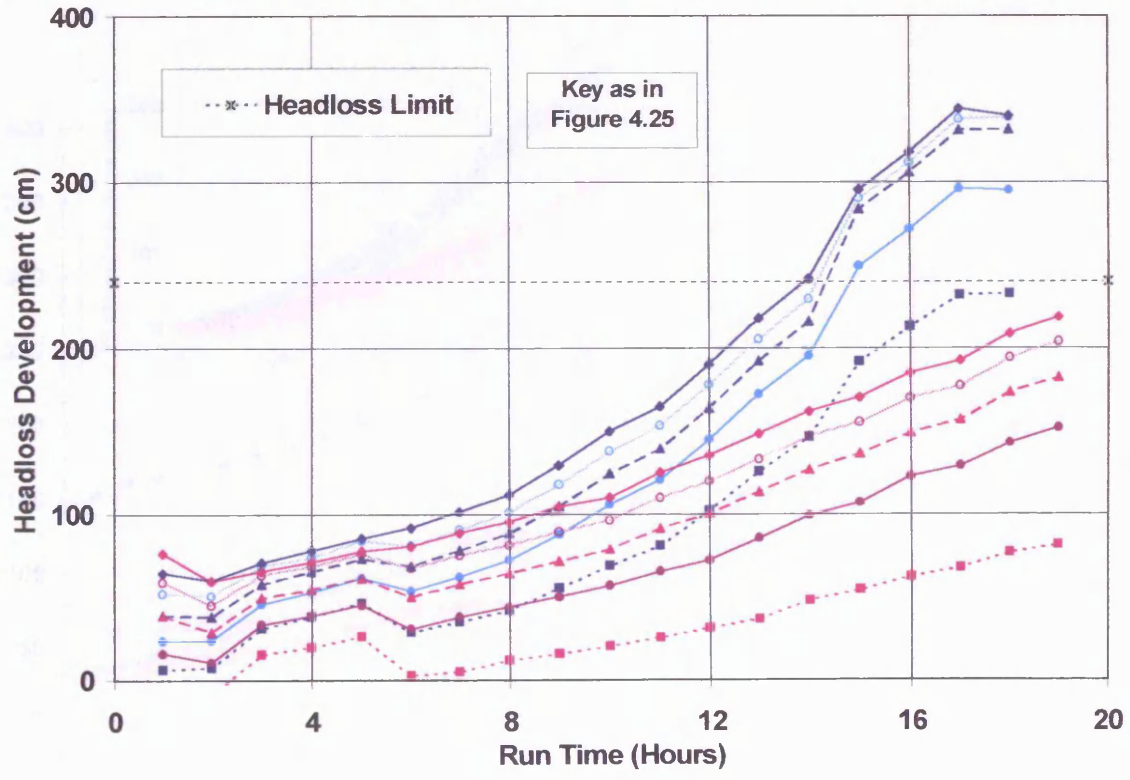

Figure 4.26 Headloss development as a function of bed depth; raw water turbidity 20 NTU, bed depth 70 cm.

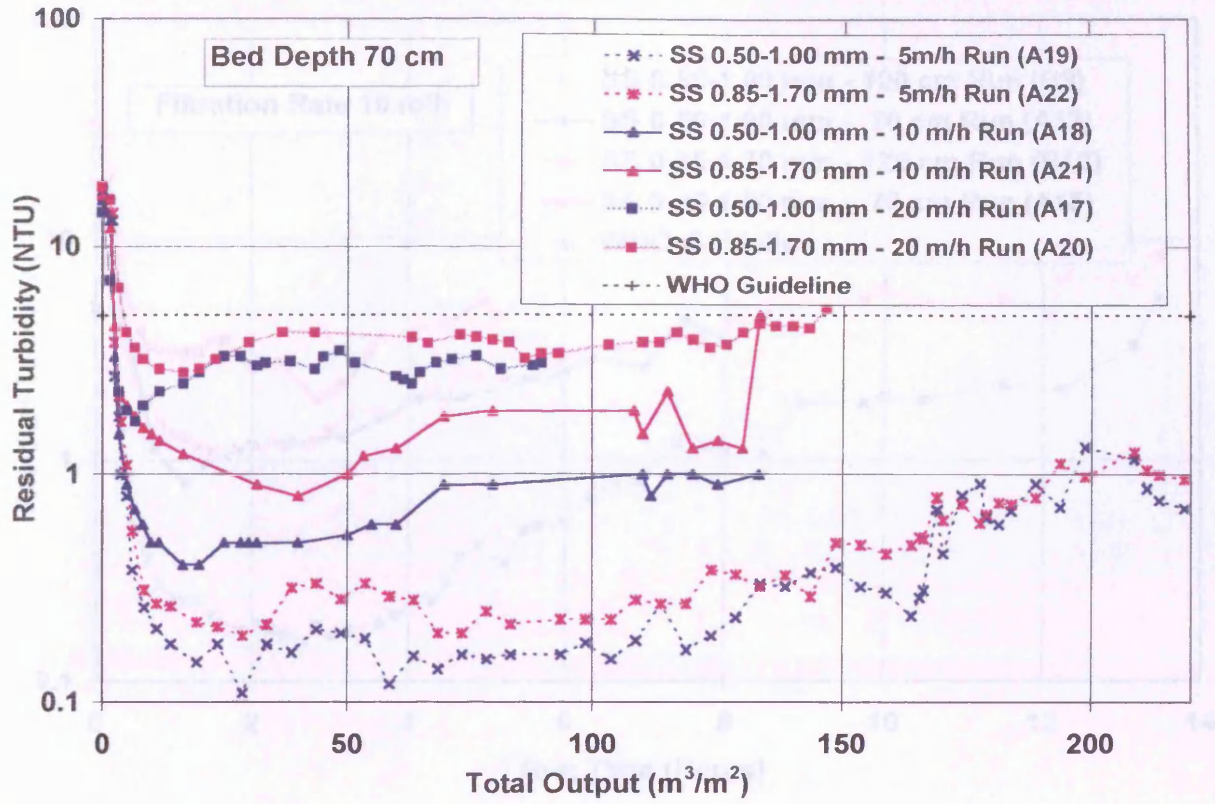

Figure 4.27 Turbidity reduction as a function of media size and filtration rate; raw water turbidity 20 NTU.

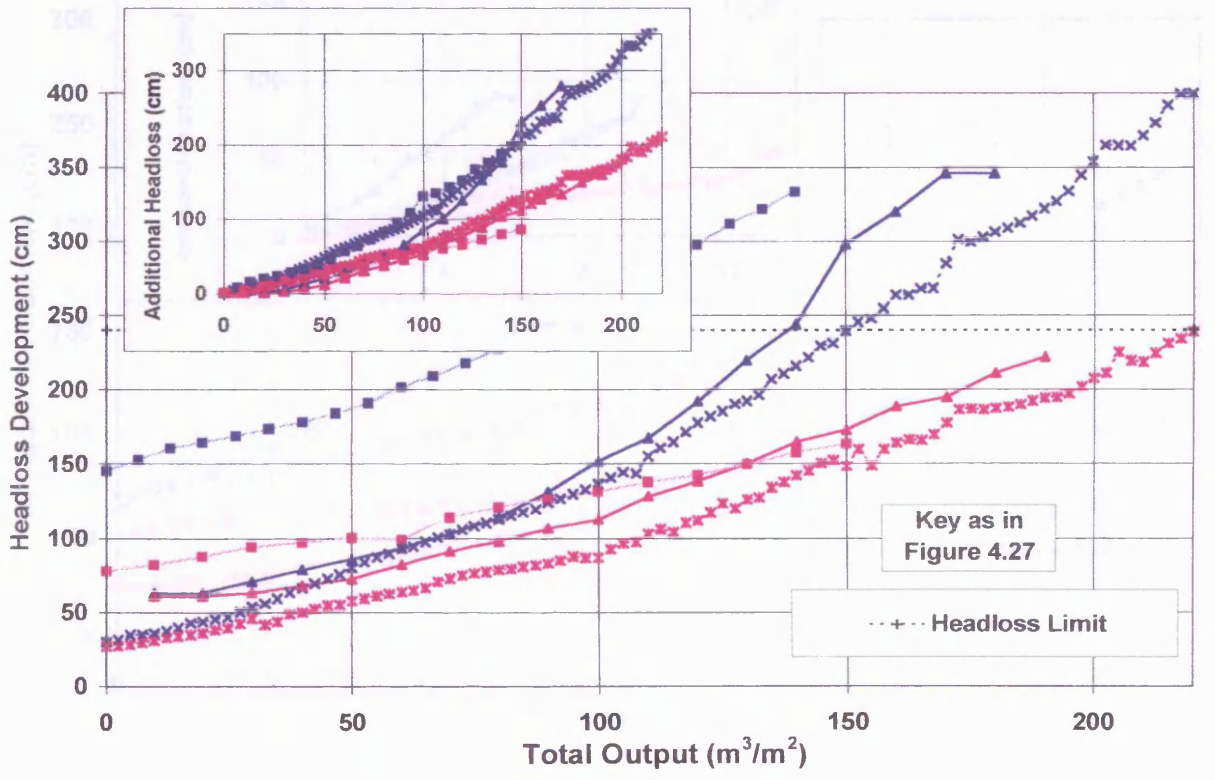

Figure 4.28 Absolute and additional headloss development as a function of media size and filtration rate; raw water turbidity 20 NTU.

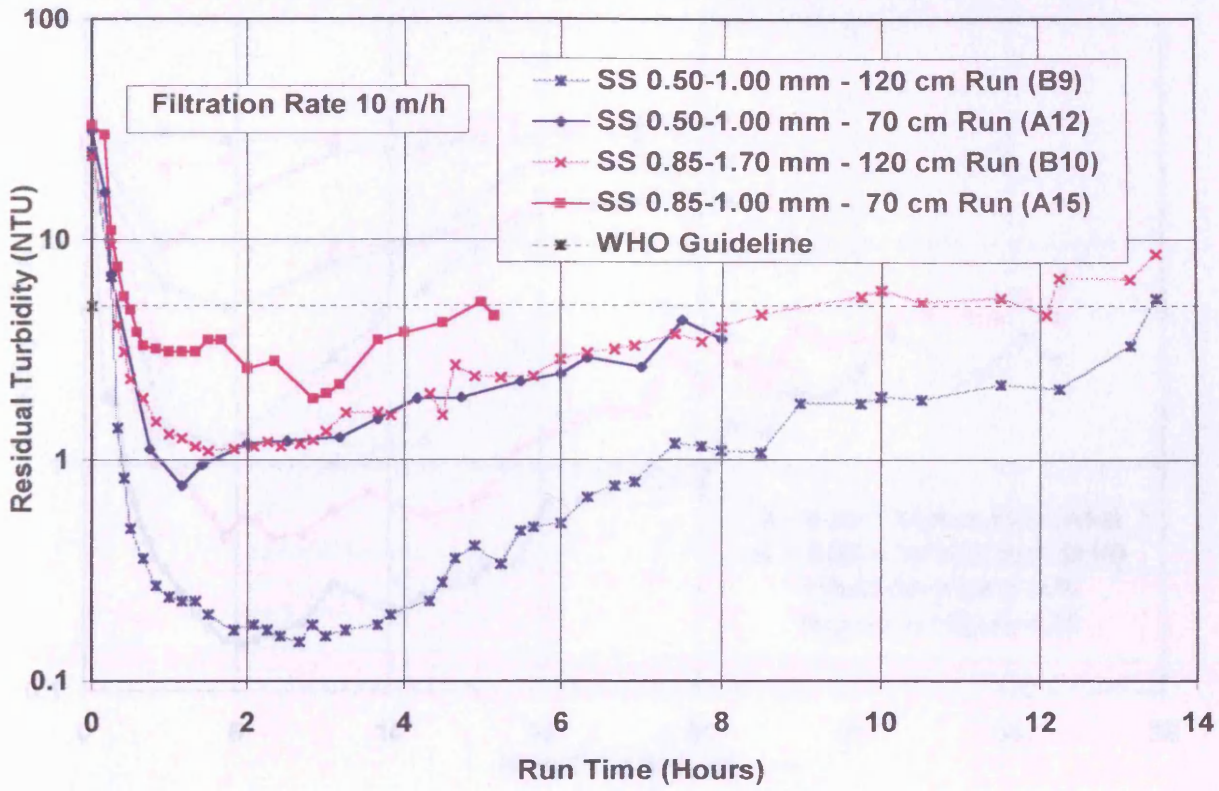

Figure 4.29 Turbidity reduction as a function of media size and bed depth; raw water turbidity 35 NTU.

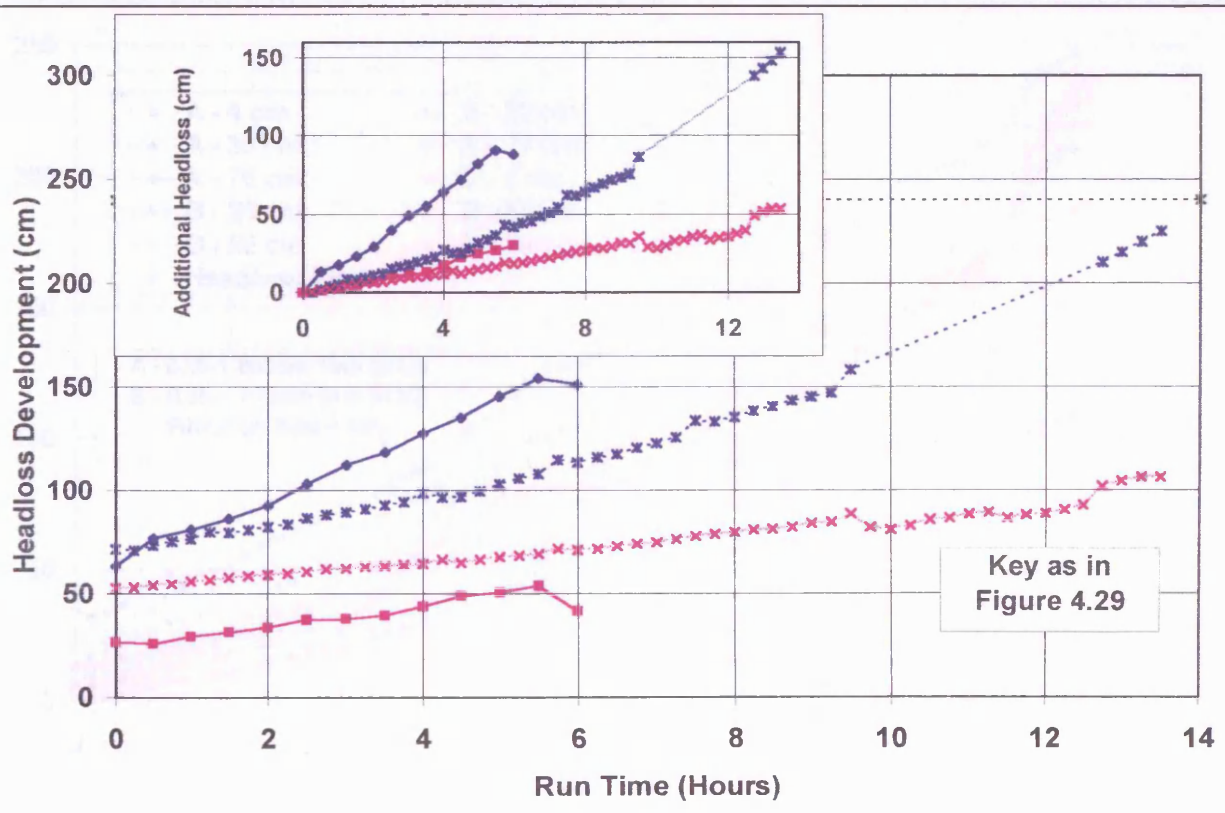

Figure 4.30 Absolute and additional headloss development as a function of media size and bed depth; raw water turbidity 35 NTU.

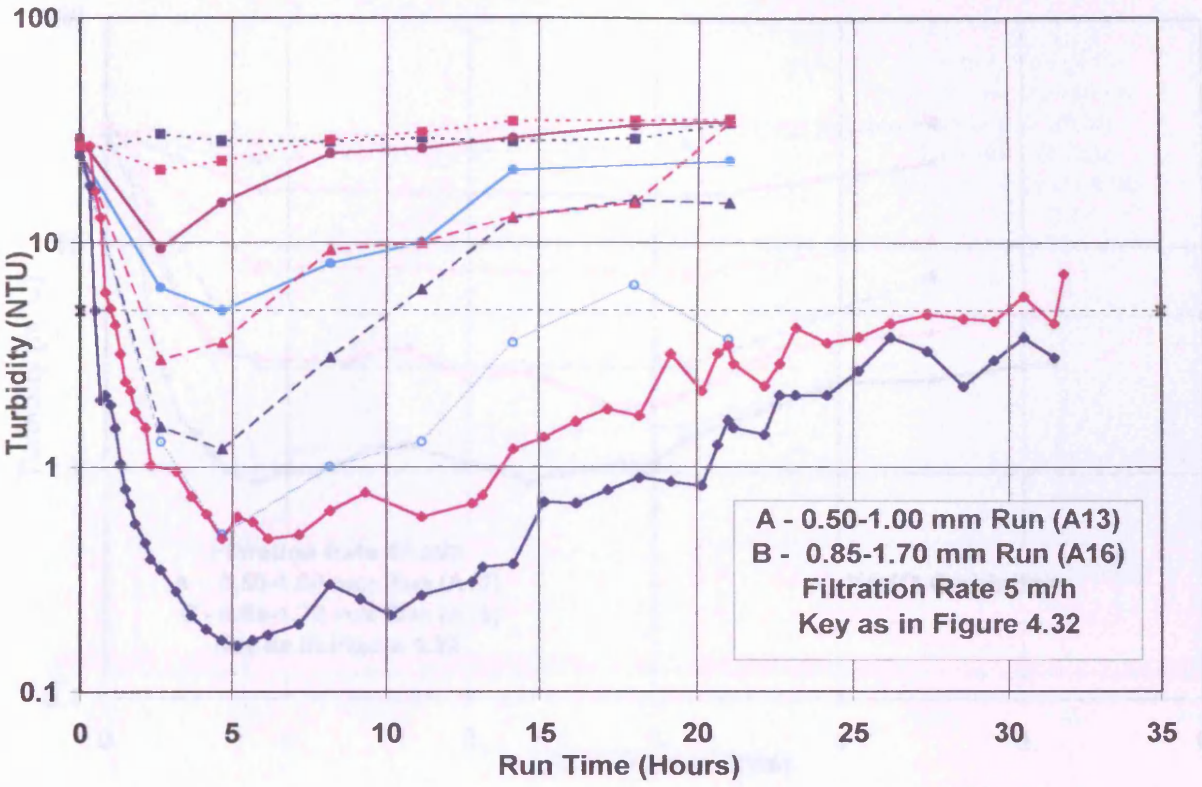

Figure 4.31 Turbidity reduction as a function of depth; raw water turbidity 35 NTU, filtration rate 5 m/h.

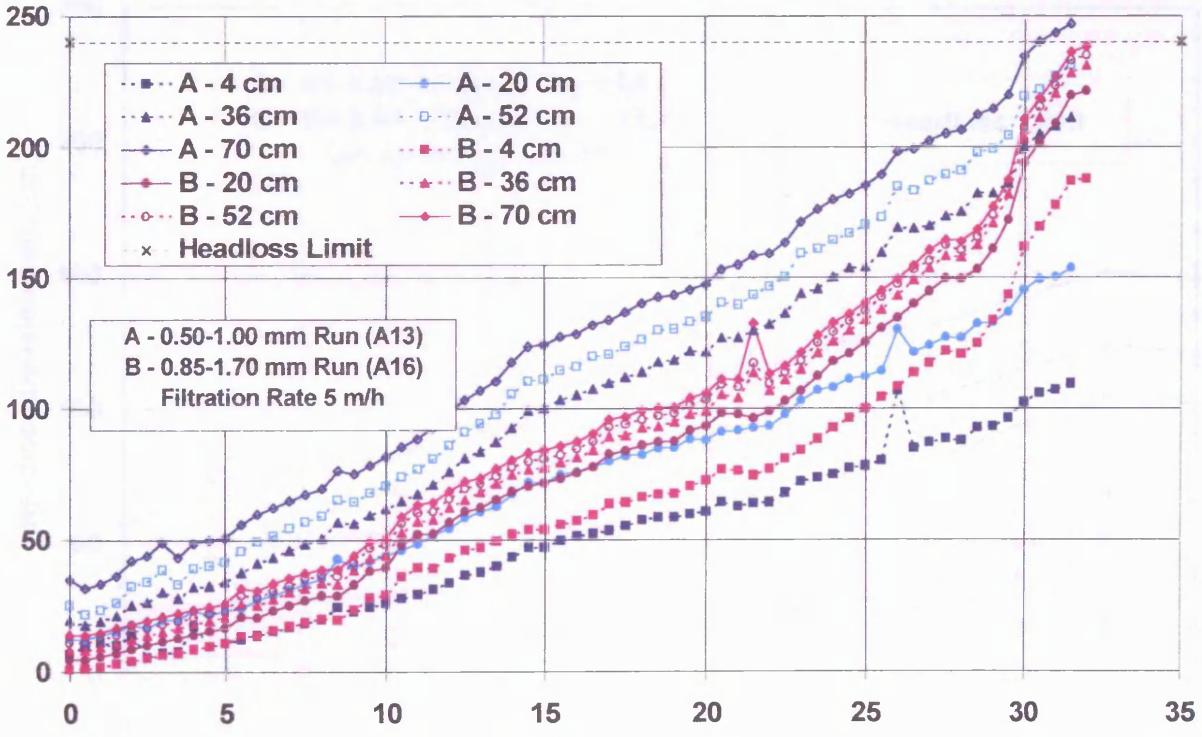

Figure 4.32 Headloss development as a function of depth; raw water turbidity 35 NTU, filtration rate 5 m/h.

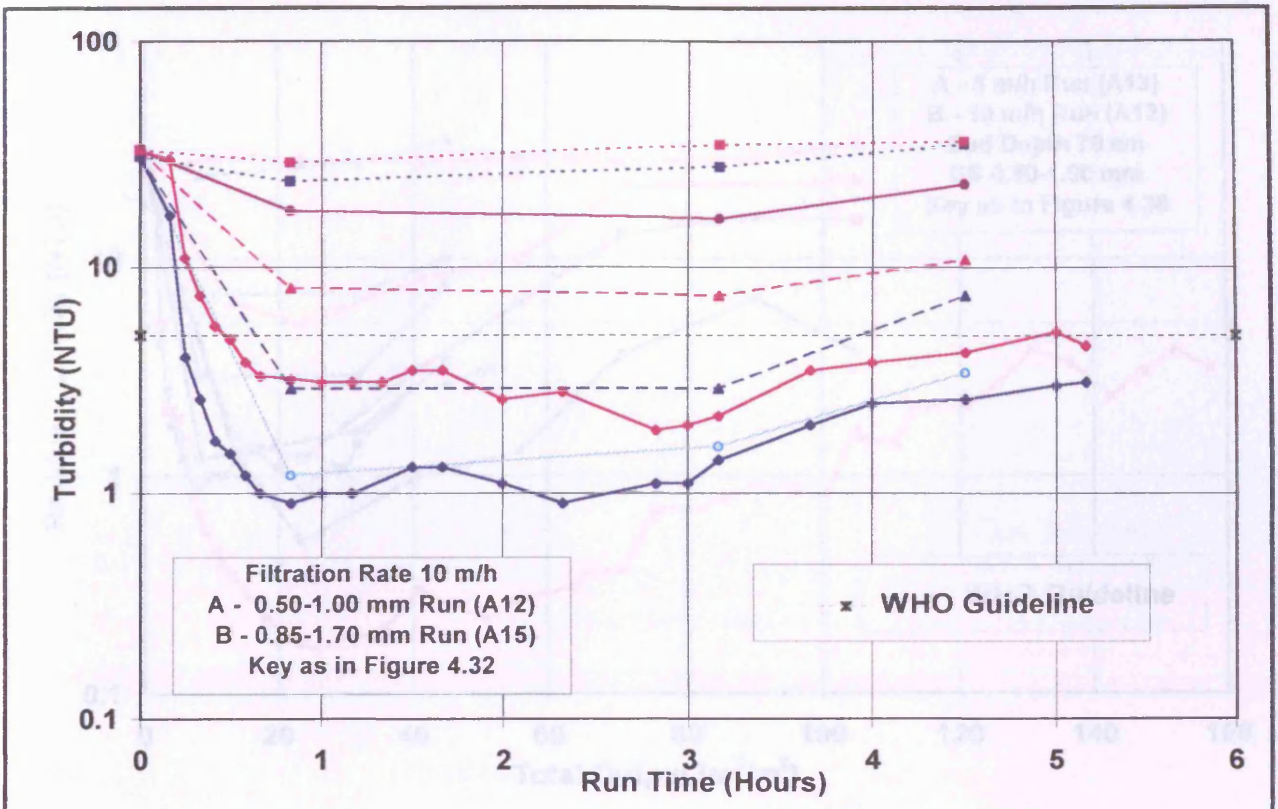

Figure 4.33 Turbidity reduction as a function of depth; raw water turbidity 35 NTU, filtration rate 10 m/h.

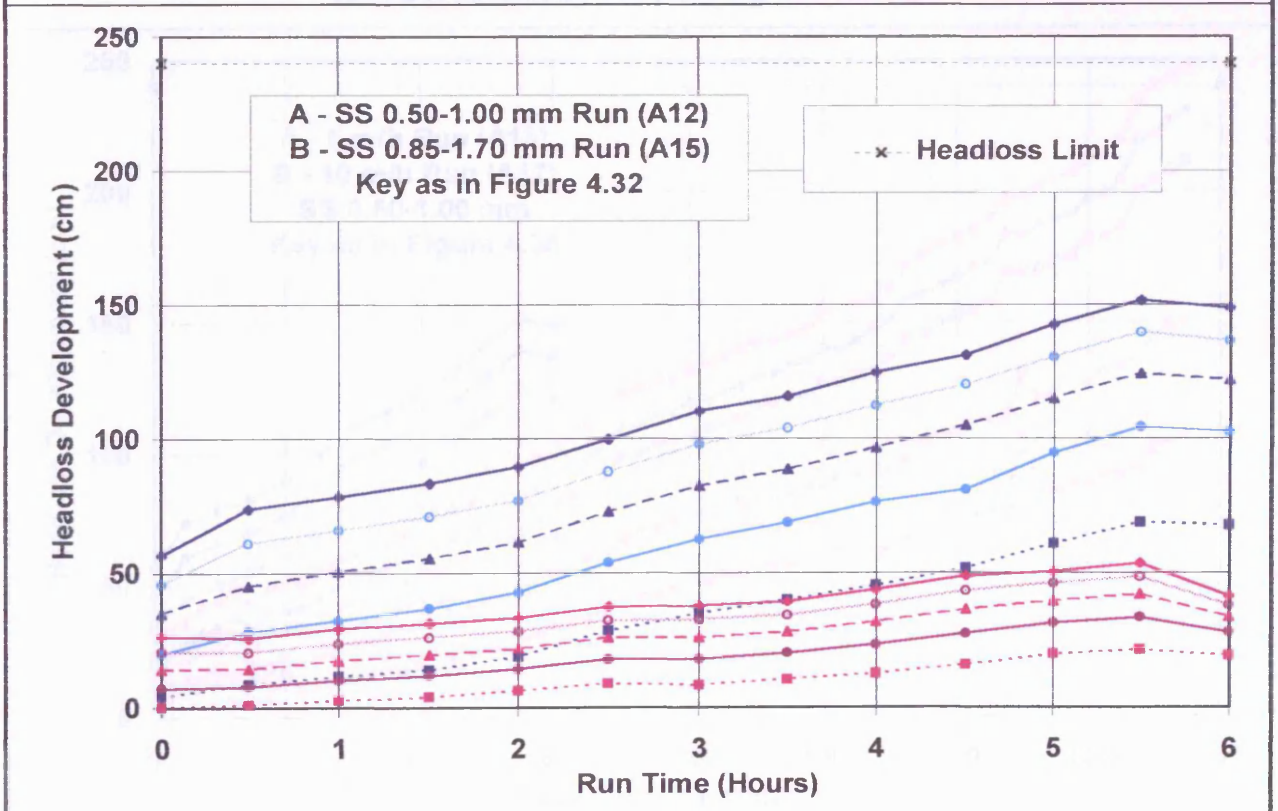

Figure 4.34 Headloss development as a function of depth; raw water turbidity 35 NTU, filtration rate 10 m/h.

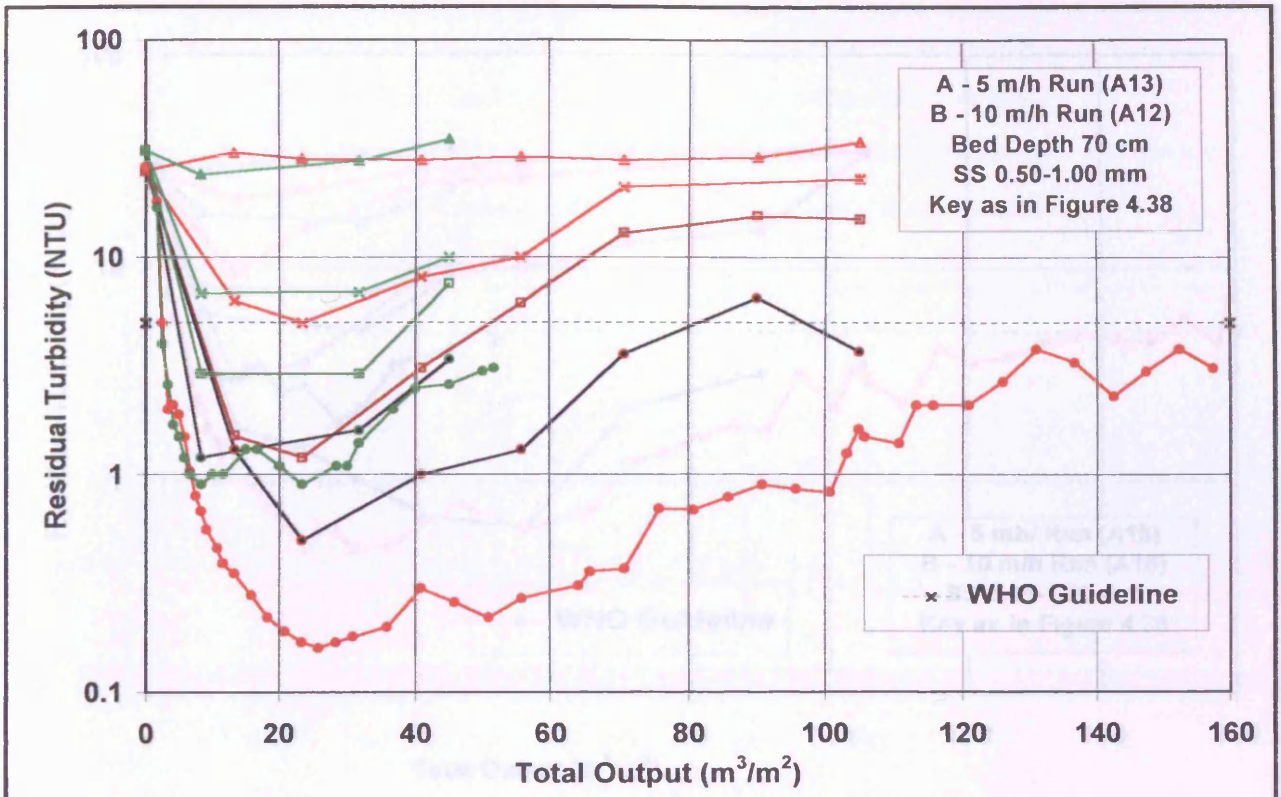

Figure 4.35 Turbidity reduction as a function of depth & filtration rate; raw water turbidity 35 NTU, sand size 0.50-1.00 mm.

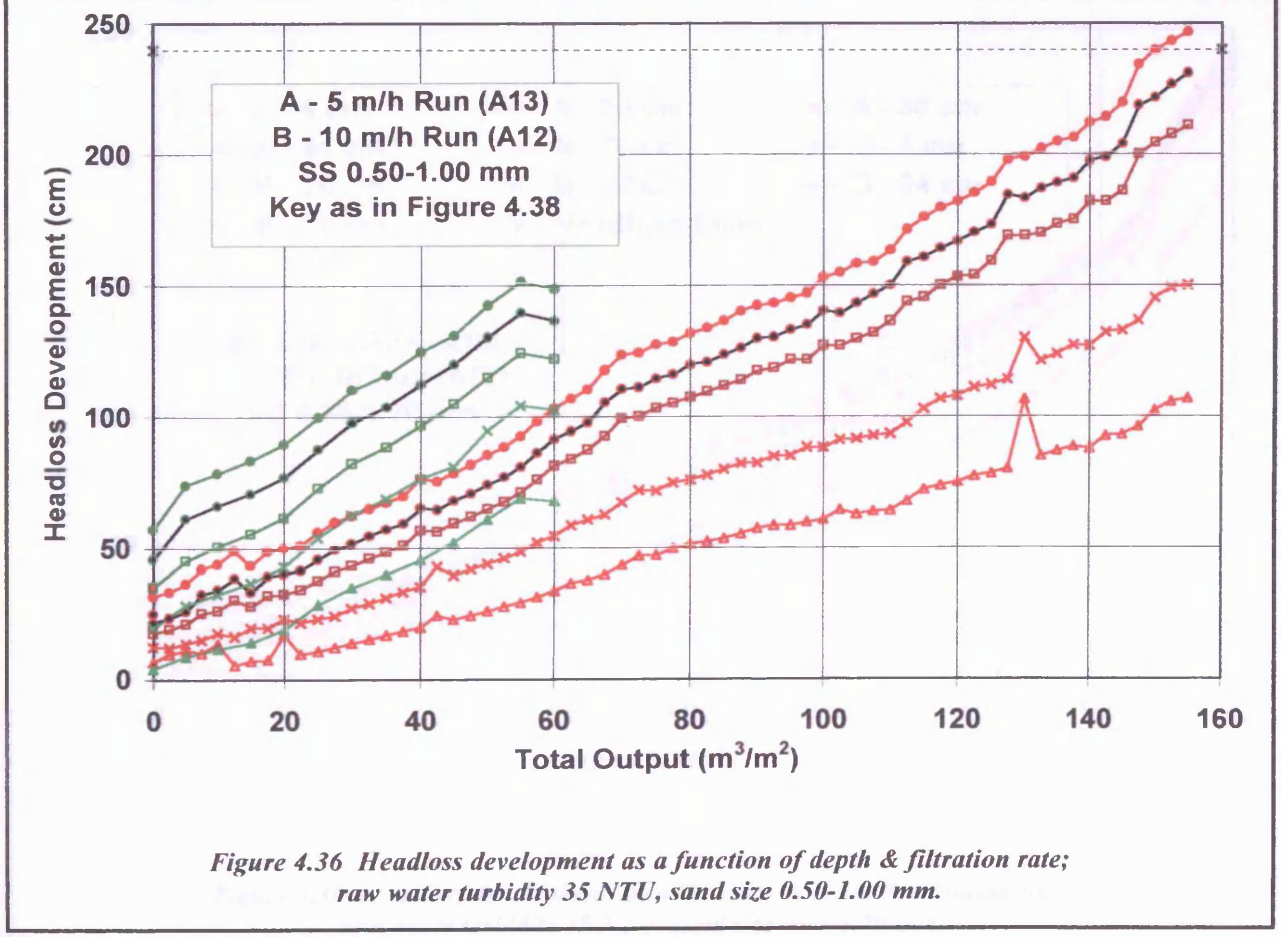

Figure 4.36 Headloss development as a function of depth & filtration rate; raw water turbidity 35 NTU, sand size 0.50-1.00 mm.

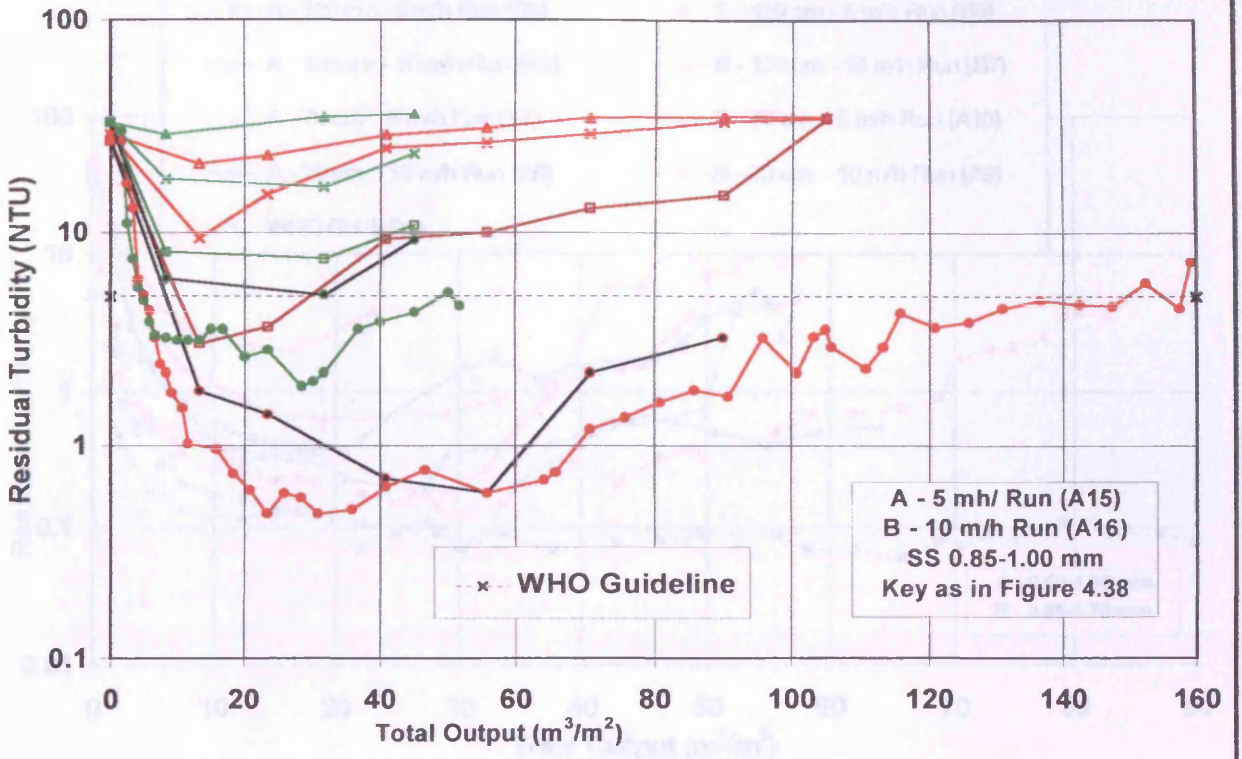

Figure 4.37 Turbidity reduction as a function of depth & filtration rate; raw water turbidity 35 NTU, sand size 0.85-1.70 mm.

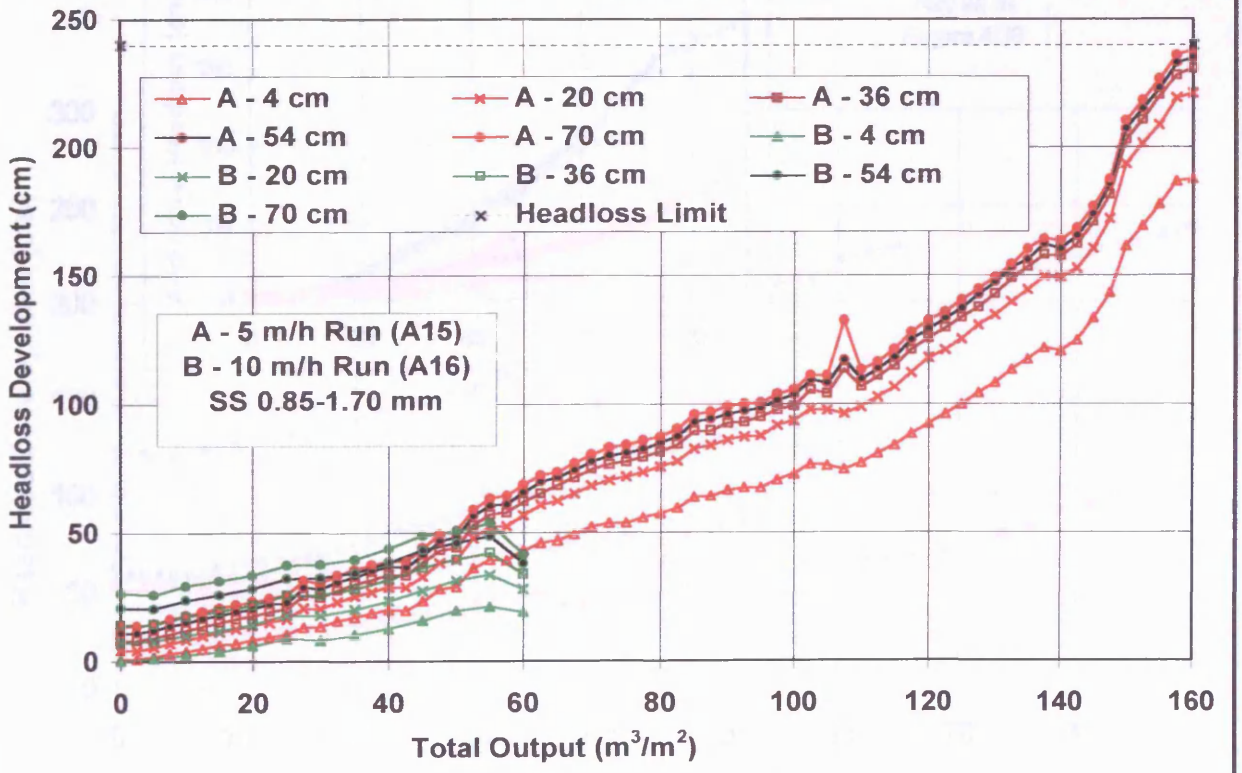

Figure 4.38 Headloss development as a function of depth & filtration rate; raw water turbidity 35 NTU, sand size 0.85-1.70 mm.

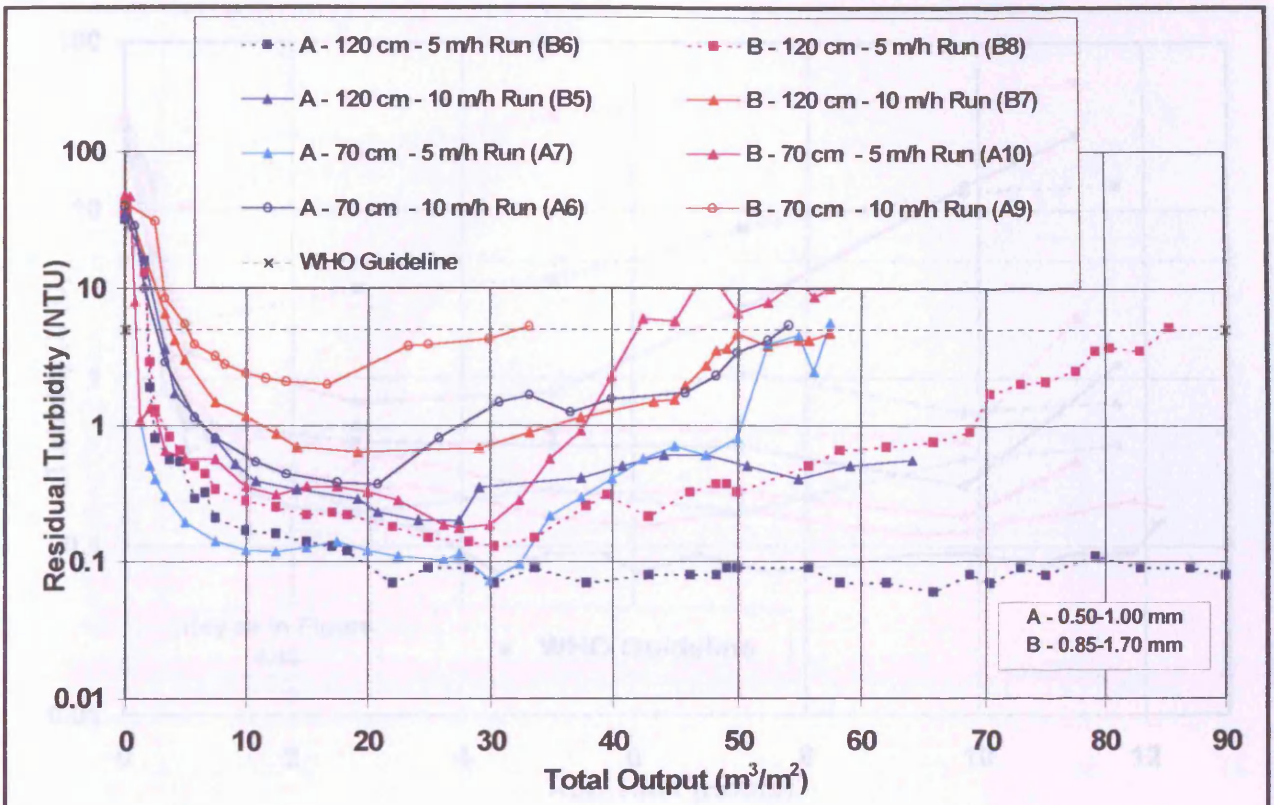

Figure 4.39 Turbidity reduction as a function of media size, bed depth & filtration rate; raw water turbidity 50 NTU.

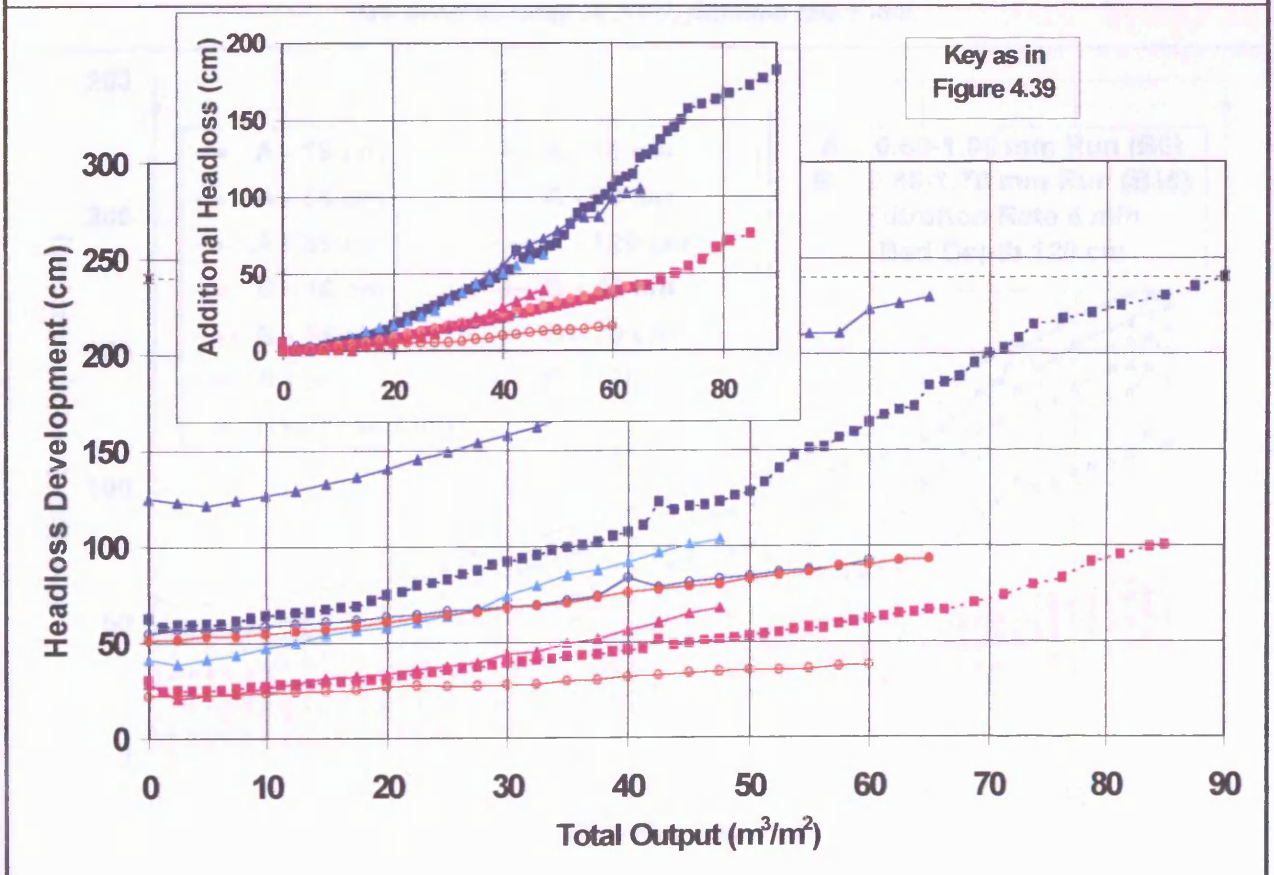

Figure 4.40 Absolute and additional headloss development as a function of media size bed depth & filtration rate; raw water turbidity 50 NTU.

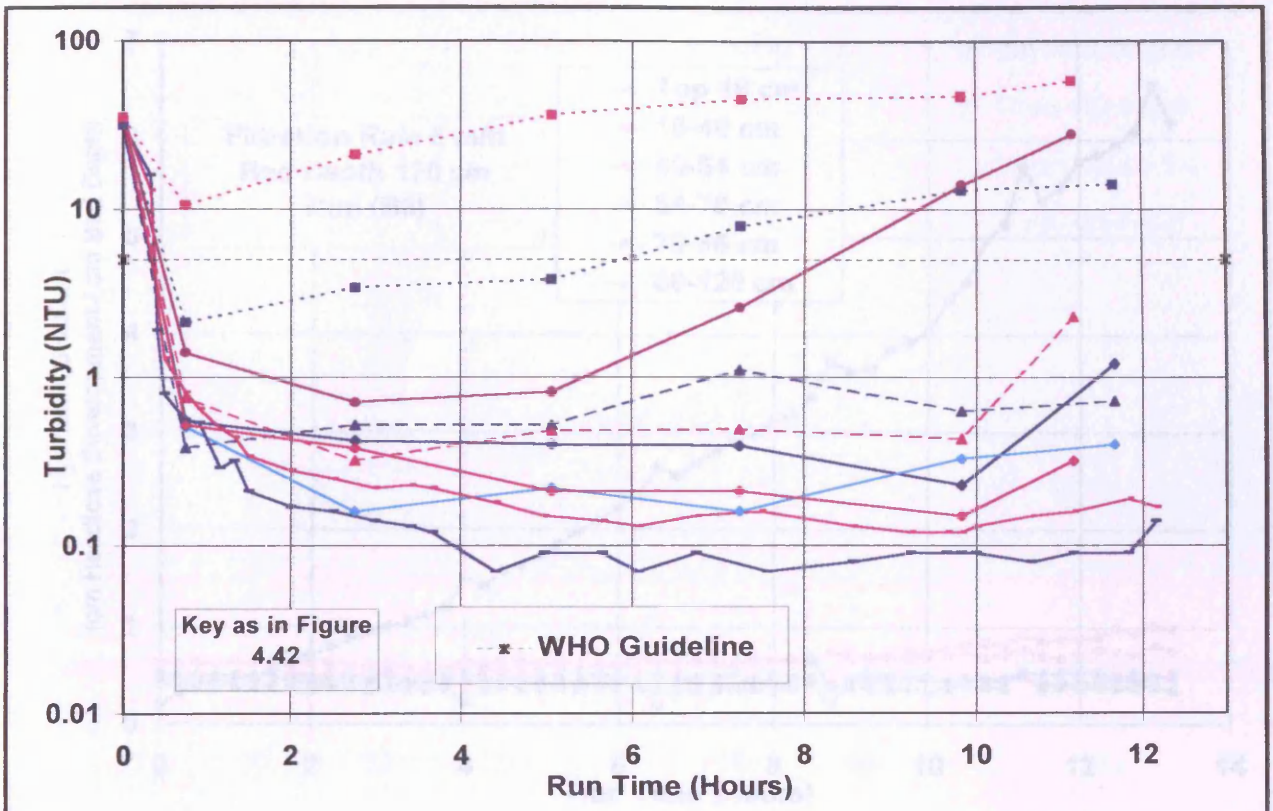

Figure 4.41 Turbidity reduction as a function of depth; raw water turbidity 50 NTU, filtration rate 5 m/h.

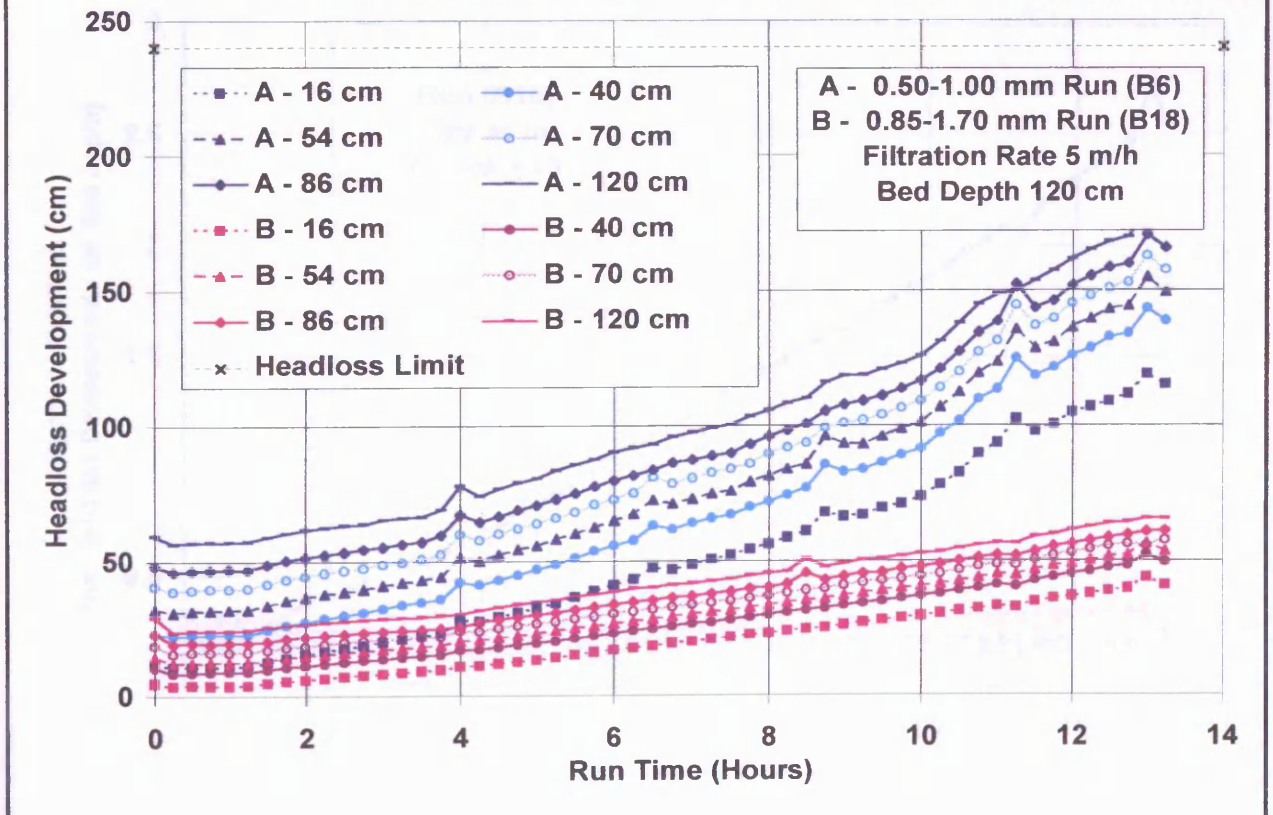

Figure 4.42 Headloss development as a function of depth; raw water turbidity 50 NTU, filtration rate 5 m/h.

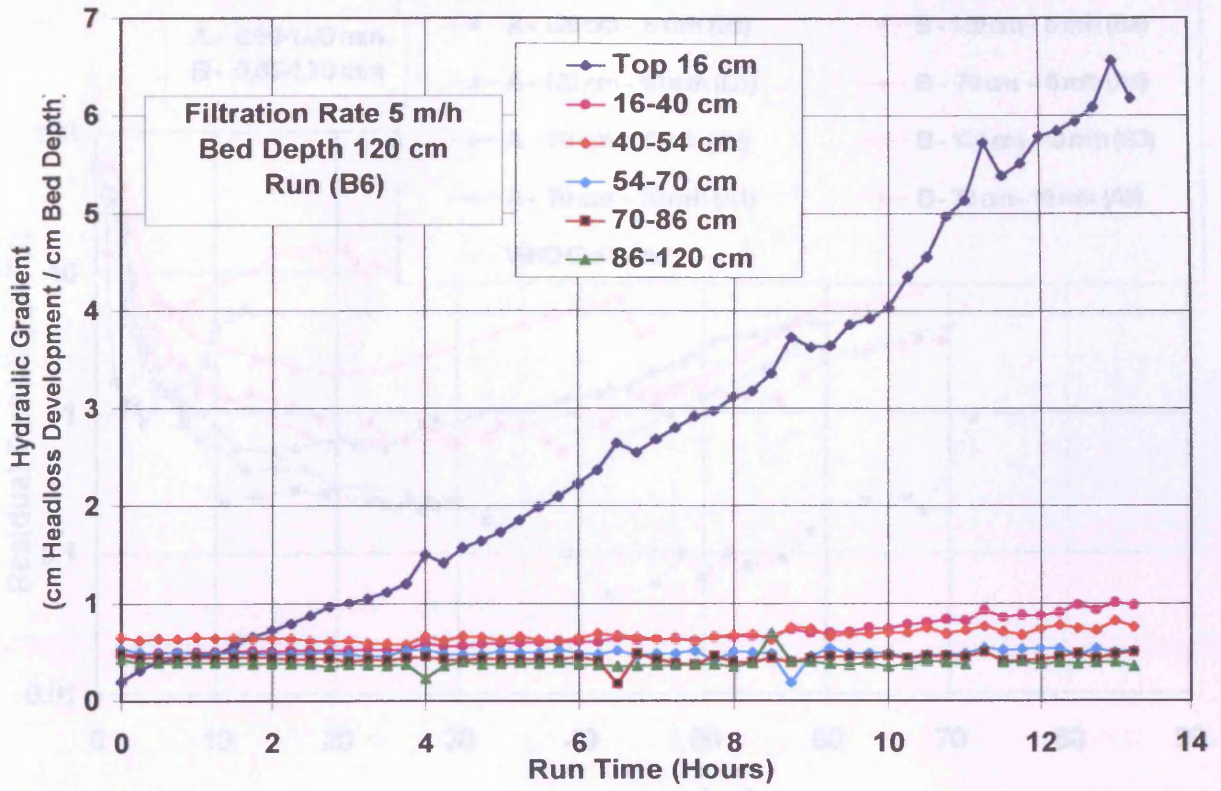

Figure 4.43 Hydraulic gradient as a function of depth; raw water turbidity 50 NTU, sand size 0.50-1.00 mm.

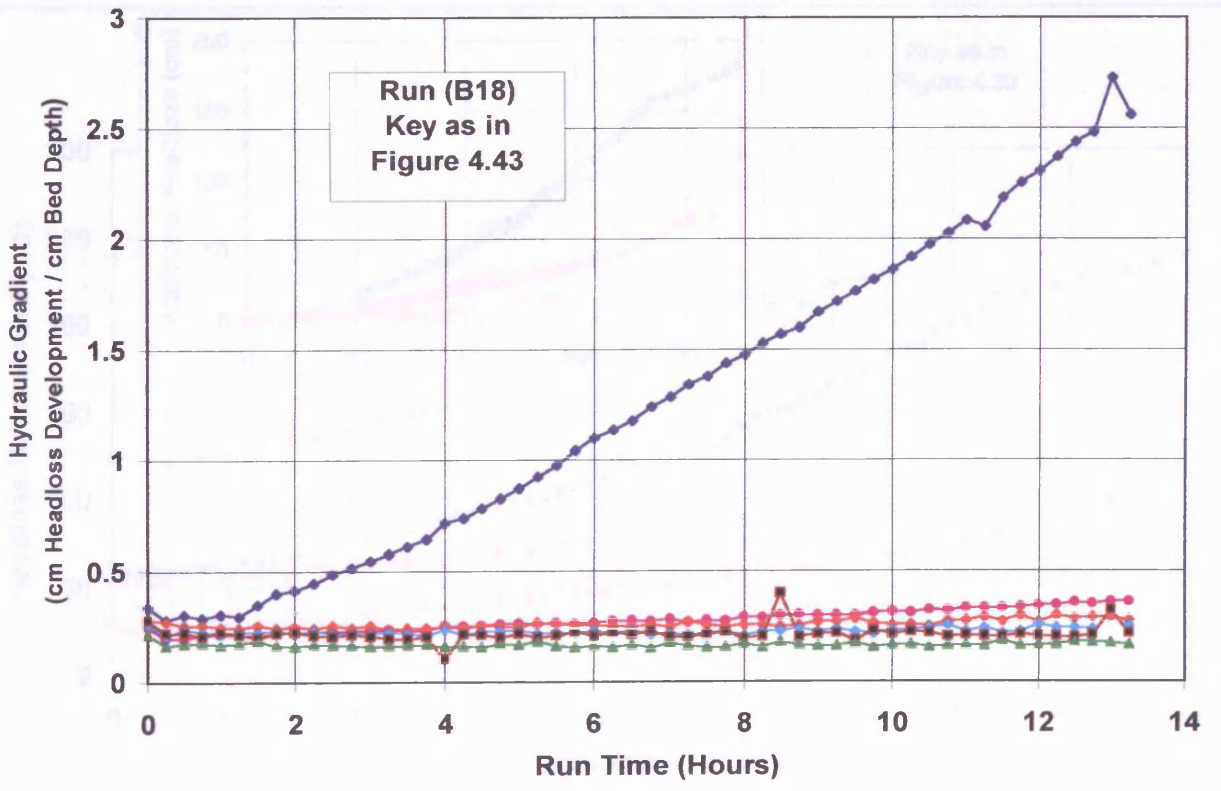

Figure 4.44 Hydraulic gradient as a function of depth; raw water turbidity 50 NTU, sand size 0.85-1.70 mm.

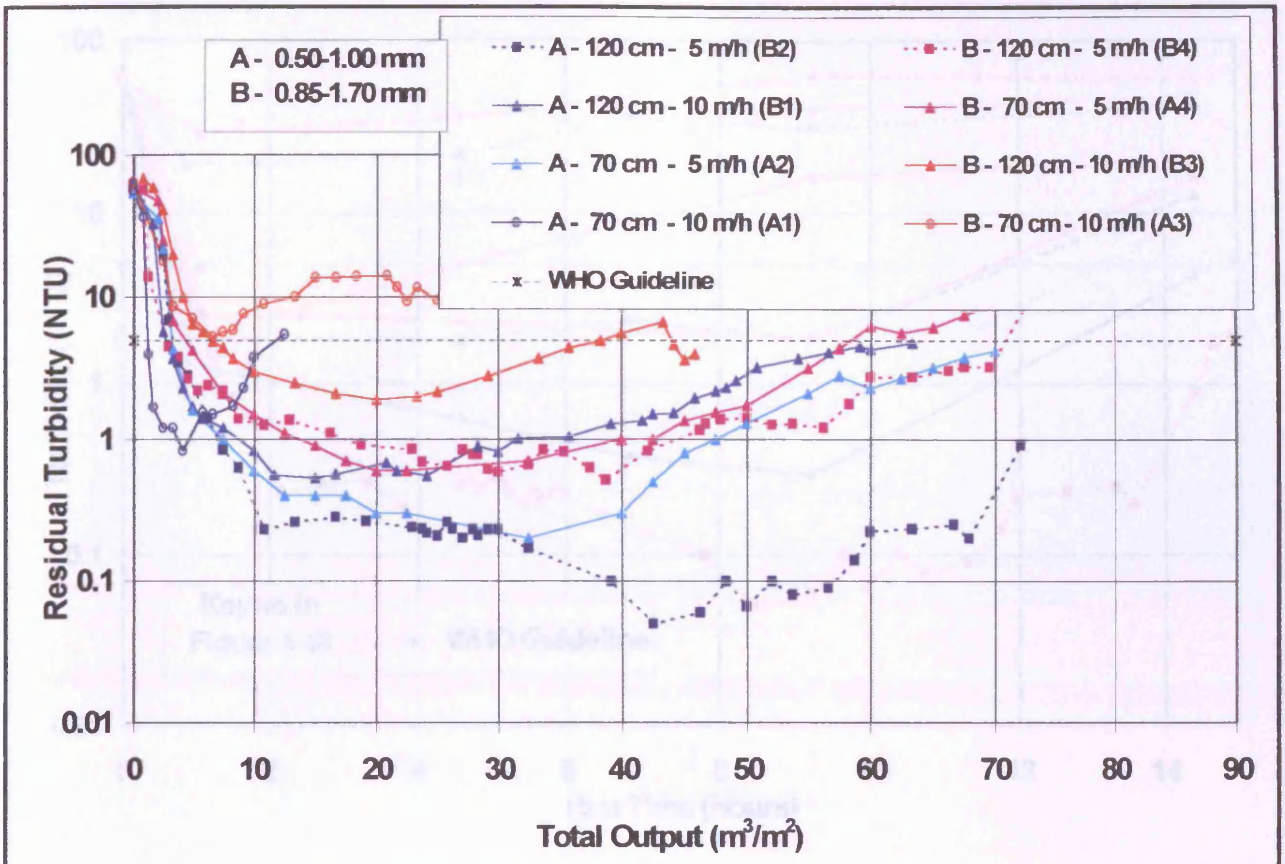

Figure 4.45 Turbidity reduction as a function of media size, bed depth & filtration rate; raw water turbidity 75 NTU.

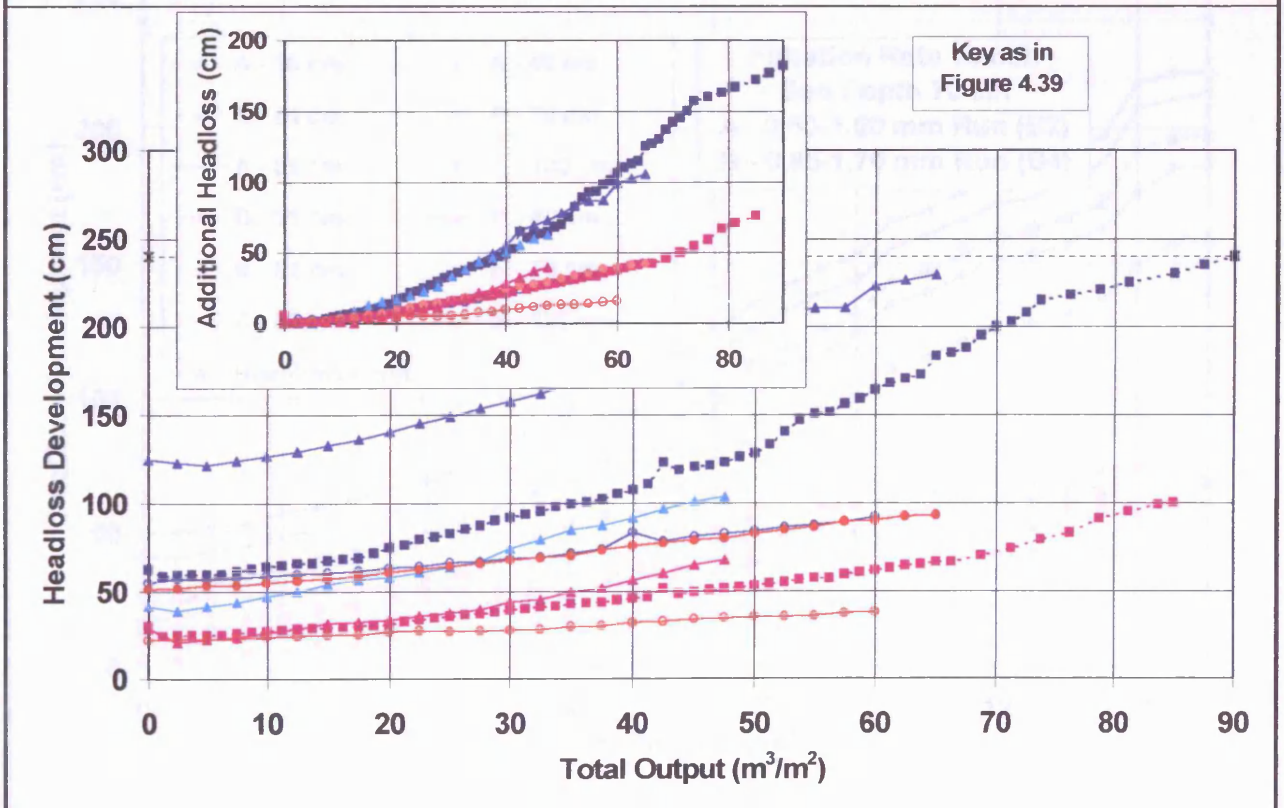

Figure 4.46 Absolute and additional headloss development as a function of media size, bed depth & filtration rate; raw water turbidity 75 NTU.

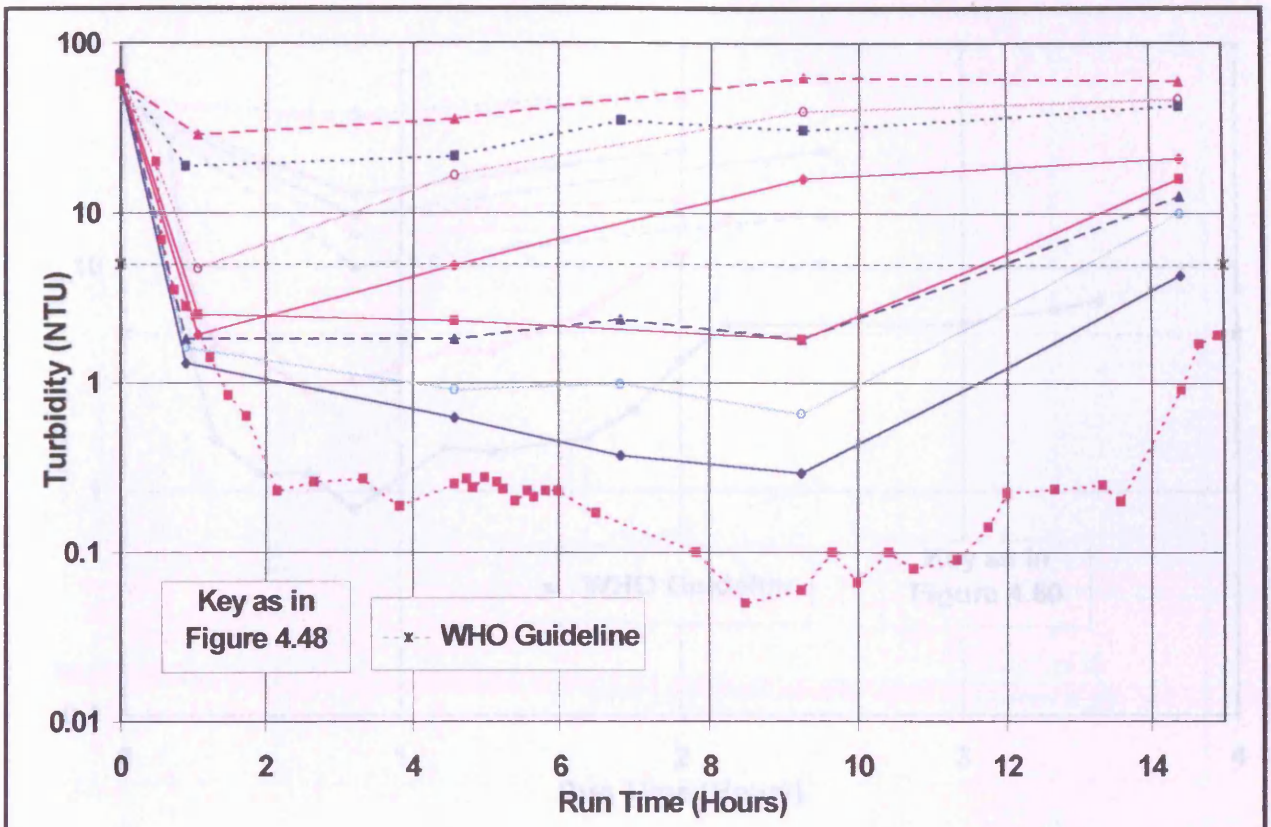

Figure 4.47 Turbidity reduction as a function of depth; raw water turbidity 75 NTU, filtration rate 5m/h, bed depth 120 cm.

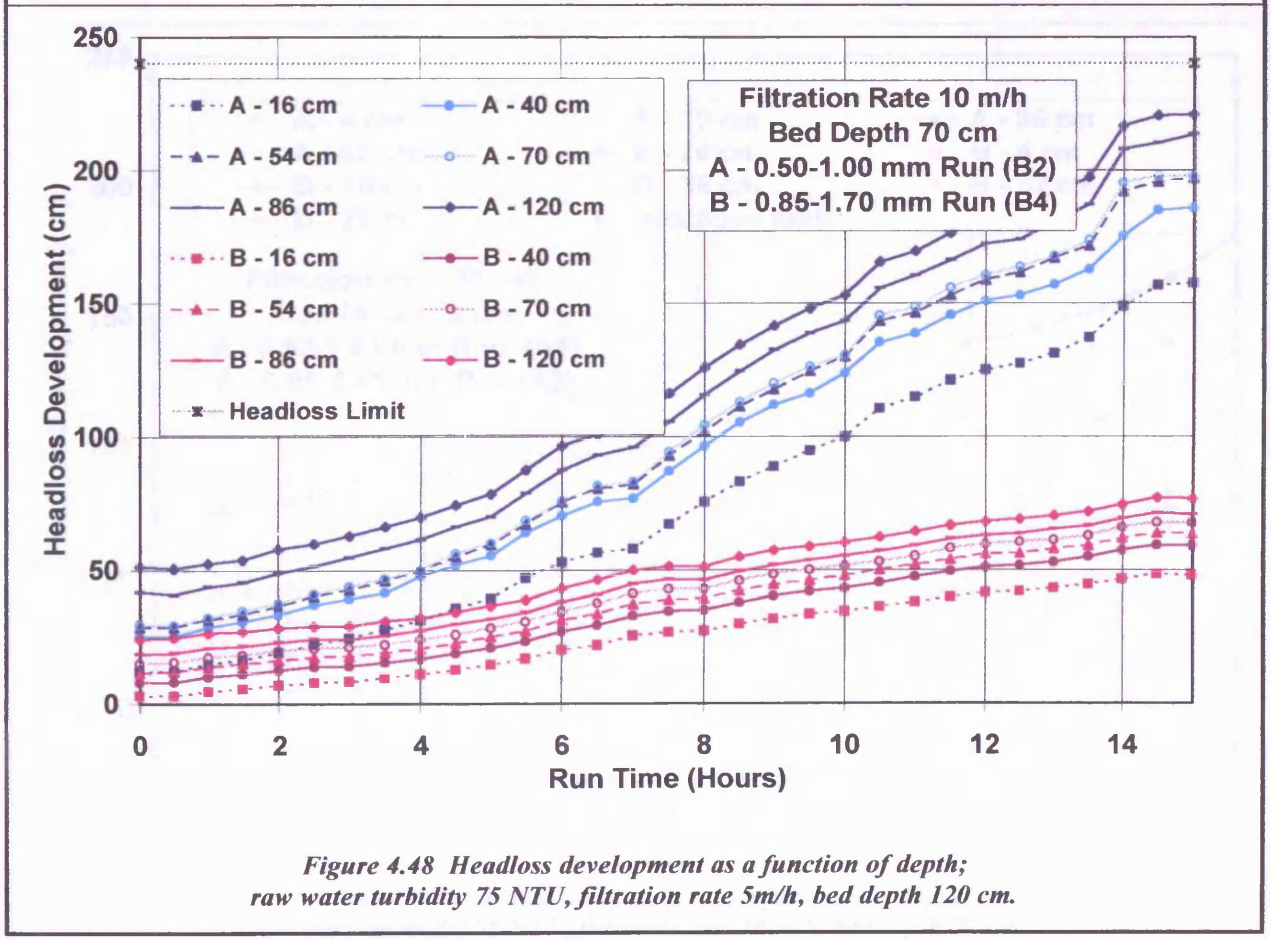

Figure 4.48 Headloss development as a function of depth; raw water turbidity 75 NTU, filtration rate 5m/h, bed depth 120 cm.

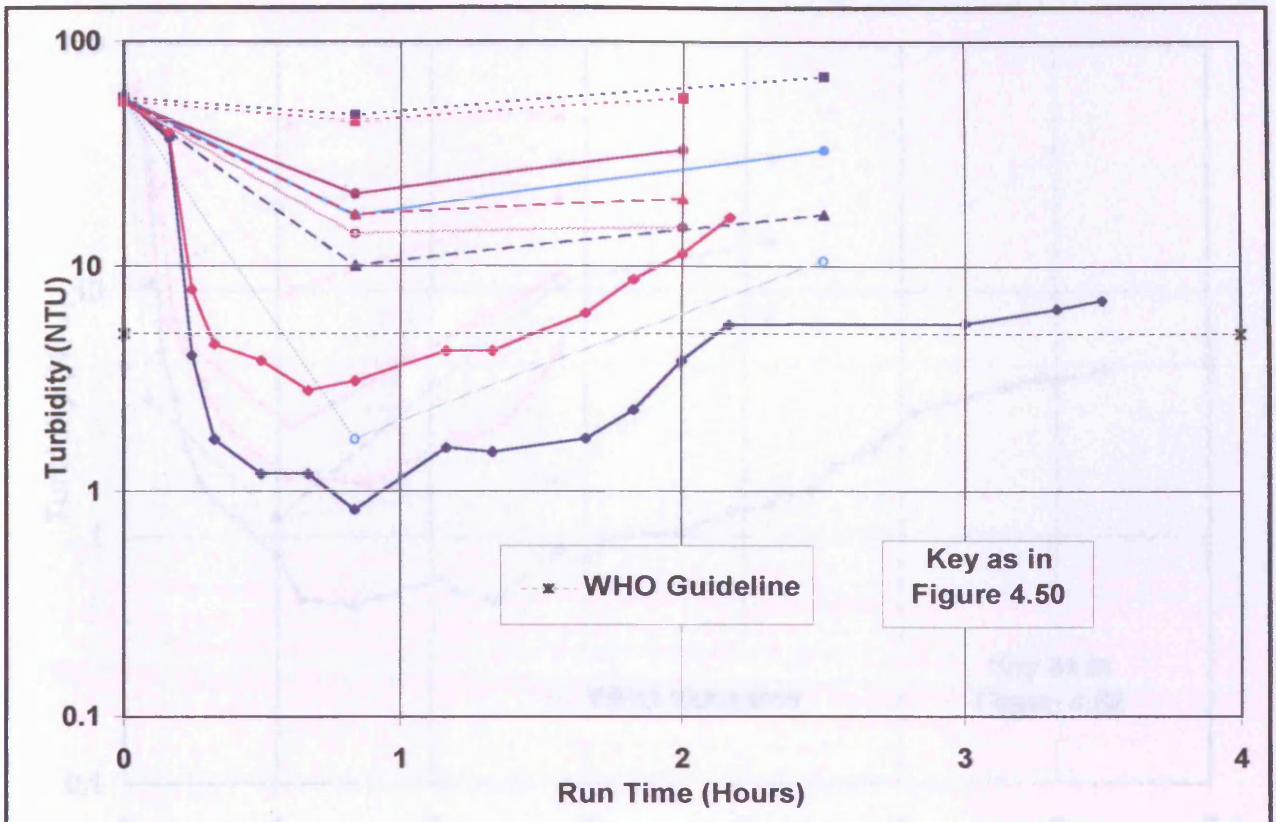

Figure 4.49 Turbidity reduction as a function of depth;
raw water turbidity 75 NTU, filtration rate 10 m/h, bed depth 70 cm.

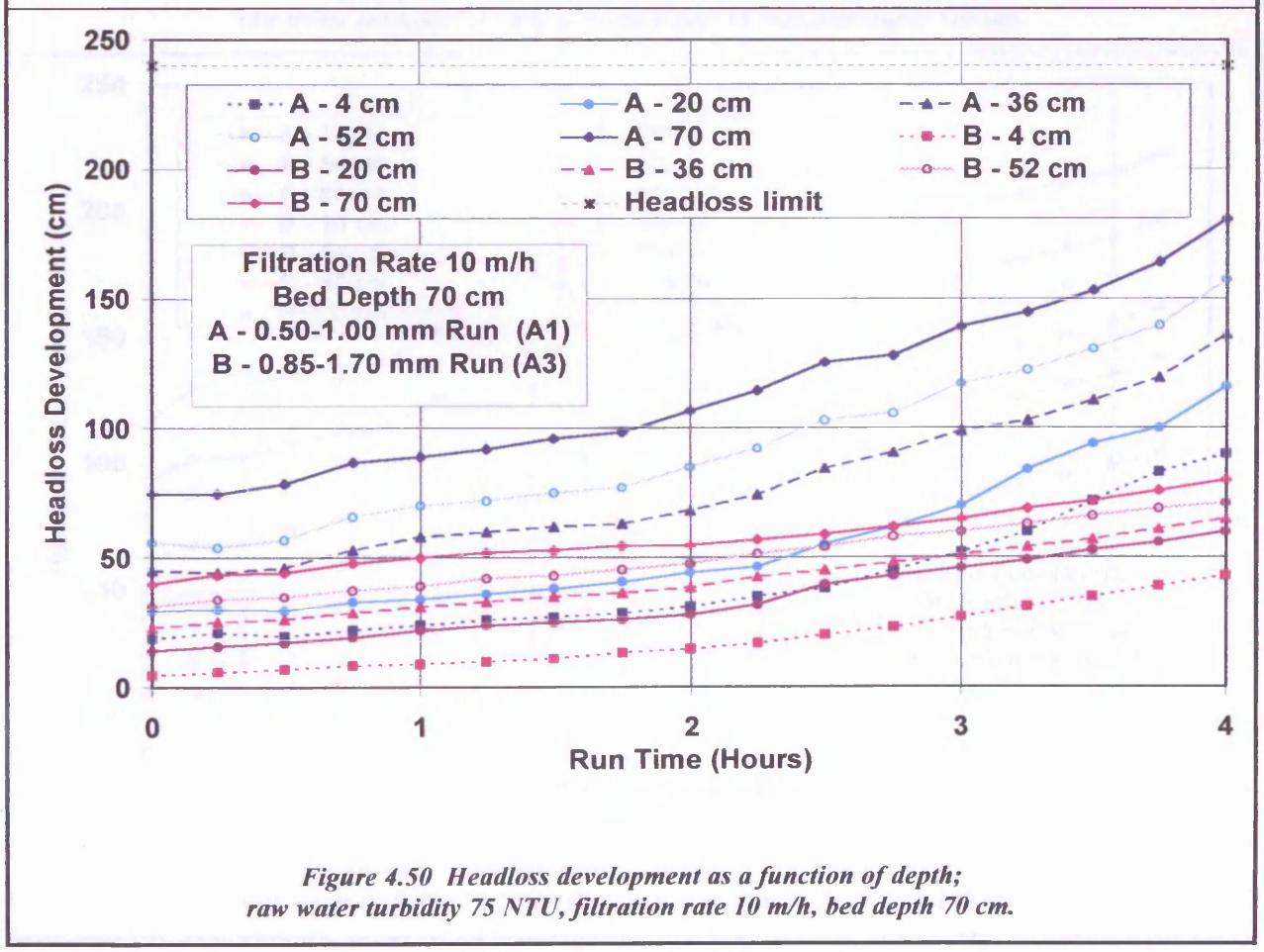

Figure 4.50 Headloss development as a function of depth;
raw water turbidity 75 NTU, filtration rate 10 m/h, bed depth 70 cm.

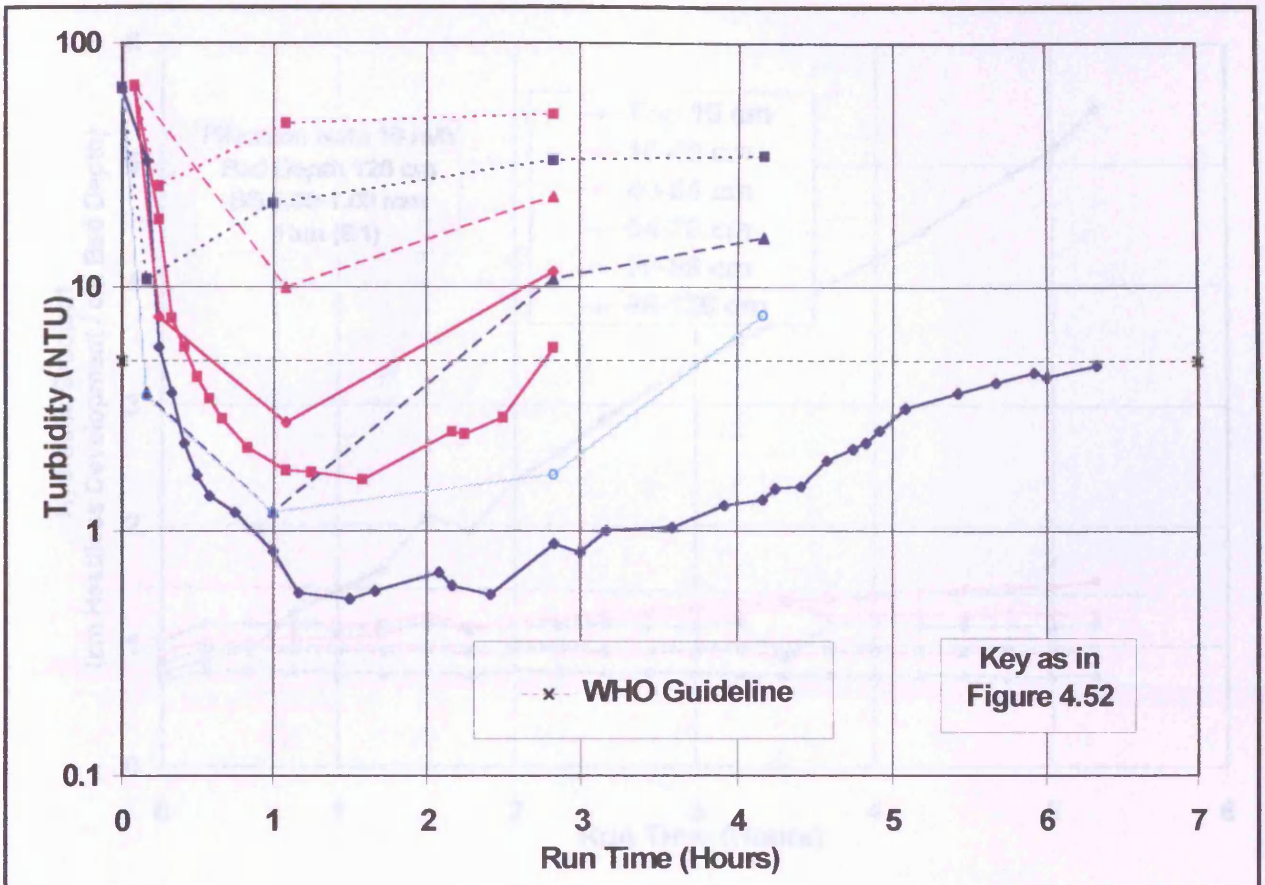

Figure 4.51 Turbidity reduction as a function of depth;
raw water turbidity 75 NTU, filtration rate 10 m/h, bed depth 120 cm.

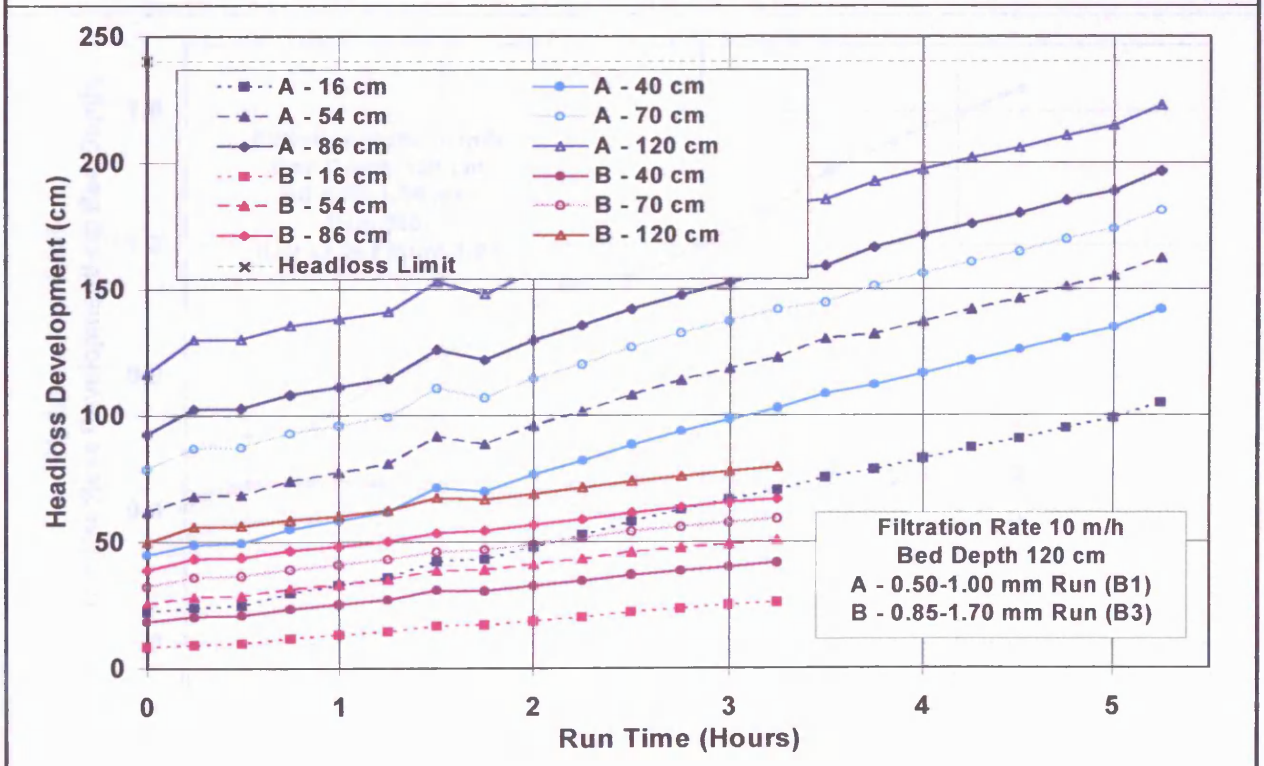

Figure 4.52 Headloss development as a function of depth;
raw water turbidity 75 NTU, filtration rate 10 m/h, bed depth 120 cm.

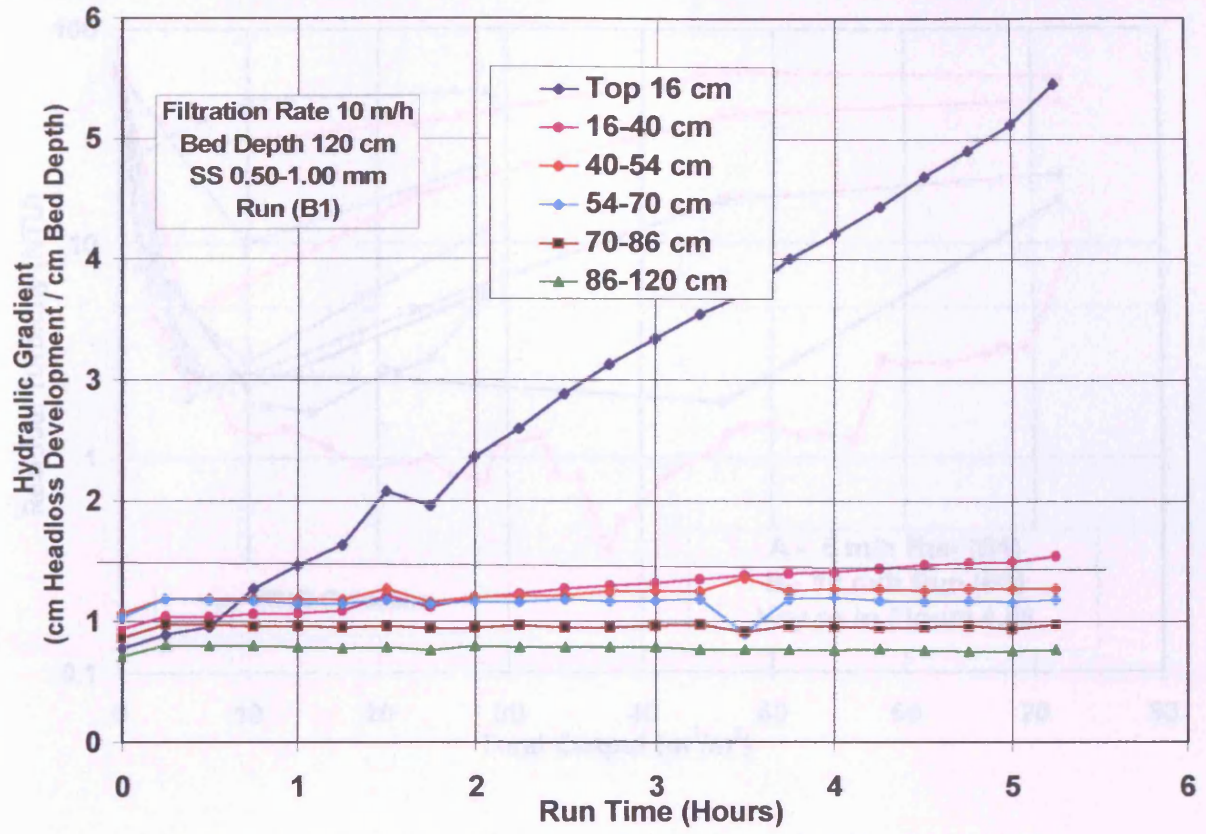

Figure 4.53 Hydraulic gradient as a function of depth; raw water turbidity 75 NTU, sand size 0.50-1.00 mm.

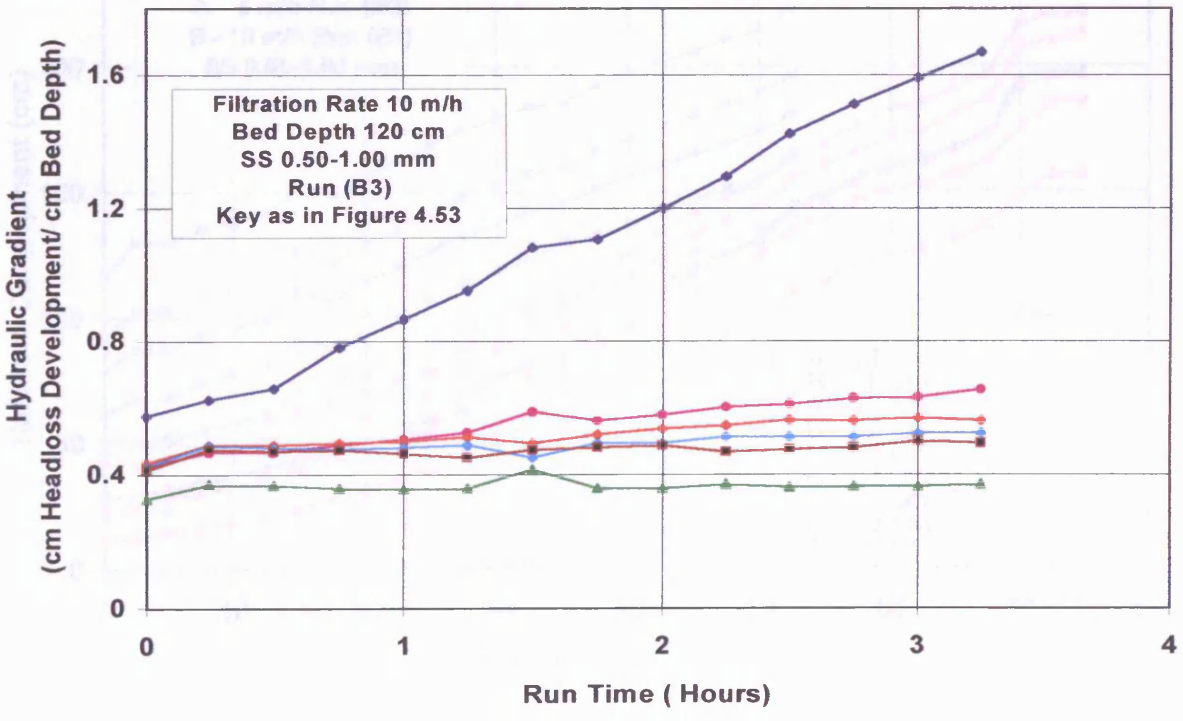

Figure 4.54 Hydraulic gradient as a function of depth; raw water turbidity 75 NTU, sand size 0.85-1.70 mm.

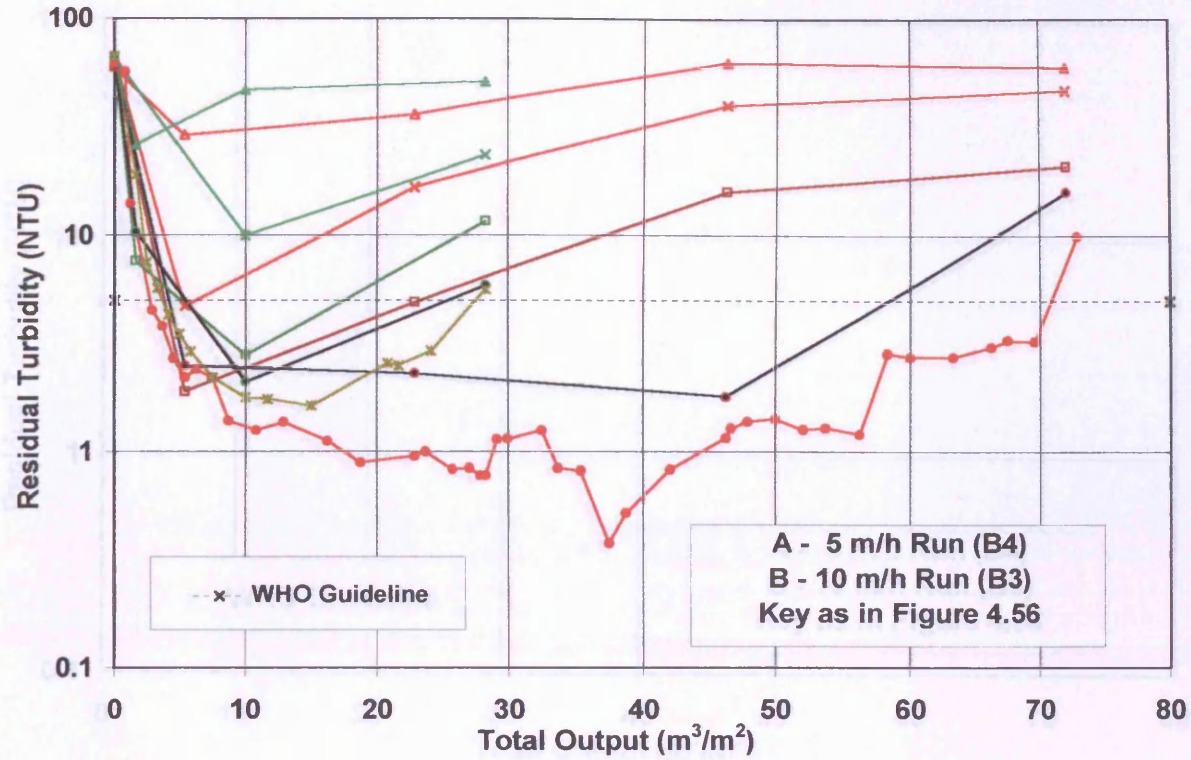

Figure 4.55 Turbidity reduction as a function of depth & filtration rate; raw water turbidity 75 NTU, sand size 0.50-1.00 mm.

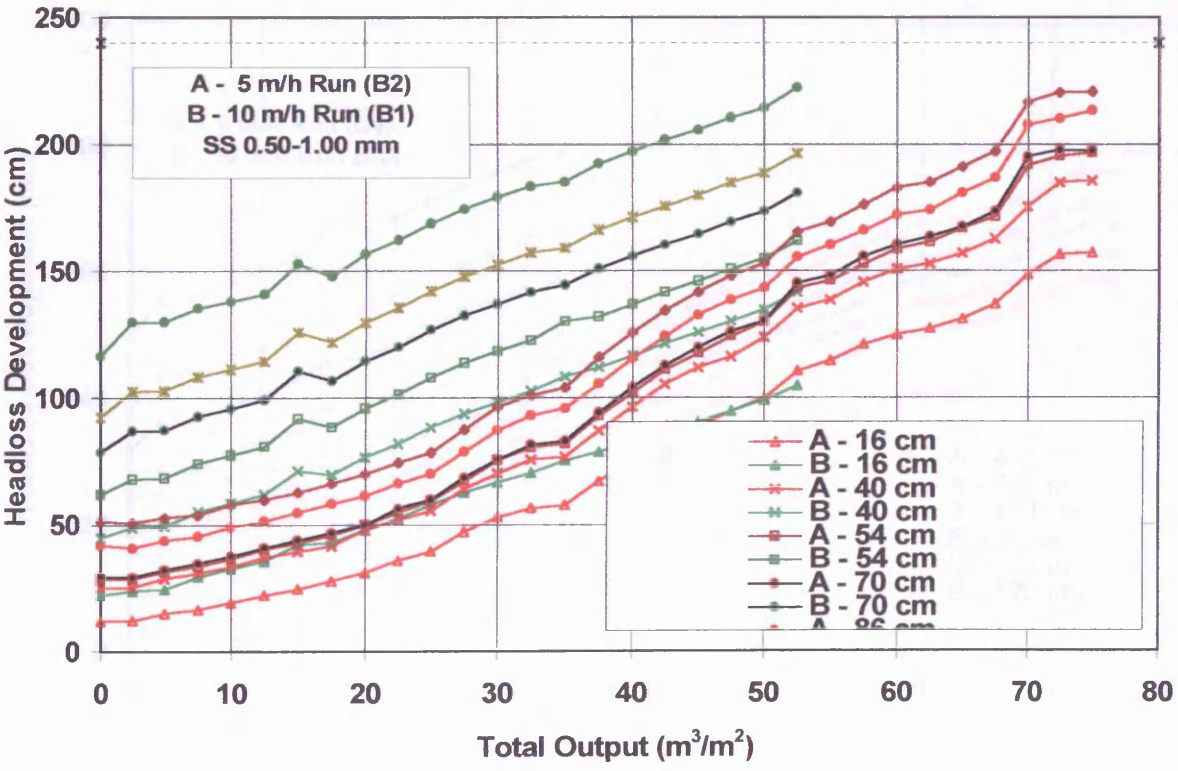

Figure 4.56 Headloss development as a function of depth & filtration rate; raw water turbidity 75 NTU, sand size 0.50-1.00 mm.

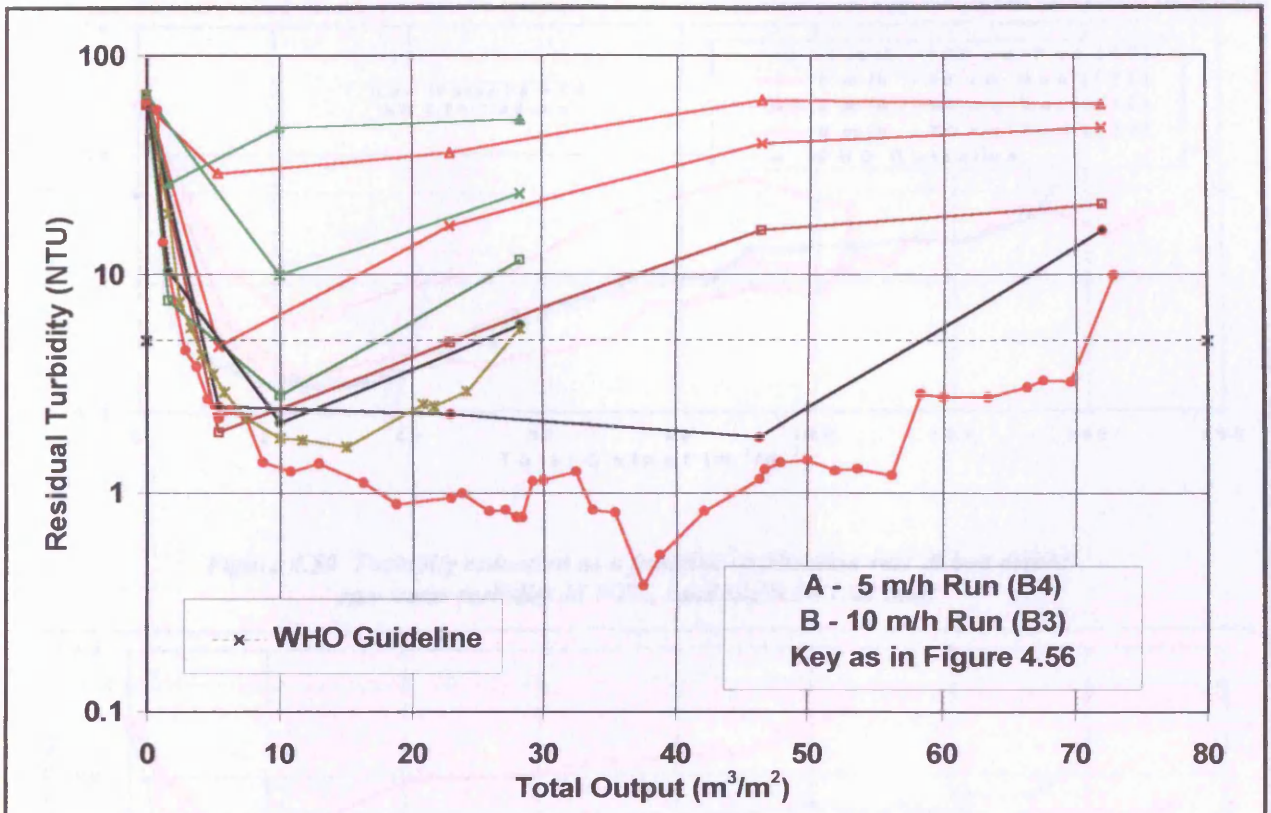

Figure 4.57 Turbidity reduction as a function of depth & filtration rate; raw water turbidity 75 NTU, sand size 0.85-1.70 mm.

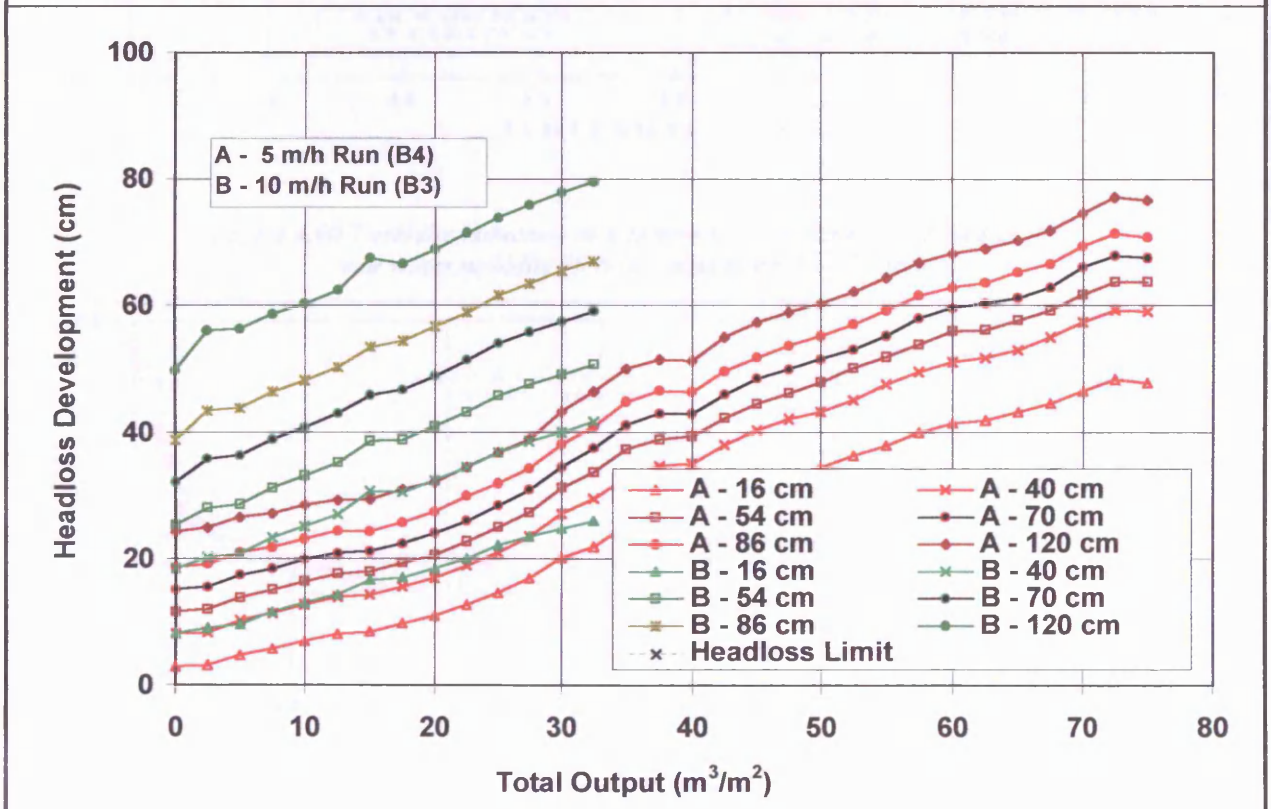

Figure 4.58 Headloss development as a function of depth & filtration rate; raw water turbidity 75 NTU, sand size 0.85-1.70 mm.

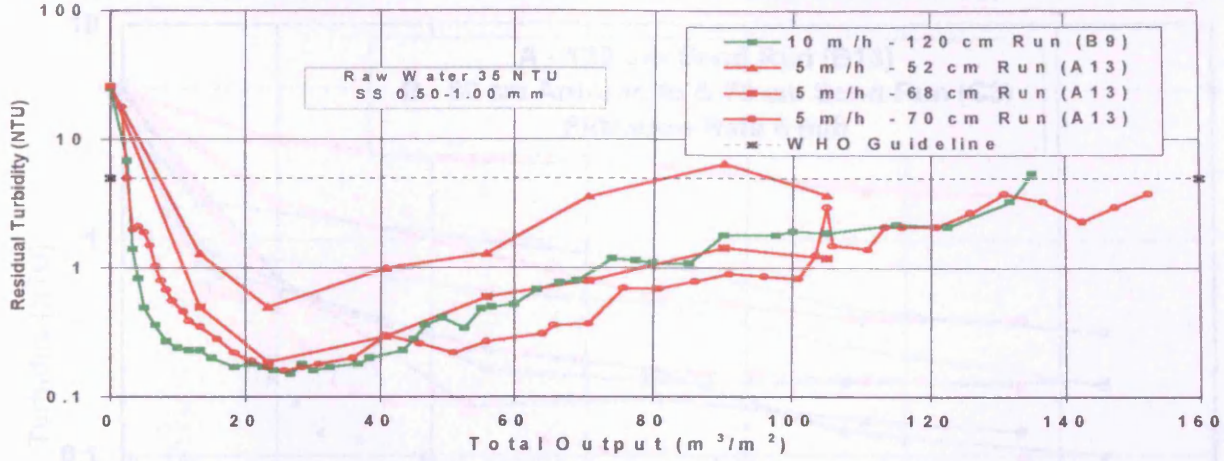

Figure 4.59 Turbidity reduction as a function of filtration rate & bed depth; raw water turbidity 35 NTU, sand size 0.50-1.00 mm.

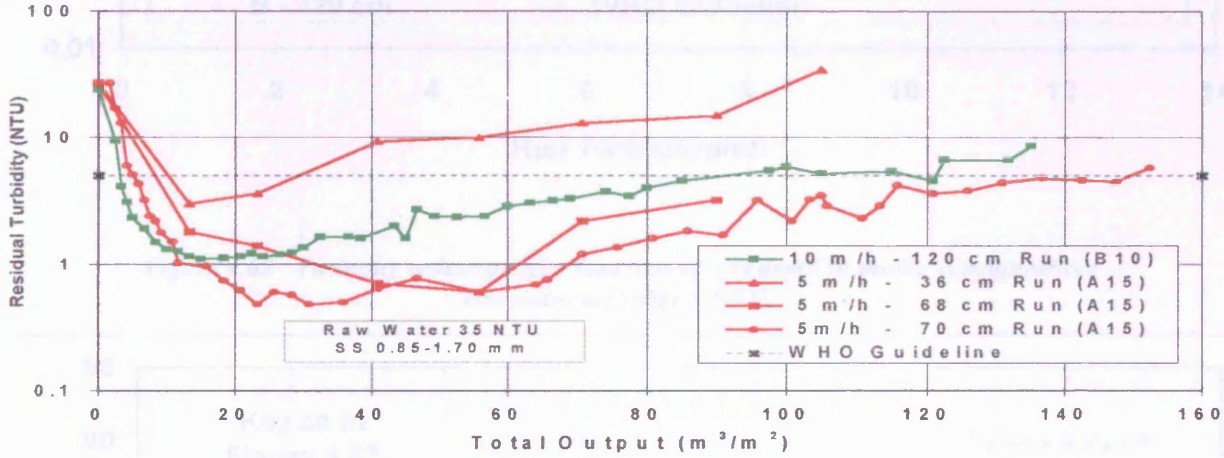

Figure 4.60 Turbidity reduction as a function of filtration rate & bed depth; raw water turbidity 35 NTU, sand size 0.85-1.70 mm.

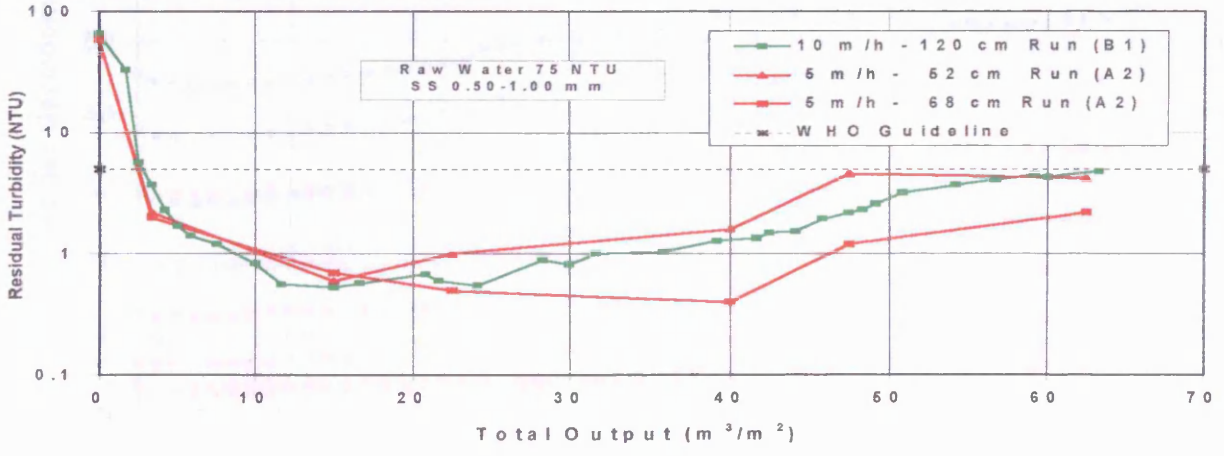

Figure 4.61 Turbidity reduction as a function of filtration rate & bed depth; raw water turbidity 75 NTU, sand size 0.50-1.00 mm.

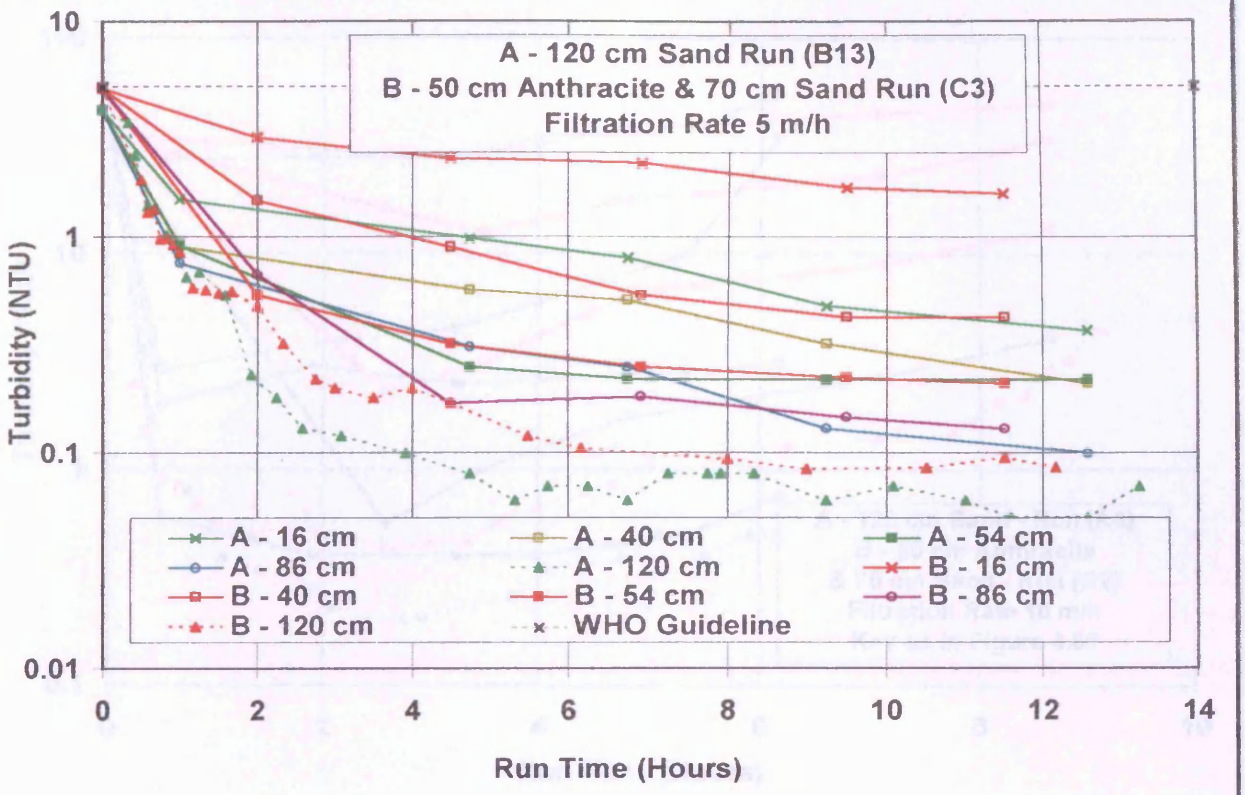

Figure 4.62 Turbidity reduction as a function of bed depth & media configuration; raw water turbidity 5 NTU.

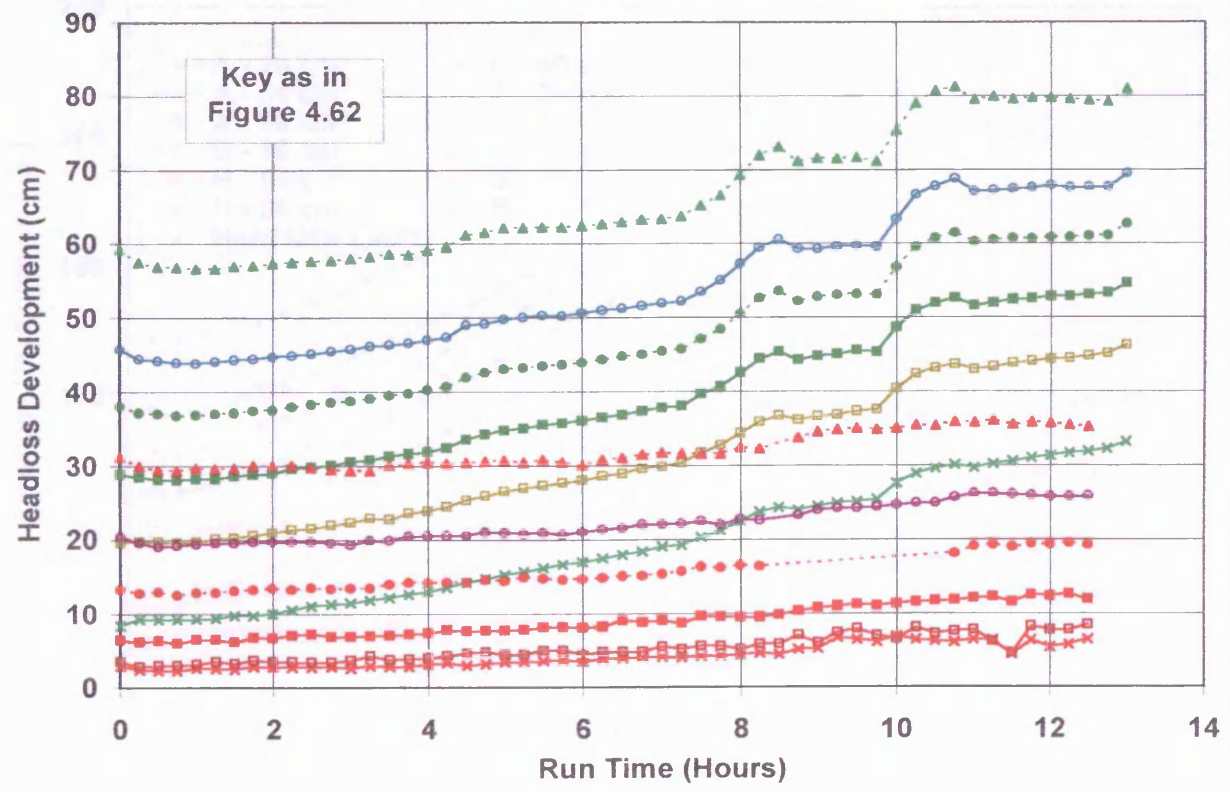

Figure 4.63 Headloss development as a function of bed depth & media configuration; raw water turbidity 5 NTU.

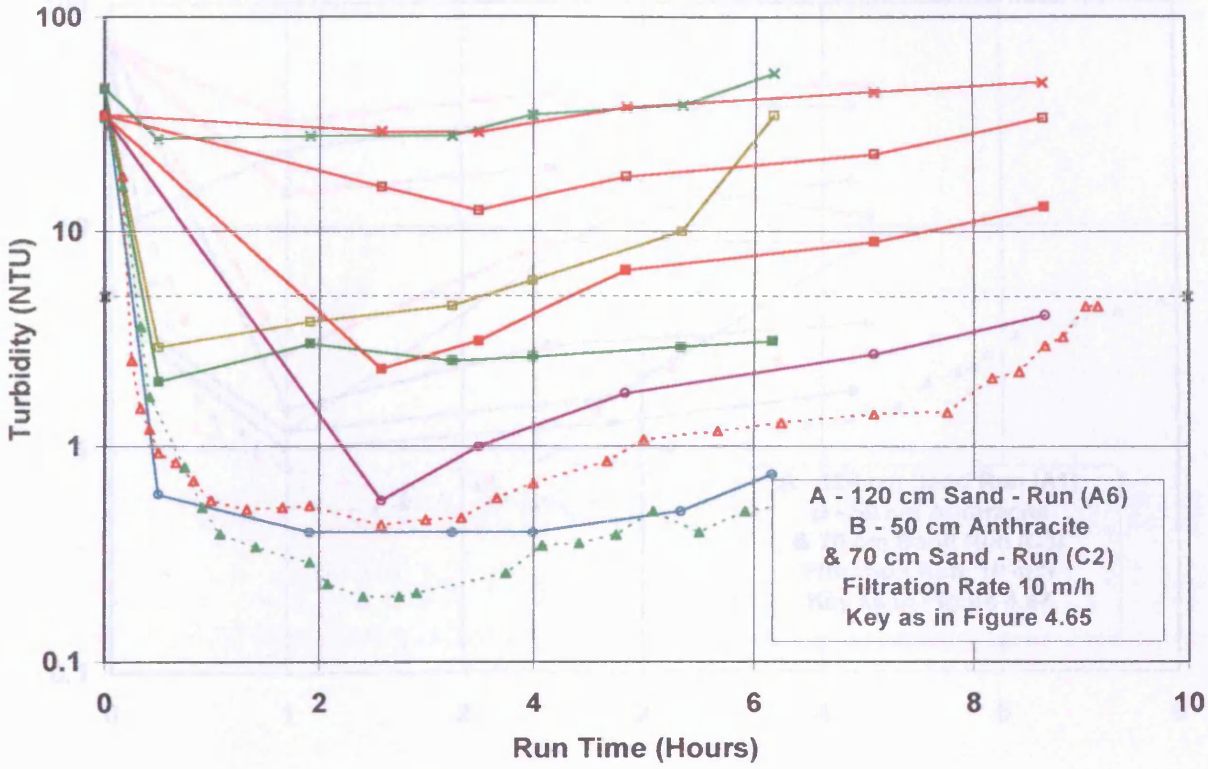

Figure 4.64 Turbidity reduction as a function of bed depth & media configuration; raw water turbidity 50 NTU.

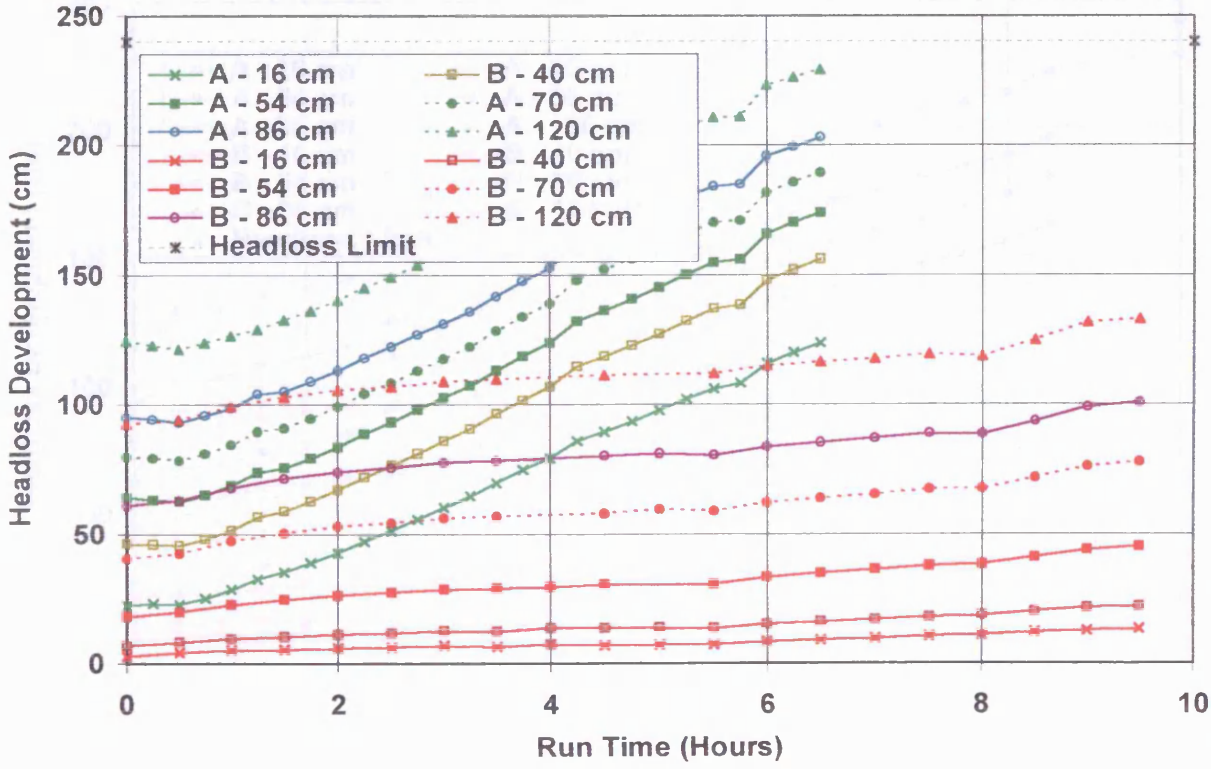

Figure 4.65 Headloss development as a function of bed depth & media configuration; raw water turbidity 50 NTU.

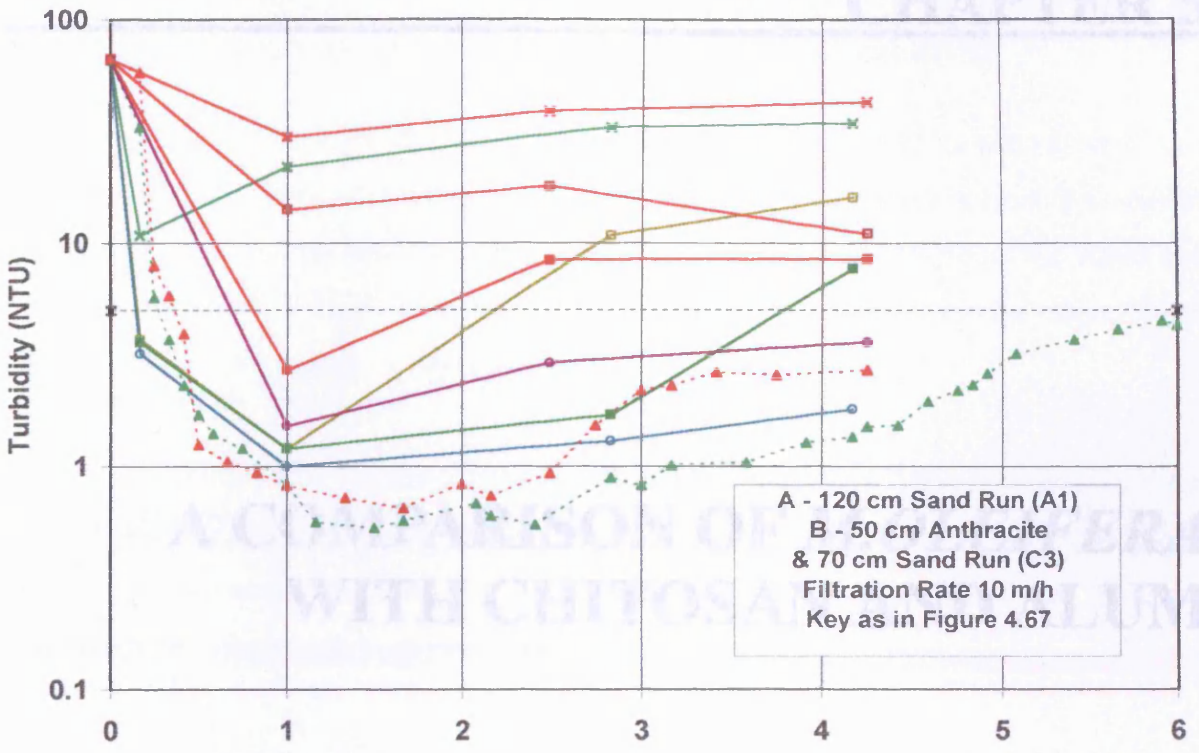

Figure 4.66 Turbidity reduction as a function of bed depth & media configuration; raw water turbidity 75 NTU.

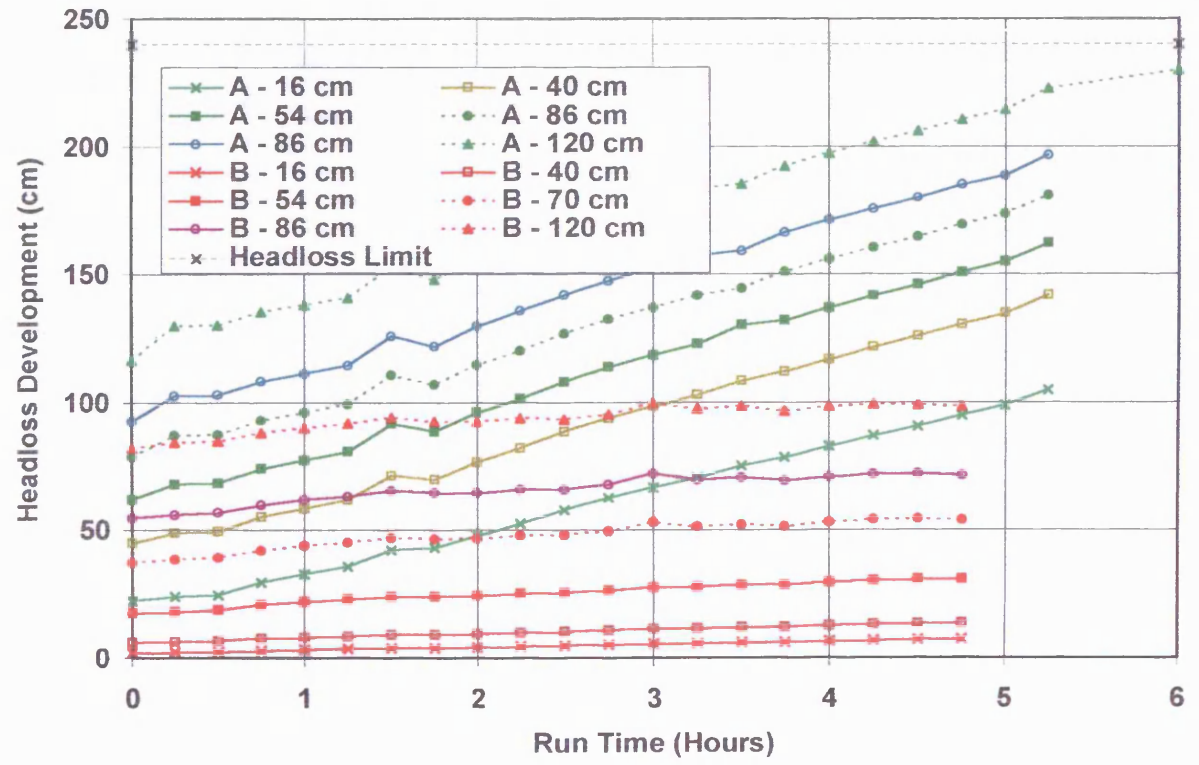

Figure 4.67 Headloss development as a function of bed depth & media configuration; raw water turbidity 75 NTU.

CHAPTER 5

A COMPARISON OF *M.OLEIFERA* WITH CHITOSAN AND ALUM

5.0 A comparison of *M.oleifera* with chitosan and alum

The objective of this element of the study was to compare the effectiveness and modes of floc formation using *M.oleifera* seed, chitosan and alum. Chitosan was chosen because it is another naturally derived coagulant but with contrasting characteristics to *M.oleifera* seed. Alum was investigated because it is the most commonly used coagulant worldwide, and may therefore be used as 'baseline' for introducing new coagulants for water treatment.

The study was conducted using the pilot plant described in Chapter 3. The media configuration used consisted of a single media bed of 70 cm sand (sand size 0.50-1.00 mm) and a dual media bed of 50 cm anthracite and 70 cm sand. Two filtration rates were considered viz.: 5 and 10 m/h, and two raw water turbidities viz.: 5 and 75 NTU.

For the comparison of *M.oleifera* seed with chitosan, the raw water used consisted of a kaolin solution suspended in deionised water. The pH ranged from 6 to 7.5, and there was no alkalinity present. Whereas for the comparison with alum, the raw water consisted of kaolin solution suspended in tap water. The reason for this is that for alum-induced floc formation to occur, the water must contain sufficient alkalinity, as explained in Section 2.2.2. It is estimated that 1 mg/L of alum requires 0.5 mg/L alkalinity as $CaCO_3$ in order for the hydroxide precipitate to form (Tebbutt, 1998). The raw water used contained 100 mg/L of alkalinity as $CaCO_3$ and had a pH of 6.5-7.5. Details of the preparation of chitosan and alum solutions are given in Section 3.1. Preliminary jar tests were undertaken to obtain the optimum dose for each coagulant prior to conducting the filter runs. The raw water used in the jar tests was the same as that used in the filter columns.

5.1 Chitosan

Chitosan is a modified natural carbohydrate derived from chitin. Chitin was first discovered in mushrooms in 1811 by Professor Henri Braconnot in Nance in France (Vanson, 1997; FatAbsorb, 1997). It has since been found in crustacean shells, making up between 20-40% of the total shell weight (Schulz & Okun, 1992). It is also present in the shells of some insects, zooplankton and in the cell walls of numerous fungi. This ubiquitous compound is considered to be the second most abundant natural polymer on earth after cellulose (Brzeski, 1987).

Chitin is a linear polymer of chitobiose, an aminopolysaccharide. It has a molecular weight of about 10^6 daltons. It is virtually insoluble in water and organic solvents, but is soluble in mineral acids. Chitin is a waste product of the shellfish industry. It is estimated that for every 1 kg of crabmeat 6 kg of shell is produced. Indiscriminate disposal of the shells can result in a serious pollution problem due to the release of ammonia and nitrates into the soil (Leffler, 1997). Using chitin in water treatment is an environmentally attractive option as it solves a waste disposal problem in coastal areas. However, the conversion of chitin to chitosan is relatively costly. It consists of dissolving chitin in HCl, to remove compounds other than chitosan, the remaining material is then deacetylated with sodium hydroxide to remove N-acetyl-D-glucosamine to produce chitosan (Kawamura, 1991a; Sigma, 1998). Furthermore, conversion of the solid chitosan flakes into a solution that can be dosed into the water, requires high intensity mixing in an acidic solution, which adds to the overall production costs of this coagulant. Cost estimates obtained during this research for chitosan ranged from £5-20/kg (Murcott & Harleman, 1993b; Stead, 1998; Higashimaru Feeds Ltd., 1999; Xiamen Pharmaceutical Factory, 1999).

A major benefit of chitosan, as in the case with *M.oleifera* seed, compared to other coagulants is that the sludge is biodegradable, non-toxic and relatively compact, it is therefore easily disposed of and may be used or sold as a fertilizer. There are many other economic advantages to using indigenous products, such as utilising the local labour force, bolstering the economy and a reduction in the use of foreign exchange. A specific advantage of using chitosan, is that the solution remains active for long periods, since no bacterial activity or degradation will occur in an acidic solution. A 5 year old chitosan solution was found to be effective (Schulz and Okun, 1992), whereas *M.oleifera* solutions became ineffective within about 8-12 hours.

Chitosan is approved by the US Environmental Protection Agency (USEPA) for use in water treatment at concentrations of up to 10 mg/L (Kawamura, 1991a). It has also been used as a coagulant aid in Japan since 1950 (Kawamura, 1981), and there have been successful pilot studies in Norway on its use in the treatment of dairy wastewater. The advantage of using it on such waters is that the non-toxic sludge can be used as a food additive (Ratnaweera & Selmer-Olsen, 1996). Other researchers have conducted work into the use of chitosan in the treatment of drinking water with varying degrees of success. Details of the main studies conducted are listed in Table 5.1.

Reference	Raw Water Characteristics[1]	Conditions of Testing	Coagulant(s) & Dose (mg/L)	Residual Turbidity (NTU)
Kawamura (1991a)	Turbidity: 2.5 NTU, alkalinity: 200 mg/L, pH 8.1	Jar test: 15 sec at 120 rpm, 7.5 mins at 70 rpm, 7.5 mins at 40 rpm, 5 mins at 25 rpm, 20 mins settling	Primary coagulant - alum: 17 coagulant aid - chitosan 0.15	0.4
Kawamura (1991a)	Turbidity: 51 NTU, alkalinity: 112 mg/L, pH 8.2	Jar test: 15 sec at 120 rpm, 7.5 mins at 70 rpm, 7.5 mins at 40 rpm, 5 mins at 25 rpm, 20 mins settling	Primary coagulant - alum: 30. coagulant aid - chitosan 0.2	1
Kawamura (1991a)	Turbidity: 15 NTU, alkalinity: 28 mg/L, pH 7.1	Jar test: 15 sec at 120 rpm, 7.5 mins at 70 rpm, 7.5 mins at 40 rpm, 5 mins at 25 rpm, 20 mins settling	Chitosan: 5.0	1
Kawamura (1991a)	Turbidity: 10 NTU, alkalinity: 30 mg/L, pH 7.0	Jar test: 15 sec at 120 rpm, 7.5 mins at 70 rpm, 7.5 mins at 40 rpm, 5 mins at 25 rpm, 20 mins settling	Chitosan: 5.0	0.8
Kawamura (1991a)	Turbidity: 2.3 NTU, alkalinity: 114 mg/L, pH 8.0	Pilot plant, direct filtration, dual media consisting of anthracite and sand. Filtration rate 8 mpg/sq ft	Primary coagulant - alum: 8, coagulant aid - chitosan: 1.4	0.1
Kawamura (1991a)	Turbidity: 1.1 NTU, alkalinity: 282 mg/L, pH 8.4	Pilot plant, direct filtration, dual media consisting of anthracite and sand. Filtration rate 8 mpg/sq ft	Chitosan: 0.8	0.15
Kawamura (1991b)	Turbidity: 66 NTU	Jar test: 10 sec at 100 rpm, 7.5 mins at 60 rpm, 7.5 mins at 40 rpm, 5 mins at 20 rpm, 20 mins settling	Chitosan: 1, 2, 4 & 8	12, 6, 5.3 & 4.9
Kawamura (1991b)	Turbidity: 66 NTU	Jar test: 10 sec at 100 rpm, 7.5 mins at 60 rpm, 7.5 mins at 40 rpm, 5 mins at 20 rpm, 20 mins settling, filtered through Whatman No. 1	Chitosan: 1, 4 & 8	1, 0.7 & 0.6
Murcott & Harleman (1993b)	Turbidity: 1.5 NTU, alkalinity: 26 mg/L, pH 7.4	Jar test: 4 mins at 150 rpm, 30 mins at 50 rpm, 30 mins settling	Primary coagulant - chitosan: 0.5, coagulant aid - bentonite [2]: 9	0.17
Murcott & Harleman (1993a)	Turbidity: 0.56 NTU, alkalinity: 4.2 mg/L, pH 6.8	Jar test: 4 mins at 150 rpm, 30 mins at 50 rpm, 30 mins settling	Primary coagulant - chitosan: 1.0, coagulant aid - bentonite: 8	0.3
Huang & Chen (1996)	Bentonite suspension[3], turbidity: 35 NTU	Jar test: 2 mins at 100 rpm, 20 mins at 30 rpm, 10 mins settling	Chitosan: 0.5	0.56

Huang & Chen (1996)	Kaolinite suspension[3], turbidity: 25, 100, 500 & 1000 NTU	Jar test: 2 mins at 100 rpm, 20 mins at 30 rpm, 10 mins settling	Chitosan: 0.05, 0.10, 0.50 & 0.8	15, 21, 18 & 20
Huang & Chen (1996)	Kaolinite suspension[3], turbidity: 25, 100, 500 & 1000 NTU	Jar test: 2 mins at 100 rpm, 20 mins at 30 rpm, 10 mins settling	Primary coagulant - chitosan: 2, 3, 7, 11, coagulant aid - bentonite: 200, 300, 700 & 1100	0.5, 0.8, 1.5 & 2.5
Jahn (1981)	Turbidity: 3200, 1400, 500 & 70 NTU	Jar test: 1 mins at 100 rpm, 9 mins at 40 rpm, 10 mins settling	Chitosan: 1.00, 1.00, 0.25 & 0.25	10, 10, 25 & 18
Jahn (1981)	Turbidity: 3200, 1400, 500 & 70 NTU	Jar test: 1 mins at 100 rpm, 9 mins at 40 rpm, 10 mins settling	Primary coagulant - alum: 20, 20, 5, 8, coagulant aid - chitosan: 0.15, 0.10, 0.10 & 0.05	4, 3, 5 & 10

Table 5.1 List of previous researchers' work using chitosan in the treatment of drinking water.

[1] Natural raw water used except where noted.

[2] Bentonite is a fine-grained inorganic clay of the mineral montmorillonite that assists in increasing the rate and efficiency of coagulation.

[3] Raw water made up in electrolyte suspension (10^{-2} M $NaCl_4$).

5.1.1 Discussion of previous research on chitosan

Huang & Chen (1996) concluded from their study that chitosan formed floc by the 'bridging' mechanism rather than charge neutralisation at pH 7 (which was the pH of the raw water used in this study). This was deduced by measuring the zeta potential of the coagulated particles, where it was found that there was no charge reversal, at or above, the optimum dose. If the 'patch' mechanism of charge neutralisation was in operation, the coagulated particles would have a neutral charge at optimum removal conditions, and a positive charge when the coagulant was over-dosed, as described in Section 2.2.2. Another indication that the 'bridging' mechanism is applicable is that it is effective as a coagulant aid with low doses of alum on low turbidity waters. Alum forms relatively small floc in such waters, and the chitosan polymer can be used to adhere, or 'bridge', the floc together to form larger more 'settleable' floc (Schulz & Okun, 1992). Furthermore, the molecular weight of chitosan (the sample used was 1^6-2^6 daltons) indicates a high molecular weight polymer. This size polymer is considered to be a 'bridge' former, since attachment of the chain onto a particle would generally result in a portion of its length protruding into the solution, enabling attachment to other particles.

The main conclusion drawn from Table 5.1, is that chitosan is very effective at particulate removal with high turbidity waters (100-3200 NTU), as a primary coagulant. It is less effective in the treatment of low turbidity waters (1.5-66 NTU), particularly when used as the sole coagulant. It has been used as a coagulant aid in conjunction with alum, and as a primary coagulant with bentonite as a coagulant aid to produce relatively high quality filtrate. Murcott & Harleman (1993a) did not achieve good results when chitosan was used as the sole coagulant (although no data was given), but effectiveness improved with the addition of bentonite as a coagulant aid.

5.1.2 Results and discussion from the comparative studies on chitosan & *M.oleifera* seed.

The results of the comparative studies on these two coagulants, in terms of turbidity removal and headloss development, with various media configurations and filtration rates are illustrated in Figures 5.1-5.15. There were three main differences observed between the performance of the two coagulants:

- Turbidity removal was greater with chitosan than with *M.oleifera* seed.
- Headloss developed at a faster rate with chitosan than with *M.oleifera* seed.
- Chitosan formed floc at a faster rate and to greater size than *M.oleifera* seed.

Raw waters with turbidities of 5 and 75 NTU were reduced to < 0.1 NTU, when treated with chitosan in dual and single media beds. When treated with *M.oleifera* in a single media bed, a 75 NTU raw water produced a filtrate of < 5 NTU (Figures 5.1). In a dual media bed 75 and 5 NTU raw waters produced filtrates of <1 NTU (Figure 5.4) and < 0.1 NTU (Figure 5.7) respectively.

Figure 5.2 shows rapid development of surface headloss with chitosan in a single media bed. At the termination of the run, about 70% of the total headloss had developed in the top 4 cm. The next section of the bed, between 4-20 cm depth developed about 24% of the headloss. The remaining 50 cm of the bed played a very small part in the removal process. The inset in Figure 5.2 indicates that the top 4 cm of the bed reduced the turbidity by 50%. The next 20 cm was responsible for producing filtrate with a turbidity of less than 1 NTU. A similar trend is seen in the dual media bed (Figure 5.50), in that surface removal dominated, with about 70% of the headloss developing in the top 16 cm. The headloss development rates were considerably lower with *M.oleifera* in all three treatment conditions. Figure 5.3 indicate that only 50% of the headloss accumulated in the 4 cm of the single media bed. In the dual media beds (Figures 5.6 and 5.9), the headloss in the entire anthracite layer was < 15%. Thus indicating a significant reduction in the surface removal with *M.oleifera* in the dual compared to the single media bed. Whereas with chitosan the dual media bed was less effective in reducing surface removal, although the total bed headloss was reduced, effecting a longer filter run.

The difference in deposit morphology, shown by the headloss data in the beds treated with *M.oleifera* and chitosan, lies in the variation in the size of the floc formed, and the attachment mechanism to the filter media. *M.oleifera* produced small compact pin floc, and chitosan produced larger floc. Plates 9 and 10, show samples of floc produced in jar tests using the two coagulants. A relatively high turbidity water of 300 NTU was used in order to show the difference in floc morphology. The chitosan floc are clearly much larger than the *M.oleifera* floc. This variation in the size is considered to be due to the mode of particle destabilisation and floc formation. *M.oleifera* forms floc by the 'patch' mechanism of charge reversal, as

described in Section 2.7.3, since the active proteins are of low molecular weight and a high charge density (Tauscher, 1994; Gassenschmidt *et al.*, 1995). In comparison, the chitosan polymer induces destabilisation and floc formation by the 'bridging' mechanism at pH 7 (described in Figure 1.4) (Huang & Chen, 1996). Large molecular weight polymers rapidly induce the formation of large floc with strong bonds (LaMer & Healy, 1963; AWWA, 1992).

It is postulated that chitosan formed floc by 'sweep' flocculation. The dosing solution used in the pilot study was made up in 1% acetic acid, and had a pH of 3.5; the raw water had a pH of 6-6.5. The rise in the pH of the chitosan solution on dosing into the raw would have caused the chitosan to precipitate out of solution forming solid material, which could then enmesh particulate matter. This mechanism is analogous to that induced by metal salt (cf. coagulation and flocculation with alum in Section 2.2.2).

There is evidence of both these mechanisms of floc formation operating in the pilot plant. The indication that precipitation and enmeshment occurs, is firstly that floc formation was very rapid. Jar tests conducted in the laboratory using chitosan, on relatively high turbidity waters (75 NTU), indicated that floc formed during the rapid mix stage. This is in contrast to *M.oleifera* floc, which only formed after about 10 minutes of slow mixing. In the filter columns, it was found that some chitosan formed floc prior to entering the columns. In contrast *M.oleifera* floc only formed when in contact with the filter media, i.e. discrete particles entered the bed and not floc. Furthermore, addition and mixing of the coagulant into the raw water in the filter columns occurred in the clear narrow plastic tube, described in Section 3.3.1. The inside of the tube became coated in a white material, which may have been precipitated chitosan and entrapped colloidal matter. This very rapid floc formation brought about by chitosan addition, coupled with the high removal rates achieved even with low turbidity raw waters suggests that 'sweep' floc coagulation was occurring.

The evidence that chitosan was also acting as a 'bridge' forming polymer, is that small floc formed initially in the jar tests continued to grow as the slow mix continued. Precipitation induced floc were 'bridged' together to form larger floc. If 'sweep' flocculation was the only mechanism of particle destabilisation and aggregation, then the resulting floc would have been weak. Whereas the floc formed in jar tests and the filter columns were very strong. Chitosan also formed strong bonds between the particles/floc and the media in the filter beds, and consequently high washwater flow rates were required to backwash the bed.

A consequence of forming large, strong floc is that attachment to the filter media occurs rapidly (Stump & Novak, 1979). Particle attachment to the filter media tends to be autocatalytic due to the effect of bed ripening (Kau & Lawler, 1995; M. Moran *et al.*, 1991), and therefore removal in the surface layers continues. The water that enters the subsequent layers of the bed, will have a reduced particulate load. The particles that have been destabilised to a greater extent will be removed in the surface layers, leaving the more stable particle to enter the deeper sections of the bed. *M.oleifera* floc are much smaller than chitosan floc, as seen in Plates 9 and 10. Deeper particulate penetration would have occurred and therefore removal would not have been restricted to the surface layers of the bed; accordingly the headloss development rate is much lower.

The effect of filtration rate on the removal process with the two coagulants was investigated. Figures 5.10-5.15 illustrate turbidity removal and headloss development at filtration rates of 5 and 10 m/h with *M.oleifera* seed and chitosan. There are three main features to these graphs:

- Chitosan was more effective in terms of turbidity removal at a filtration rate of 10 than at 5 m/h (Figure 5.10). This contrasts with *M.oleifera* seed where turbidity removal was far superior at 5 than at 10 m/h (Figure 5.11). Higher filtration rates are generally utilised in treatment works as a matter of necessity, although often at the expense of final water quality. The total headloss at 5 m/h was almost double that at 10 m/h for the same total output, demonstrating the extent of surface removal and lack of deep bed filtration occurring at the lower rate. In contrast, Figures 6.14 and 6.15 indicate that the total headloss development rate at 40 m^3/m^2 with a filtration rate of 5 m/h was double that at 10 m/h.

- Headloss development rates for chitosan and *M.oleifera* seed (Figures 5.12-5.15), indicate that there was a greater distribution of deposited particles throughout the bed at a filtration rate of 10 than 5 m/h. There was, however, a difference in the deposit morphology with the two coagulants. *M.oleifera* seed produced a greater distribution of particulate deposit at both filtration rates than did chitosan. Essentially, the majority of particulate removal occurred in the surface layers with chitosan even at the higher filtration rate of 10 m/h.

The effectiveness of particulate removal during filtration is influenced by filtration rate for two reasons. Namely, the effect on the extent of particle aggregation and the length of the retention

time of the coagulated water in the filter bed. The rate of particle aggregation and floc formation is determined by the intensity (G) and duration (t) of the rapid mix stage. Calculation of the G value in the rapid mix stage of the pilot plant is complex, since the coagulant was injected into the flow of water, causing turbulence and hence mixing. The G value in the subsequent rapid flow section in the narrow tubing, can be calculated but does not reflect the full mixing regime encountered. The G value was higher at 10 than 5 m/h. The t values at these filtration rates were 1 and 2 minutes respectively. The variation in the removal rates at the two flow rates, as shown in Figures 5.10 and 5.11, was caused by more optimal conditions for floc formation using chitosan at 10 m/h, and *vice versa* with *M.oleifera* seed. High molecular weight polymers, such as chitosan, generally require higher G values in the rapid mix stage (Yeh & Ghosh, 1981). A less intense mixing period may result in polymers becoming wrapped around particles, by secondary adsorption, thus causing restabilisation, as seen in Figure 2.4, Step 2B. Additionally, the high viscosity of the chitosan solution can result in an uneven distribution of the chemical throughout the water, leading to destabilisation of some particles, whilst others may not have come into contact with the polymer. The reduced mixing intensity at 5 m/h did not have the same effect when using *M.oleifera* seed. Ndabigengesere & Narasiah (1996) found that *M.oleifera* seed did not require a rapid mix stage prior to a period of flocculation and sedimentation in jar tests in order to ensure efficient removal. This was considered to be as a result of the low molecular weight of the *M.oleifera* active proteins (approximately 6×10^3 daltons), suggesting that a lower G value would be sufficient to induce flocculation. Although the treatment regime was different to that used in the filter columns, it can be assumed that the G value during the rapid mix stage is of less importance for *M.oleifera* seed, than was the case with chitosan.

Furthermore, at 5 m/h, t was 2 minutes compared to 1 minute at 10 m/h. The extended mixing period at 5 m/h, may have caused floc break-up, since chitosan floc formed so rapidly. Floc induced by 'bridge' formers, once fragmented, do not easily reform even if more quiescent condition subsequently prevail (Gregory, 1996). Fragments of floc can become restabilised, as described in Figure 2.4, Step 4B. Removal rates are thus reduced. *M.oleifera* floc, however, were not formed so rapidly and therefore unlikely to experience breakup. In Addition, floc formed by charge neurtalisation may readily reform following rupture (Gregory, 1996).

A further effect of a higher filtration rate is the reduced retention time in the filter bed, and the consequent reduction in in-pore flocculation. As discussed in Section 4.4.1, in-pore

flocculation was considered to be a significant factor with *M.oleifera* seed and therefore the retention time in the bed was important. Whereas with the high molecular weight chitosan polymer, in-pore flocculation was less significant. Therefore the reduced retention time in the filter bed was not detrimental to the removal process.

Higher filtration rates would also affect attachment and detachment mechanisms in the filter bed, as discussed in Section 4.4.1. Chitosan forms strong floc and readily attaches to the filter media, as described above. Higher filtration rates would therefore be advantageous in preventing surface removal from dominating. Since the *M.oleifera* induced bonds are weaker, higher filtration rates, could cause break-off of particles from collector surfaces and premature breakthrough of turbidity if the filter bed is not deep enough, resulting in reduced filter run times, as seen in Figure 5.1.

Headloss development with chitosan was high at both filtration rates considered, and was the cause of run termination in both cases, as seen Figure 5.12 and 5.13. It was expected from theory that the total headloss would be greater with higher filtration rates, since Darcy's Law established that headloss development across the filter bed was proportional to filtration rate, described in Section 4.2.1 (Ives, 1980; Tebbutt, 1998). This was found to be the case with *M.oleifera* seed, as seen in Figures 5.14 and 5.15. This effect was not seen with chitosan due to the dominance of surface filtration; a visible thick layer of floc formed on the surface of the filter bed. Higher filtration rates brought about deeper penetration of the particles and therefore greater use of the filter bed and a lower rate of headloss development. This occurred with chitosan and not *M.oleifera* seed, due to the size of the chitosan polymer ($1x10^6$-$2x10^6$ daltons (Stead, 1999)); Yeh and Ghosh (1981) considered polymers > $1x10^6$ daltons to be too large for use in DF and CFF for the fact that they lead to excessive headloss development. The use of such polymers may still be appropriate if sufficiently high filtration rates and large media are used, which can prevent surface accumulation of the floc. Furthermore pressure rather than gravity-fed filters would need to be employed, since these can operate with higher headlosses. The formation of mud balls, a problem encountered with high pressures across the filter bed, can be overcome by providing sufficient space above the filter bed for full bed fluidisation during backwashing. This would allow a high backwash flow rate which, in conjunction with an air scourer, would cause the breakup and removal of the mud balls.

The overall results obtained using chitosan indicated that it was an excellent coagulant in terms of turbidity removal and volume treated. However, these are not necessarily the only considerations in the choice of coagulant. Costs of the coagulant, pH adjustment chemicals, sludge disposal, etc. must also be considered. If the coagulant was to be used in developing countries, with relatively unskilled labour, then the complexity of preparation and use may be a problem. The production cost of many natural coagulants is difficult to estimate. With *M.oleifera* seed it is dependent upon the management techniques employed in the cultivation of the tree, the type of soil the tree is grown, the cost of local labour, the variety of tree that is grown etc. Other costs such as shelling, grinding and sieving of the seeds to produce the powder would also need to be considered. Some expertise is required in the preparation of both these coagulant solutions. *M.oleifera* seed powder production is potentially more labour intensive in terms of shelling and grinding of the seeds. The location of the treatment works would also need to be considered. Coastal areas with an abundance of shellfish waste are likely to produce chitosan at a relatively low price. *M.oleifera* seed is likely to be more available and cheaper in tropical and subtropical regions.

5.1.3 Comparison of the results obtained from the laboratory study with previous research on chitosan

The levels of turbidity removal obtained during the operation of the pilot plant using chitosan, as seen in Figures 5.1-5.9, are far greater than those obtained by other researchers, indicated in Table 5.1. In addition, the optimum dosages required were, in many cases, an order of magnitude lower. Figures 5.1 and 5.4 indicate that a 75 NTU water was reduced to < 0.1 NTU for over 8 hours at a dose of 0.11 mg/L. Other authors found conducting jar tests, a 70 NTU raw water was reduced to 18 NTU with a dose of 0.25 mg/L, and to 10 NTU when chitosan was used as a coagulant aid in conjunction with alum (Jahn, 1981). Furthermore, in the laboratory study a raw water of 5 NTU was reduced to < 0.1 NTU with a dose of 0.03 mg/L of chitosan (see Figure 5.7). Jar tests conducted by Kawamura (1991a) reduced the turbidity of a 2.5 NTU raw water to 0.4 NTU, with chitosan as a coagulant aid in conjunction with alum. Even with the dual media bed, operated in the DF mode, a raw water turbidity of 2.3 NTU was only reduced to 0.1 NTU, at a dose of 1.5 mg/L of chitosan as a coagulant aid in conjunction with alum (Kawamura, 1991a).

The difference in turbidity removal rates and optimal doses can be explained by the difference in treatment conditions. The majority of tests considered in Table 5.1 were conducted as jar tests; comprising a rapid mix, a slow mix and a settling stage. Chitosan floc formed rapidly in jar test and the filter columns in the laboratory study. It is therefore postulated that floc break-up could occur during the flocculation stage, if the mixing time extended for too long. Huang & Chen (1996) found that extended mixing of chitosan floc resulted in their rupture and subsequent folding back of the polymer over the surfaces of the particles, causing charge reversal, restabilisation and a reduction in particulate removal. It is thus postulated that inclusion of a flocculation stage in floc formation with chitosan may be detrimental to the treatment process, and therefore, removal rates in the filter columns operated in the CFF mode were greater than in the jar tests and pilot plants using conventional treatment and DF.

The lower dose requirement found in this study compared to those reported in Table 5.1, is caused by the difference in optimal floc size for CFF and conventional treatment. In order to achieve efficient removal with the former treatment mode, lower doses are used to form small 'filterable' pin floc. Whereas with conventional treatment, floc are ideally large and 'settleable', and therefore a higher coagulant dose is required to achieve optimal floc formation. The reduction in the optimal dose requirement with CFF compared to conventional treatment, is one of the major advantages of this treatment process (Schulz & Okun, 1992).

The other differences between this study and the studies summarised in Table 5.1, were the alkalinity and the pH values of the raw waters. The raw water used in this study consisted of deionised water with pH values of between 6.0-6.5; whereas for the other studies varying amounts of alkalinity were either added or present naturally in the water, and the pH ranged from 6.8-8.4. To investigate the effect of increasing the alkalinity of the water, $NaHCO_3$ was added to the raw water in the laboratory study during the course of a filter run. This is shown in Figure 5.16. The raw water turbidity used was 5 NTU, and an initial dose of 0.50 mg/L reduced the turbidity to < 0.1 NTU. After 7 hours into the run, 112 mg/L of $NaHCO_3$ was added to the raw water tanks, the effect of which was to increase the residual turbidity to about 4 NTU. When the chitosan dose was increased to 0.1 mg/L, the residual turbidity decreased to 1 NTU. Essentially, the effect of increasing the alkalinity of the raw water was to reduce removal efficiency. It is known that high ionic concentrations cause polymer strands to coil, thereby reducing their length and effectiveness at 'bridging' (Yeh & Ghosh, 1981;

Montgomery, 1985). Thus the presence of alkalinity caused the chitosan polymer to be less effective in turbidity removal, and therefore a higher dose was required.

The iso-electric point of chitosan is at pH 8, at lower pH values the polymer is positive and at higher pH values it is negative (Huang & Chen, 1996). Thus the polymer would have had a higher positive charge in this study than in the other studies, resulting in stronger attachment forces between the polymer and the particles and a lower dose requirement. Huang & Chen (1996) concluded from their work that pH has little effect on the coagulation of chitosan, but analysing their results it would appear that in fact pH affects the dose requirement. For a raw water turbidity of 30 NTU at pH 4, the optimum dose was about 0.4 mg/L whereas at pH 7 the optimum dose was 2 mg/L. Contrary to their conclusions, it is suggested that the pH and the alkalinity of the raw water do affect particle removal and optimal doses of chitosan required.

5.2 Aluminium sulphate

The earliest records of the use of alum as a coagulant date back to 77 A.D., where it was used in conjunction with lime to make 'bitter water potable' (Faust & Aly, 1983). It is the most commonly used coagulant for use in water and wastewater treatment throughout the world (Bratby, 1980; Tebbutt, 1998). However, as a consequence of the health concern associated with its use, the EC (1985 Drinking Water Directive) set a MAC of 200 μg/L and a guideline value of 50 μg/L. The UK, as a Member State incorporated this into national law in the Water Supply (Water Quality) Regulations 1989 and the Water Supply (Water Quality) (Amendment) Regulations 1991 and set a prescribed value or concentration (PVC) of 200 μg/L for treated waters (Severn Trent Water, 1999). The WHO, however, does not consider alum to be a chemical of significance to health, but recommends that concentrations should be < 200 μg/L in drinking water (WHO, 1996). The arguments for its use in water treatment are firstly, it is a very effective coagulant. Secondly, the majority of the chemical will form an insoluble precipitate, and is then removed in the solid-liquid separation stage; therefore, only low levels are expected to be present in the treated water. Thirdly, it has been found that higher levels of aluminium can be ingested through our food than in the water supply (although aluminium can be more bioavailable in water than food (Gray, 1994)). Tebbutt (1998) notes that latterly alum is regaining its popularity in some treatment works.

Alum has many advantages over other chemical coagulants, as discussed in Section 2.2.2. At a price of £55 per ton (Laporte Absorbents, 1998), it is one of the cheapest coagulants available in the UK. It is readily available in most developed countries, as well as being fast acting and very effective on waters with low turbidity and/or high colour. However, many developing countries must import alum at considerable cost, if it is not manufactured locally. Alum also has the disadvantage of creating voluminous floc causing rapid filter clogging, being pH and alkalinity dependent, and potentially leaving residual in the treated water.

5.2.1 Mechanism of floc formation and previous research on alum

The aqueous chemistry of aluminium salts is complex. The hydrolysing aluminium ion dissociates into Al^{3+} in water and then hydrates to form aqua-metal complexes. Then, during a

series of hydrolytic reactions monomeric, dimeric and polymeric species are formed. There are two mechanisms of destabilisation of particles using alum. Firstly, the monomeric, dimeric and polymeric species remain in solution long enough to attach to particles and cause destabilisation by charge neutralisation. Secondly, an amorphous precipitate is formed by the reaction of the aluminium ion with the hydroxide in the water, causing entrapment of the particulate matter. In this case the hydrolysis products are not the end products, but rather an intermediary of the eventual precipitation of the metal hydroxides. The particles are thus destabilised by charge neutralisation and by enmeshment. As to which mechanism dominates, is dependent upon the pH of the water and the dose of coagulant used (Gregory, 1978), as discussed in Section 2.2.2 and shown in Figure 2.8.

There is a plethora of research on the use of alum in water treatment, but it is beyond the scope of this research to review it in its entirety. Rather, consideration is given to the use of alum in CFF and DF, in order to compare with the results obtained from the laboratory study. The results of studies comparing *M.oleifera* with alum in conventional treatment process are also considered.

Typically in the UK, DF and CFF are used for the treatment of upland coloured waters. These waters tend to have an increase in organic matter during the autumn months. In order to obtain water of an acceptable quality, in such situations, high doses of alum must be used. The concern is then that the filter runs become shorter and uneconomical due to media clogging (Graham *et al.*, 1992). Moreover, the residual aluminium in the treated water may exceed the UK limit of 200 μg/L. There is therefore a reluctance with many operators towards using high doses of alum in CFF treatment (Britton & Cochrane, 1989). Consequently, some researchers do not recommend the use of alum with CFF (Edzwald *et al.*, 1982 & 1987), while others recommend doses of no higher than 2.5 mg/L (Coccagne, 1987). Accordingly, there has been a shift towards using dual coagulants: alum as a primary and a polyelectrolyte as a secondary coagulant. Alternatively, alum is replaced with a polyelectrolyte, which produces less voluminous floc when destabilising humic material (Packham, 1973). The drawback is that polyelectrolytes are less effective than alum, unless high concentrations are used. This is undesirable from an economic point of view.

In full scale trials in Malawi, comparisons were made between *M.oleifera* seed and alum (Sutherland *et al.*, 1994). The treatment works consisted of two upflow contact clarifiers,

followed by rapid gravity filters. The influent water entered a central chamber where chemical addition, rapid mixing and preliminary flocculation occurred. The coagulated/flocculated water entered a secondary mixing zone. The decrease in upflow velocity promoted a combination of suspended floc blanket formation and settling, with the clarified water passing upwards through the blanket into a collecting launder. A flow rate of 60 m^3/h was maintained throughout the works. The raw water turbidity during the trial period was about 300 NTU. With a dose of 75 mg/L of *M.oleifera* seed, a filtrate turbidity of < 3 NTU was maintained for over 6 hours. With a dose of 50 mg/L of alum, a filtrate turbidity of < 1 NTU was produced for over 6 hours (EERG, 1993). These results indicated that the two coagulants produced comparable turbidity removal within a conventional treatment train.

Comparative jar tests using *M.oleifera* seed and alum on laboratory prepared low turbidity waters, consisting of 2 minutes rapid mix, 15 minutes slow mix and 20 minutes settling, indicated that alum was marginally more effective than *M.oleifera* seed in terms of turbidity removal (McConnachie, 1993). Comparative tests on a natural raw water (with a pH of 8.5) in Sudan found that 200 mg/L of alum reduced a 470 NTU water to 11 NTU, and a 40 mg/L dose reduced a 75 NTU water to 3 NTU. Whereas *M.oleifera* seed reduced the same waters to 16 and 10 NTU with doses of 200 and 50 mg/L respectively (Jahn, 1991).

5.2.2 Comparative removal rates in the laboratory study using alum and *M.oleifera* seed

The comparative study of these two coagulants was made using the dual media beds and a low loading rate of 5 NTU at 5 m/h and a relatively high loading rate of 75 NTU at 10 m/h. A summary of the main results obtained are shown in Figures 5.17-5.22. The main observations were:

- Turbidity removal and the rate of ripening with alum were greater than with *M.oleifera* seed for both operating conditions. This was particularly so with the higher loading rate.

- The headloss development rate was higher with alum than with *M.oleifera* seed, for both operating conditions. The distribution of headloss development was similar with the two

coagulants at the lower loading rates. With the higher loading rates, the headloss distribution throughout the bed was greater with *M.oleifera* seed.

The mode of floc formation induced by alum is different to that using *M.oleifera* seed, and the subsequent extent of removal and the deposit morphology are different. Plate 11 shows a sample of high turbidity water (about 300 NTU) after jar testing using alum. Although individual floc are not visible in this plate, a layer of sludge at the base of the jar can be seen. Thus showing the nature of the large settleable floc produced by alum. Metal salts are generally considered to be more effective at particulate removal in low turbidity waters than are polyelectrolytes (Kawamura, 1991b). The aqua-metal complexes enmesh particles into their structure. Since floc formation with alum is a chemical not a physical process, it is independent of the number of collisions between the coagulant and the particulates. Whereas, the 'patch' mechanism of charge neutralisation (as induced by *M.oleifera* seed) operates more effectively with higher particular concentrations, to increase the collision rate.

This study shows that *M.oleifera* seed and alum had similar removal rates with the lower turbidity water (5 NTU), as seen in Figure 5.17, and substantially different rates with the higher turbidity water (75 NTU), as seen in Figure 5.20. For the low turbidity water, a filtrate turbidity of < 0.1 NTU was achieved for longer than 11 hours with both coagulants. With the higher turbidity water, however, *M.oleifera* seed produced a filtrate of < 5 NTU, and alum < 0.1 NTU for 2 hours.

There are a number of possible explanations for this variation in performance of the two coagulants, as shown in Figures 5.17 and 5.20. The filtration rates as well as the raw water turbidities were different. As discussed in Section 5.1.3, the filtration rate affects the mixing intensity prior to the water entering the bed and thus the efficiency of particle aggregation. Metal coagulants require an intense mixing period after addition, flash mixing, to ensure the formation of the hydrolysed precipitate and its adsorption onto the particulates (Amirtharajah & Tusler, 1986; Kawamura, 1991b). It is conceivable that at 10 m/h, mixing conditions were optimised for alum, whereas at 5 m/h the mixing intensity was too low for optimal floc production, and therefore removal was only marginally greater than with *M.oleifera* seed. It is reported in the literature that the optimum G value for metal salts should be 300 s^{-1} and t = 1 minute (Collins *et al.*, 1987; Ashan *et al.*, 1996). At lower than optimal G values, the coagulant is not dispersed uniformly, causing local over- and under-dosing to occur. The floc

thus formed have a low density, and are weak and voluminous (Twort *et al.*, 1985). Thus removal rates may have improved if the mixing intensity was higher, regardless of the flow through the filter media (as discussed in section 5.1.2 using chitosan).

The intensity of the rapid mix stage with *M.oleifera* seed is not critical, since the reactions that occur are not hydrolytic. The filtration rate would, however, affect the retention time in the filter bed and the extent of in-pore flocculation. It was observed from jar testing that *M.oleifera* floc took longer to form compared with alum, chitosan and other cationic (synthetic) polyelectrolytes. Other workers confirm this (Jahn, 1991; McConnachie, 1993). Therefore, higher removal rates were achieved at 5 than at 10 m/h when using *M.oleifera* seed and not with alum, due to the increase in filter bed retention time.

Ripening of the filter bed in Figures 5.17 and 5.20 was faster with alum than *M.oleifera* due to the larger floc produced. Moreover, the addition of alum increased the solid load on the filter, due to the formation of the precipitate. The deposition of floc in the bed causes an increase in the surface area available for subsequent particle attachment. Rapid filter bed ripening is a significant factor in the treatment process where the initial filtrate produced after backwashing is discarded due to its low quality. Optimal removal conditions, as defined in Section 2.5, occurred to a greater extent with alum than *M.oleifera* seed. This can be seen at the higher loading rates, T_1 and T_2 (as defined by Figure 2.12), were closer to occurring simultaneously with the use of alum than with *M.oleifera* seed.

The use of alum in conjunction with CFF and DF is not recommended for a number of reasons: Firstly, alum floc do not form strong bonds with the filter media, and since relatively high filtration rates are used, the floc will detach from the media surfaces (Adin & Rebhun, 1974). Secondly, these coagulants tend to form voluminous floc, which lead to surface blinding of the filter if a clarification stage is not included prior to filtration. The literature reports that where single media filter beds are used in CFF, the filter runs become inefficient, especially with alum as the coagulant (Graham *et al.*, 1992). The headloss development rates in the runs with alum in this research were unexpectedly low. This was particularly so at the higher loading rate, as shown in Figure 5.21, where headloss was expected to be the cause of termination. Although the headloss development in the top 16 cm of the bed was 18% of the total headloss development with alum as compared to 12% with *M.oleifera* seed, the entire bed was active.

The main factor in the low headloss development rate with alum was considered to be the dual media bed.

The use of a deeper layer of anthracite may have increased the turbidity removal rates with alum, as has been suggested by other workers, whilst maintaining a relatively low rate of headloss development. A study by Logsdon *et al.* (1993) found than a filter bed of 2 m of anthracite was more effective than a 0.9 m dual media bed when using alum and polyelectrolytes. This study concluded that the greater pore volume allowed deeper penetration of the particulates and consequently a lower headloss development rate. Furthermore, the extended depth of anthracite provided a storage area for the voluminous floc, thus reducing surface filtration and early filter bed clogging. This was not considered to be the case with *M.oleifera* seed, as described in Section 4.5.1. Rapid headloss development was not an issue and therefore a deeper layer of sand at the expense of the anthracite layer was considered to be potentially beneficial.

The greater headloss development with alum than *M.oleifera* seed, as seen by comparing Figures 5.18 with 5.19, and 5.21 with 5.22, was expected, since higher removal rates were achieved with alum. Additionally, larger floc were produced with alum than with *M.oleifera* seed. Examination of Figure 2.8 shows that conditions in the pilot plant (raw water pH of 7.5 and alum dose of 15 - 17.5 mg/L) would have induced floc formation by 'sweep' coagulation and to a lesser extent, by charge neutralisation. Therefore the large voluminous floc which formed caused surface clogging and removal to occur, and rapid rise in headloss development rate. This trend of rapid headloss increases with alum was also found by other workers (Grutsch & Mallatt, 1977; Rebhun *et al.*, 1983; Nasser *et al.*, 1995).

For the 5 NTU raw water treated at 5 m/h, the turbidity removal in the top 16 cm of the bed was marginally greater with alum than with *M.oleifera* seed, as seen in the insets of Figures 5.18 and 5.19. This was expected since floc formation was more rapid and the floc were larger. The removal rate with *M.oleifera* seed was higher in the section of bed between 16-40 cm, indicating that more of the bed was active than was the case with the use of alum. The difference in particulate deposition patterns between alum and *M.oleifera* seed was not as great as between chitosan and *M.oleifera* seed. Indicating that *M.oleifera* floc were more similar in size and/or attachment characteristics to alum floc than chitosan floc.

5.3 Conclusions of the study comparing coagulants

The following conclusions were drawn from the comparative studies between chitosan and *M.oleifera*, and alum and *M.oleifera*.

- Chitosan was effective in the treatment of water with a turbidity of 75 NTU at 10 m/h in dual and single media filter beds. Whereas, *M.oleifera* only effectively treated the 75 NTU water in the dual media bed. The 5 NTU water was effectively treated by both coagulants at 5 m/h. Optimisation of the mixing intensities and media configurations may have improved turbidity removal and deposition morphology with chitosan, since surface clogging was found to be a serious problem in all the treatment conditions considered.

- Chitosan is known to form floc by the 'bridging' mechanism (Huang & Chen, 1996), evidence for which was the strong inter-particle and particle-collector bonds apparent in this study. Chitosan also formed floc by enmeshment into a precipitate. The chitosan solution is made up in acid because it is only soluble at low pH, but on dosing into a raw water with a pH of 6-7, the chitosan is precipitated out, forming solid material that entraps the particles by the 'sweep' floc mechanism. Evidence for this was the rapid floc formation encountered in the jar tests and the pilot plant, coupled with the high removal rates with the low turbidity raw water.

- The effect of increasing the filtration rate was to improve removal rates with chitosan, but the opposite was the case with *M.oleifera*. This was considered to be due to the variation in mixing intensity prior to the water entering the bed, and the retention time in the filter bed. Lower filtration rates effected a lower mixing intensity for a longer period. This resulted in larger chitosan floc forming and blocking the surface layer of the bed. Further mixing of the water beyond the point of destabilisation may cause restabilisation by the long chained polymer folding over the surface of the particle. Therefore extended mixing periods would have resulted in restabilisation and reduced turbidity removal.

- Increases in filtration rates were detrimental to removal with *M.oleifera*, due to the reduction in the retention time in the filter bed; this would have reduced in-pore flocculation, a significant factor in removal with *M.oleifera* due to its low molecular weight. Also, since attachment

bonds formed by *M.oleifera* are comparatively weak, higher filtration rates could cause the detachment of previously removed particles, thereby effecting early breakthrough of turbidity.

• Headloss development was considerably higher with chitosan than with *M.oleifera*. This was a consequence of higher removal rates, larger floc, formation of a precipitate and stronger attachment bonds to the collector surfaces with chitosan. These factors combine to give increased removal in the surface layers of the bed. Further optimisation work on the use of chitosan in CFF therefore should include pH adjustment of the raw water to ensure chitosan remains in solution. Smaller floc would form which could penetrate deeper into the bed.

• As natural coagulants, *M.oleifera* seed and chitosan have many advantages, especially for developing counties. These include a potential reduction in costs due to the use of local indigenous materials; use of local labour for production; use of the sludge as a fertiliser. The optimum choice of coagulant would depend on location of the treatment works, availability of labour, type of water to be treated and the treatment process in use.

• Alum was effective in the treatment of waters with a turbidity of 75 NTU at 10 m/h, and also of 5 NTU at 5 m/h. *M.oleifera* seed was less effective at the higher loading rates. This was due to the difference in floc formation mechanisms and in the mixing intensities. Optimisation of the mixing intensities prior to the water entering the bed could potentially improve turbidity removal with alum.

• Filter bed ripening occurred more rapidly with alum than with *M.oleifera* seed, (at both raw water turbidities and filtration rates.) This was due to the formation of larger alum floc, acting as better collectors for the in-coming particles. This may be significant if the initial filtrate is discarded until the working stage is reached.

• The relatively low rate of headloss development in the dual media bed tests, with alum was considered to be due to the anthracite layer. Surface removal was minimal and headloss distribution throughout the bed was similar to that encountered with *M.oleifera*.

• The headloss development rate was greater with alum than with *M.oleifera* for two reasons. Removal rates were higher, and also alum produced a more voluminous floc. Larger floc cause

clogging of the pores to a greater extent than the smaller *M.oleifera* floc, and therefore preventing deep bed filtration from occurring.

◆ Thus alum and chitosan were more effective as coagulants than *M.oleifera* seed in terms of turbidity removal. But the rates of headloss development were considerably lower with *M.oleifera*. Since the choice of optimum coagulant is determined by cost effectiveness of treatment as well as the quality of the filtrate, the duration of the filter run is an essential consideration. Therefore the use of coagulants that effect rapid headloss development can be very costly due to the reduction in output before backwashing is required. The optimum choice of coagulant is therefore dependent upon the particular raw water being treated, the filtration rate and the design of the filter bed in terms of the headloss which can be tolerated before termination is necessary. Clearly other factors such as cost and availability of the coagulant are also essential factors in determining the optimum coagulant. If metal coagulants such as alum are used then the cost of pH adjustment chemicals must also be considered. In addition chitosan and alum produced a more voluminous sludge than *M.oleifera* seed. A consequence of charge neutralisation by the 'patch' mechanism is that the subsequently formed floc consists of particles that are in close proximity of each other. Thus the floc contain less water than would be present in floc formed by the 'bridging' mechanism or 'sweep' flocculation. Dewatering and subsequent disposal are therefore less complex with *M.oleifera* sludge than chitosan and alum.

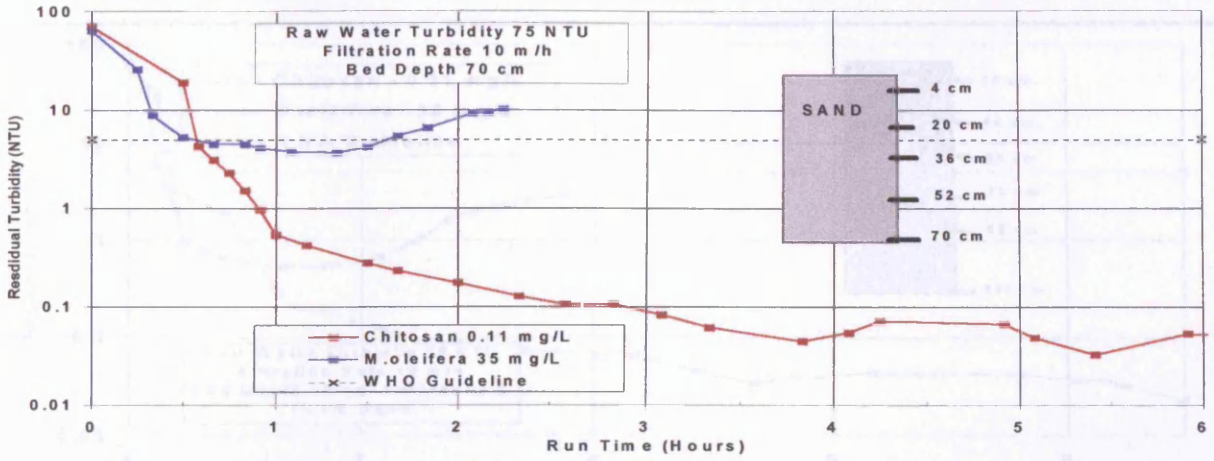

Figure 5.1 Comparative turbidity reduction with M.oleifera seed & chitosan; raw water turbidity 75 NTU, 70 cm sand.

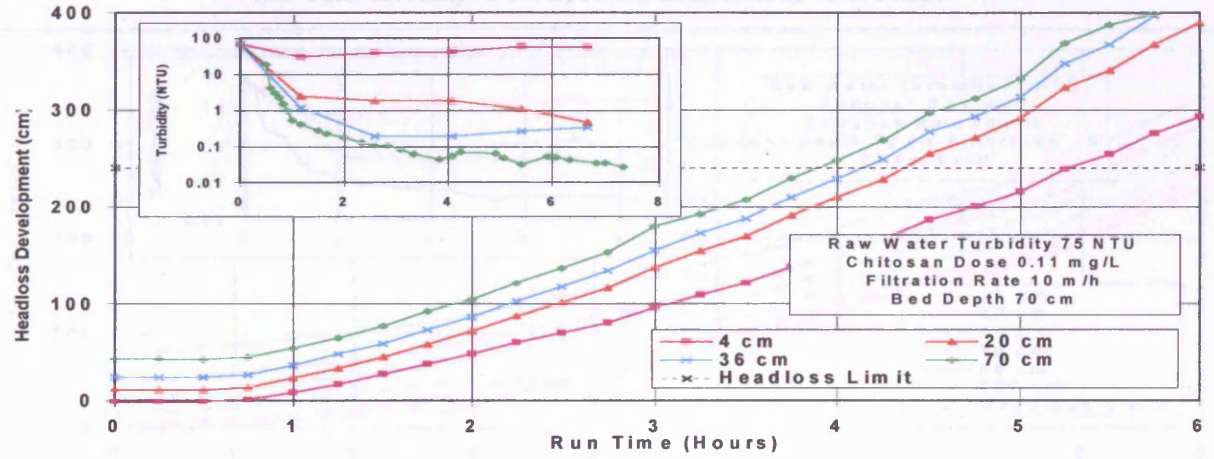

Figure 5.2 Headloss development and turbidity reduction as a function of bed depth; using chitosan, raw water turbidity 75 NTU, 70 cm sand.

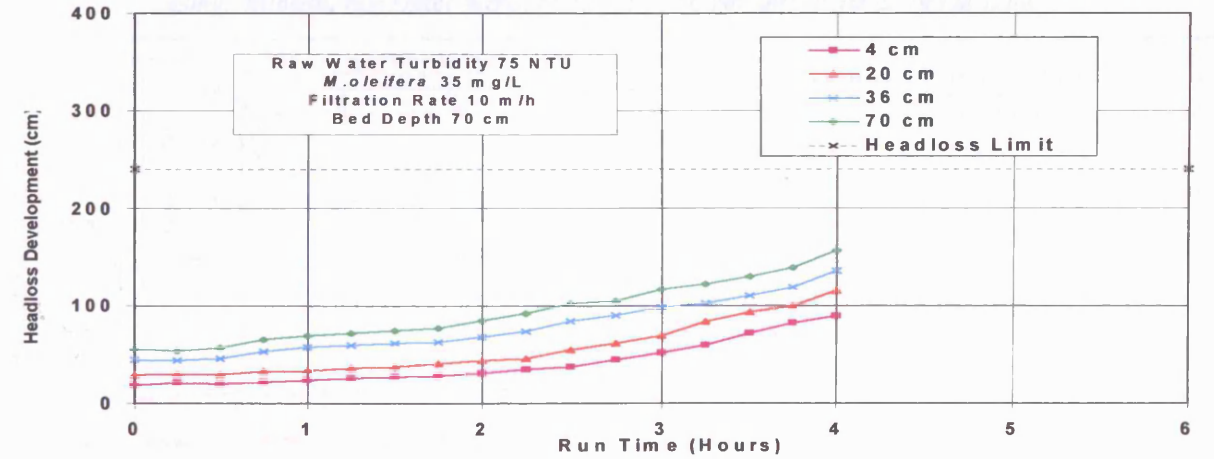

Figure 5.3 Headloss development as a function of bed depth; using M.oleifera seed, raw water turbidity 75 NTU, 70 cm sand.

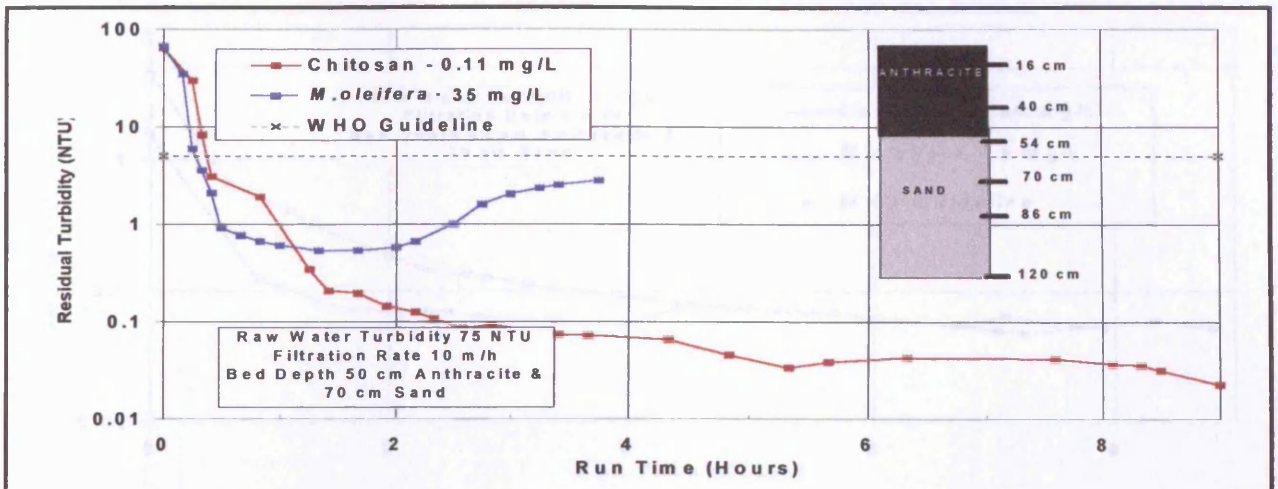

Figure 5.4 Comparative turbidity reduction with M.oleifera seed & chitosan; raw water turbidity 75 NTU, 50 cm anthracite & 70 cm sand.

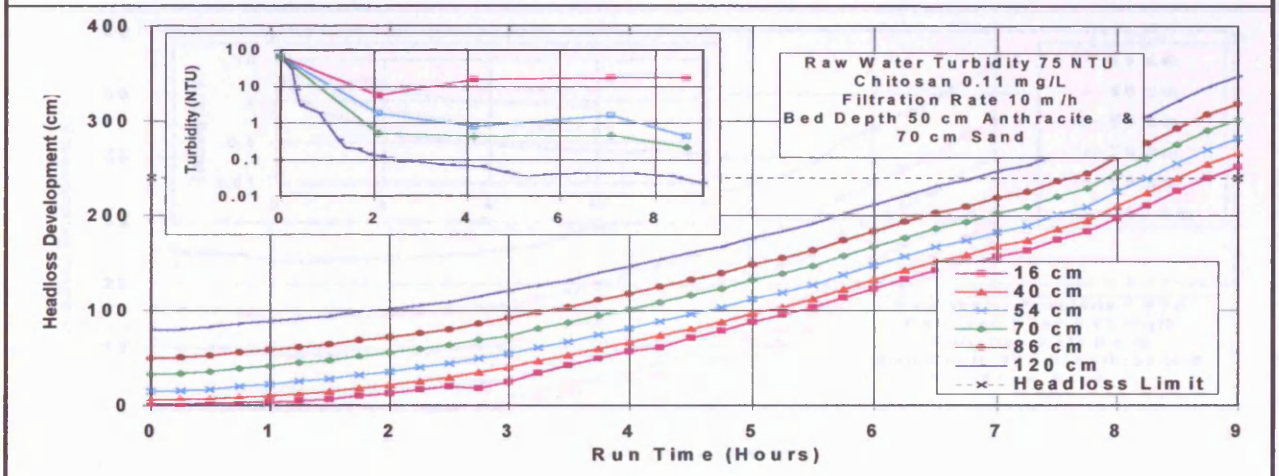

Figure 5.5 Headloss development & turbidity reduction as a function of bed depth; using chitosan, raw water turbidity 75 NTU, 50 cm anthracite & 70 cm sand.

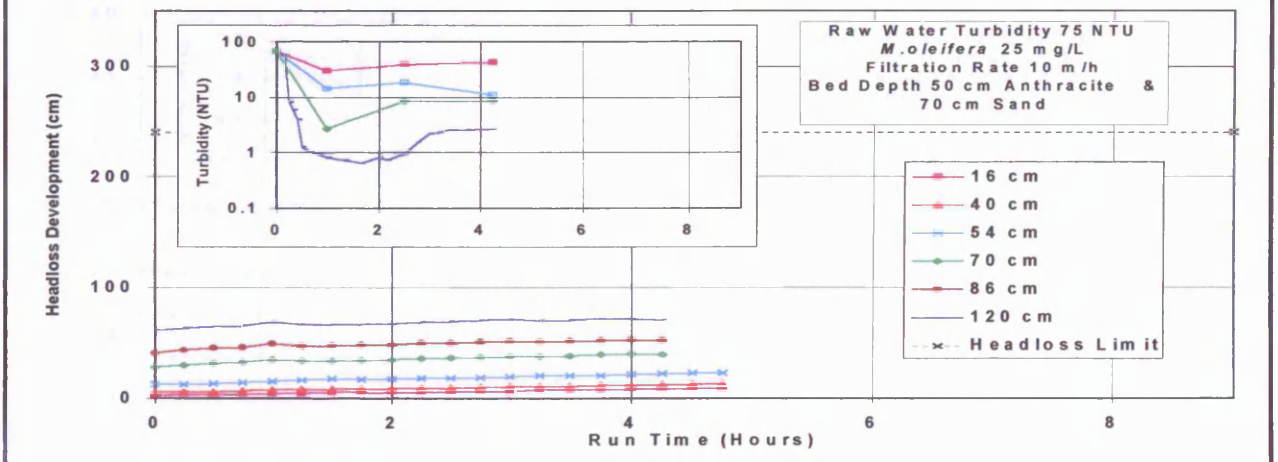

Figure 5.6 Headloss development and turbidity and reduction as a function of bed depth; using M.oleifera seed, raw water turbidity 75 NTU, 50 cm anthracite & 70 cm sand.

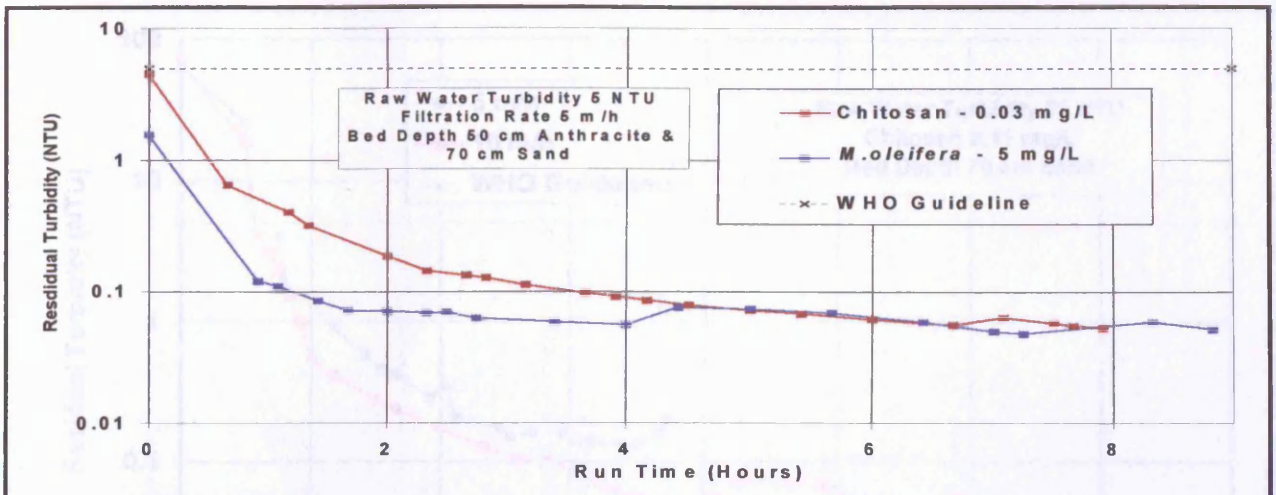

Figure 5.7 Comparative turbidity reduction with M.oleifera seed & chitosan; raw water turbidity 5 NTU, 50 cm anthracite & 70 cm sand.

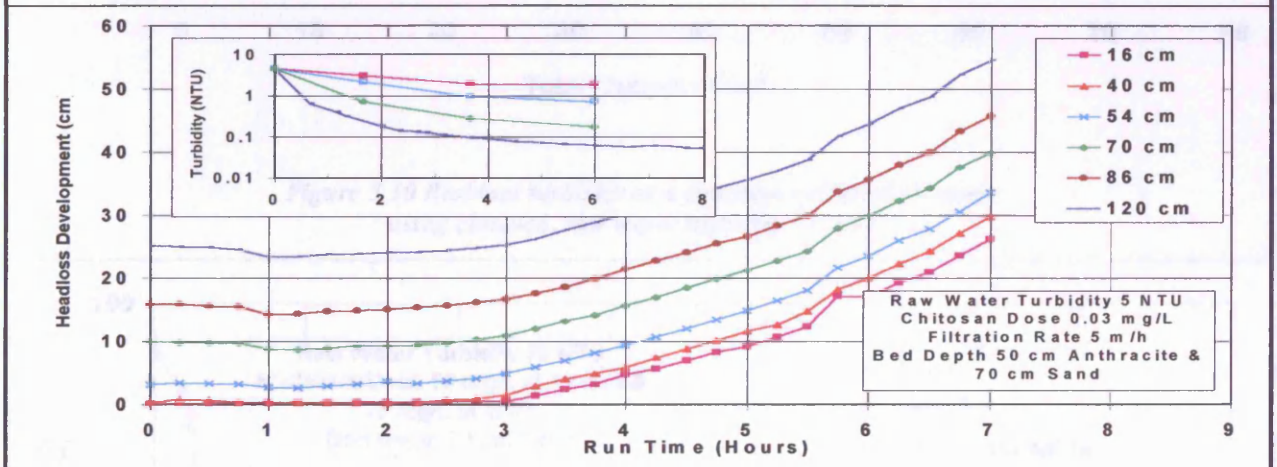

Figure 5.8 Headloss development and turbidity reduction as a function of bed; using chitosan raw water turbidity 5 NTU, 50 cm anthracite & 70 cm sand.

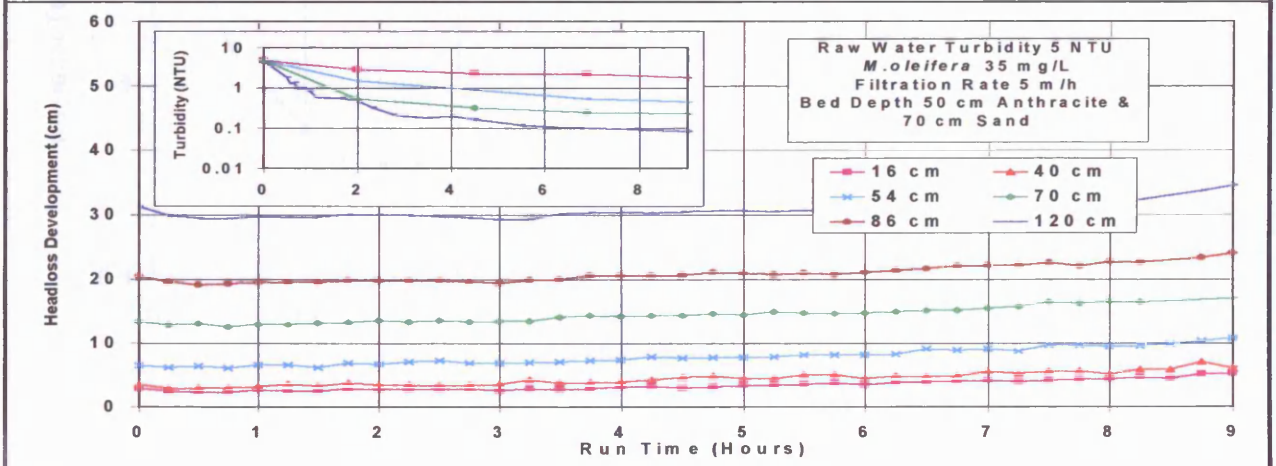

Figure 5.9 Headloss development and turbidity reduction as a function of bed; using M.oleifera seed, raw water turbidity 5 NTU, 50 cm anthracite & 70 cm sand.

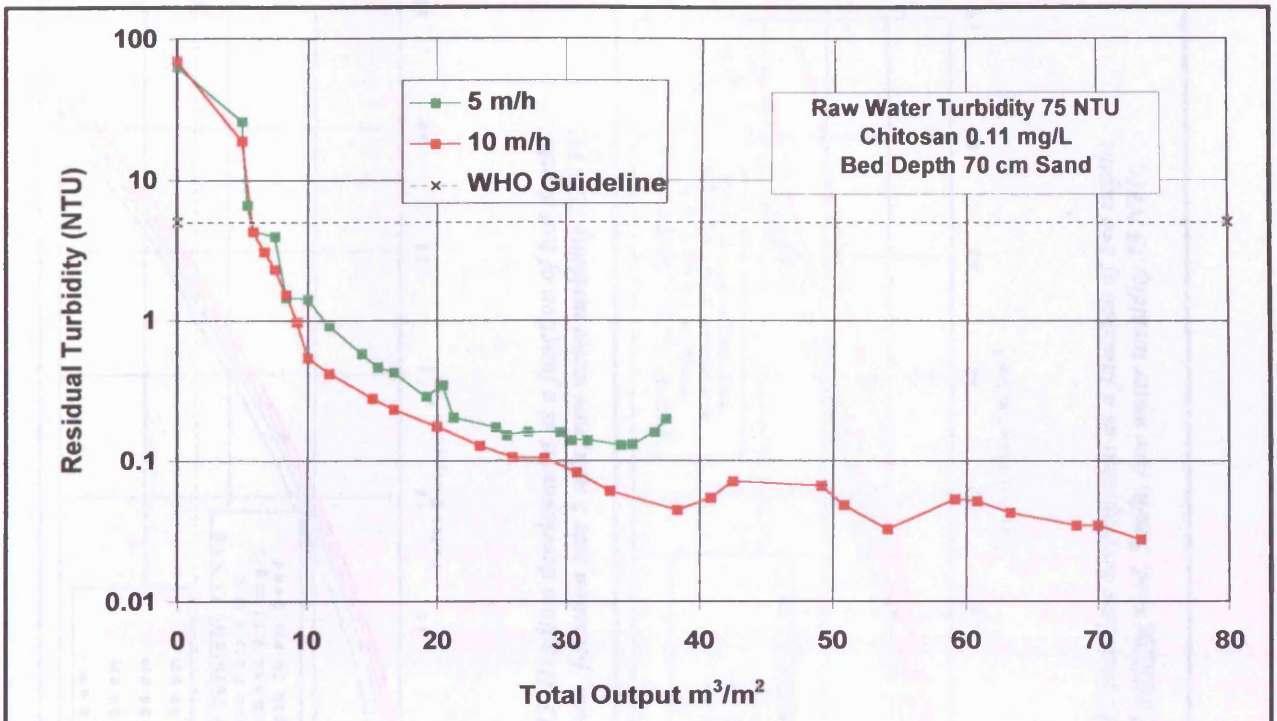

Figure 5.10 Residual turbidity as a function of filtration rates; using chitosan, raw water turbidity 75 NTU.

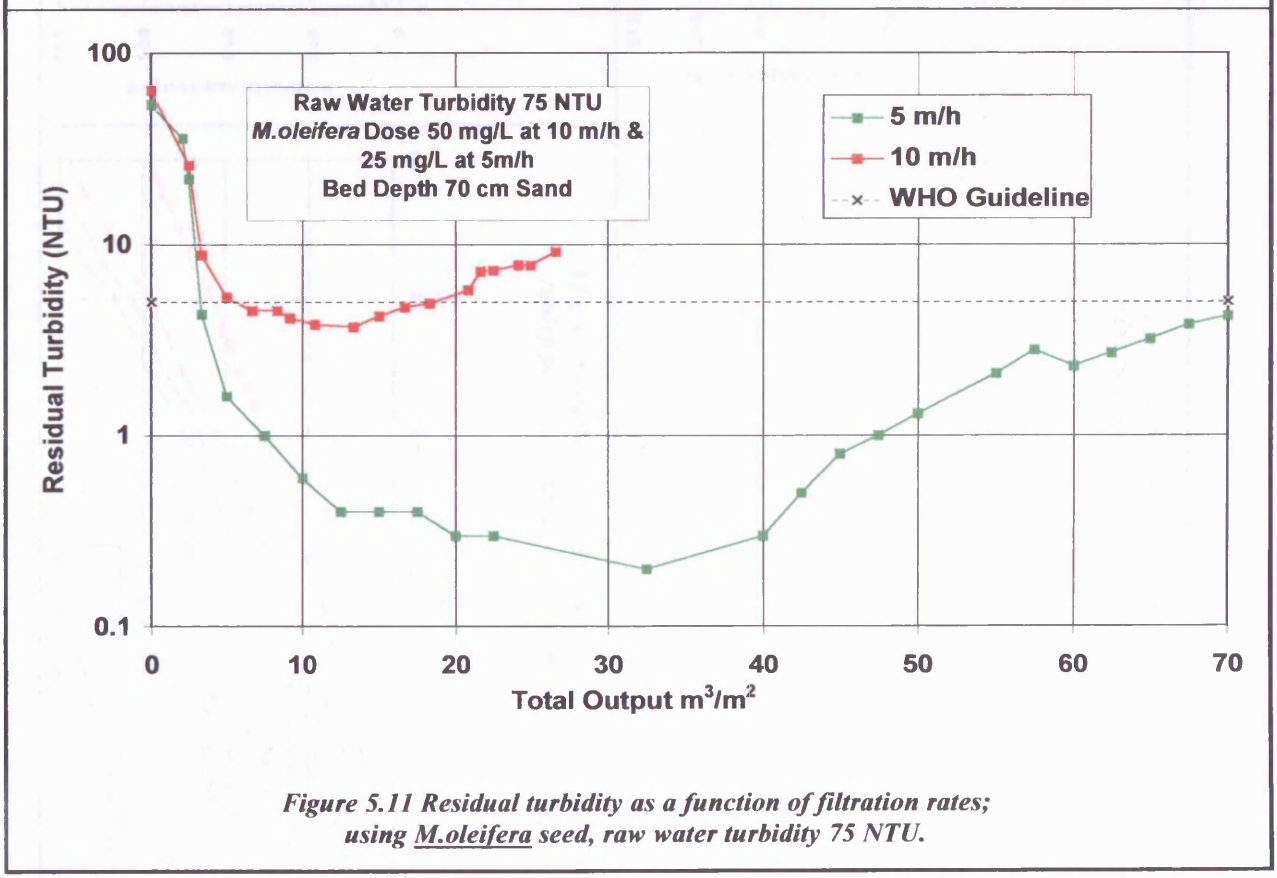

Figure 5.11 Residual turbidity as a function of filtration rates; using M.oleifera seed, raw water turbidity 75 NTU.

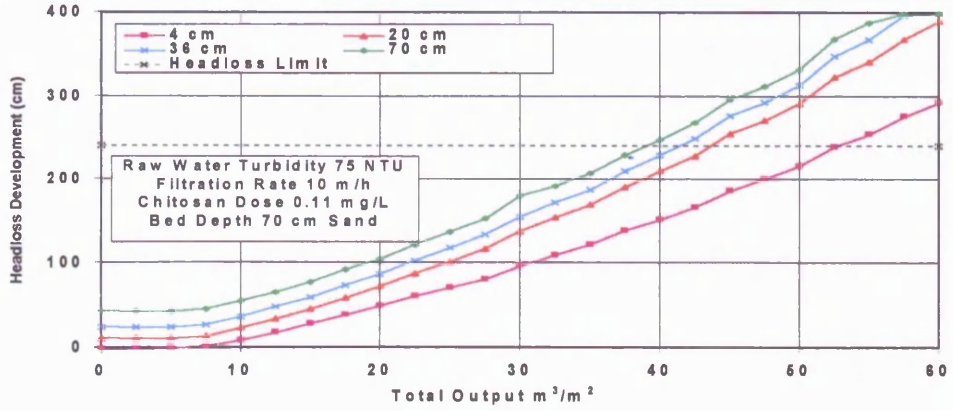

Figure 5.12 Headloss development as a function of bed depth; using chitosan, filtration rate 10 m/h, raw water turbidity 75 NTU

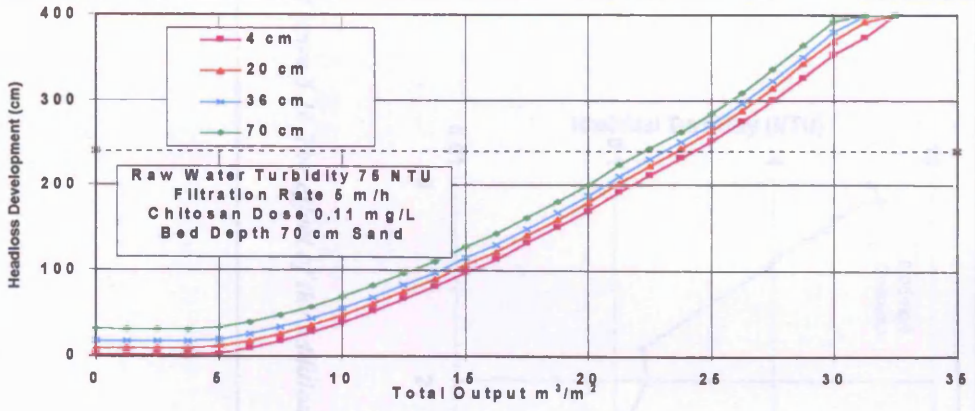

Figure 5.13 Headloss development as a function of bed depth; using chitosan, filtration rate 5 m/h, raw water turbidity 75 NTU.

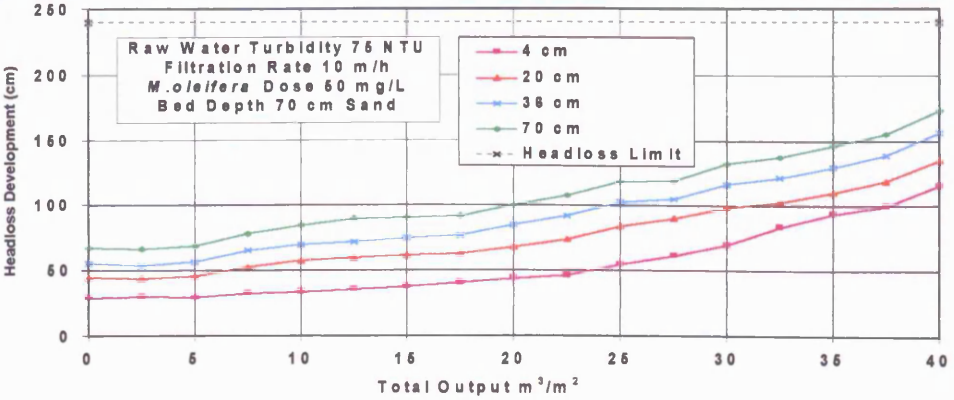

Figure 5.14 Headloss development as a function of bed depth; using M.oleifera seed, 10 m/h, raw water turbidity 75 NTU.

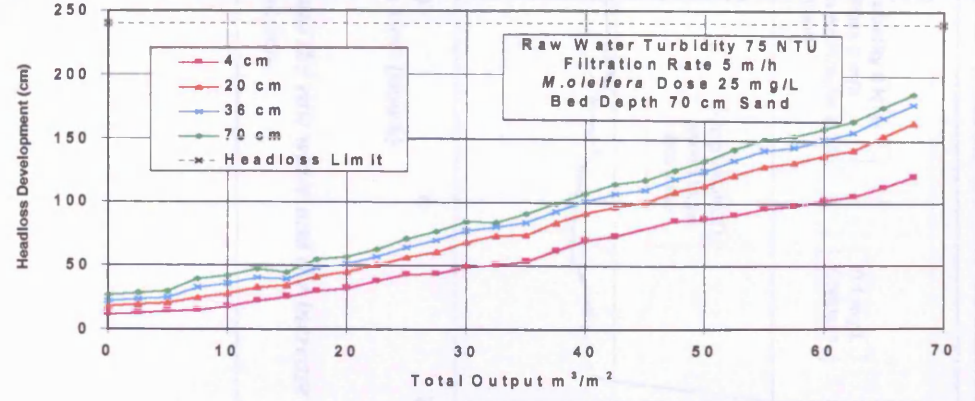

Figure 5.15 Headloss development as a function of bed depth; using M.oleifera seed, 5 m/h, raw water turbidity 75 NTU;

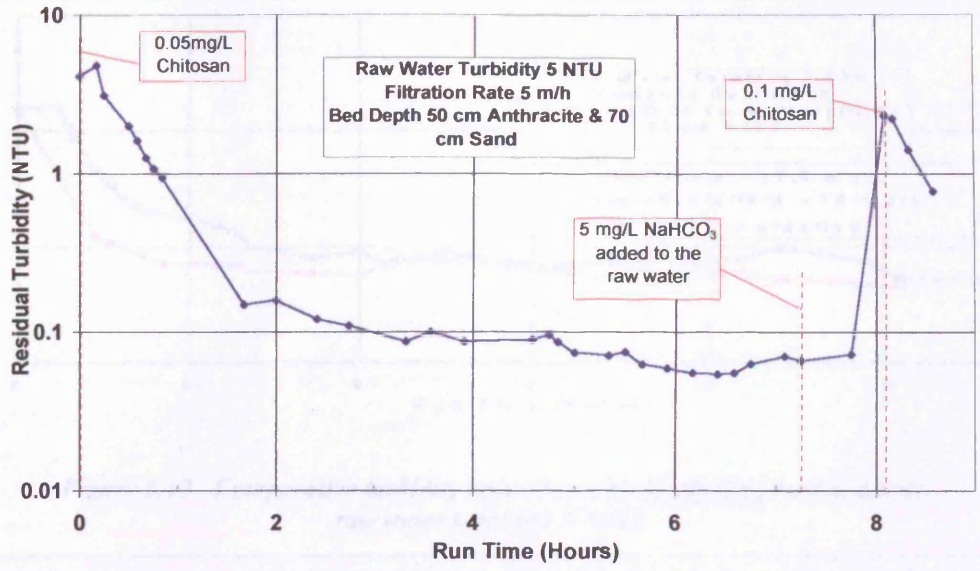

Figure 5.16 The effect of the addition of alkalinity into the raw water and the increase in dose on turbidity reduction.

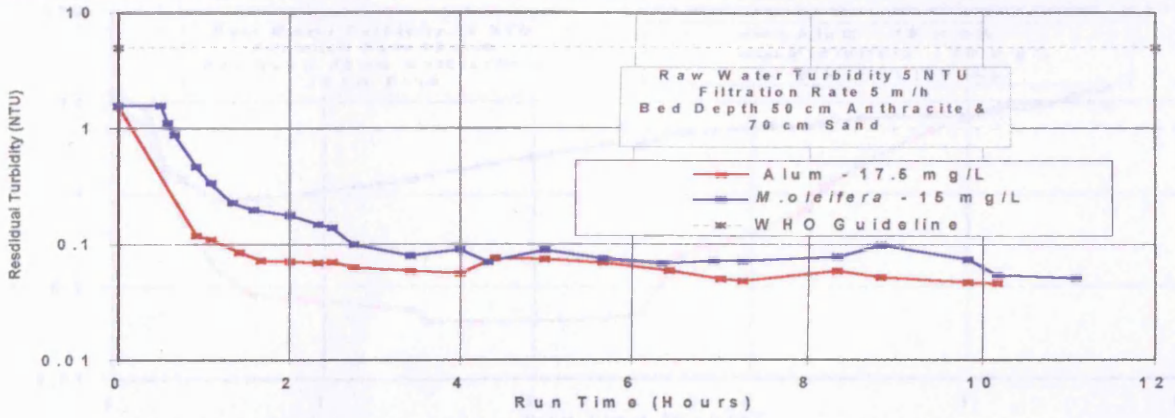

Figure 5.17 Comparative turbidity reduction with M.oleifera seed & alum; raw water turbidity 5 NTU.

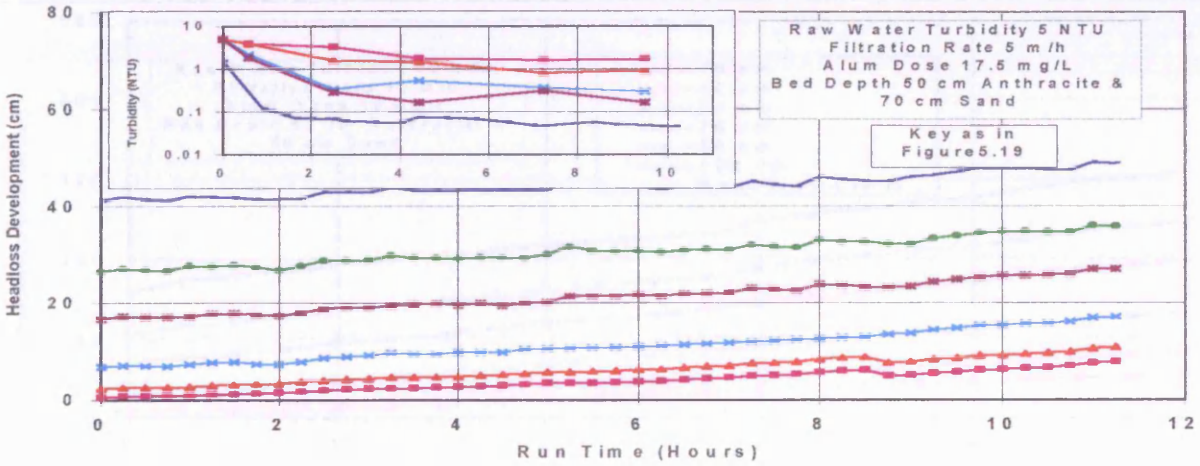

Figure 5.18 Headloss development and turbidity reduction as a function of bed depth; using alum, raw water turbidity 5 NTU.

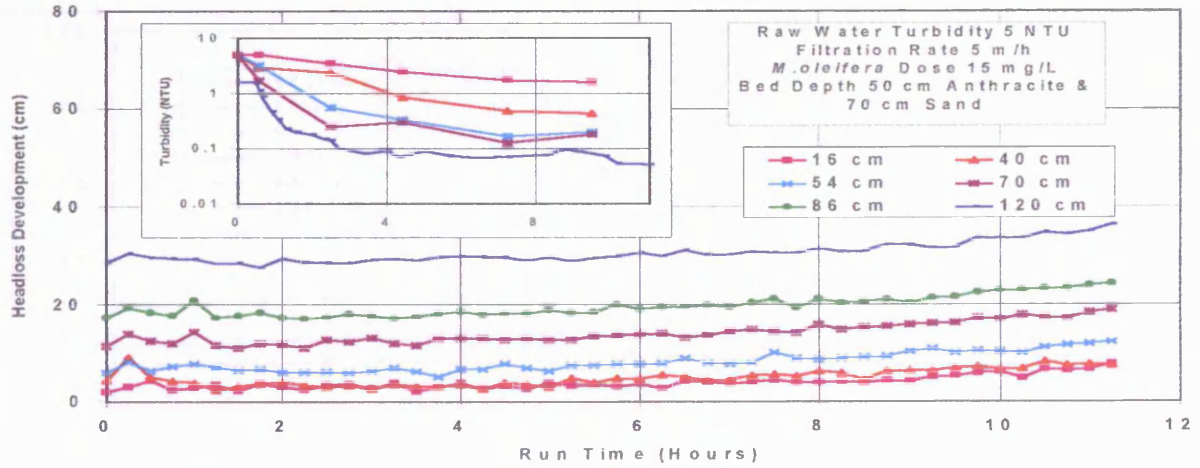

Figure 5.19 Headloss development and turbidity reduction as a function of bed depth; using M.oleifera seed, raw water turbidity 5 NTU.

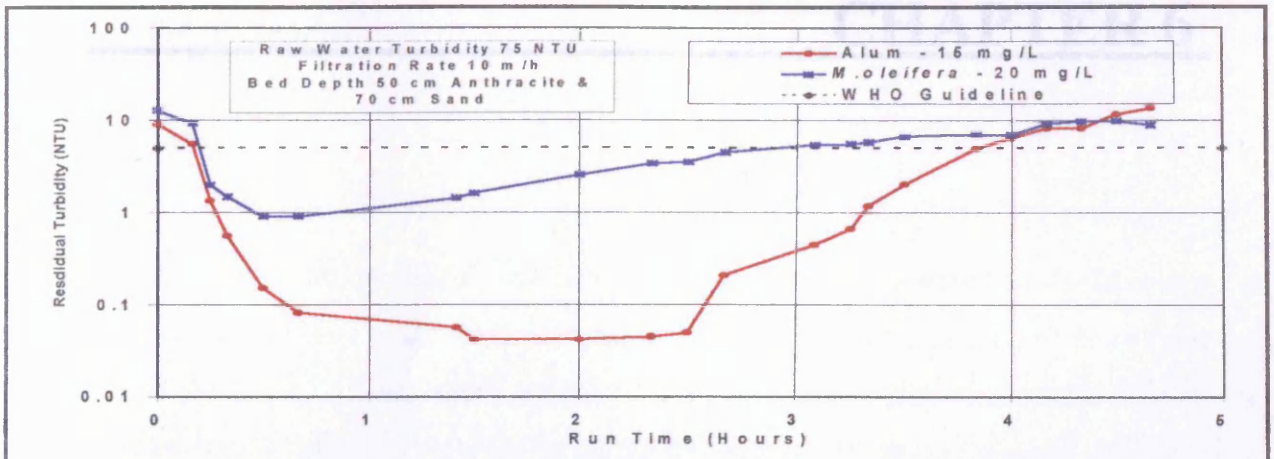

Figure 5.20 Comparative turbidity reduction with M.oleifera seed & alum; raw water turbidity 75 NTU.

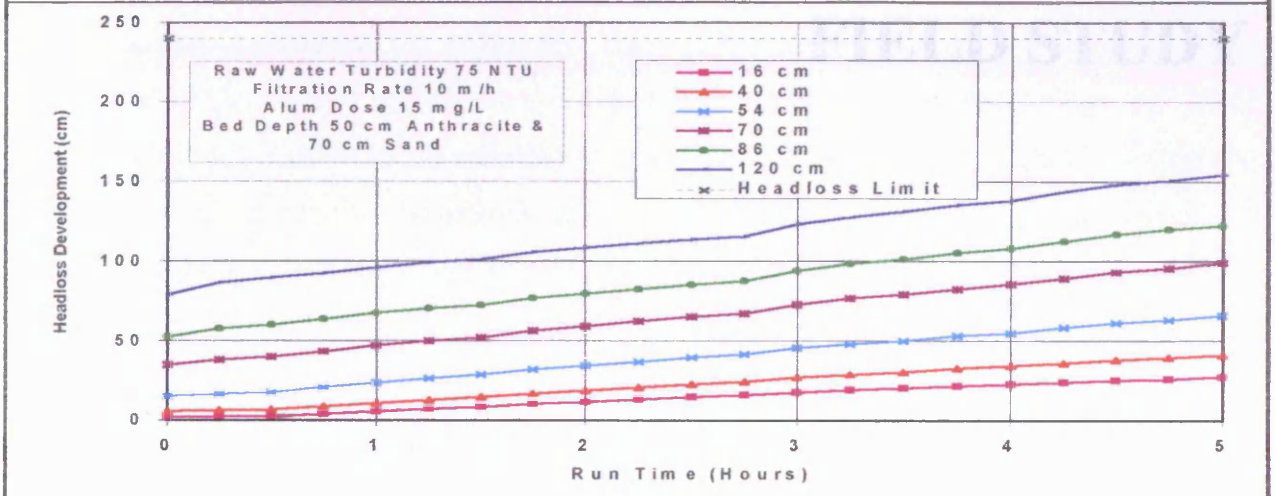

Figure 5.21 Headloss development as a function of bed depth; using alum raw water turbidity 75 NTU.

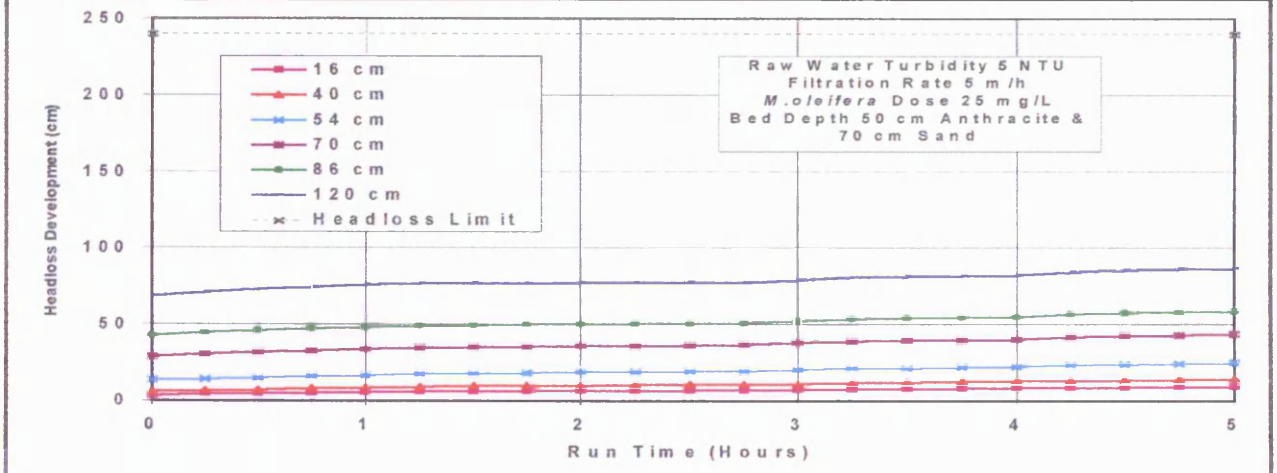

Figure 5.22 Headloss development as a function of bed depth; using M.oleifera seed, raw water turbidity 75 NTU

CHAPTER 6

FIELD STUDY

6.0 Field Study

6.1 Introduction

All the laboratory studies reported in Chapters 4 and 5 were conducted in the laboratory at Leicester University on a small scale pilot rig to examine the process of CFF in conjunction with *M.oleifera*, used an artificially prepared raw water consisting of a kaolin suspension. Natural raw waters are more complex involving interactions of various organic and inorganic compounds. Turbidity in such waters can be caused by clay, silt, plankton, microorganisms and dissolved organics, etc. Compounds that colour waters are also present in natural waters, and are difficult to replicate in the laboratory. Thus CFF field trials using a natural water were conducted to complement the laboratory work. It was decided that the pilot facility at Felindre WTW in South Wales under the auspices of Dwr Cymru Welsh Water, would be an ideal site to conduct a field study. It offered the opportunity to investigate the effectiveness of *M.oleifera* seed in treating a low turbidity (2-20 NTU), coloured water containing organic material. The pilot plant consisted of four filter columns which could be operated in parallel, allowing the investigation of various physical parameters, using one raw water source. Not only were the variables considered in the laboratory study investigated, but the effect of the addition of a separate flocculation stage was also considered. Thus a direct comparison could be made of the operation of this pilot plant in the CFF and DF modes. Furthermore, *M.oleifera* seed could be compared directly with ferric aluminium sulphate (Ferral 2060), the coagulant used in the main works. It was also possible to investigate bacterial and organic matter removal rates using *M.oleifera* seed.

6.1.1 Specific objectives

- To compare the results obtained from operating the laboratory pilot rig using a synthetic water with a natural raw water rather than an artificially prepared model raw water, in terms of optimisation of the physical parameters of operation.

- To compare effectiveness of turbidity removal using *M.oleifera* seed and a proprietary chemical coagulant, Ferral. As the coagulant of choice, Ferral is highly effective in the

treatment of the waters at Felindre WTW. Therefore a comparison of the removal rates achieved with Ferral and *M.oleifera* is a rigorous test for the latter.

- To investigate bacterial removal rates using *M.oleifera* seed. This parameter has previously been investigated using conventional treatment, but not for CFF and DF.

- To investigate removal of organic compounds using *M.oleifera* seed. This is an important parameter because generally small chain polymers, such as *M.oleifera,* are not as effective as metal coagulants at removing such compounds. Furthermore, removal of organic compounds has not been studied previously using *M.oleifera* seed as the coagulant.

- To investigate whether the use of *M.oleifera* seed introduces significant residual organic material to the treated water. As the whole seed is used in the preparation of the dosing suspension, potentially much of the soluble material not active in the clarification process may be present in the treated water as a residual. This is significant for two reasons. Firstly, the organic matter can act as a substrate for pathogens present in the water or the distribution system. Secondly, Chlorination of organic compounds may lead to the formation of CBP.

- As a consequence of the above, the dissolved organic carbon and the THM content of the final water were measured.

- To directly compare the effectiveness of removal rates using *M.oleifera* seed in CFF and DF. The main works at Felindre is operated as DF, but it was considered by the operators that the flocculation stage may in fact be superfluous to the treatment process.

6.2 The Towy Scheme

Felindre WTW is situated about 10 miles north of Swansea and is at the centre of a system called the Towy Scheme. This scheme was developed in the 1970's to supply drinking water to the population in and around Swansea, Port Talbot and Neath. It was later extended to supply Llanelli and Kidwelly to the west and Bridgend, Llantrisant and the outskirts of Cardiff to the east. The Towy Scheme has six main constituents, as illustrated in the schematic in Figure 6.1 and described below:

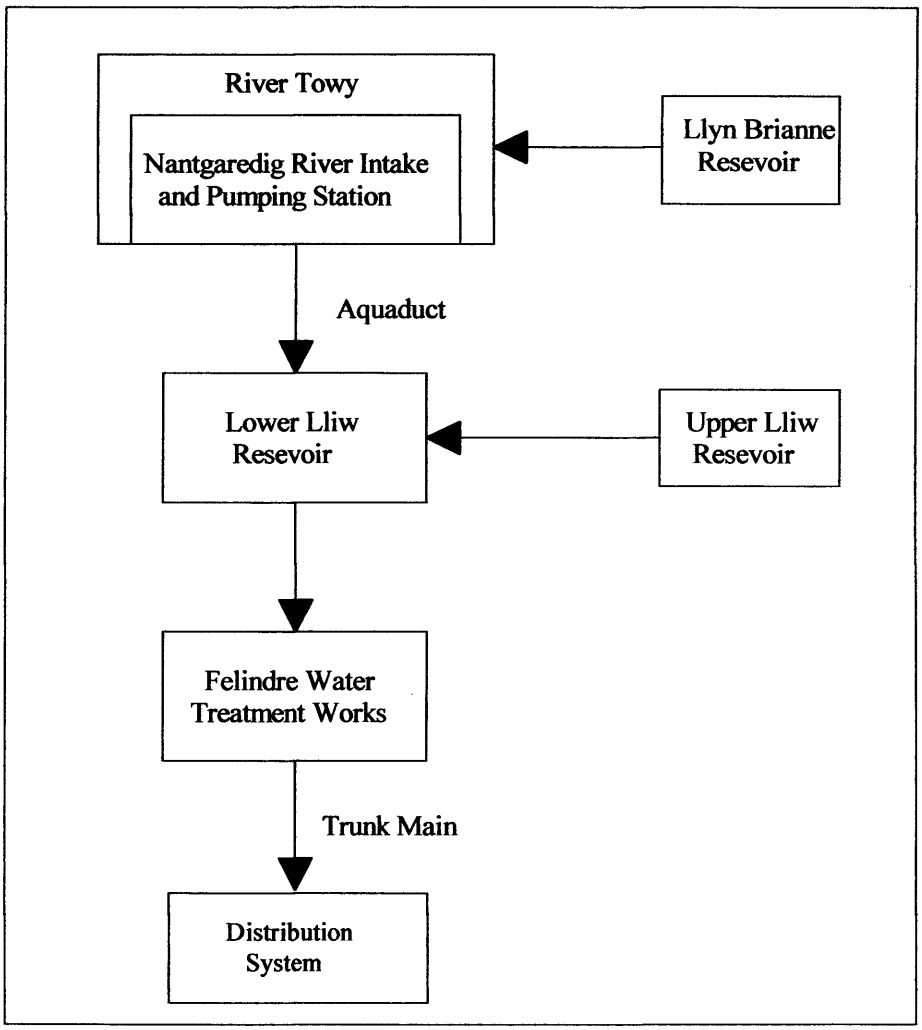

Figure 6.1 Schematic of the Towy Scheme.

• Llyn Brianne storage reservoir - This clay core reservoir is situated 300 m above sea level. It stores 64,300,000 m^3 of water, which is an eight day supply. This is necessary in case of a

pollution incident in the river or failure of the pumping station farther down the line. Its function is to regulate the flow into the River Towy.

- Nantgaredig river intake and pumping station - Water is taken out of the River Towy at this point and pumped at a rate of 250,000 m^3 into an aqueduct which flows in to the Lower Lliw Reservoir.

- Lower and Upper Lliw Reservoirs - Raw water from the river Towy is stored at the Lower Lliw Impounding Reservoir with a capacity of 1221,000 m^3; thus providing a minimum of seven days supply against the possibility of failure of the aqueduct or a major pollution incident in the River Towy. The Upper Lliw provides additional storage for water collected from the local catchment. Both these reservoirs can be seen in Plate 12.

- Felindre WTW - This is discussed in Section 6.2.1 in greater detail.

- Trunk main - This carries the treated water from Felindre WTW to the various service reservoirs ready for distribution.

6.2.1 Felindre WTW

The treatment works is illustrated schematically in Plate 13. It has a maximum output of 220,000 m^3/day. The treatment process consists of coagulation, flocculation, filtration and disinfection, and is thus defined as DF for the purposes of this study. On entering the treatment works the water is dosed with the coagulant Ferral 2060, and hydraulic mixing ensures the even distribution of the chemical throughout the raw water. The coagulated water then passes into the raw water contact tanks, which contains baffles to ensure a slow mix stage for floc formation.

Twelve rapid gravity filters are the next stage in the treatment process. Each filter is capable of processing up to 25,000 m^3 per day at an average filtration rate of between 3.5-6.2 m/h. The filters consist of a treble layer of filter media over a double layer of support media.

After filtration the water enters the filtered water contact tank, with a capacity of 2,700 m^3, for the disinfection stage. Chlorine is dosed at the inlet of the tank, at about 1.2 mg/L, with a

contact time of about 30 minutes. At the outlet from the tank, sulphur dioxide is added at a concentration of about 0.2 mg/L (sulphonation). This reduces the free chlorine concentration to an acceptable level. Ammonia is then added to the water at a concentration of 0.2 mg/L. This reacts with the free chlorine to form chloramines. The advantage of having these in the treated water rather than chlorine is that they form very effective disinfectant residuals, that remain effective for long periods of time. They are also more efficient at disinfecting sediments and biofilms in the distribution system, and preventing the chlorine from reacting with organic compounds causing taste problems. Lime is added to raise the pH of the final water to about 8.7, thus stabilising the water and minimising its corrosive action on iron pipework. Finally phosphoric acid is added to reduce the ability of the water to dissolve lead. The water then enters one of two treated water service reservoirs, with a combined capacity of 150,000 m^3. This would be sufficient to supply water for up to 12 hours in the event of a complete shutdown of the works.

Cleaning of the filters involves air scouring the filter bed and then backwashing with the raw water. The washwater is sent to the washwater recovery tanks, consisting of 6 hopper-bottomed tanks with a storage capacity of 500 m^3. The water is dosed with 2 mg/L of polyelectrolyte to aid settling. The rapid flow of the water into the tanks ensures an even distribution of the coagulant. Floc is formed by the gentle agitation achieved by the tank filling. A 30 minute settlement time is allowed. The supernatant is then pumped via a floating arm outlet to the Works' inlet for recycling and full treatment. The sludge formed at the base of the tank is pumped to the sludge thickening tanks where further dewatering occurs. Finally, the sludge is pumped to drying lagoons on site, where it dries to form a thick cake, which can be disposed of more easily (Welsh Water, 1997).

Ferral 2060, manufactured by Laporte, is essentially aluminium iron sulphate. Its exact chemical formula is variable because it is manufactured by the extraction of ferric and aluminium from clay using sulphuric acid. The proportion of these chemicals is approximately in the ratio of 2:8. Magnesium is also present, which does not affect treatment. Ferral contains about 50-70 g Al_2O_3 (water soluble aluminium) per kg of solution and 10-30 g Fe_2O_3 (water soluble iron) per kg of solution. The manufacturer recommends a dose range of 1-100 mg/L as product. The usual dose at the Works is between 14-18 mg/L as product, giving an average annual expenditure on Ferral of about £106,000. Posifloc PW92, the polyelectrolyte used in the washwater recovery tanks. It is an anionic polyacrylamide produced by O.C.I. Ltd.

The media in the filter beds consists of support media and filter media. The base layer of the support media consists of 75 mm of coarse gravel in the size range 13.2-6.7 mm, followed by another 75 mm layer of finer gravel in the size range 6.4-2.5 mm. The first layer of the filter media is 425 mm of silica sand, classified as BS mesh size 14/25 (sand size range 0.60-1.18 mm), with a UC of 1.3-1.7. The second layer is 280 mm of anthracite classified as BS grade 2 (sand size range 1.2-2.5 mm), with a hardness of between 2.75-3.25 on the Moh Hardgrove Index, a UC of < 1.5 and a specific gravity of between 1.35-1.45. The sand and the anthracite were supplied by Waterforce UK. The top layer is 15 mm of granular activated carbon (GAC), is graded as PK 3-5 (sand size range 3-5 mm), with a UC of < 1.5 and an apparent bulk density of < 300 g/L. The carbon is manufactured and supplied by Norrit Ltd.

6.2.3 The raw water at Felindre WTW

The bed rock in the Towy Scheme catchment area consists mainly of old red sandstone and some limestone. There tends to be a high concentration of humic and fulvic substances in the ground around the reservoirs which feed into the system. These are responsible for the relatively high colour at certain times of the year of the raw water at Felindre WTW. Acid rain has a significant effect on the water quality of Llyn Brianne. Research undertaken by the Environment Agency (UK) into reasons why game fish were not surviving in Llyn Brianne and its outlets, revealed that the pH of the water was too low. Consequently, in the six months prior to the study, lime had been dosed into two tributaries of the reservoir in order to raise the pH of the water (Owen, 1997). The water also tends to have high concentrations of manganese and iron at intervals throughout the year. The source of these metals is thought to be the Upper Lliw reservoir, since the water pumped from the River Towy does not contain significant concentrations of either of these.

Table 6.1 indicates that the raw water at Felindre WTW is of relatively low turbidity and high colour throughout the year. It can also be seen that the raw water quality is variable, since the source is an impoundment reservoir and seasonally dependent. The Lower Lliw reservoir buffers the effect of increases in turbidity in the River Towy, by reducing and delaying the effect for a few days. The colour in the water is caused by the leaching out of humic substances from the soil and the presence of algae. The filamentous algae *asterionella* is present in the water throughout the year, but the numbers are limited by the lack of phosphorus and/or

nitrogen in the water. In spring and early summer these nutrients become available as a result of runoff from the land, and mixing of the lake strata due to temperature inversions in the lake. In the spring the water temperature rises and the light intensity increases; all of which cause an increase in algal concentrations. Numbers of *asterionella* were relatively low during the early part of the study but increased towards the end, promoted by the rapid increase in temperature and reduction in rainfall in late June and July. When these filamentous algae are present in large numbers, they form a thick mat on the surface of the filter bed, causing very rapid increases in headloss, shorter filter runs, and consequently more frequent backwashing. Other occasional problems with the raw water include high manganese and iron concentrations, which can exceed the maximum allowable concentrations in the final water supply (Owen, 1997).

Water Quality Parameter	Raw Water Quality Throughout 1997	Raw Water Quality (during study period)			Treated Water Quality (during study period)			UK PVC[1]
		May 1997	June 1997	July 1997	May 1997	June 1997	July 1997	
Turbidity (NTU)	1-10	1.1-6.0	2.0-8.5	0.8-1.7	0.1	0.1	0.2- 1.0	0.4
Colour (^{0}H)	2-30	15-30	15-25	15-25	< 2	< 2	2	20
DOC (mg/L)	0.9-5.7	1.6-3.4	2.0-3.6	1.2-3.2	1.3-1.7	1.1-1.7	1.0-1.5	nsl
pH	6.5-8	6.3-7.7	7.4-7.5	7.1-7.5	7.8-8.0	7.6-8.1	7.7-8.6	6.5-8.5
Manganese (mg/L)	0.01-0.06	0.019-0.038	0.040-0.063	0.032-0.046	0.013-0.022	0.010-0.063	0.007-0.600	0.05
Iron (mg/L)	0.10-0.40	n/a	n/a	0.135-0.198	0.024-0.340	0.029-0.042	0.010-0.049	0.3
Aluminium (mg/L)	0.02-0.2	0.031-0.203	0.065-0.160	0.031-0.097	0.035-0.057	0.030-0.720	0.024-0.088	nsl
Total coliforms /100 ml	10-1400	10-650	110-750	6-330	0	0-1	0-2	0
E.coli/100 ml	0-800	10-350	70-710	0-150	0	0-1	0-2	0
HPC [2] (1 day at 37^{0}C /ml)	0-3000	1980	50-440	1-1290	n/a	n/a	1	nsl

Table 6.1 Raw and treated water quality at Felindre WTW.

[1] The Prescribed Value or Concentration (PVC) are the limits set by the UK Water Supply (Water Quality) Act 1989 (Severn Trent Water, 1999), but most WTW would aim to achieve lower levels.

[2] Heterotrophic Plate Count (see Section 6.3.2.3)

n/a : not available.

nsl : no set limit.

6.3 The pilot plant and its operation

6.3.1 Description of the pilot plant

Schematics of the plant can be seen in Figures 6.2 and 6.3, illustrating the operation of the plant in the CFF and DF modes. The plant consisted of two 0.1 m^3 header tanks, which had baffles placed in them to create flocculation tanks when operated as DF. The tanks were raised 3 m above the ground to allow the 4 columns to be gravity fed. Connecting the tanks to the columns were 12 m delivery pipes, which in common with all the connecting pipework consisted of 12.5 mm braided pipe. The columns were constructed of μPVC and were 3 m in length with an internal diameter of 0.15 m (as can be seen in Plate 14). Each column outlet is connected to a flow controlling tank, 0.4 m^3 in volume; the flow being controlled by a float valve. This setup allowed a constant flow into the columns regardless of the extent of headloss development. The outlets from each of the tanks were connected to rotameters with flow controlling taps which allowed the flow rate to be set. Taps were located after each rotameter to allow sampling, and the remaining flow went to waste.

Chemical dosing was achieved by the use of variable speed peristaltic pumps connected to a power supply. The various doses were achieved by adjusting the voltage on the power supply, and also by altering the concentration of the coagulant. The dosing pumps were calibrated before each run. Mixing of the raw water and the coagulant was achieved using a static mixer in the pipe immediately after dosing.

Headloss was measured by the use of clear plastic tubing connected to the top and bottom of each column. In two of the columns the pressure differential was measured directly from the difference in height of the water in the tubes. In the other two columns pressure differential cells were used to measure headloss. The pressure readings were converted into an electrical signal in the cell, and then into centimetres of headloss which was shown on a display unit.

Filter backwashing was achieved by connecting water from the mains to the outlet piping on each column via manually controlled valves. The upward flow of water through the bed fluidised and cleaned the filter media. This was continued until the turbidity of the washwater was < 2 NTU. The media used in the pilot plant was identical to the sand and anthracite used

in the main plant (see Section 6.2.2). The depths used were single media beds of 70 and 120 cm of sand, and a dual media bed of 70 cm of sand and 50 cm of anthracite.

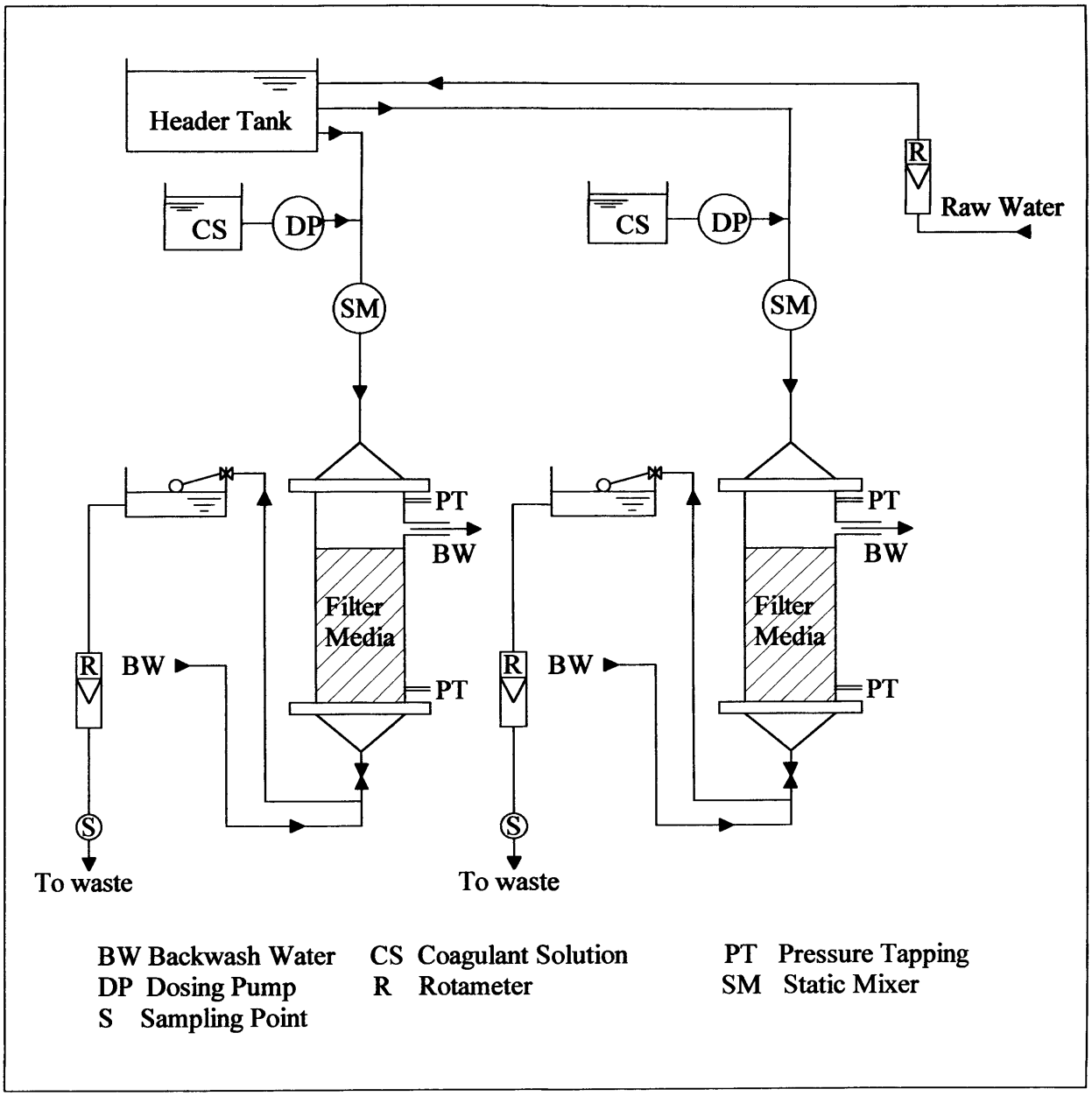

Figure 6.2 Schematic of the pilot plant in CFF study.

(Second twin column set-up is identical to that illustrated).

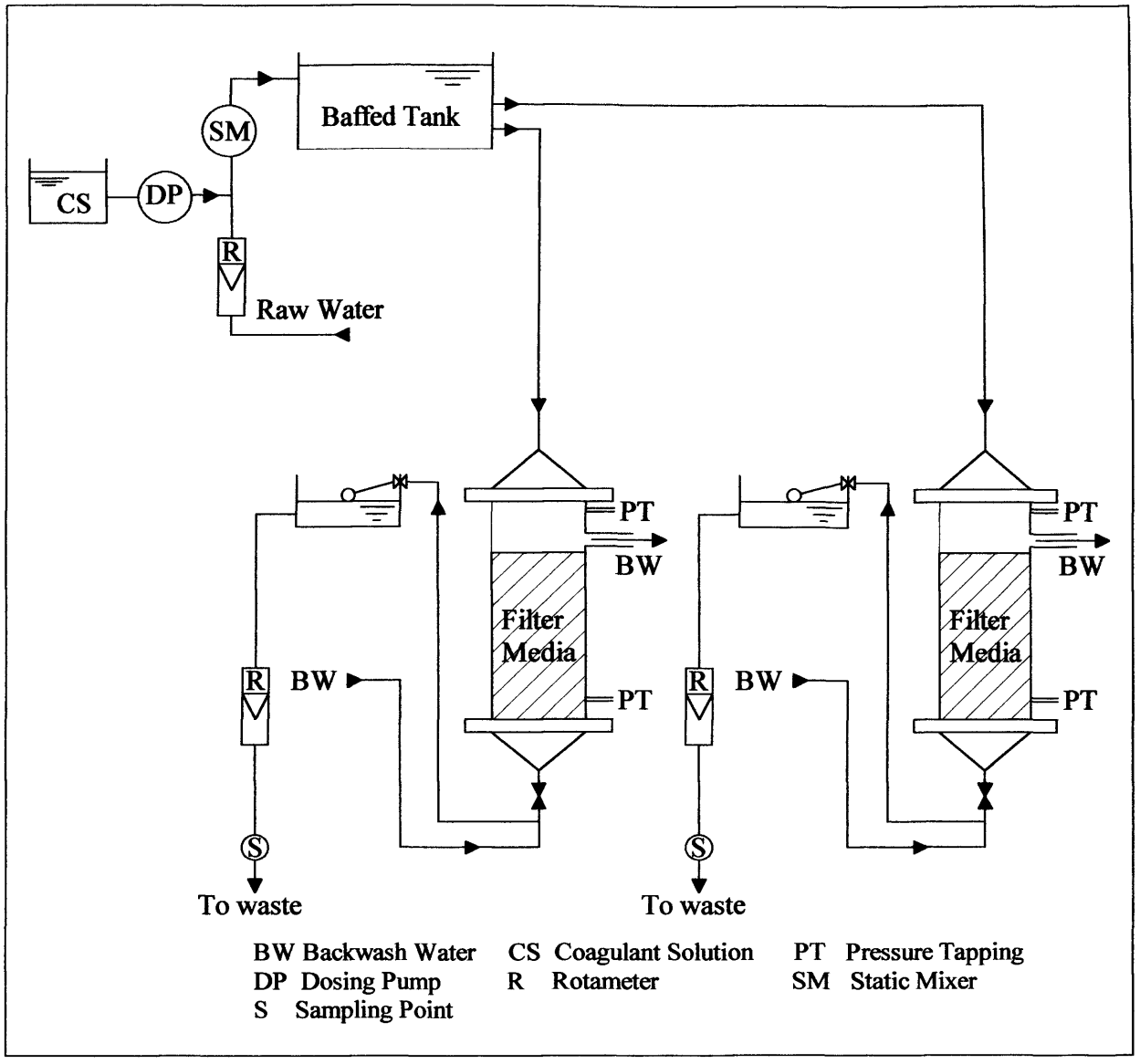

Figure 6.3 Schematic of the field pilot plant in the DF study.

(Second twin column set-up is identical to that illustrated).

6.3.2 Operation of the pilot plant

Prior to each filter run, the pilot columns were backwashed, to remove both the particulate matter and air trapped in the filter media. The filter media was then consolidated by several sharp taps on the sides of the columns. The water manometers and the pressure differential cells were bled to remove any trapped air. The header tanks were filled with the raw water, using the same source as the Works, i.e. the Lower Lliw. The turbidity, pH, temperature and conductivity of the raw water were measured and recorded on entry into the tanks. The taps attached to the rotameters were set to the desired flow rate and the run begun. The filtration

rates used were 5 m/h (88 L/hr) and 10 m/h (176 L/hr). In the DF mode the raw water was dosed with the coagulant then entered one of the two baffled flocculation tanks via a in-line static mixer in the pipe. The residence time in the tank was 68 minutes at a filtration rate of 5 m/h. In the CFF mode, the flocculation tanks had the baffles removed, and dosing of the coagulant occurred as the flow left the tank. The dosed water then passed through the static mixer, and into the delivery pipes. The residence time in the pipes (12 m in length and 12.5 mm internal diameter) was 4 minutes at a filtration rate of 5 m/h. This was found to be sufficient to permit coagulation of the water during jar testing, as discussed later.

Headloss readings were taken immediately after commencement of flow through the filter bed, in order to obtain the clean bed headloss measurements. Turbidity measurements of the filtrate were also taken prior to the addition of the coagulant. The coagulant dosing pumps were then set to the desired dosing rate and activated. Turbidity and headloss measurements were taken at between 30-60 minute intervals throughout the course of the run. Treated and raw water samples were taken once during each run for DOC and bacterial analysis, and on selected runs for THM formation potential.

When the plant was operated as DF, the dosed water entered the static mixer immediately prior to entering the baffled tanks, where flocculation occurred. Subsequently the water passed through the delivery pipes into the columns. It was considered that this 'third' mixing stage may induce shearing and floc disruption. As this could not be avoided, its effect was assessed. Samples of water were taken from the flocculation tank (near the outlet from the tank to ensure flocculation had occurred for the full 30 minutes) and compared with samples taken from the FWZ immediately above the filter bed. It was found on examination that the floc from the sample above the filter bed were slightly larger than the floc from the tank, suggesting the flow through the delivery pipe aided the flocculation process and did not disturb it.

Simulation of the three mixing stages (coagulation, flocculation and potential mixing in the delivery pipes) in the pilot plant was undertaken using a jar stirrer in the laboratory. Again a water sample was taken from near the outlet of the flocculation tank, rapid mixed in the jar stirrer at 257 rpm for 4 minutes and then slow mixed at 30 rpm for 20 minutes. Thus simulating flocculation in the filter bed after the potentially disruptive effect of the rapid flow through the delivery pipes. The mixing rate of 257 rpm was used because it was the maximum speed obtainable on the jar stirrer. The slower mixing rate of 30 rpm was based on jar tests

undertaken in the laboratory to simulate conditions in the filter beds of the Works and the pilot plant. Floc breakup did occur to some extent during rapid mixing, but generally reformed during subsequent slow mixing. It was therefore concluded that the flow rate through the delivery pipe did not cause long term floc disruption.

It was not possible to use the same termination criteria in the field study as was used in the laboratory pilot studies for a number of reasons. The raw water turbidity was occasionally below 5 NTU and therefore the filtrate would not have exceeded the limit of 5 NTU even after operation for long periods of time. A terminal headloss of 2.4 m was not practical because the filter columns were gravity fed, and therefore clogging of the filter media resulted in the reduction in flow rates to very low levels before the headloss reached 1.7-2.3 m. Furthermore, using the same termination criteria as the laboratory study would have led to runs exceeding 24 hours at a filtration rate of 5 m/h. This would have resulted in the need to use extra personnel to operate the plant. As this was not possible it was decided that all runs would be terminated after 10 hours, or when the extent of bed clogging resulted in a reduced flow rate. It was considered that operating the plant for such periods of time was sufficient to show trends in the filtration process, although the total output of the runs could not be compared.

Jar tests were undertaken to obtain the optimum coagulant dose for each set of conditions in the pilot plant. Each test consisted of placing 6 one-litre samples of the raw water under the jar stirrers and adding a range of coagulant doses. The samples were then mixed for 2 minutes at 257 rpm and slow mixed at 30 rpm for varying lengths of time to simulate the hydraulic conditions within the FWZ in the column, as shown in Table 6.2. Immediately after mixing, the samples were poured through a glass funnel containing two sheets of Whatman No. 1 filter paper, into a collection vessel. Turbidity readings of the final water were taken and the estimated optimum dose found. When operating the pilot plant, a range of doses around this optimum was used in order to find the actual optimum for the plant. Routinely two filter papers are used by the Works' operators to simulate DF (Owen, 1997), and this was therefore used to simulate conditions in the pilot plant.

	Depth of media (cm)	
Filtration rate (m/h)	**70**	**120**
5	18 mins	12 mins
10	9 mins	6 mins

Table 6.2 Mixing times used in the jar tests based on the depth of the FWZ and the filtration rates.

6.3.3 Preparation of the coagulant solutions

Preparation of the seed suspension consisted of making up 3 L batches of 1% suspension. This involved using 3 litres of tap water and 30 g of seed powder. The seed source and method for preparation of the powder are given in Section 3.2.1. The powder was placed into a mortar, then a small amount of tap water was added and made into a paste. The remaining water was then added and the suspension thoroughly mixed. It was then passed through two layers of muslin cloth in order to remove the larger insoluble remains of the seed. The resulting suspension was allowed to stand for 5 minutes and the clearer supernatant taken off and used as the coagulant suspension.

Preparation of the Ferral solution consisted of diluting the Works' stock solution with tap water to the desired strength, depending on the required dosage and flow rate through the pilot plant. Working solutions of 1-3% of the stock solution were made up in batches of 5L.

6.3.4 The parameters investigated in the study

Various parameters were measured during the course of the field study. Turbidity was measured in the laboratory at Felindre WTW; bacteria, DOC, organics detectable by gas chromatography and CBP were analysed for at the Hyder Environmental Laboratory at Bridgend in Mid-Glamorgan. Full descriptions of the methods of sample collection, storage and analyses used can be found in Appendix 4.

6.3.4.1 Turbidity

The importance of measuring this parameter is stated in Section 1.1. Turbidity was measured throughout the run to monitor the effectiveness of the treatment process, using 20 ml samples in a HACH turbidimeter (2100P). The calibration of the meter was checked once a week using Gelex Secondary Standards provided with the instrument. The PVC for turbidity used at Felindre WTW is 4 NTU, although in practice runs are terminated when the filtrate turbidity reached 1 NTU.

6.3.4.2 Headloss development

The headloss was measured across each column at 30-60 minute intervals throughout the course of the filter runs. Headloss measurements were made using simple piezometric tubes at the top and bottom of the column.

6.3.4.3 Bacterial analysis

Ideally, water treatment is completed with a disinfection stage to render the water potable. However, this is not always the case in developing countries. Removal of bacteria during filtration is thus very important. The microbiological content of a water is important because many communicable diseases are waterborne, e.g. typhoid fever, bacterial or amoebic dysentery, giardiasis, ineffective hepatitis and poliomyelitis. It is not practical to monitor water for all pathogenic microorganisms, therefore surrogate measurements are used. The commensal bacterial flora of human intestines are used for this purpose, such as total coliforms, faecal coliforms, *Escherichia coli*, faecal streptococci and enterococci. The Heterotrophic plate count (HPC) is used to give a general indication of water quality. Coliforms and *E.coli* are considered to be the most sensitive indicators of faecal contamination (HMSO, 1984).

In this study, the Membrane Filtration Method was used for the enumeration of coliforms and *E.coli*. This involved filtering a measured volume of water through a membrane. The pore size is such that the organisms to be enumerated are retained on the surface of the membrane and this is then placed in a selective media for the organisms sought, and then incubated under controlled conditions for a fixed period of time. The number of colonies that then develop on the media is used to enumerate the number of indicator organisms present (HMSO, 1984). The PVC for total coliforms and *E.coli* in the UK Water Supply Act (1989) is 0 numbers/100 ml in 95% of the samples.

The HPC includes bacteria which are pathogens, non-pathogen and opportunistic pathogens, as well as moulds, yeast, etc. There is therefore no direct correlation with the number of pathogenic organisms present, but it is an indication of the microbiological quality of the water sample. The main value of colony counts lies in comparing the results of repeated samples from the same supply and identification of their spatial and temporal variation. A rise in the HPC

following incubation, at 37°C is important since it includes possible human pathogens (WHO, 1996). The Water Supply Act (1989) gives a guideline value of < 10 colony counts at 37°C and states that there should be no significant rise in the counts above the normal background levels.

6.3.4.4 Dissolved Organic Carbon

DOC is defined as the organic carbon which passes through a 0.45 μm silver or glass fibre filter. DOC represents a quantification of the chemically reactive fraction. The majority of the material making up the DOC ranges in size from 1 nm-0.45 μm. It is also possible for some viruses and ultra small bacteria to pass through the filter and be included in the DOC fraction.

Waters with high DOC tend to be coloured due to the presence of natural organic material (NOM), generally imparting a green/yellow/brown colour, which cause aesthetic, taste, odour and health problems. NOM consists of humic and fulvic substances, which originate from peat in the soil and decaying organic matter (Montgomery, 1985). These can act as a substrate for the growth of potentially pathogenic organisms and can lead to the formation of THM following chlorination (Rook, 1974). NOM may also form complexes with certain metals, thereby increasing their solubility and bioavailability to humans.

NOM makes up about 50% of the total dissolved organic carbon in most natural waters, and as described previously consists of humic and fulvic substances, generally in the ratio of 4:1. Humic substances are high molecular weight compounds with low charge densities, and can be colloidal or soluble depending on the pH of the water. The fulvic acids are low molecular weight compounds with relatively high charge densities. The concentration of humic substances in most rivers and stream is in the range of 1.0-4.0 mg/L. About 30% of the DOC consists of hydrophilic acid, a relatively unknown group of compounds consisting of complex polyelectrolytic acids with carboxyl and hydroxyl functional groups. Carbohydrates make up about 10% of the DOC, and are important because they act as substrate for bacteria and fungi. This group originates from decaying plant material releasing the polysaccharides, which are broken down by soil microbes to release simple sugars and oligosaccharides. These are then leached out of the soil and into the aquatic system and are generally highly correlated to the algal concentrations. The remaining 10 % of the DOC consists of carboxylic and amino acids, and hydrocarbons. The latter only make up about 1% of the DOC in water, but are

nevertheless a very important constituent for two reasons. Firstly, methane - the simplest hydrocarbon, contributes significantly to the carbon cycle of all aquatic systems, and therefore contribute to the growth of many organisms in the water. Secondly, many hydrocarbons are of anthropogenic origin, such as chlorinated hydrocarbons, and are of potential health hazards (Thurman, 1986).

The DOC of the raw water at Felindre WTW ranged from 2 to 3.5 mg/L during the study, which is relatively low for a surface water. This is due to the oligotrophic nature of the water source at Llyn Brianne. Raw water DOC can vary from 0.5 mg/L for some ground waters to about 30 mg/L for highly coloured waters. It is generally considered that the DOC of the raw water should be less than 6 mg/L for direct or contact flocculation filtration to be effective treatment processes (Wiesner, 1992). The organic material in water has a stabilising effect on the colloidal matter. The charged functional groups on many organic compounds can impart stability to the suspended matter in the water, as well as interacting directly with the coagulant (Jiang *et al.*, 1996); thus creating a high coagulant demand, and consequently a large volume of material is introduced into the bed. The effect of this is a rapid rise in the headloss development rate if there is no sedimentation stage prior to filtration.

6.3.4.5 Organics detectable by gas chromatography

Awareness has grown in the last 20 years of possible toxicological effects and carcinogenic risks associated with the long term effects of drinking water containing trace quantities of organic compounds. Following their initial detection, there was a rapid development in analytical techniques to quantify them; paralleling this were epidemiological and toxicological studies to asses their health risks (WHO, 1993). The detection, identification and quantification of these trace organic compounds was greatly aided by the introduction of gas chromatography/mass spectroscopy (GC/MS). Thousands of organic contaminants present at nanogram to microgram concentrations were detected in water; originating from industry, agriculture or occurring naturally in the soil and therefore present in untreated waters.

The use of *M.oleifera* seed as a coagulant may also result in the addition of natural organic material into the treated water, albeit in very small quantities of residual coagulant. GC/MS was used to give a profile of organic compounds present in the final water.

6.3.4.6 Chlorination by-products

Chlorination is the predominant method of disinfection of drinking water for a number of reasons. It is a strong oxidising agent, relatively inexpensive, highly soluble in water and has a residual activity in the water supply. Chlorine combines with water to form hypochlorous acid, hypochlorite ions and hydrogen. The hypochlorous acid reacts with ammonia and amines in the raw water to form chloramines which can persist in the distribution system.

The major disadvantage of using chlorine is its reaction with certain organic compounds present in the water. Inorganic and organic molecules, suspended particles and microorganisms can exert a chlorine demand, and therefore higher doses are required to ensure biocidal activity. Previous research has indicated that halogenated hydrocarbons are formed during the disinfection stage (Rook, 1974; Bellar *et al.*, 1974). One such group, the THM are methanes where three hydrogen atoms are replaced with halogens, and these have been investigated extensively. The THM analysed for in this research were chloroform, bromodichloromethane, bromoform and dibromochloromethane. Other CBP also exist in treated waters such as dihaloacetonitriles (Trehy & Bieber 1983) and di- and trichloroacetic acid (Christman *et al.*, 1983; Miller & Uden, 1983). In many cases only chloroform and trichloroacetic are analysed for in chlorinated waters, as these two compounds will indicate the extent of formation of all DBP (WHO, 1996). Other organic compounds were also analysed for in the final water, these were carbon tetrachloride, tetrachloroethylene and trichloroethylene

Chloroform, bromodichloromethane, bromoform and carbon tetrachloride are considered to be probable human carcinogens, and tetrachloroethylene and dibrochloromethane possible human carcinogens based on tests on animals. Trichloroethylene is unclassifiable due to lack of data (Chung, 1993).

Chloroform (trichloromethane) is a colourless liquid that is slightly soluble in water. It is used as a fumigant and as a solvent for adhesives, pesticides, oils and rubber and is therefore found in the atmosphere and has also been detected in some foods. It is absorbed following oral, inhalation and dermal exposure. Bromoform and dibromochloromethane are readily absorbed by the gastrointestinal tract, and long term exposure can cause damage to the liver and kidneys (WHO, 1996). Bromodichloromethane can cause renal adenomas and adenocarcinomas.

Carbon tetrachloride, trichloroethylene and tetrachloroethylene are released into the environment by various industrial processes. The main reservoir for carbon tetrachloride is the atmosphere, although it can be adsorbed onto organic matter in the soil, from where it can migrate into the ground water. IARC concluded that it is a human carcinogen and therefore the WHO has set a guideline value of 2 μg/L and the PVC in the UK is 3 μg/L for drinking water. Trichloroethylene is highly reactive and highly mobile in soil and may be leached into the ground water, where it can degrade to form more toxic compounds. It is mainly used as a cleaning agent and is therefore often found in wastewaters. Acute exposure to high concentrations of trichloroethylene can cause depression of the central nervous system (Granjean *et al.*, 1955). Although to date there is no conclusive evidence of its carcinogenicity to humans, a provisional WHO guideline has therefore been set at 70 μg/L and the UK PVC is 30 μg/L in drinking water. Tetrachloroethylene is also used in cleaning processes and is found in wastewaters. It can be biodegraded by microorganisms to potentially toxic compounds. It is considered to be a potential human carcinogen (IARC, 1987), the WHO guideline has been set at 45 μg/L and the PVC in the UK is 10 μg/L in drinking water.

6.4 Results & discussion

Various parameters were measured during the course of the filter runs to give an indication of the effectiveness of the filtration process and the mode of action of the two coagulants. It should be noted that the turbidity removal is indicated by C/C_o, where C is effluent turbidity (NTU) and C_o is the influent turbidity (NTU). This was considered to be a more informative method of displaying the data due to the natural variation in the raw water turbidity throughout the study period. The residual turbidity as a function of bed depth is also plotted and is shown in the inset of some of the graphs. The additional headloss was used rather than the total headloss because of the variation in clean bed headloss rates in the four columns. Using the total headloss data would have resulted in an unfair comparison of the rates of headloss development in the four columns.

The DOC content of the final water should be considered with caution, since the majority of organic matter in water is of no health significance. A rise in the DOC of the treated water does not necessarily indicate that it is unfit for human consumption. The analyses undertaken using the GC/MS for various organic compounds and for THMs are of note as concentrations of specific compounds are given.

Elevated iron and manganese concentrations can occasionally be a problem at Felindre WTW, as can be seen from Table 6.1. Levels of 0.34 and 0.60 mg/L respectively can be found in the treated water, and the UK PVC for these metals is 0.30 and 0.05 mg/L. These high concentrations did not coincide with the days when filter runs using *M.oleifera* were conducted. Furthermore, the method for manganese analysis was not reliable, and consequently removal rates for these two metals using *M.oleifera* seed cannot be reported.

A list of all the runs undertaken in the field study is shown in Appendix 3; the runs are coded, such that 'M' indicates all runs conducted using *M.oleifera* seed and 'F' indicates all runs conducted using Ferral.

6.4.1 Optimisation of the hydraulic variables

The physical characteristics of filtration investigated in the field study were bed depth, media configuration and filtration rate; specifically single media beds of 70 and 120 cm of sand, and dual media beds consisting of 50 cm anthracite over a layer of 70 cm sand (sand size in all cases 0.60-1.18 mm), and filtration rates of 5 and 10 m/h. These parameters were chosen in order to compare removal rates with that achieved in the laboratory study.

6.4.1.1 Particulate removal

Comparative turbidity removal and headloss development data for hydraulic variables are presented in Figures 6.4-6.17. In all cases it can be seen that with the optimum dose, turbidity removal was consistently > 95% ($C/C_o < 0.05$) for longer than 11 hours. This entailed the production of a final water with a turbidity of < 1 NTU. It was found that *M.oleifera* was effective, regardless of media configuration and filtration rate.

Clear dose responses can be seen for each media configuration and filtration rate (Figures 6.4, 6.6, 6.8, 6.12 and 6.14). For example, Figure 6.4 indicates that with a bed depth of 70 cm and a filtration rate of 5 m/h, the optimal dose was between 50 and 70 mg/L. Considering the headloss development data for the same run, Figure 6.5, it can be seen that after 9 hours of operation a headloss of about 100 cm was reached at a dose of 50 mg/L. Using a 70 mg/L dose, however, a headloss of 170 was reached after 9 hours. The deeper bed of 120 cm, shown in Figures 6.7, the same trend can be seen. The highest removal rates were achieved at doses of 70-90 mg/L, but the high headlosses at 80 and 90 mg/L, would make 70 mg/L the optimum dose. Thus the benefit of using a lower dose is not only a potential reduction in coagulant costs, but also a greater total output before termination, due to the reduction in headloss development rate. In the dual media bed considered in Figures 6.8 and 6.9, the optimal dose was considered to be 70 mg/L based on the highest removal rate. The headloss development rate was consistently low in the dual media configuration, and was therefore not significant in the choice of coagulant. Using doses well in excess of the optimal dose for each filter condition resulted in increases in turbidity of the treated water. A similar trend was found in the laboratory results (Figures 4.1 and 4.4), and indicates that overdosing leads to particle restabilisation.

A comparison of the laboratory and field results revealed the following:

- Turbidity removal rates were very similar in the field and the laboratory study using the same raw water turbidity; the highest removal rates were achieved in the 120 cm bed of the small sand in the laboratory study, shown in Figure 4.5.

- Higher coagulant doses were required to achieve similar removal rates in the field study than in the laboratory study. This can be seen by comparing the optimal dose required in Figures 6.4, 6.6 and 6.8 with the laboratory results using similar media configurations in Figures 4.5 and 4.62.

- Turbidity removal rates with three media configurations in the field study were very similar, in contrast to the laboratory study, where the bed depth and configuration affected the removal rates.

- Headloss development trends were the same in both studies: higher filtration rates and deeper beds gave higher clean bed and lower additional headlosses. But the actual rate of headloss increase for all media configurations and filtration rates were higher in the field study than the laboratory study.

- Higher filtration rates in the field study result in reduced turbidity removal, as was the case in the laboratory study.

It is considered that the organic matter present in the low turbidity raw water at Felindre WTW was the cause of the high coagulant requirement, compared to that required for the low turbidity waters investigated in the laboratory. The DOC of the raw water throughout the study period ranged between 2 to 3.5 mg/L, as seen in Table 6.1. It was found by Wiesner (1992) that charged functional groups adsorbed onto organic compounds, had a stabilising effect on colloidal matter in the water, due to the hydrophilic nature of organic molecules, as described in Section 2.1.1. NOM has been found previously to exert a high coagulant demand (Jiang *et al.*, 1996), and therefore the natural raw water in the field study required a much higher dose than in the laboratory study to effect turbidity removal.

The turbidity removal rates were found to be very similar in the two studies, except for the 120 cm of the smaller media in the laboratory study. As can be seen in Figure 4.5, the residual turbidity in this case was < 0.1 NTU. This was considered to be due to the high total collector surface area for this media configuration. The similarity in the removal rates of the two studies indicates that the kaolin suspension used simulates a natural raw water with a good level of consistency.

The highest turbidity removal in the laboratory study was consistently found to be the single media bed of small media at a depth of 120 cm, although at low loading rates there was not much variation in performance of all the media configurations. In the field study, there was no difference in the removal rates with the three media configurations, as can be seen in Figure 6.10. Thus it would seem that the increase in collector surface area had no effect on turbidity removal rates in the field study. Therefore it can be assumed that the maximum possible removal was attained in the shallow bed of 70 cm, unlike the laboratory study where increases in the depth, improved the removal rates.

The effects of media configuration on headloss development rates in the field pilot rig, however, were very similar to that in the laboratory rig, as expected from filtration theory and explained in Section 4.5.1. The effect of dual media and deeper beds was to reduce the rate of additional headloss, due to the larger interstitial pores and greater distribution of the deposited material respectively, as seen in Figure 6.11. The optimum choice of media configuration would thus be the dual bed, since removal was equal to that in the single media beds, but with the advantage of a greatly reduced headloss development rate. The rate of increase in headloss was higher in the field study in all the media configurations for two reasons. Firstly, the organic matter, consists of large molecules which form large voluminous floc that can rapidly clog the filter pores. Secondly, the high coagulant doses would also clog the filter pores.

Two media configurations were also considered at a 10 m/h flow rate: the single media 70 cm bed and the dual media bed, the removal and headloss accumulation rates being shown in Figures 6.12-6.15. A comparison of the turbidity removal with the two media configurations and filtration rates at their optimum doses are shown in Figures 6.16, the corresponding headlosses are shown in Figure 6.17. Greater removal rates for both media configurations occurred at 5 than 10 m/h; a similar trend to that found in the laboratory results. This was

considered to be due to the longer retention times in the bed and the consequent increase in in-pore flocculation, as discussed in section 4.1.1.

The additional headloss at 5 m/h was greater than at 10 m/h due to better utilisation of the bed depth and the reduction in surface clogging at the higher rate, as was the case in the laboratory study. Lower rates of headloss development are of more significance with gravity fed filters, as a rapid increase can result in a lower total output of filtrate.

It is therefore concluded that the importance of the dual media beds can be seen most clearly at the higher filtration rate of 10 m/h: removal rates remain high, with no sign of breakthrough after 10 hours of operation, and the headloss development rates remain very low. These benefits were not realised to the same extent in the laboratory study because of the difference in the chemistry of the raw water and the lower coagulant dose, leading to a lower headloss development rate. Treatment of the natural raw water containing organic material, specifically large organic molecules, benefited from the larger storage capacity of the anthracite layer.

The optimal dose was lower with higher filtration rates (using the same media configuration). This was considered to be a consequence of the greater inter-pore mixing intensity at the higher filtration rates, leading to a greater number of particle collisions, and ultimately higher rates of in-pore flocculation at a lower dose. Similar results were found by McConnachie (1993), where jar tests using various mixing intensities in the slow mix stage, indicated that higher rates resulted in a reduction in the optimum dose of *M.oleifera* seed coagulant.

6.4.1.2 Bacterial removal

The influence of hydraulic variables on the bacterial content of the final water treated with *M.oleifera* seed was investigated by taking and analysing samples once during the working stage of selected runs, and comparing these with the bacterial concentrations of the raw water. Table 6.3 and Figures 6.18 show the results for the lowest dose to achieve maximum bacterial removal rates (shown as a percentage of the raw water bacterial concentration) for each media configuration. (The raw data and percentage removal of three groups of bacteria using various media configurations are shown in Appendix 5: Tables A5.1-A5.3.)

The rates of coliform and *E.coli* removal rates were almost identical for the three media configurations considered. The HPC results indicated a difference in performance of the media configuration: 120 cm of sand achieved the greatest removal, with a small reduction in the dual media bed and a further reduction with the 70 cm bed. Although the HPC is not necessarily of any health significance, the results indicate that potentially bacterial removal rates may be lower in the shallower bed. The collector surface area may have an effect on bacterial removal rates, even if the turbidity removal was not affected.

Determinand	70 cm Sand (60 mg/L)	120 cm Sand (70 mg/L)	70 cm Sand & 50 cm Anthracite (60 mg/L)
HPC (1 day at 37°C/ml)	91	100	97
Total Coliforms /100 ml	99	100	100
***E.coli* /100 ml**	100	100	100

Table 6.3 Bacterial reduction at the optimum dose of M.oleifera seed using CFF.

(Filtration rate - 5 m/h).

It had been estimated by the WHO that thermotolerant bacterial (*E.coli*) numbers should be reduced by 80% after rapid filtration and by 99.9% after a combination of coagulation, settling and rapid filtration (WHO, 1996). The treatment process used in this study consisted of coagulation and filtration, *E.coli* and total coliform removal rates with the optimal dose and media configuration were 100%. This indicates that the combination of *M.oleifera* and CFF can reduce bacterial numbers to levels not only acceptable by the WHO standard but greater than that achieved by other workers using different treatment regimes: Treatment of water with a turbidity of about 1000 NTU and a *M.oleifera* dose of 200 mg/L achieved a one log reduction in bacterial concentrations (Jahn & Dirar, 1979). A 2-3 log reduction of bacterial numbers in a 1:10 dilution of raw sewage was achieved using a dose of 100 mg/L (Folkard & Grant, 1989). A 1-2 log reduction of total coliforms was achieved on Blue Nile water with a turbidity of 2000 NTU using 200 mg/L of *M.oleifera* (Madsen *et al.*, 1987). It is difficult to compare optimal doses, since the majority of previous work was conducted on high turbidity raw waters, requiring higher doses of coagulant than those used in this study. The excellent

bacterial removal rates achieved in this study must be interpreted with caution, as the raw water used was of relatively good microbiological quality. The total coliform counts ranged from 64-750, whereas surface water bacterial numbers can be considerably higher. For example, total coliform numbers of up to 8,000 have been found in rivers in Uganda and up to 3100,000 in canals in Indonesia (Feacham, 1980).

It has been shown that *M.oleifera* seeds contain an antimicrobial element, the glucosidic mustard oil, 4 (α-L-Rhamnosyloxy) benzyl isothiocyanate (Das *et al.*, 1957; Eilert *et al.*, 1981). But the reduction in bacterial numbers after treatment with *M.oleifera* is considered to be only due to coagulation and filtration of the bacteria in the same manner as any other negatively charged particle in the water. For water treatment purposes the highest concentration of *M.oleifera* used as a coagulant is about 200 mg/L. However, in order to be an effective disinfectant, concentrations of about 100,000 mg/L would need to be used (Jahn, 1988; Jahn, 1986; Eilert *et al.*, 1981).

It is clear then that bacterial removal does occur to a significant level with this natural water using *M.oleifera* in conjunction with CFF. But only due to coagulation and filtration, and in order to achieve microbiologically safe of potable quality, a disinfection stage would need to be included in the treatment process.

6.4.1.3 The organics in the final water

The increase in DOC of the water treated with *M.oleifera* seed as a function of the hydraulic variables is shown in Table 6.4 and in Figures 6.19 and 6.20. (The raw data for the DOC of the raw and final waters is shown in Appendix 5: Table A5.4).

The DOC of the treated water was consistently higher than in the raw water, due to the addition of extraneous material from the seed suspension which contained a high concentration of organic matter. Also the higher the dose used the higher the increase in DOC of the final water, as shown by the regression analysis of the data in Figure 6.19. The R^2 value was 0.69, indicating a positive although moderate correlation between the dose and the increase in DOC of the final water. It is possible that the use of higher doses resulted in greater removal of the organic matter, as was the case with turbidity removal. But this effect would have been masked by the addition of greater amounts of seed material at the higher doses.

The DOC of the seed suspension was 880 mg/L; dosing at 50-90 mg/L would result in the addition of between 4.4-7.9 mg/L DOC. However, it can be seen from Table 6.4, that the final water DOC is less than the raw water DOC plus the calculated DOC from the seed material, therefore there must be some removal occurring in the filter bed.

Dose (mg/L)	70 cm Sand			120 cm Sand	70 cm Sand & 50 cm Anthracite		
	Run M52-55	Run M18- 20	Run M88 & 89	Run M48 & 52	Run M58- 60	Run M7-19	Run M85-86
90	3.13	-	3.43	3.04	3.03	-	3.13
80	2.97	-	2.98	2.62	2.73	-	-
70	2.74	-	-	2.69	2.71	-	-
60	2.50	3.02	-	2.08	2.69	2.59	2.18
50	2.26	2.25	-	-	-	2.24	-
DOC of Raw Water	3.56	2.92	-	2.07	2.96	2.92	2.1

Table 6.4 The increase in DOC (mg/L) after treatment with M.oleifera seed using CFF.

(-) = analysis not undertaken

The range of doses of *M.oleifera* used was large, and it was therefore anticipated that the range of DOC concentrations in the final water would be correspondingly large. This was not the case: with a dose range of 50-90 mg/L, the DOC ranged from 5.82 to 6.69 mg/L, in the 70 cm bed); a dose range of 60-90 mg/L, resulted in a final DOC of 4.15-5.11 mg/L and 4.28 to 5.72 mg/L in the single media bed of 120 cm and the dual media bed respectively. Although a greater amount of extraneous material is added at higher doses, the removal of the original organic matter was greater, therefore the organic matter present in the final water was not proportional to the dose. This would indicate that *M.oleifera* seed was effective in the removal of the 'original' DOC in the raw water, despite adding to the final DOC content. The use of the presscake (the portion of the seed which remains after the oil has been expelled) or the proteins containing the polyelectrolyte were extracted and used as the coagulant, the DOC of the treated water would be significantly lower, as described in Section 2.7.

The increase in DOC at each dose varied with the three media configurations. This indicates that the extent of removal of the organic matter was different with each media configuration. This contrast with the turbidity removal results, where the media configurations did not have

an effect. Figure 6.20 indicates that the greatest increase in DOC occurred in the 70 cm bed, followed by the dual media bed, and the lowest rate of increase was found in the 120 cm bed. It would thus appear that the collector surface area was influencing the DOC removal rate. This is due to the greater stability of the dissolved organic matter compared to the particulate matter (Wiesner, 1992). The greater stability is caused by the hydrophilic nature of most of the organic molecules such that water molecules bind to their surfaces and act as a barrier to collisions with other particles. DOC removal therefore requires a higher amount of in-pore flocculation, this occurs to a greater extent in beds with greater collector surface area.

A GC/MS was used to analyse the organic matter found in the water treated with *M.oleifera* seed. The compounds detected and their concentrations are listed in Table 6.5. It is assumed that some of these organic compounds originated from the seed suspension, as they are not found in the raw or treated water at Felindre WTW (Owen, 1997), nor were they found in the water treated with Ferral. Table 6.5 may not be a complete list of the organic compounds present in the treated water, but rather a list of compounds that are extractable in dichloromethane, amenable to GC/MS analysis and present in concentrations higher than 0.1 μg/L (compounds which are too volatile to be separated from dichloromethane or pass through a GC, or are thermally unstable will not be detected).

Determinand	Concentration (μg/L)
1-Isothiosyanato Propane	0.66
1-Isothiosyanato Butane	0.18
2-Isothiosyanato Butane	0.19
Methyl Phenol Isomer	0.17
(Isothiocyanatomethyl) Benzene	1.87
4-Methyltetrahydro-1, 3-Oxazine-2-Thione	0.19
4-Hydroxy Benzeneacetic Acid, Methyl Ester	1.52

Table 6.5 Concentration of organics found in the water treated with M.oleifera seed.

(Using CFF, filtration rate - 5m/h, 70 cm sand & 50 cm anthracite).

As discussed in Section 6.4.1.2, the seeds contain the glycosidic mustard oil, 4 (α-L-Rhamnosyloxy) benzyl isothiocyanate, which is responsible for the seed's antimicrobial

properties. Previous workers have found this compound in the seeds at concentrations of up to 10% (Kjær *et al.*, 1979) and 8-10% when extracted using ascorbic acid (Eilert *et al.*, 1981). The seed solution prepared for the pilot study had a pH range of 6.8-7.4, and therefore the extraction rate of the isothiocyanate was not expected to be very high in this neutral solution. But Table 6.5 indicates that 64% by weight of the organic compounds detected in the final water are derivatives of isothiocyanate. This further indicated that much of the 'original' DOC was removed in the filter bed, but the seed suspension added organic matter to the final water.

Methyl phenol, which was found in concentrations of 0.17 μg/L, is known to occur occasionally in the raw water. It can be introduced into the aqueous environment as a result of anthropogenic activity or the breakdown of certain organic material, which was the most probable source in this case. The significance of the phenol family is that they combine with chlorine to give cholorophenols that have a low taste and odour threshold. Occasionally there are taste and odour complaints concerning the treated water from Felindre WTW, but none occurred during the study period; nor were phenol compounds found in the Works' final water or the sample treated with Ferral. The taste threshold for methyl phenol is 0.1 μg/L according to the WHO. The presence of the methyl phenol at concentrations of 0.17 μg/L in the final water treated with *M.oleifera* was therefore not considered to be a problem.

Benzene is known to be a human carcinogen (Merian & Zander, 1982; RIWA, 1989), but the levels found in drinking water are generally insignificant compared with the intake from food and air (ATSDR, 1990). Detection of small concentrations of benzene is not necessarily a matter of concern. It was detected in 50-60% of potable water samples in Canada, where it was considered normal, in concentrations similar to that found in this study (WHO, 1996). The 1985 EC Directive and the 1989 and 1991 UK Regulations do not set a limit for benzene. The WHO guideline value, however, is 10 μg/L and the USEPA set a maximum contamination level (MCL) of 5μg/L. Therefore a concentration of 1.8 μg/L is not considered significant. The origin of the benzene is unknown; it has never been found in the raw or treated water from the Works, nor the sample treated with Ferral. It is not a natural constituent of the *M.oleifera* seed, but it is possible that the seed had absorbed benzene from the environment. Studies have shown that fruits such as blackcurrents and peppers contained this compound due to absorption from the atmosphere (Wilson *et al.*, 1986). If this was the case the seed suspension would have contained the benzene due to its solubility, and consequently was dosed into the raw water.

The health risks from consumption of these trace organic compounds in water cannot be quantified. Often such micro-pollutants are considered of low risk, simply because of their low concentrations (Twort *et al.*, 1994), but this is not necessarily true or acceptable. However, there is no reference to them in the Merck Index (Budavari *et al.*, 1996), a chemical Encyclopedia of all known harmful compounds used by Hyder Consulting (Environmental) Ltd. Also the seeds themselves are edible and no adverse effects have been known to occur after their consumption. Nonetheless the presence of an antibacterial agent in a water treatment coagulant may pose a risk to human health, or at least be seen to be a potential problem. (Eilert *et al.*, 1981; Barth *et al.*, 1982; Grabow *et al.*, 1985). This is not likely to be significant to human health since concentrations of between 200-1000 mg/L were used in these tests. As discussed in section 2.7.3, 200 mg/L is the highest concentration used for coagulation purposes

6.4.2 A comparison of *M.oleifera* and Ferral in terms of effectiveness of treatment

In order to asses the effectiveness of *M.oleifera* seed on this natural raw water, it was compared with the proprietary coagulant in use at Felindre WTW, in terms of turbidity and bacterial removal rates and addition/removal of organic matter. The treatment mechanisms of the two coagulants are very different. There is little published data on the use of Ferral, but information obtained from the manufacturer, Laporte, indicates that it acts in the same manner as other metal coagulants (Laporte, 1997). Floc formation is induced by the formation of a precipitate, which entraps particulate and dissolved matter into its amorphous structure. For this reason the latter is considered to be effective at removal of colour and organic matter (Edzwald, 1986).

Direct comparative studies were undertaken by maintaining all the hydraulic conditions of the four columns, then dosing two columns with *M.oleifera* seed and two with Ferral.

6.4.2.1 Particulate removal

Turbidity removal for each media configuration and the corresponding additional headloss development rate for a range of doses of Ferral are shown in Figures 6.21-6.26. In each case the residual turbidity during the working stage was < 0.5 NTU, indicating that removal rates were similar to that achieved with *M.oleifera* (shown in Figures 6.4, 6.6 and 6.8). Removal rates were not as high as that achieved in the Works, where a final water turbidity of < 0.1

NTU was achieved during the study period as indicated in Table 6.1. This may have been due to two factors: the use of a separate flocculation stage and a triple media (sand, anthracite and GAC) filter bed.

In each of the media configurations, it can be seen that the highest dose used resulted in maximum removal, but that the rate of headloss accumulation was excessive. Figure 6.21 indicates that the greatest removal occurred with a dose of 22.5 mg/L in the bed of 70 cm sand. But the corresponding headloss data, Figure 6.22, shows that the additional headloss after 6 hours was 230 cm. Similarly with the single media bed of 120 cm, Figure 6.23, maximum removal was achieved at a dose of 22.5 mg/L, and the additional headloss after 5.5 hours was 240 cm, Figure 6.24. With the use of the dual media bed, the effect on the headloss development rate of using the highest dose was not critical, as can be seen in Figure 6.26. (As explained in Section 4.5.1, the main benefit of the dual media bed was to reduce the rate of headloss development.) For both depths of single media beds, the rate of headloss increase was very high, and therefore the optimum dose is dependent on the termination criteria and the treatment objectives. For consistency, it was decided to use the dose of Ferral that gave the highest particulate removal, regardless of the rate of headloss accumulation, so that a comparison could be made with *M.oleifera* runs shown in Figure 6.29 and 6.30.

The problem of excessive headloss accumulation with the highest turbidity removal rates was not encountered to the same extent with *M.oleifera*. If Figure 2.12 and the definition of optimal filtration condition described in Section 2.5 (Mints, 1966; Ives, 1969; Ives, 1982) are considered, it can be seen that T_2-T_1 is closer to zero with *M.oleifera* than Ferral. Thus it can be concluded that conditions in the filter were optimised with *M.oleifera* due to improved bed utilisation. A visual inspection of the filter media after treatment with the two coagulants, indicated that surface removal and clogging occurred to a greater extent with Ferral than *M.oleifera*. This was as expected and was the cause of the rapid rise in headloss across the bed with the former in the single media beds. The problem of surface clogging was reduced with the dual media bed, and therefore the rate of headloss development was considerably lower, as illustrated in Figure 6.25.

Figures 6.27 and 6.28 illustrate the comparative turbidity removal and the headloss development for the optimum dose for the two coagulants and each of the three media configurations. The turbidity removal rates were very similar in all cases, although marginally

higher with *M.oleifera*. The headloss development rates, however varied greatly according to the media configuration and coagulant type. The latter observation was as expected, but the former was not. Metal coagulants are generally more effective at particulate removal with lower turbidity waters than are low molecular weight polymers such as *M.oleifera*. The rate of floc formation induced by the 'patch' mechanism of charge neutralisation is affected by the number of particulates in the water, and the number of collisions, as described in Section 2.2.2. Whereas the rate of 'sweep' flocculation, the mechanism of floc formation induced by Ferral, is dependent upon the rate of formation of the hydroxide precipitate in the water, and independent of the particulate concentration. These unexpected positive results further indicate the potential for use of CFF in conjunction with *M.oleifera* seed in the treatment of low turbidity waters.

6.4.2.2 Bacterial removal

Table 6.6 and Figure 6.29 show the comparison of percentage removal rates of the three categories of bacteria with the two coagulants. (The raw data for the bacterial content of the water treated with Ferral is shown in Appendix 5: Table A5.5-A5.7). It can be seen that removal rates at the optimum dose were higher with *M.oleifera* seed than with Ferral; a further indication of the potential for use of *M.oleifera* seed on such waters. Ferral was found to be least effective in bacterial removal in the 70 cm bed, and was considered to be due to the reduction in collector surface area, as described in section 6.4.1.2.

Determinand	70 cm Sand		120 cm Sand		70 cm Sand & 50 cm Anthracite	
	M.oleifera (60 mg/L)	Ferral (20 mg/L)	*M.oleifera* (70 mg/L)	Ferral (20 mg/L)	*M.oleifera* (60 mg/L)	Ferral (17.5 mg/L)
HPC (1 day at 37°C/ml)	91	84	100	99	97	87
Total Coliforms /100 ml	99	94	100	100	100	100
E.coli /100 ml	100	96	100	100	100	100

Table 6.6 Maximum percentage bacterial removal with M.oleifera seed & Ferral.

(Using CFF, filtration rate - 5 m/h).

6.4.2.3 The DOC of the final water

The use of Ferral as the coagulant consistently reduced the DOC content of the final water, as shown in Table 6.7. (The raw data is shown in Appendix 5: Table A5.8). These results contrast with those obtained from analysis of the water treated with *M.oleifera* seed, where the DOC consistently increased. The DOC removal rates achieved with Ferral were comparable with those achieved in the Works, thus indicating that mixing conditions and doses used were optimised, in terms of destabilising the dissolved organics using Ferral.

Dose (mg/L)	Sand 70 cm		120 cm Sand	Sand 70 cm & Anthracite 50 cm	
	Run F9 & 12	Run F36	Run F3 & 4	Run F14 & 15	Run F33 & 34
17.5	-	-	0.77	1.88	0.91
20	0.86	0.78	0.80	-	1.01
22.5	1.00	-	-	-	-
28	-	-	-	1.86	-
DOC of Raw Water	3.14	-	2.04	3.15	2.1

Table 6.7 The decrease in DOC (mg/L) after treatment with Ferral.

(Using CFF, filtration rate - 5m/h).

(-) = analysis not undertaken.

In order to investigate the effect of dose on the DOC removal rate, the Ferral dose was plotted against the decrease in DOC, as shown in Figure 6.30. Regression analysis of the data gave an R^2 of 0.18, indicating that the extent of correlation was very low. It can therefore be assumed that the Ferral dose had no significant effect on the DOC removal rates, unlike the turbidity removal.

The media configuration affected the rate of DOC removal as can be seen in Figure 6.31, such that the highest removal rate was achieved in the dual media bed and the lowest removal with the 70 cm bed. Although a greater number of samples would need to be taken to validate these results, and to come to any firm conclusions. Note that the ordinate in Figures 6.30 and 6.31 is the decrease in DOC, whereas in Figures 6.19 and 6.20 it is the increase in DOC in the final water. These results contrast with the turbidity removal results using Ferral, as seen in Figure 6.27 where the media configuration had little effect on the removal rates. One explanation for

the greater organic removal with the dual media bed is that mixing conditions in the large pores of the anthracite layer allowed the formation of floc that enhanced the removal of organic matter. Shear forces would be greater in the single media beds, due to the smaller pores and higher interstitial velocities. These factors would have a greater effect on the Ferral than the *M.oleifera* floc, due to their delicate nature. Comparative jar tests conducted using the two coagulants produced floc with differing sizes and strengths. Measured microscopically, *M.oleifera* floc were generally about 1 mm in diameter, whereas Ferral floc were about 3 mm. Furthermore, the *M.oleifera* floc were found to resist breakup to a greater extent than did the Ferral floc, when the floc samples experienced high shear forces as a result of rapid mixing in jar tests. Another factor is that the larger pores of the anthracite layers allowed the capture of a greater volume of material. The voluminous Ferral floc containing the relatively large organic molecules could be retained to a greater extent in the anthracite layer than in the sand layer. The positive results in terms of DOC removal achieved with Ferral are significant, because traditional DF and CFF are not considered to be as effective as conventional treatment in the removal of organic matter. Generally large doses of coagulant are required to destabilise organic material and therefore the sludge tends to be voluminous, requiring a separate sedimentation stage for effective treatment (Edzwald, 1986).

6.4.2.4 THM formation potential

THMs are members of the family of organohalogen compounds that are formed in water by the reaction of chlorine with certain organic compounds. Generally the best method of controlling their formation is the removal of their organic precursors during the treatment process (Federal Register, 1982). As the water treated with *M.oleifera* and Ferral contained organic matter, it was considered important to measure the THM formation potential.

Table 6.8 lists the Chlorination by-products and their concentrations found in the water treated in the pilot plant and the Works, and also various limits and guideline values. The PVC in the UK are set for the total THM concentrations in the treated water, since the compounds generally occur together and it is more convenient to consider them a group. Other countries have limits for individual compounds. The WHO combine both these criteria by using a fractionation approach to determine the total THM concentration (WHO, 1996; Tebbutt, 1998). This consists of summing the ratio of the concentration of each compound to its guideline value, and the value obtained should be less than 1. Other halogenated hydrocarbons

such as carbon tetrachloride, tetrachloroethylene and trichloroethylene were analysed for, but their concentrations were below the detection limit and therefore well below the UK's PVC. There are two main observations from the results of the THM analysis. Firstly, the water coagulated with *M.oleifera* seed gave very similar results to that coagulated with Ferral, for all determinands considered. Secondly, all the results were well below the standards set by the UK and the guidelines set by the WHO. If the fractionation approach is employed, the total obtained after 24 hours contact time with chlorine with doses of 60 and 90 mg/L of *M.oleifera* was 0.10 and 0.12 respectively; whereas with doses of 17.5 and 20 mg/L of Ferral the figure was 0.10, and for the Works' final water the value was 0.45. The temperature that the chlorinated water is stored at is important, as higher temperatures lead to higher rates of THM formation (Townsend & Tripp, 1995). The water samples were stored at room temperature (20-26^{o}C), but temperatures of 30-40^{o}C could be experienced in many tropical and subtropical countries during the summer months. This would potentially lead to higher levels of THMs in the final water. Also in slightly alkaline conditions, the THM formation rate increases (Eaton *et al.*, 1995). Adjustment of the pH of the samples was not attempted, but the values were noted. The pH of the samples treated with Ferral was 6.5 and those treated with *M.oleifera* was 7.2. Therefore conditions in the samples treated with *M.oleifera* were likely to elevate THM levels compared to Ferral.

Chloroform, bromodichloromethane and dibromochloromethane concentrations all increased with the higher chlorine contact time for both coagulants. These results were expected because the longer contact time would allow the reaction to go to completion. The concentration of bromoform was below the detectable limits, and would indicate that if the precursors were present in the treated water, that their concentrations were very low. It is estimated that only about 1% of the DOC in a water is made up of hydrocarbons (Taylor, 1986), and the halogenated methanes which form the THMs are only a portion of these hydrocarbons. As the DOC concentration of the raw water during the test period was relatively low, as discussed previously, then it was not expected that the THM level would be high.

Determinand	*M.oleifera* (90 mg/L)		Ferral (20 mg/L)		*M.oleifera* (60 mg/L)		Ferral (17.5 mg/L)		Works Final Water (17.5 mg/L) [1]	Detection Limit	UK Limit	WHO Guideline
(µg/L)	1 Hr	24 Hrs	1 Hr	24 Hrs	1 Hr	24 Hrs	1 Hr	24 Hrs	24 Hrs	(µg/L)	(µg/L) [2]	(µg/L) [3]
Trichloromethane (Chloroform)	3.16	5.97	3.01	5.47	3.02	5.52	3.09	5.23	34.50	2.092	-	200
Bromodichloro-methane	1.11	3.97	1.01	3.13	1.03	2.99	1.15	3.25	14.34	0.86	-	60
Tribromomethane (Bromoform)	<1.02	<1.02	<1.02	<1.02	<1.02	<1.02	<1.02	<1.02	<1.02	1.018	-	100
Dibromochloro-methane	<0.29	1.32	<0.29	1.08	<0.29	1.20	<0.29	1.13	2.58	0.29	-	100
Total THMs	<5.58	<12.28	<5.33	<10.70	5.36	10.73	5.55	10.63	51.42		100	[4]

Table 6.8 Comparative THM formation with M.oleifera seed & Ferral and two media configurations.

(Using CFF, filtration ate 5 m/h, bed depth 70 cm sand).

[1] Obtained from Felindre WTW sample analysis data.

[2] Water Supply (Water Quality) (Amendment) Regulations 1991(Severn Trent Water, 1999).

[3] WHO Guidelines for Drinking Water Quality 1996.

[4] The sum of the ratio of the concentration of each compound to its respective guideline value should not exceed 1.

The use of *M.oleifera* seeds as a water treatment coagulant is mainly applicable to developing countries. Therefore, any risks from drinking water containing minute concentrations of THM, which have only been shown to be potentially carcinogenic, are negligible in comparison to the risks of drinking microbially contaminated water. Even in developed countries, the risks are minimal. It has been estimated that the number of deaths from cancer caused by drinking 2 litres of water per day which contain chloroform would be 1 in a million per year. As compared to 9 in a million from drinking a can of diet soda per day, and 100 per million from drinking a litre of wine per week (Taylor, 1986). Thus the risks from drinking chlorinated water must be put into perspective.

6.4.3 A comparison of the effectiveness of CFF and DF

The effectiveness of CFF and DF were compared in terms of reduction of turbidity, bacterial numbers and DOC concentrations, and the headloss development rates in the dual media filter beds. It was considered that in order to optimise removal rates using *M.oleifera* seed with very low turbidity water it may be necessary to allow a longer flocculation time to ensure a high number of particle collisions. Thus the provision of a separate flocculation stage could result in better floc formation. As was suggested in Section 6.4.2.3, it was possible that DOC removal may be enhanced with longer mixing periods. Felindre WTW is operated as a DF process, but it was suggested by the Works' operators that the flocculation stage may not be necessary to effect efficient removal (Owen, 1997). Thus this study allowed a comparison of removal rates with the two modes of operation using Ferral.

6.4.3.1 Particulate removal

A comparison of the two treatment modes in terms of turbidity removal and headloss accumulation was undertaken using the dual media beds and the two coagulants. Figure 6.32 shows turbidity removal as a function of *M.oleifera* seed dose, with the use of a flocculation tank, and Figure 6.33 is the corresponding headloss development rates. Figure 6.36 shows the turbidity removal as a function of Ferral dose and the corresponding headloss is shown in Figure 6.37.

The effectiveness of DF and CFF in terms of turbidity removal and headloss development rates can be compared by considering Figures 6.34-6.35 with Figures 6.8, 6.9, 6.25 and 6.26. The optimal dose of *M.oleifera* in DF was considered to be 70 mg/L, although the variation in performance between 60-80 mg/L was very small. The optimal dose with Ferral was found to be 22.5 mg/L in the DF mode. The optimum in the CFF mode was found to be 28 mg/L; if this higher dose had been used with DF, then potentially higher removal would be achieved. It is thus considered that the optimal doses were the same in DF and CFF for both coagulants. The removal rates were very similar in the two modes of operation, as seen in Figure 6.36, indicating that the use of a flocculation stage did not confer any benefit to the particulate removal process.

Figure 6.36 indicates that the ripening period was longer using DF than CFF, but this was caused by the longer retention time in the DF mode. Timing of the run commenced when flow from the header tank was started and therefore there was a 30 minute delay when operating in the DF mode, while the dosed water passed through the baffled tank. The duration of the runs in the two modes of operation can not be compared, since none of the four runs illustrated in Figure 6.36 and 6.37 were continued to completion. This is unfortunate since the total output from any particular mode of operation is one of the most significant parameters in a treatment works.

The headloss rates in the four cases were very low after 11 hours of operation, as illustrated in Figure 6.37. The major difference in the headloss data is that the rate of accumulation was greater with CFF than DF for both coagulants. This was considered to be a function of the ratio of surface area to volume of the captured particulate matter. This ratio would be large with discrete particles, as would occur with CFF; whereas for the same volume of material entering the bed, formed floc would have a low ratio, as would be the case with DF. It has been established by previous workers that the greater this ratio the greater the headloss development rate, because there is more resistance to flow through the bed (Darby & Lawler, 1990; Tobiason *et al.*, 1993). Accordingly, when the pilot plant was operated as DF the headloss development at the same dose was lower than when operated as CFF. Similar results were obtained by Edzwald (1986), where the use of a cationic polymer on water with a relatively high organic content produced a higher headloss in CFF than DF. A further feature of significance in Figure 6.36 and 6.37, is that for both CFF and DF, *M.oleifera* outperformed Ferral in terms of turbidity removal and headloss development rate.

6.4.3.2 Bacterial removal

Comparative bacterial removal rates when operating the plant in the CFF and DF modes are shown in Table 6.9 and Figure 6.38. (The raw data is shown in Appendix 5: Tables A5.9 & A5.10). The main conclusion from these results is that both *M.oleifera* seed and Ferral were very successful in terms of bacterial removal using both CFF and DF, since 100% removal was achieved for both total coliforms and *E.coli*. The results of the HPC indicate that potentially CFF was more effective than DF with both coagulants. Clearly more data is required to draw any firm conclusions on the variation in treatment performance using CFF and DF, although it would appear from these results that no advantage is gained by incorporating a separate flocculation stage in the treatment regime.

	M.oleifera (60 mg/L)		Ferral (17.5 mg/L)	
Determinand	**CFF**	**DF**	**CFF**	**DF**
HPC (1 day at 37°C/ml)	97	71	87	67
Total Coliforms/100 ml	100	100	100	100
***E.coli* /100 ml**	100	100	100	100

Table 6.9 A comparison of maximum percentage bacterial removal with M.oleifera seed & Ferral using CFF and DF.

(Filtration rate 5 m/h, bed depth 70 cm sand & 50 cm anthracite).

6.4.3.3 DOC of the final water

The average increase/decrease in DOC of the water after treatment using CFF and DF, and the two coagulants are shown in Tables 6.10 and 6.11 and Figures 6.39 and 6.40 (the actual DOC concentrations of the raw and final waters are shown in Appendix 5: Tables A5.11 & A512).

The increase in DOC concentration of the final water after treatment using DF was very similar to that found using CFF, as seen in Table 6.10. As was explained with the bacterial removal results, the data on the DOC concentrations of the water after treatment using DF was limited. Therefore no firm conclusion can be drawn from the comparison of the two modes of operation. Although it would appear that the addition of a flocculation stage had no effect on the rate of removal of organic matter from the water.

The data for DOC removal with Ferral in the DF mode was limited, but the removal rate was the same as with CFF, indicating that the extra mixing time did not affect removal of the organic matter.

Dose (mg/L)	DF	CFF
90	2.35	3.08
80	2.88	2.73
70	2.54	2.60
60	1.93	2.42
50	1.63	2.24

Table 6.10 A comparison of the average increase in DOC (mg/L) after treatment with M.oleifera seed using CFF and DF.

(Filtration rate 5 m/h, bed depth 70 cm sand & 50 cm anthracite).

Dose (mg/L)	DF	CFF
17.5	0.91	1.39
20	0.91	1.01

Table 6.11 A comparison of the average decrease in DOC (mg/L) after treatment with Ferral using CFF and DF.

(Filtration rate 5 m/h, bed depth 70 cm sand & 50 cm anthracite).

6.5 Conclusions of the field study

• Turbidity removal rates with *M.oleifera* seed were comparable in the field and the laboratory study, treating raw water turbidities of 2-5 NTU. This indicated the effectiveness of *M.oleifera* seed on natural low turbidity raw waters. Such waters are generally considered difficult to treat with low molecular weight polymers, therefore successful treatment of such a water indicates potential effectiveness on higher turbidity raw waters (5-75 NTU) using *M.oleifera* seed with CFF. The higher dose requirement in the field compared to the laboratory study was considered to be due to the organic content of the water. The consequence of using a higher dose and the presence of organic matter in the raw water was a more rapid rate of headloss development than was encountered in the laboratory study. As the media configuration has no effect on turbidity removal rates, the optimal configuration was based on headloss development rates to allow for the highest total output before termination. Thus the dual media bed was found to be the optimum configuration for the natural raw water, using a relatively high dose.

• Bacterial removal rates using *M.oleifera* seed were highly successful in all the media configurations considered - 100% removal of coliforms and *E.coli*, which complies with the PVC. Furthermore, if *M.oleifera* was used in a situation where disinfection was not possible, then a 100% bacterial removal rate at the optimal operational conditions, would render the water potable. Although disinfection would always be recommend, due to the possibility of regrowth of microorganisms during supply of the water. The HPC was marginally higher in the 70 cm bed, indicating that potentially a higher collector surface area may be required to achieve optimal bacterial removal. Although considering the turbidity removal rates and the rate of headloss development, the optimal media choice would be the dual media bed.

• The extraneous material in the seed suspension resulted in an increase in the DOC of the final water, such that there was a direct correlation between the dose used and the DOC of the final water. Deeper beds resulted in a lower rate of increase in DOC, and thus it was concluded that removal rates of organics benefited from the increase in filter bed contact time, unlike turbidity removal. This was identified as a potential problem when using *M.oleifera* seed in full scale treatment, and indicates the necessity of optimising mixing and removal conditions, not only based on turbidity removal rates.

• It was considered that the seed suspension added to the organic content of the final water for two reasons. Firstly, the DOC increased after treatment with *M.oleifera* seed but not with Ferral. Secondly, derivatives of isothiocyanate were found in the final water, which had not previously been found in the raw water. Furthermore isothiocyanate compounds have been found by previous worker in *M.oleifera* seed, the significance of this is that isothiocyanate, (glycosidic mustard oil), contains an antimicrobial agent with potential health implications for use in potable water treatment. It was concluded that the majority of the DOC present in the raw water was removed during the filtration process, but this was concealed to some extent by the addition of organic matter from the seed suspension. The use of the presscake as the coagulant or the protein extract in the treatment process would result in a reduction of the DOC of the final water.

• Turbidity and bacterial removal with *M.oleifera* seed was marginally greater than with Ferral, despite the difference in destabilisation and floc formation mechanisms of the two coagulants. The advantage of *M.oleifera* seed over Ferral is the lower rate of headloss development and consequently a higher total output before termination of the run. The major concern with using *M.oleifera* was the increase in the DOC of the water and the consequent increase in THM formation. Some THM compounds were not detected at all, whilst other were found in similar concentrations in water treated with *M.oleifera* seed as they were in waters treated with Ferral. However, the concentrations found were below the UK's PVC. There are other potential issues associated with an increase in DOC in the final water, such as bacterial regrowth with time and higher chlorine demand. These would need to be investigated further.

• It was expected that low turbidity water may have required a flocculation stage to enhance turbidity and bacterial removal rates, when using *M.oleifera*, due to the need for sufficient flocculation time. But the addition of a flocculation stage did not increase the removal rates. In fact the HPC were marginally higher with DF than CFF, indicating that CFF would potentially offer greater bacterial removal rates than DF. It is recognised that the data on bacterial removal is limited, and therefore no firm conclusions can be drawn.

• The overall conclusions of this field study were that *M.oleifera* seed was very effective in the treatment of the natural, low turbidity water in the CFF and DF modes of operation. Turbidity removal rates of 95% and bacterial removal of 100% compared very well with those achieved

using Ferral in the pilot plant and the Works. There was, however, an increase in the organic matter in the final water; although THM formation was not significant, other compounds were found which demand further investigation.

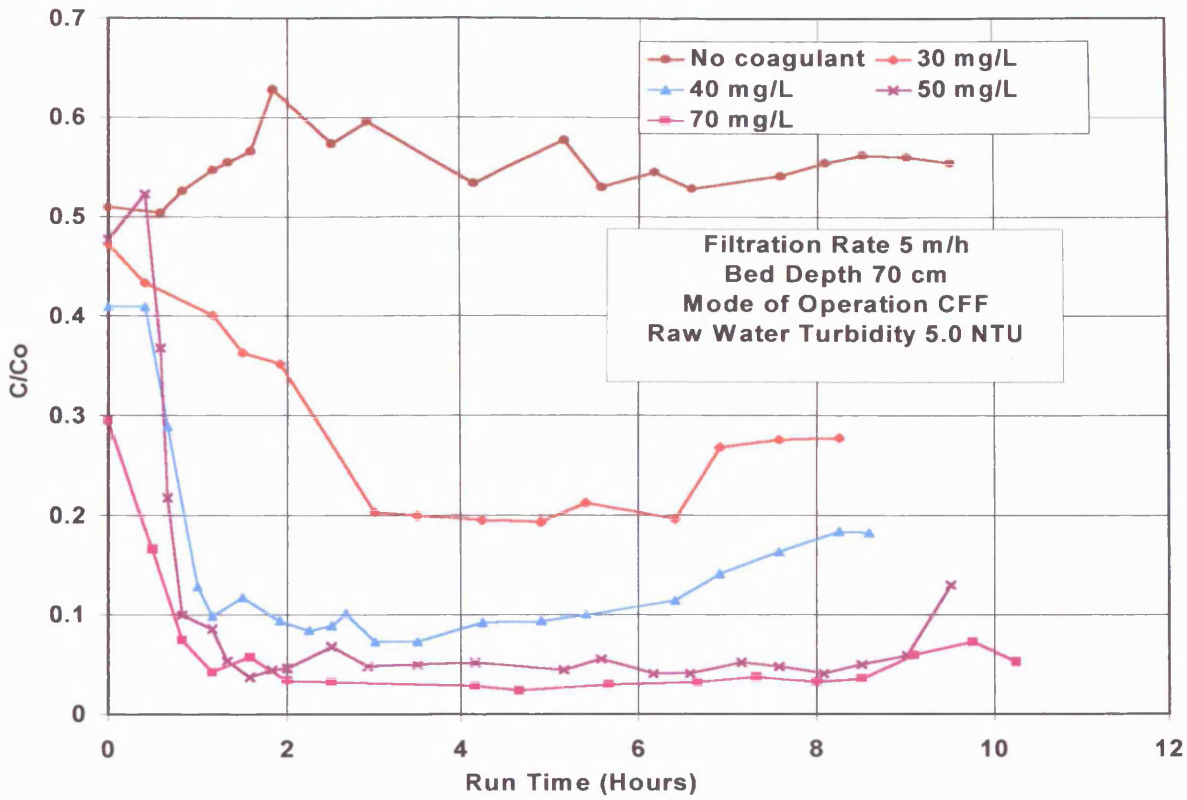

Figure 6.4 Turbidity reduction as a function of M.oleifera dose; using CFF, 5 m/h,70 cm sand.

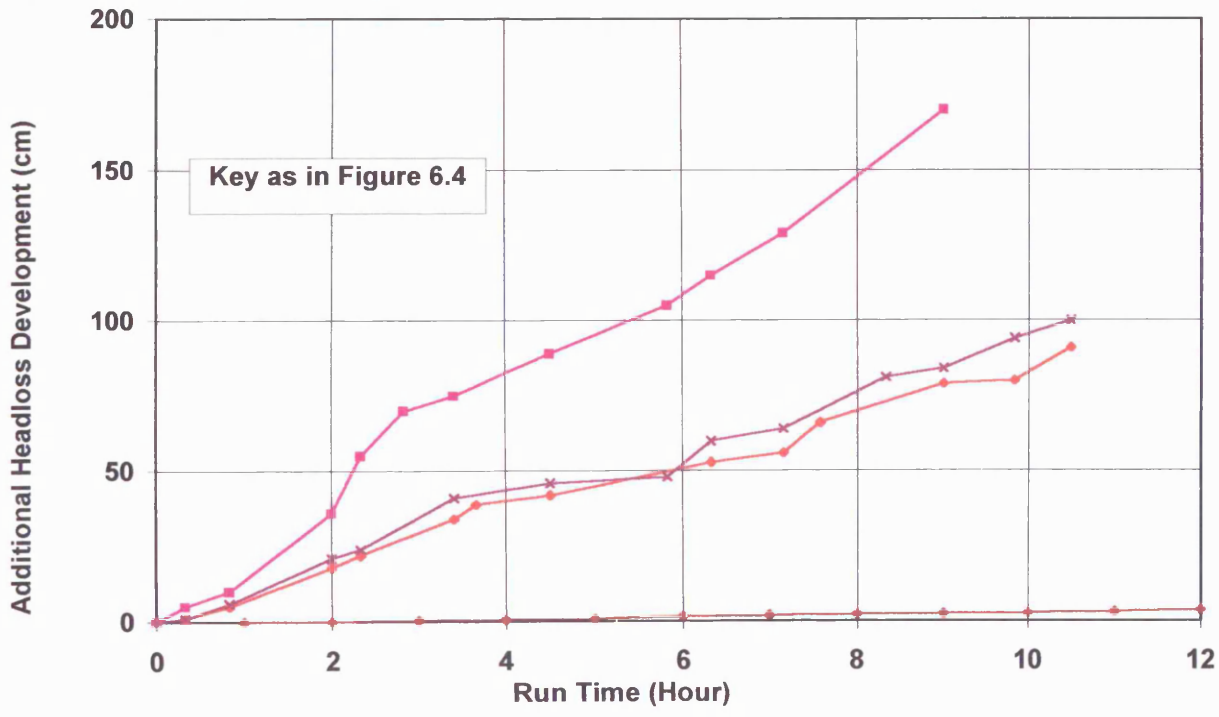

Figure 6.5 Headloss development as a function of M.oleifera dose; using CFF, 5 NTU, 5 m/h,70 cm sand.

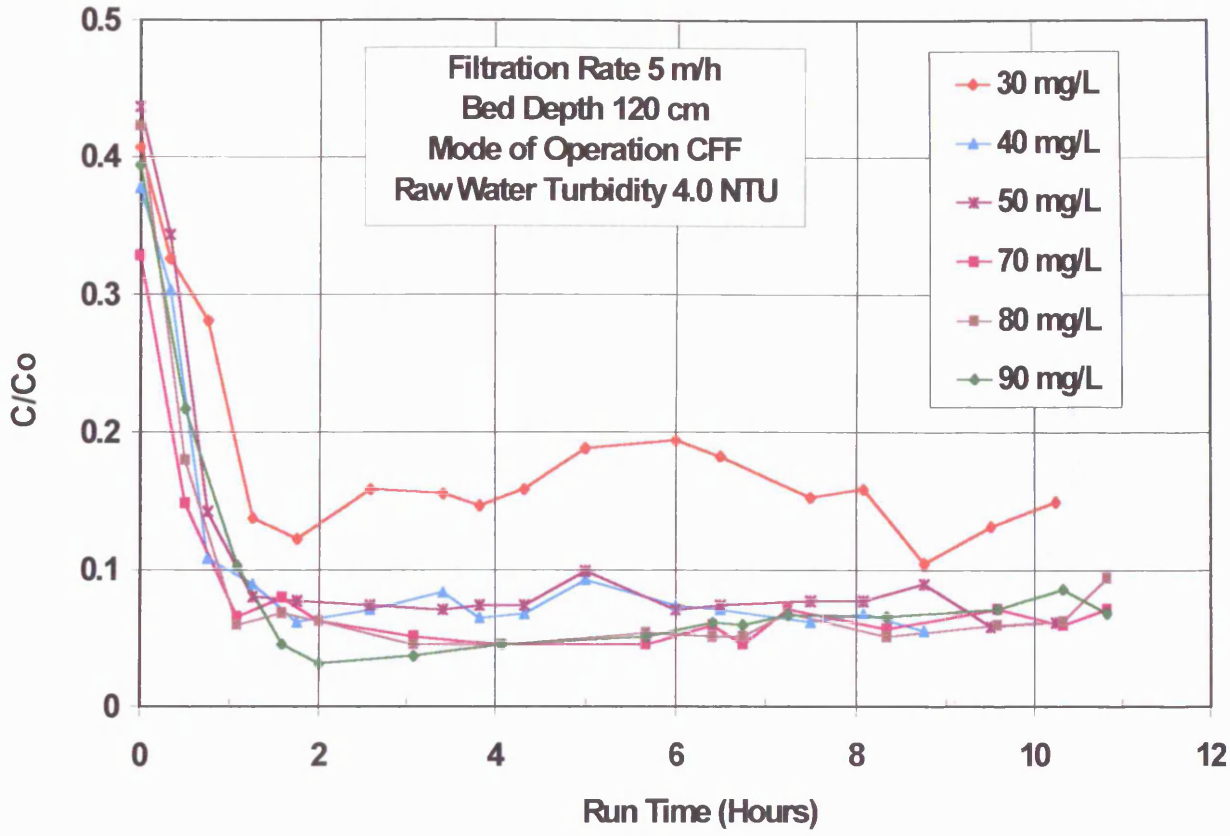

Figure 6.6 Turbidity reduction as a function of M.oleifera dose; using CFF, 5 m/h, 120 cm sand.

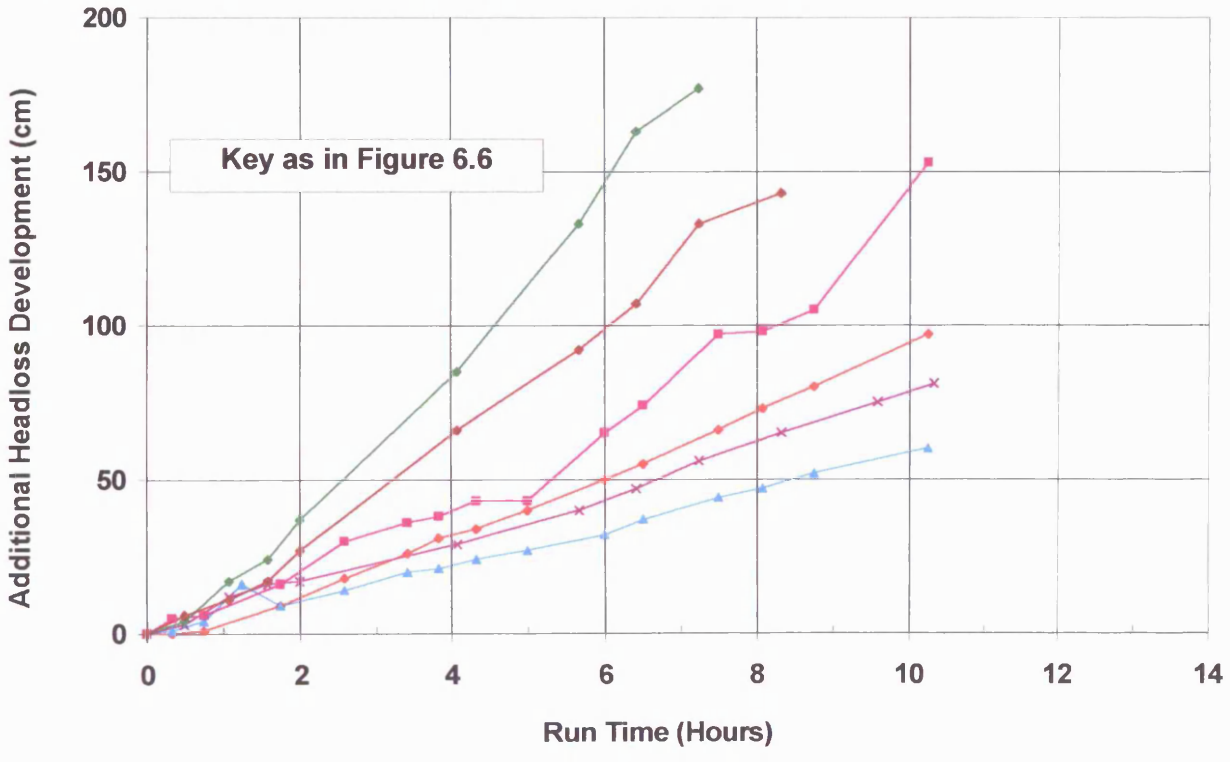

Figure 6.7 Headloss development as a function of M.oleifera dose; using CFF, 5 m/h, 120 cm sand.

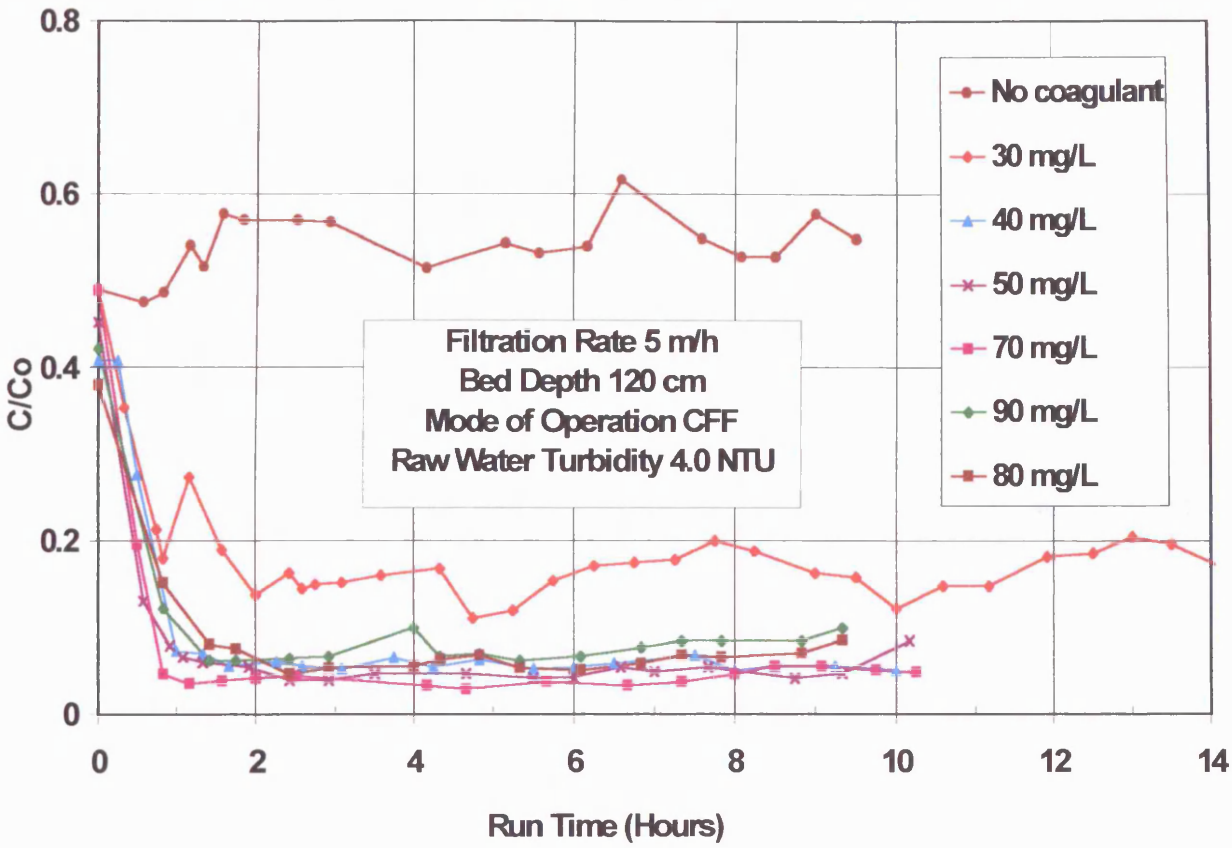

Figure 6.8 Turbidity reduction as a function of M.oleifera dose; using CFF, 5 m/h, 70 cm sand & 50 cm anthracite.

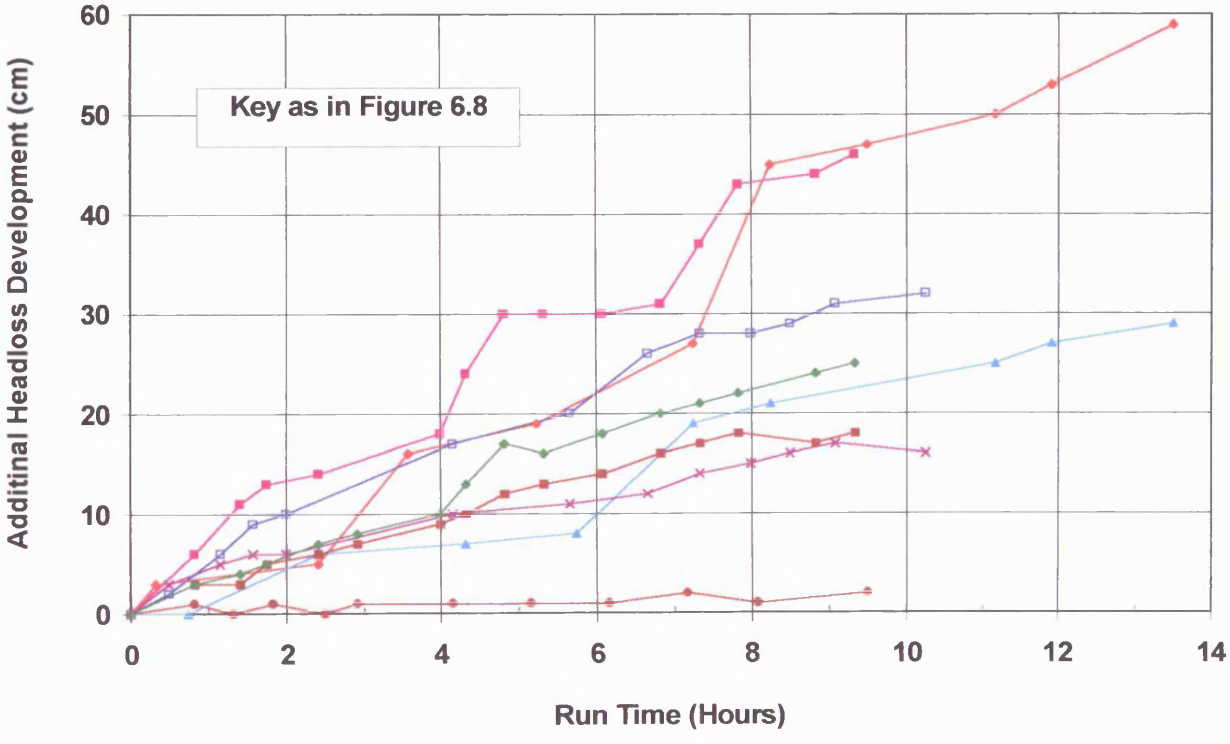

Figure 6.9 Headloss development as a function of M.oleifera dose; using CFF, 5 NTU, 70 cm sand & 50 cm anthracite.

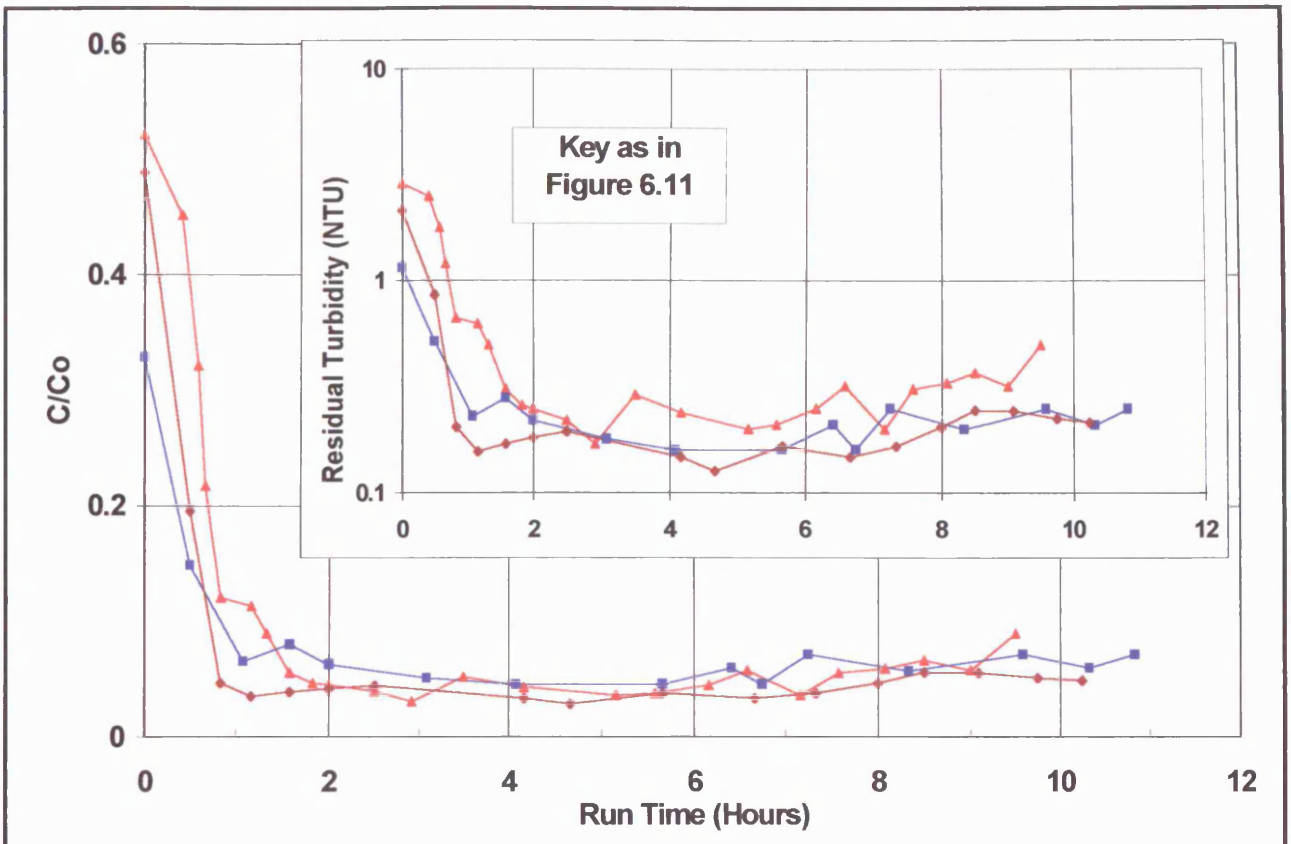

Figure 6.10 Turbidity reduction as a function of filter media; using M.oleifera and CFF, 5 m/h.

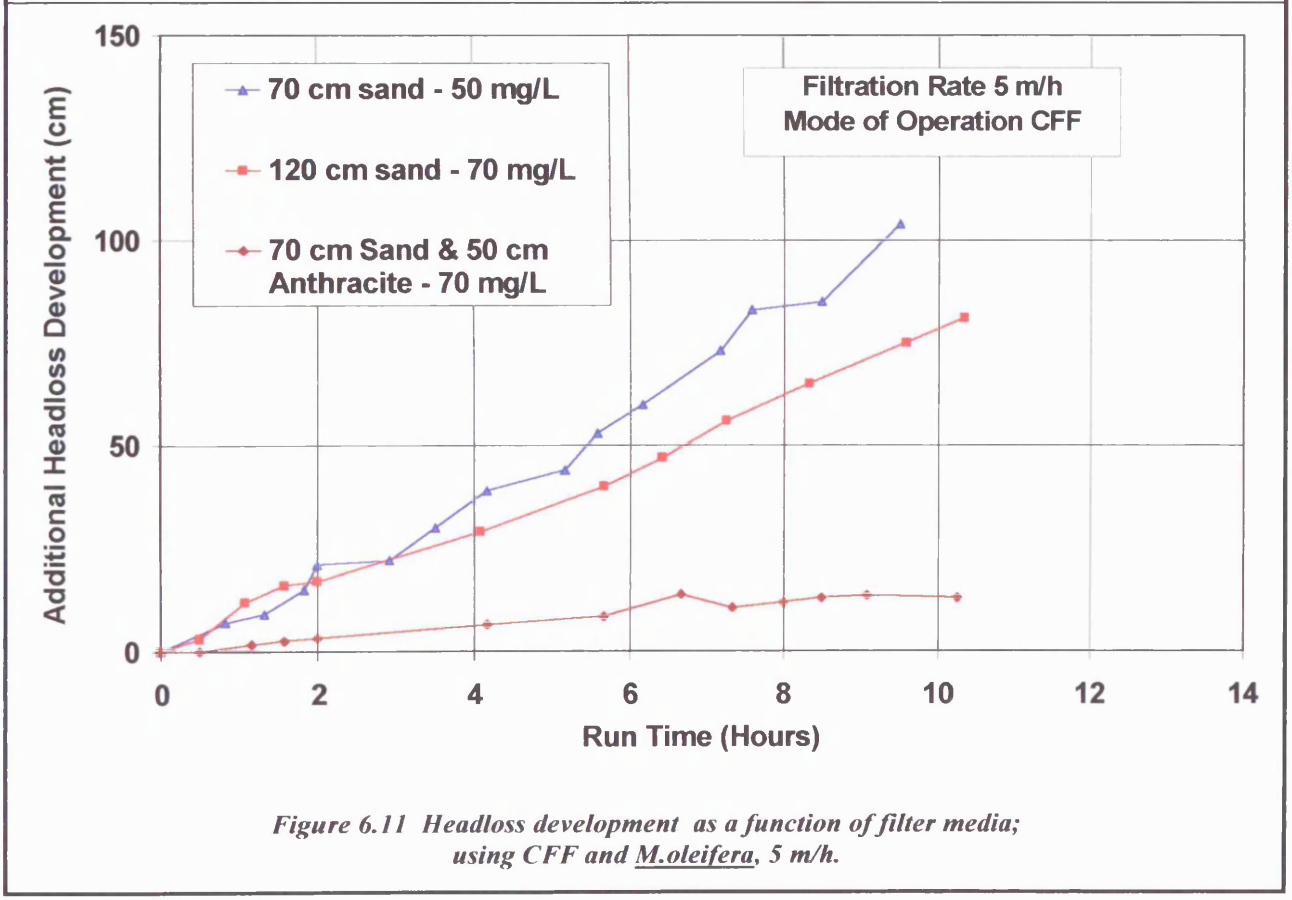

Figure 6.11 Headloss development as a function of filter media; using CFF and M.oleifera, 5 m/h.

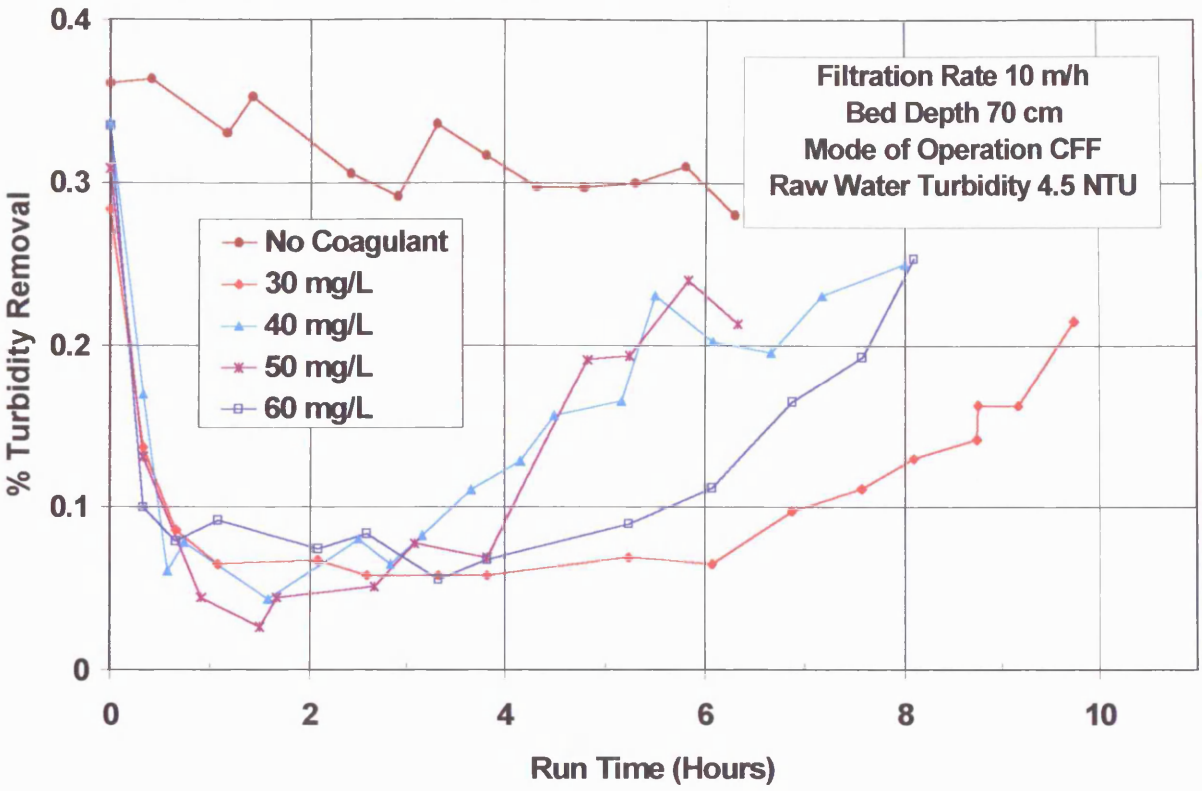

Figure 6.12 Turbidity reduction as a function of M.oleifera dose; using CFF, 10 m/h, 70 cm sand.

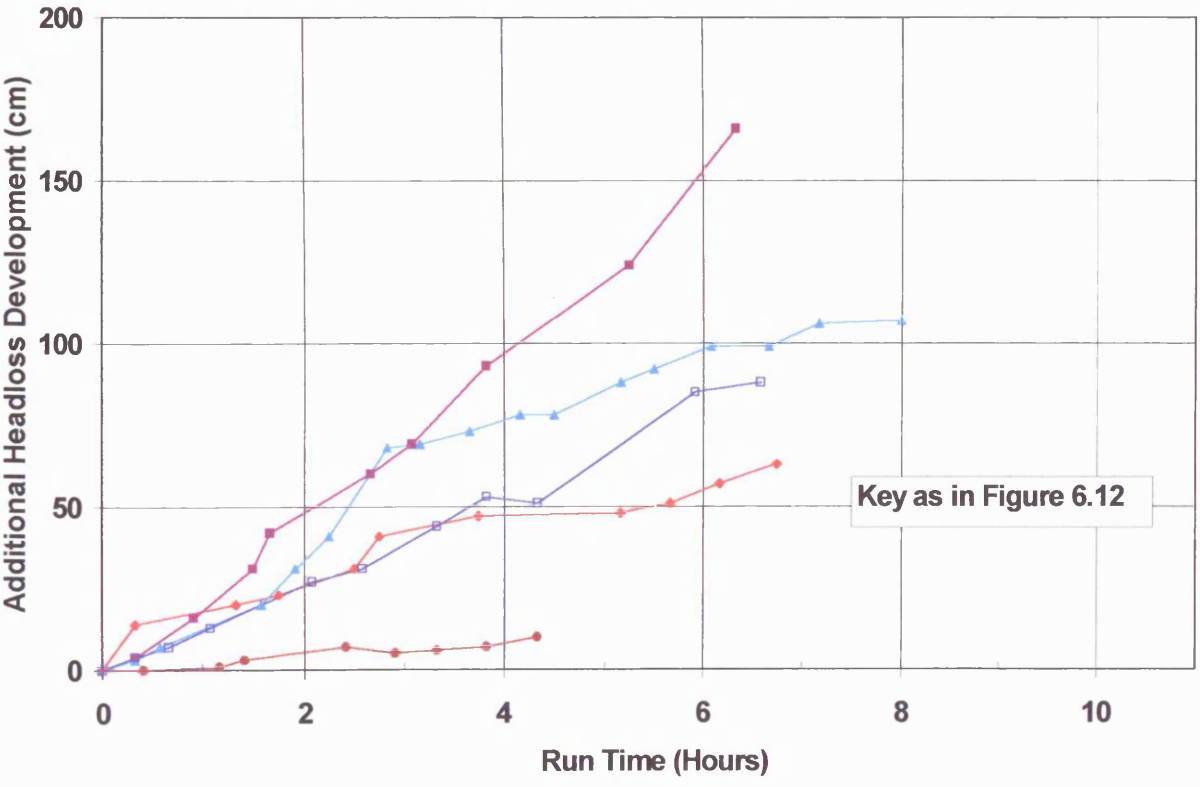

Figure 6.13 Headloss development as a function of M.oleifera dose, using CFF, 10 m/h, 70 cm sand.

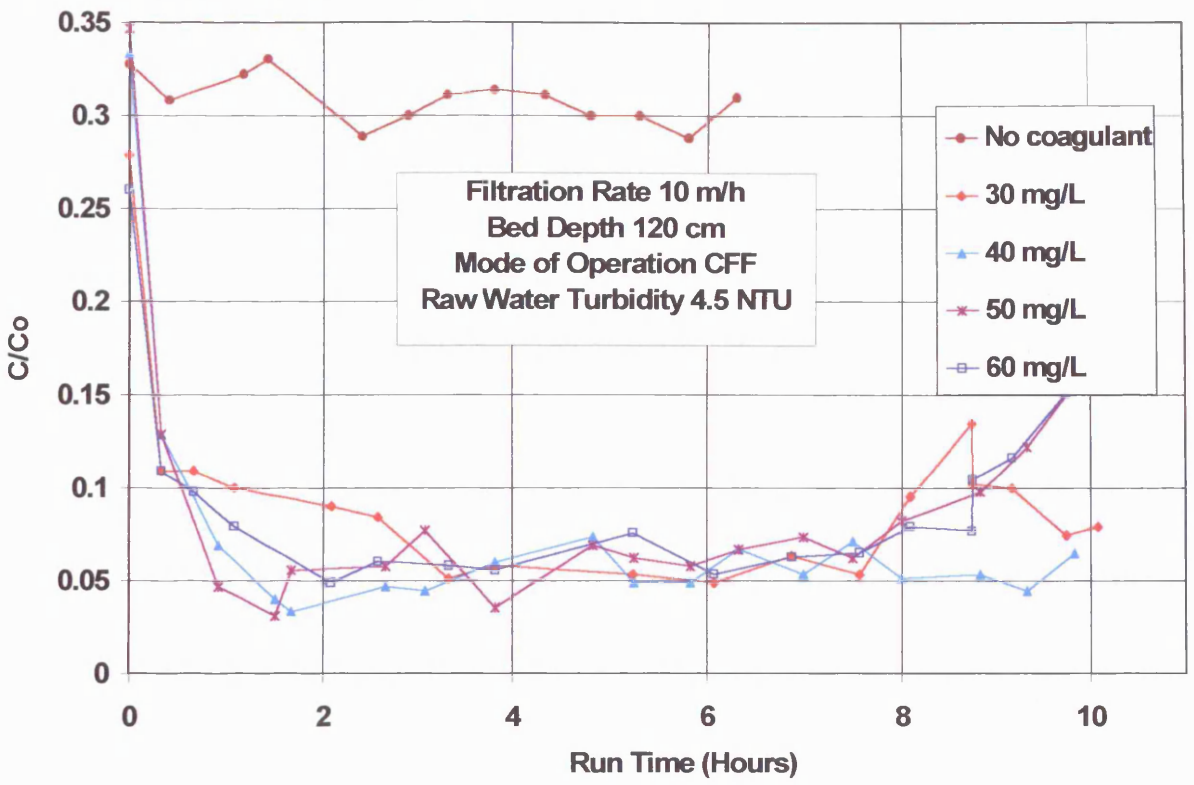

Figure 6.14 Turbidity reduction as a function of M.oleifera dose; using CFF, 10 m/h, 120 cm sand.

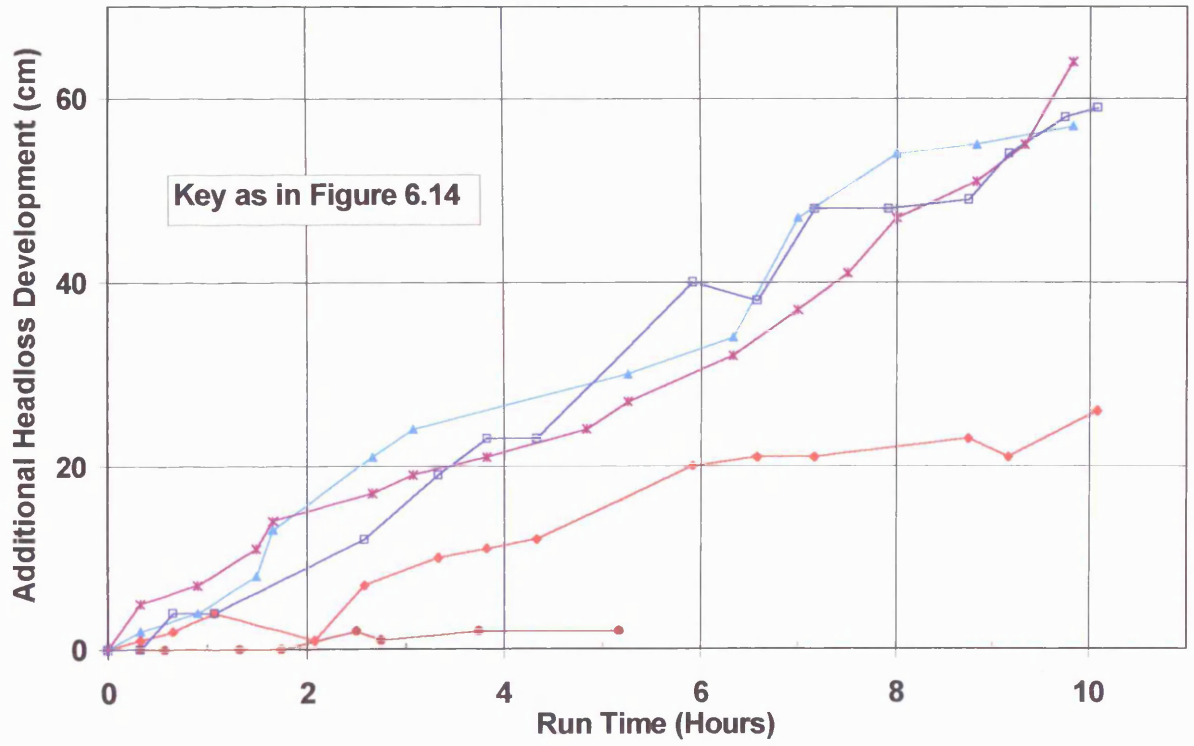

Figure 6.15 Headloss development as a function of M.oleifera dose; using CFF, 10 m/h, 120 cm sand.

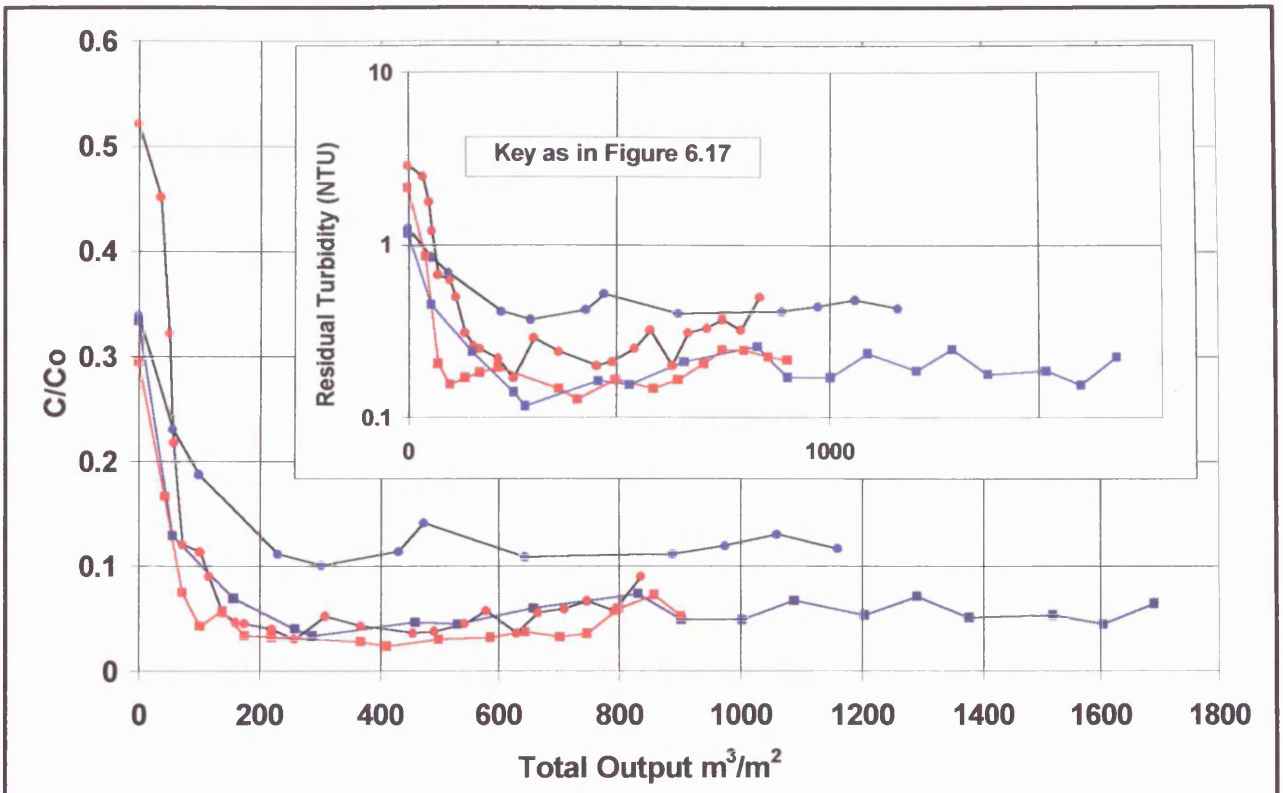

Figure 6.16 Turbidity reduction as a function of filtration rate & filter media; using CFF.

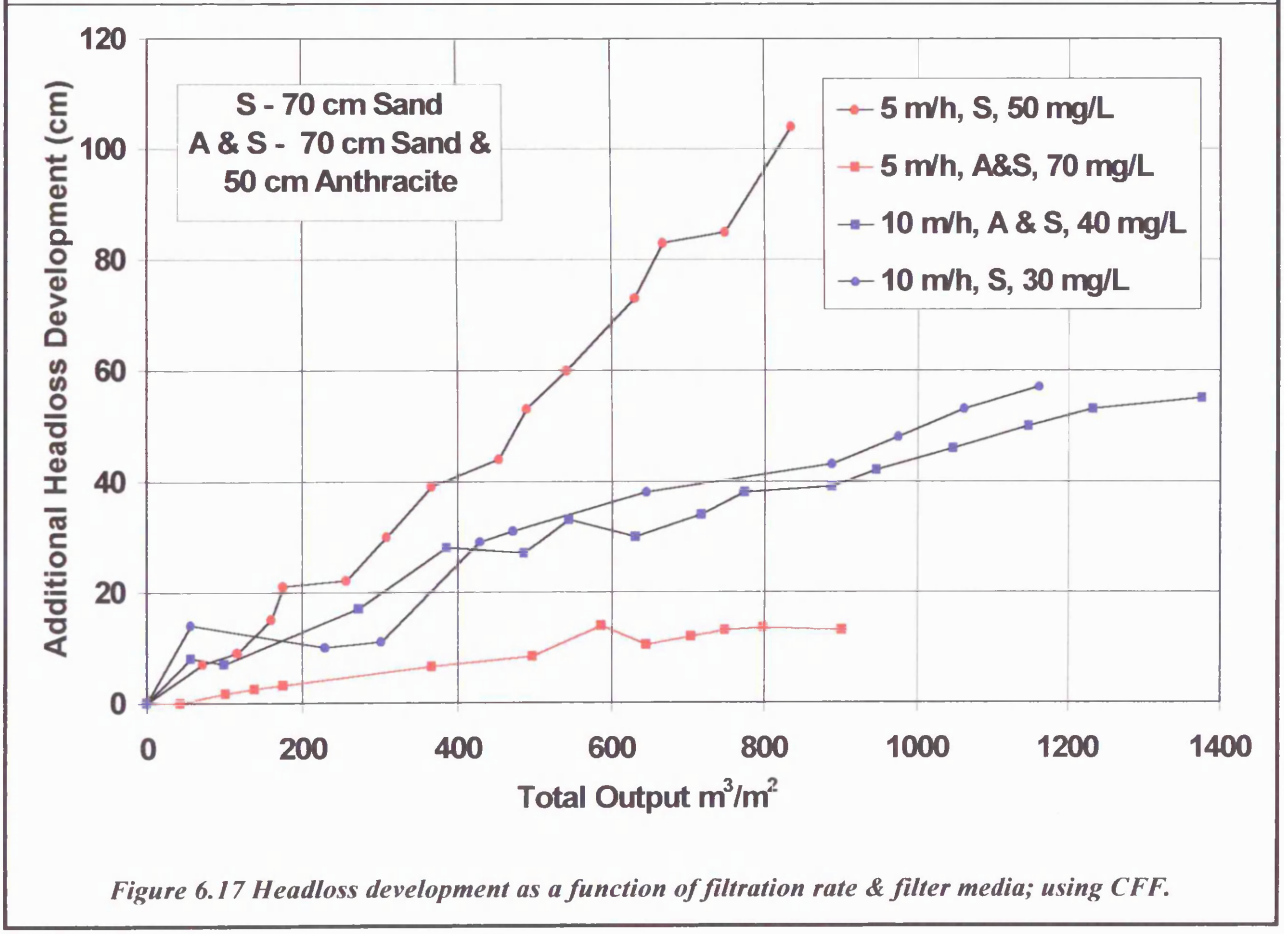

Figure 6.17 Headloss development as a function of filtration rate & filter media; using CFF.

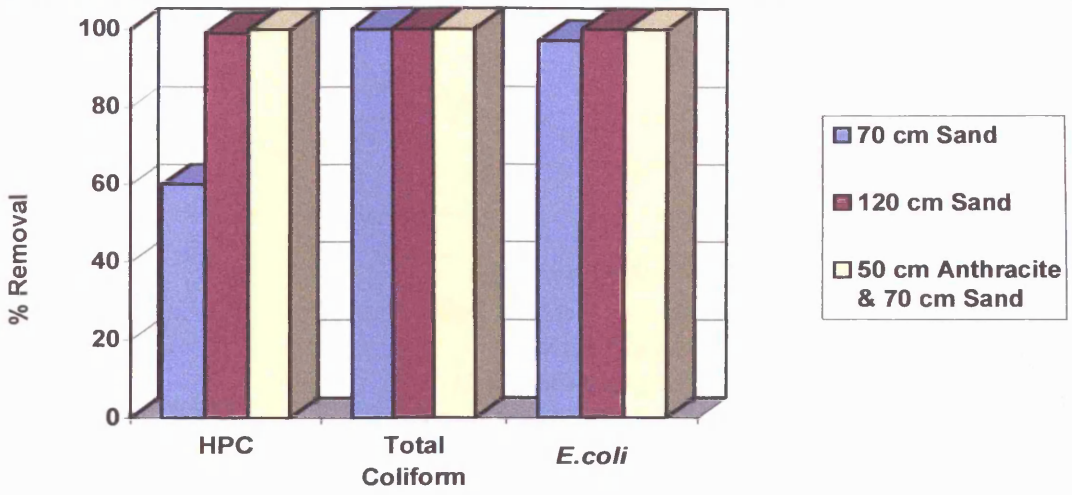

Figure 6.18 Total coliforms, E.coli and Heterotrophic bacteria reduction as a function of media configuration and M.oleifera dose.

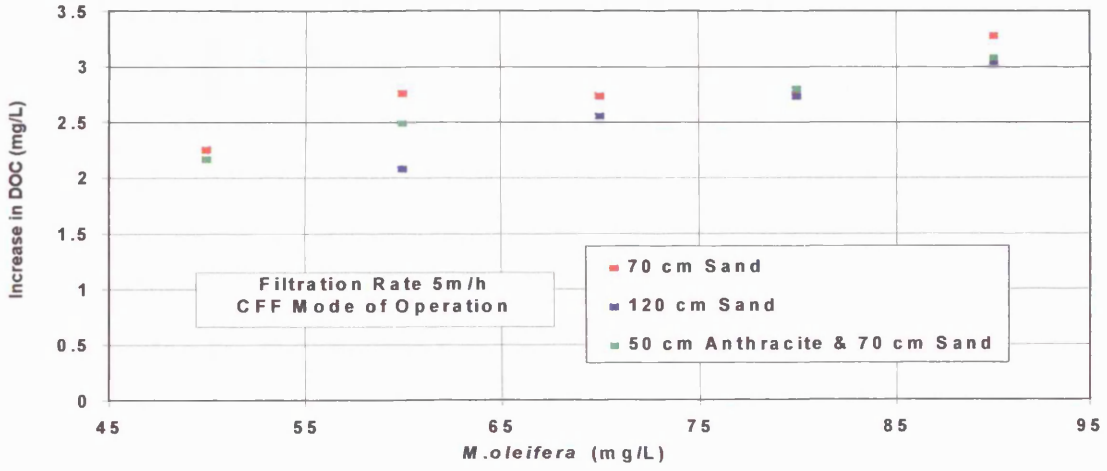

Figure 6.19 Increase in DOC after treatment with M.oleifera as a function of media configurations and the dose used.

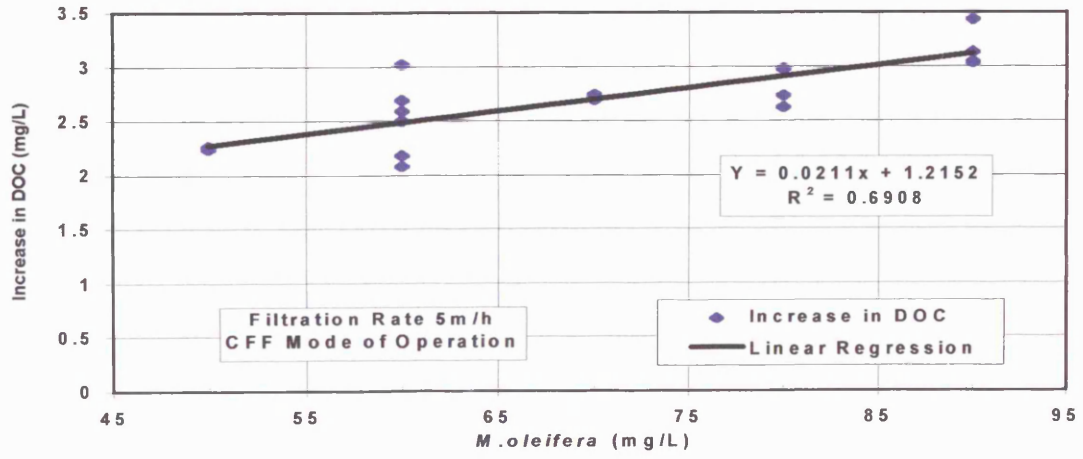

Figure 6.20 Correlation of the increase in DOC of water after treatment with M.oleifera and the dose used.

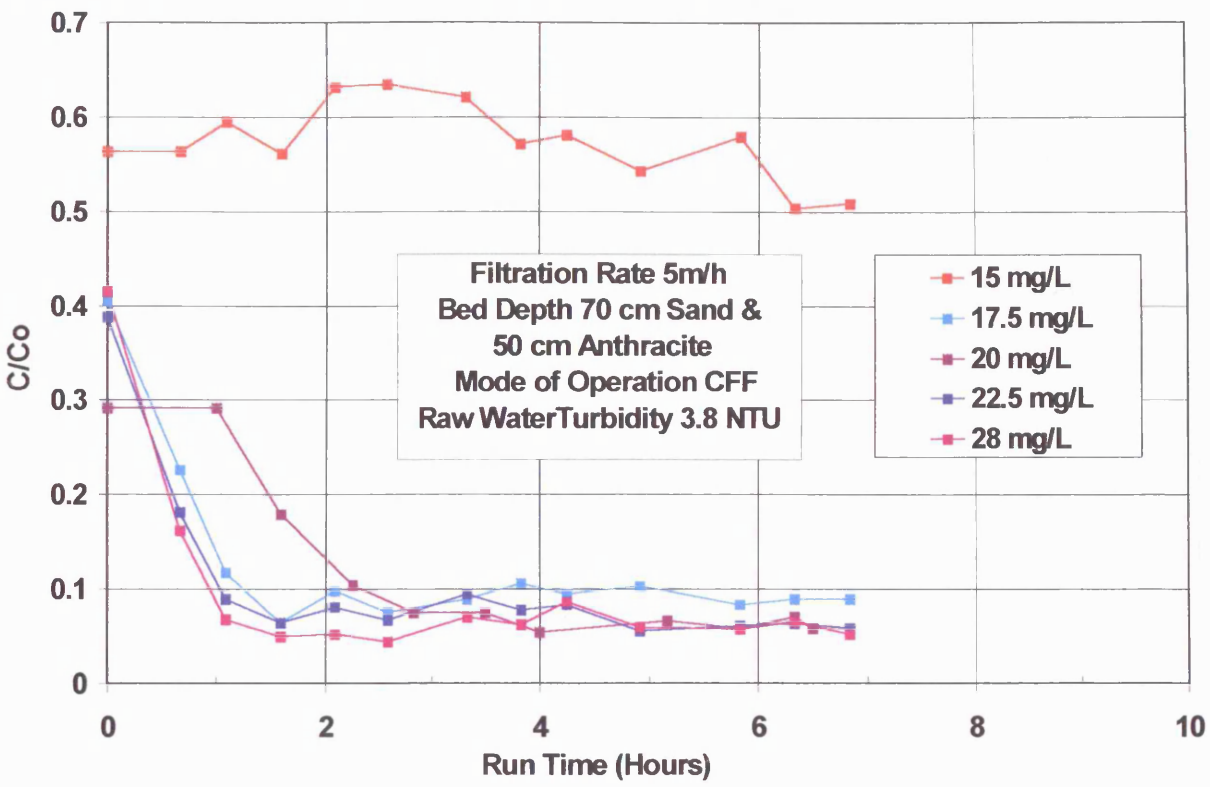

Figure 6.21 Turbidity reduction as a function of Ferral dose; using CFF, 70 cm sand.

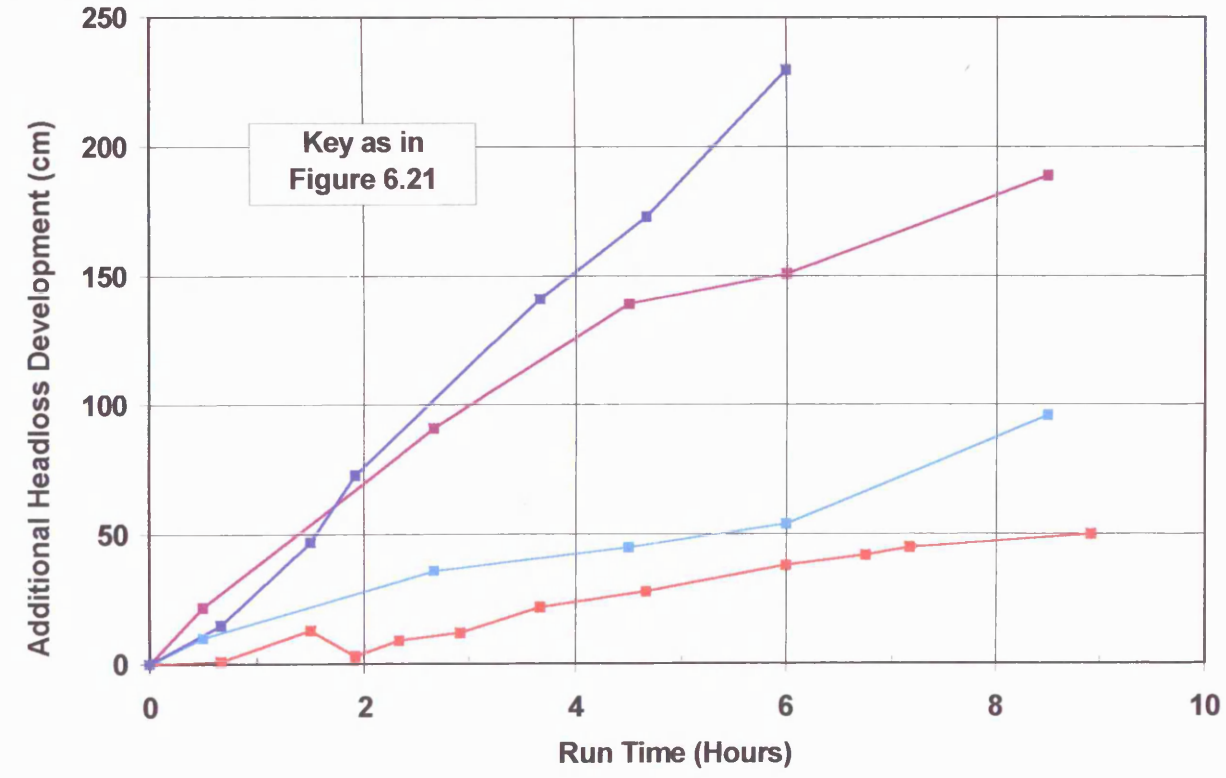

Figure 6.22 Headloss development as a function of Ferral dose; using CFF, 70 cm sand.

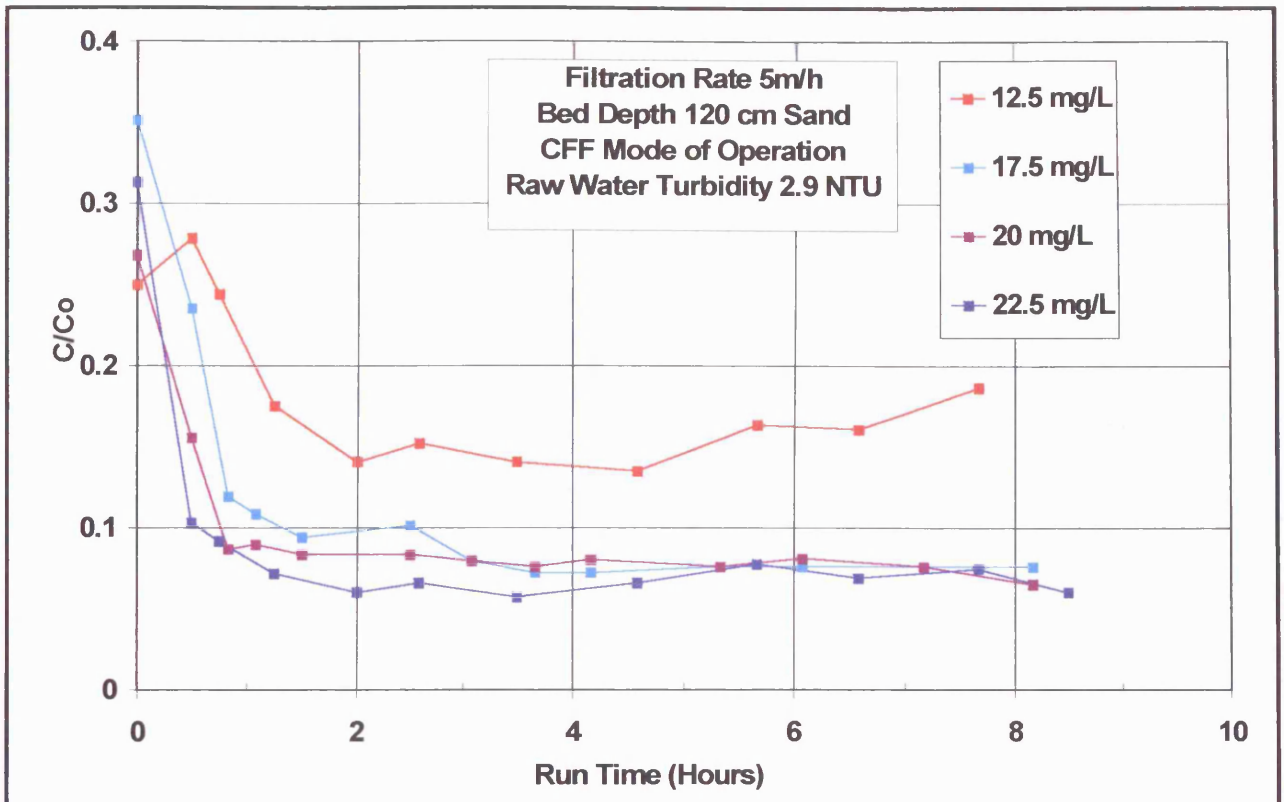

Figure 6.23 Turbidity reduction as a function of Ferral dose; using CFF, 120 cm sand.

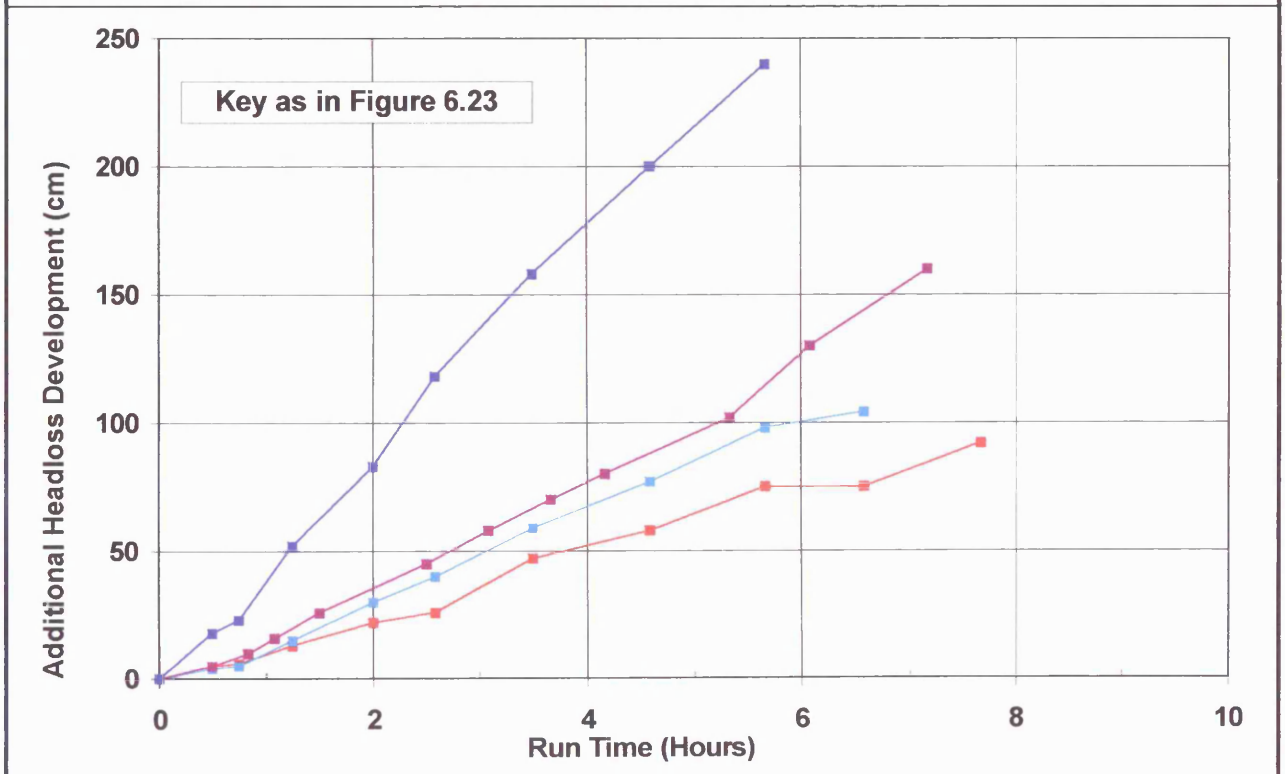

Figure 6.24 Headloss development as a function of Ferral dose; using CFF, 120 cm sand.

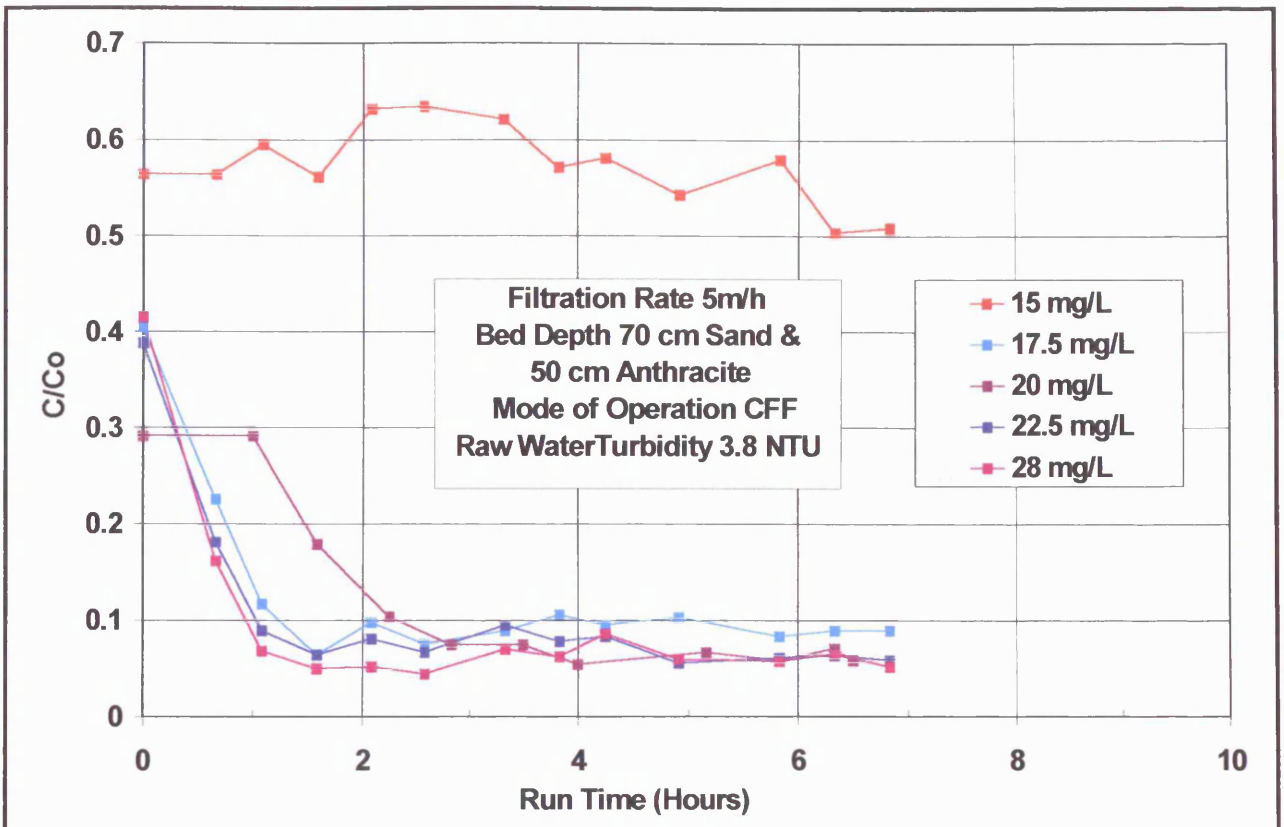

Figure 6.25 Turbidity reduction as a function of Ferral dose; using CFF, 70 cm sand & 50 cm anthracite.

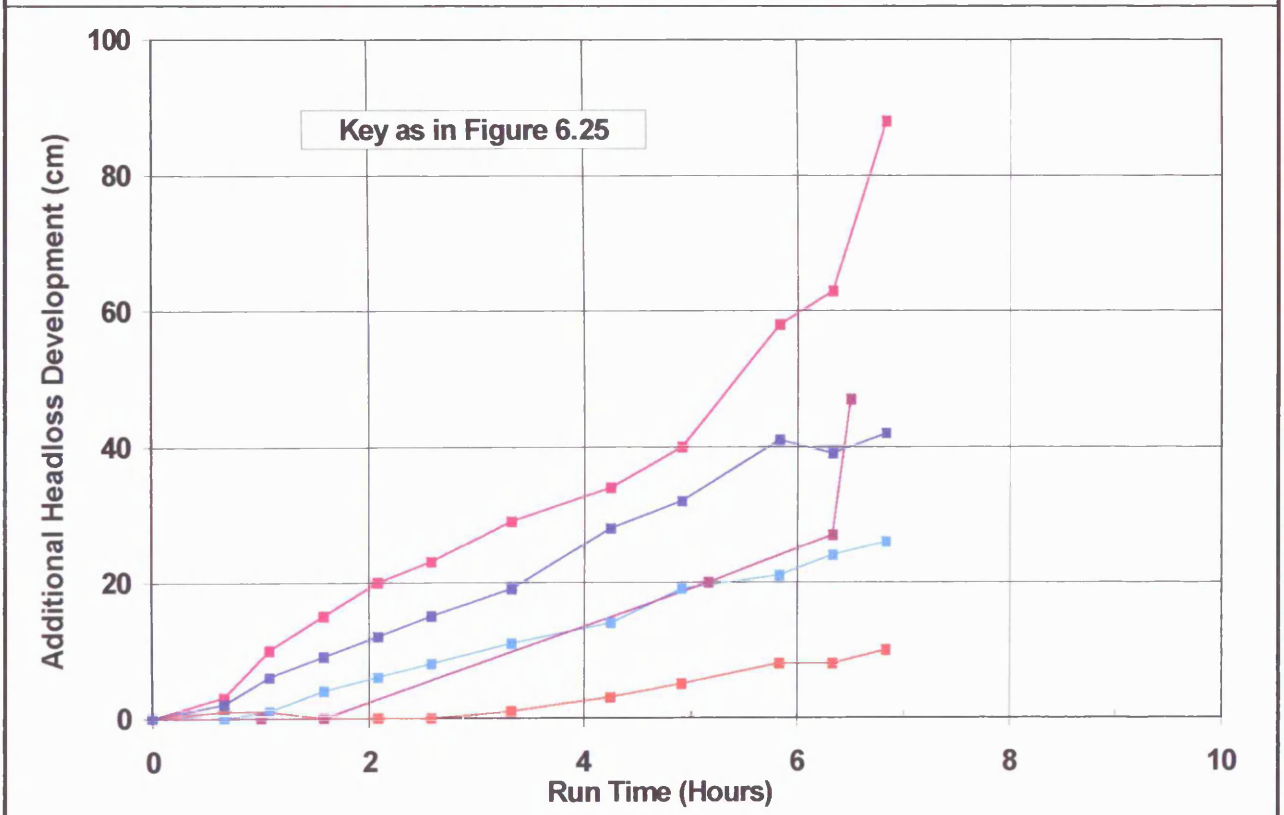

Figure 6.26 Headloss development as a function of Ferral dose; using CFF, 70 cm sand & 50 cm anthracite.

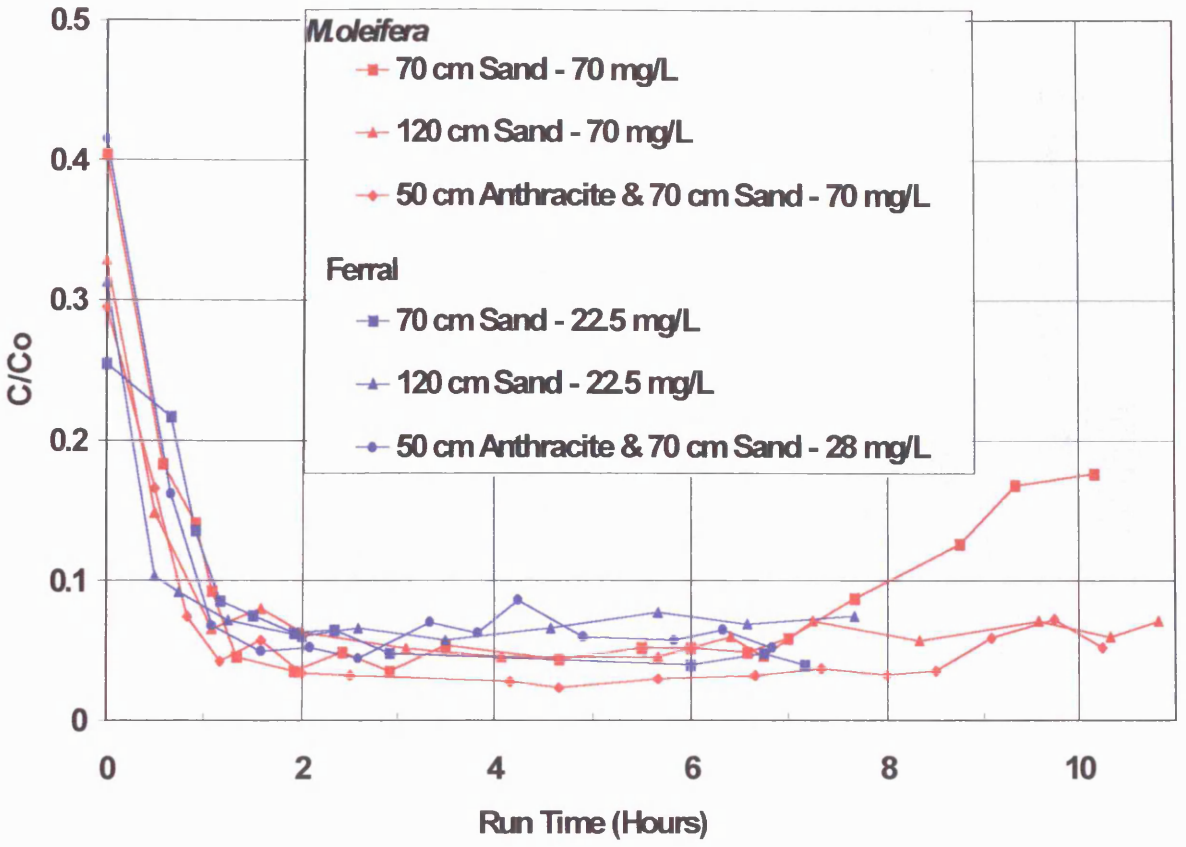

Figure 6.27 Turbidity reduction as a function of coagulant & media configuration; using CFF.

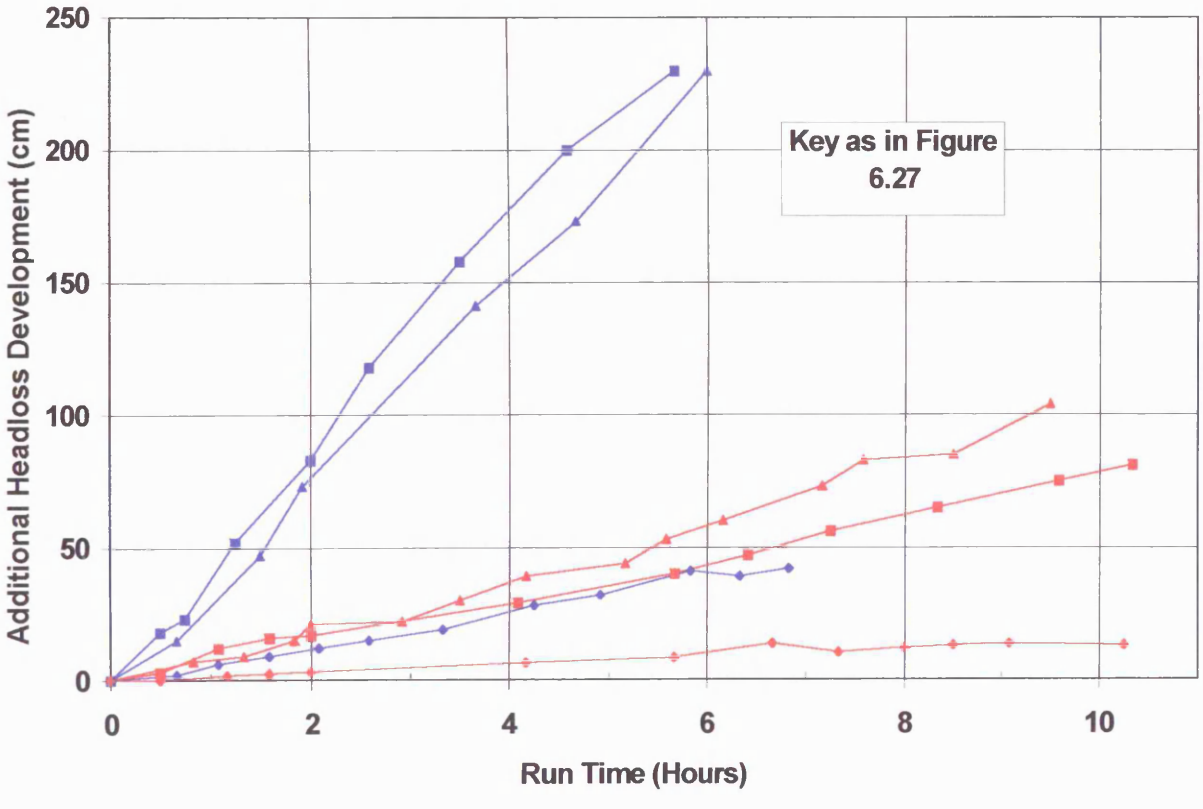

Figure 6.28 Headloss development as a function of coagulant & media configuration; using CFF.

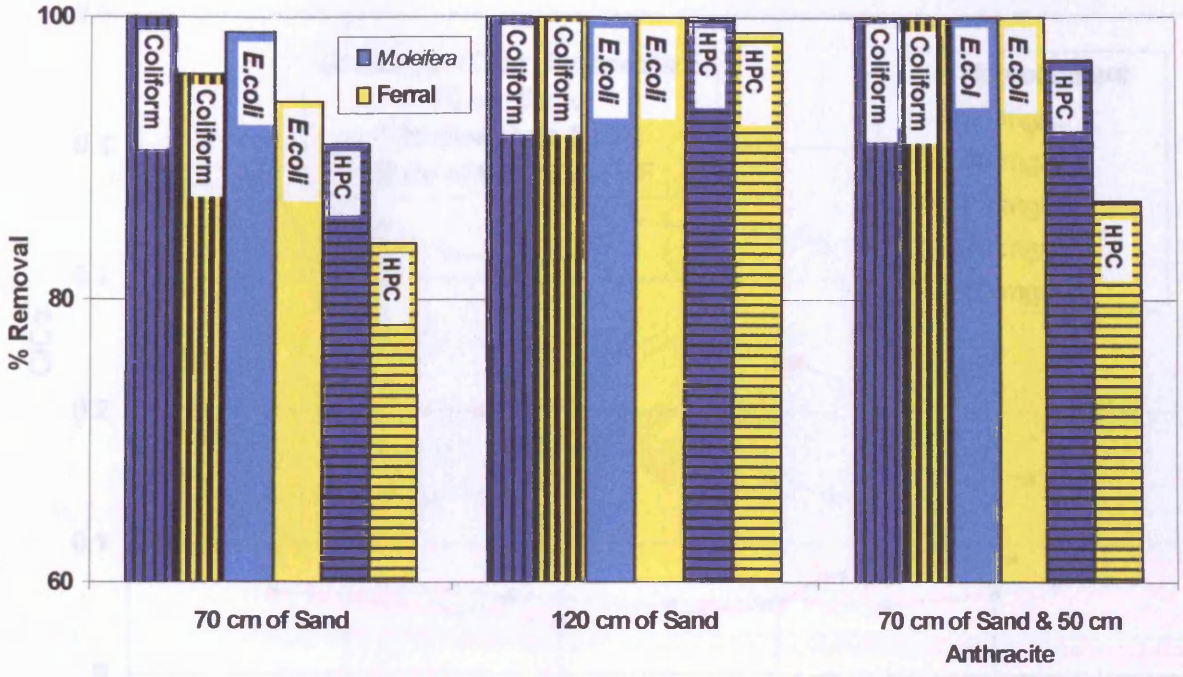

Figure 6.29 Bacterial reduction rates as a function of coagulant & media configuration; using CFF, 5 m/h.

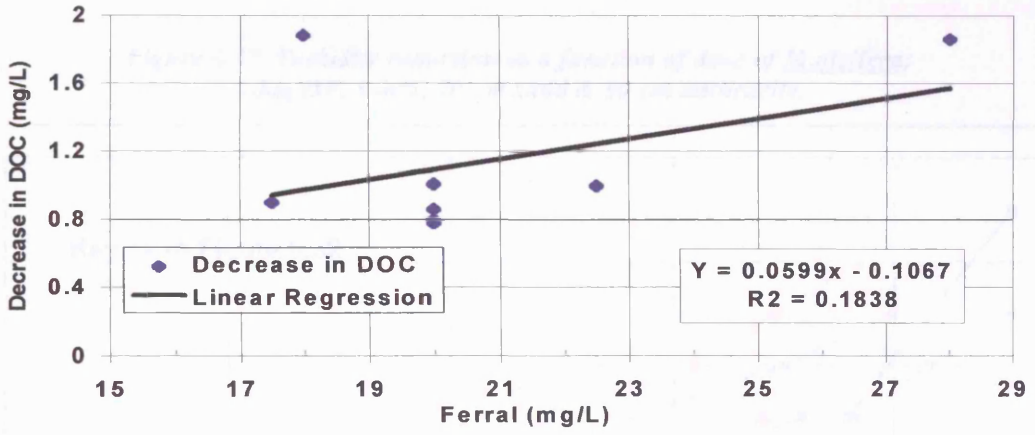

Figure 6.30 Correlation of the decrease in DOC of water after treatment with Ferral and the dose used.

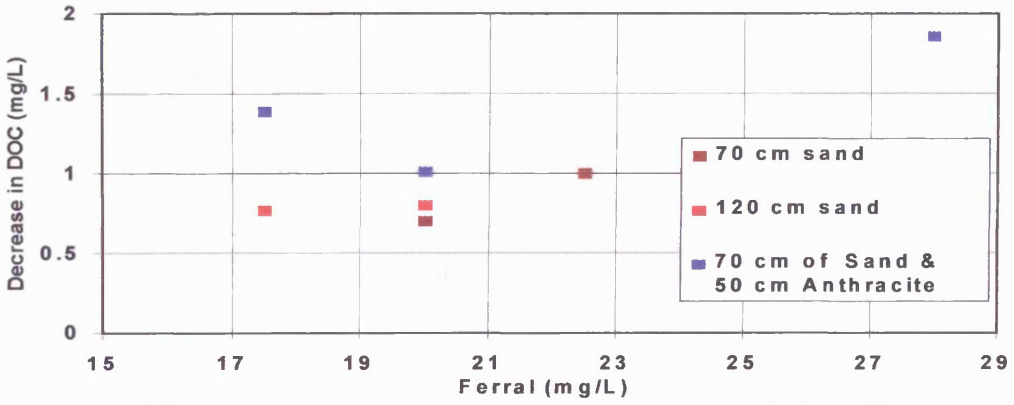

Figure 6.31 Decrease in DOC after treatment with Ferral as a function of media configurations and dose.

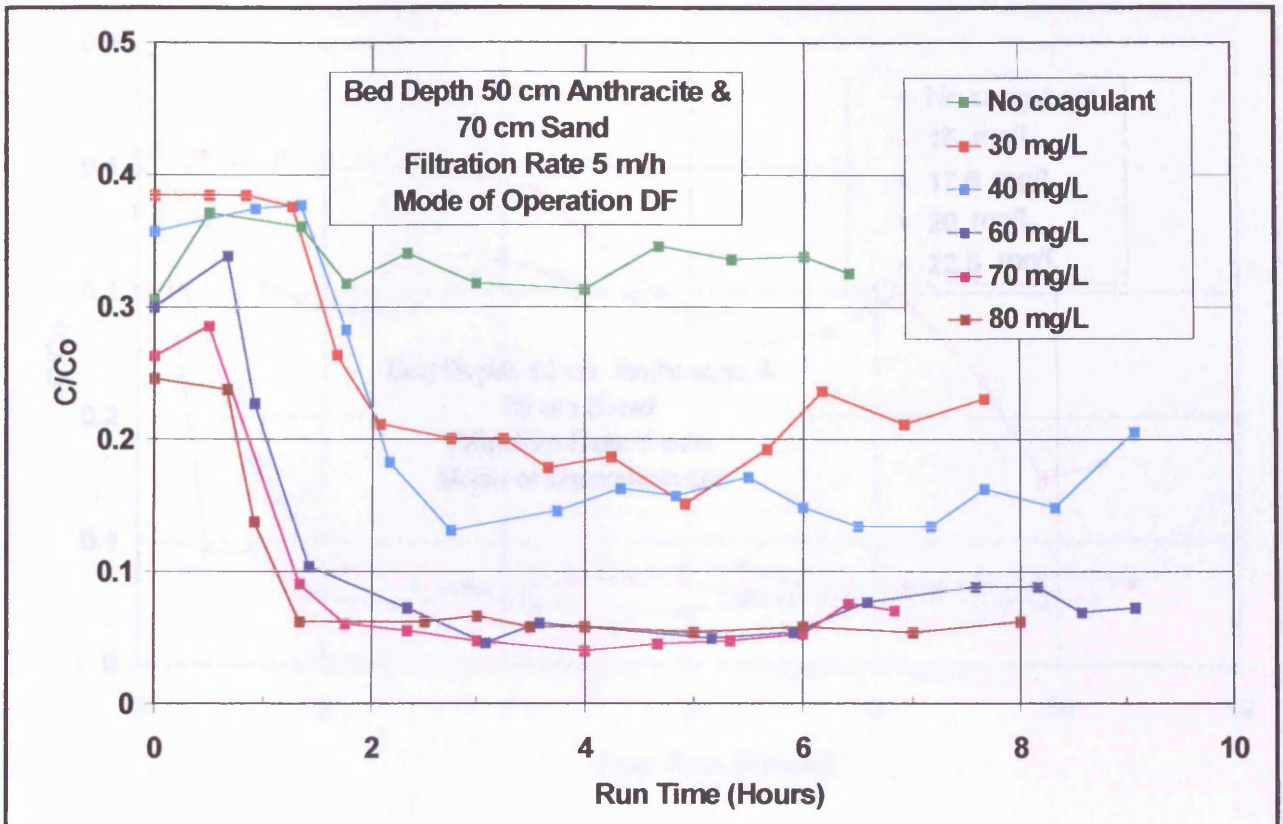

Figure 6.32 Turbidity reduction as a function of dose of M.oleifera; using DF, 5 m/h, 70 cm sand & 50 cm anthracite.

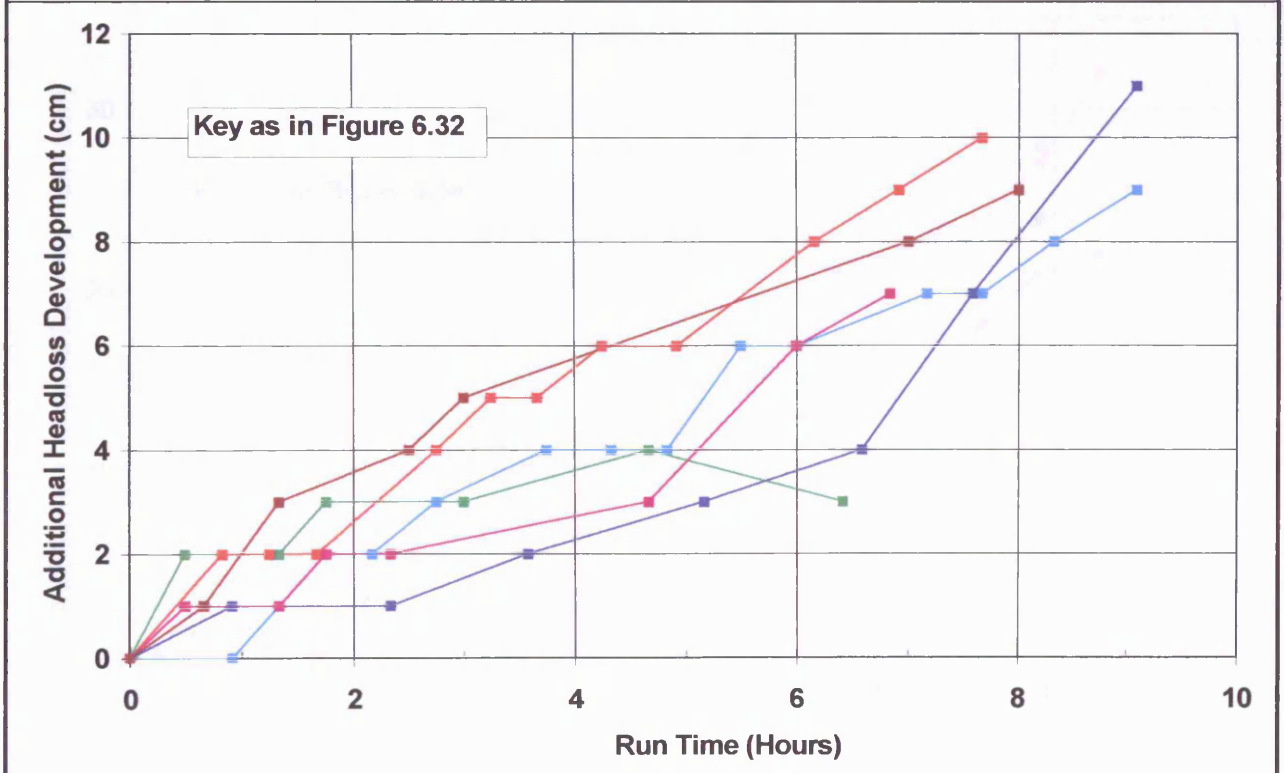

Figure 6.33 Headloss development as function of dose of M.oleifera; using DF, 5 m/h, 70 cm sand & 50 cm anthracite.

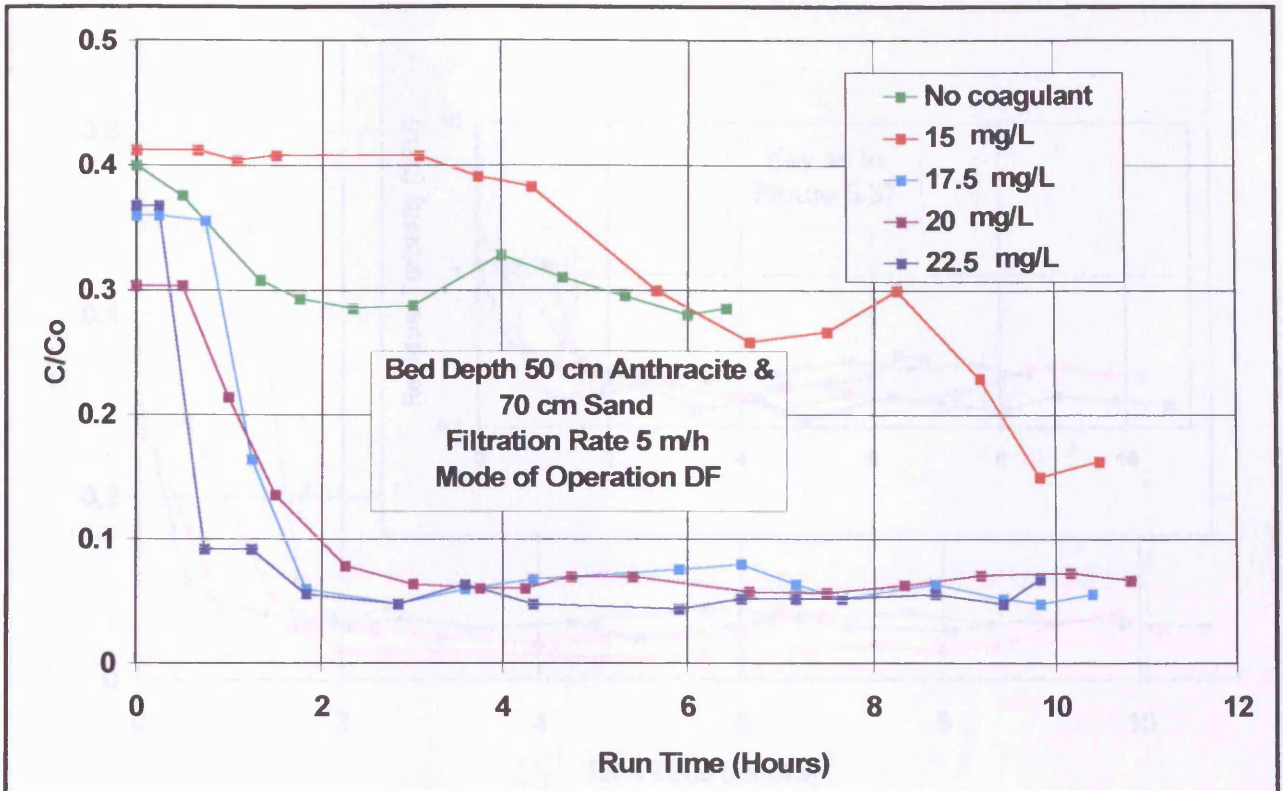

Figure 6.34 Turbidity reduction as a function of dose of Ferral; using DF, 5 m/h, 70 cm sand & 50 cm anthracite.

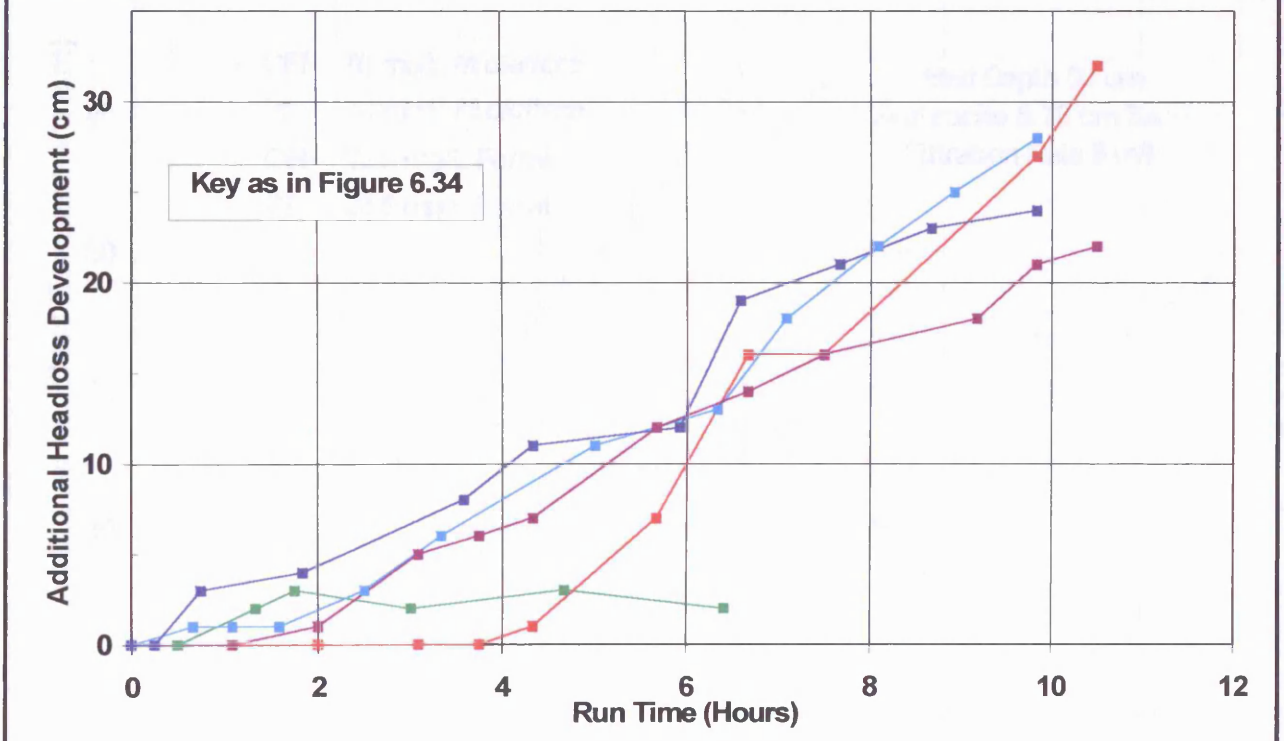

Figure 6.35 Headloss development as a function of dose of Ferral; using DF, 5 m/h, 70 cm sand & 50 cm anthracite.

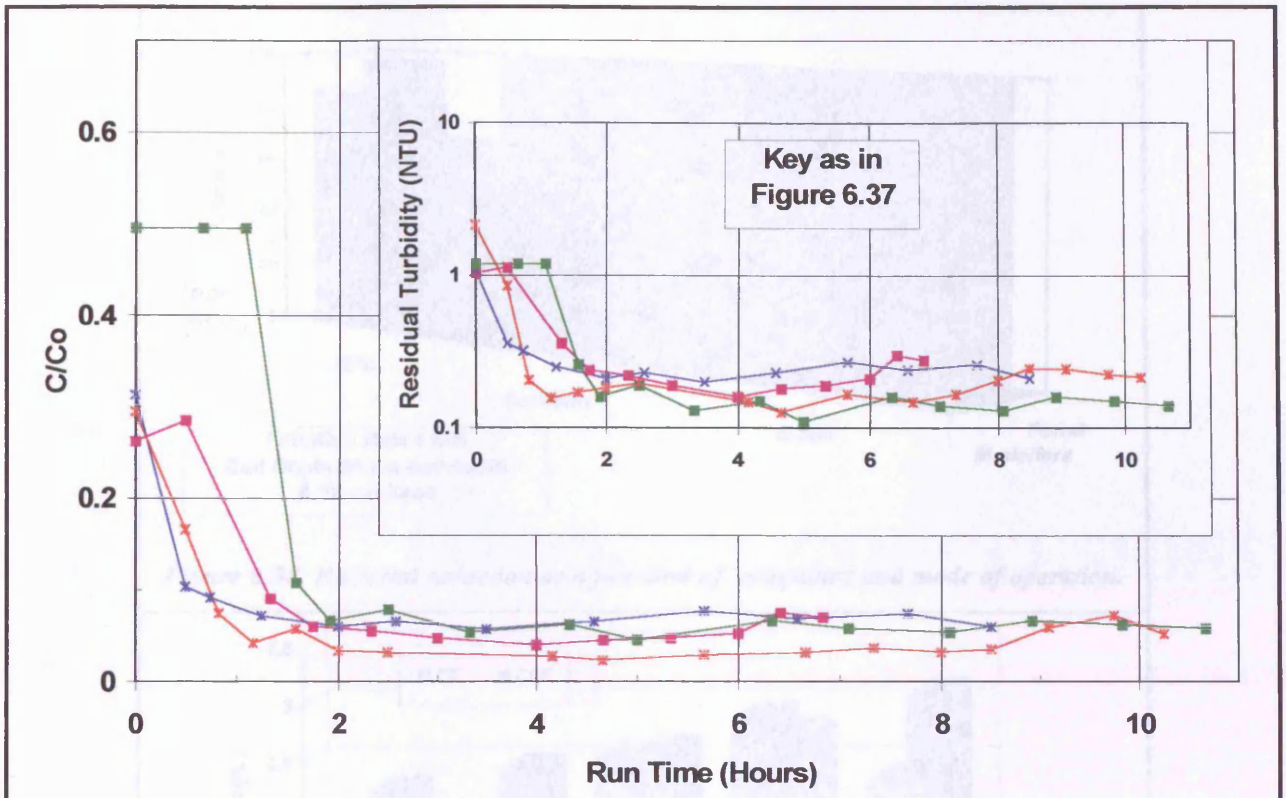

Figure 6.36 Turbidity reduction as a function of coagulant and mode of operation.

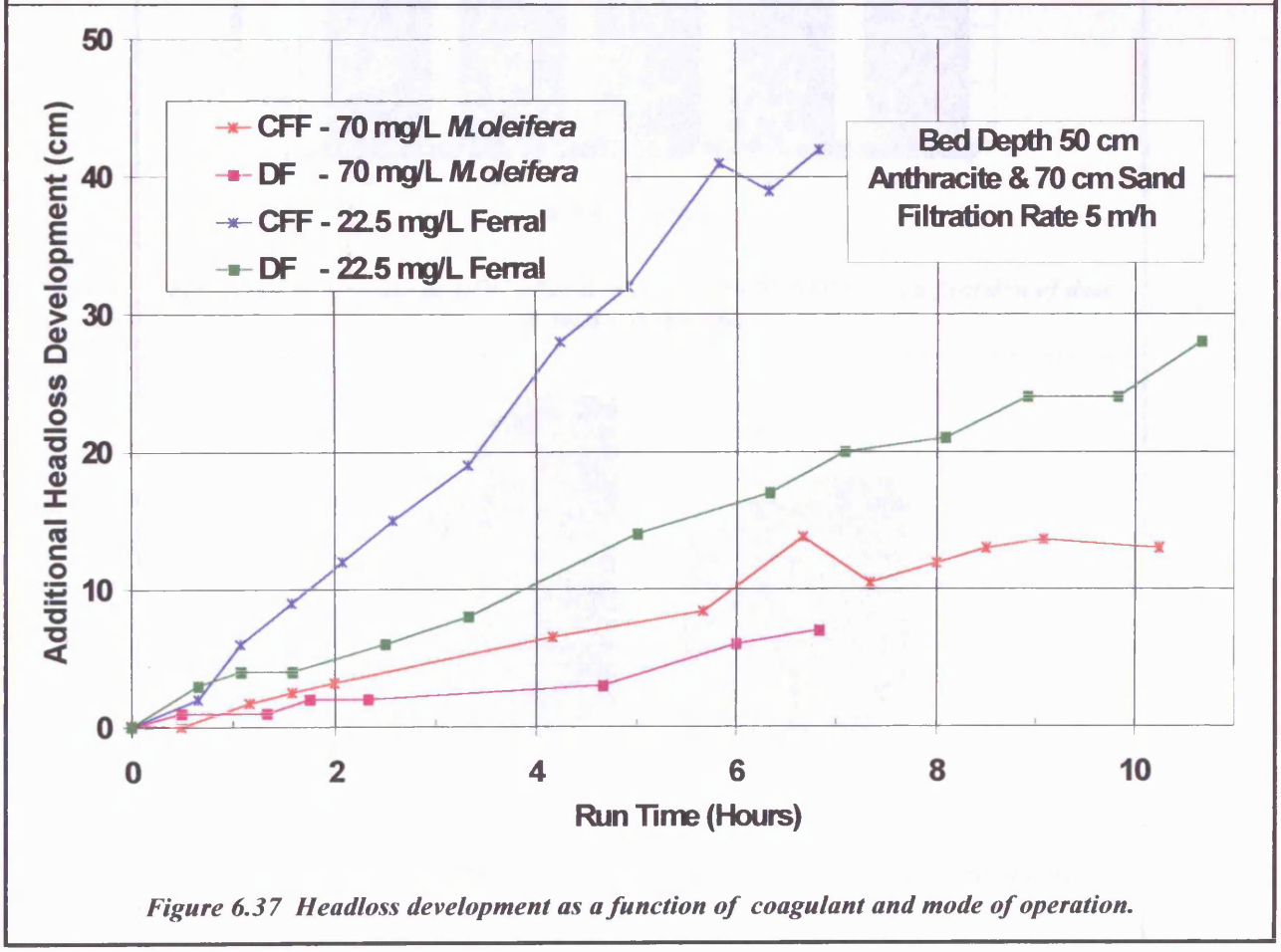

Figure 6.37 Headloss development as a function of coagulant and mode of operation.

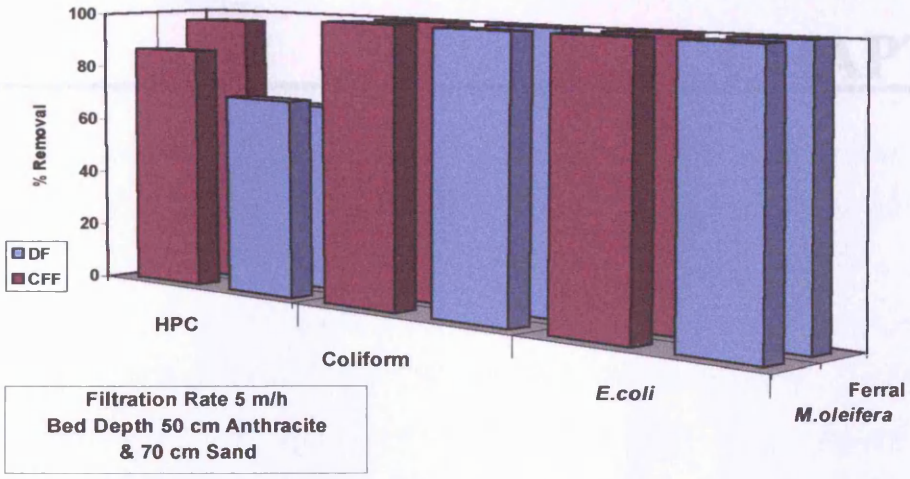

Figure 6.38 Bacterial reduction as a function of coagulant and mode of operation.

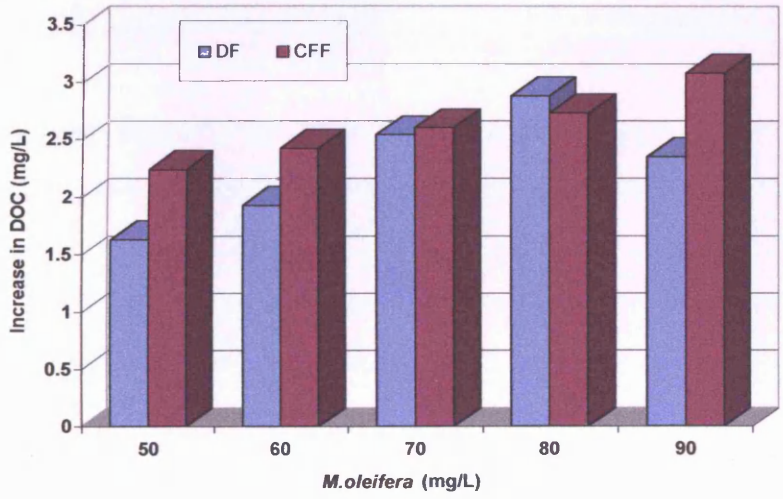

Figure 6.39 Increase in DOC after treatment with M.oleifera as a function of dose & mode of operation.

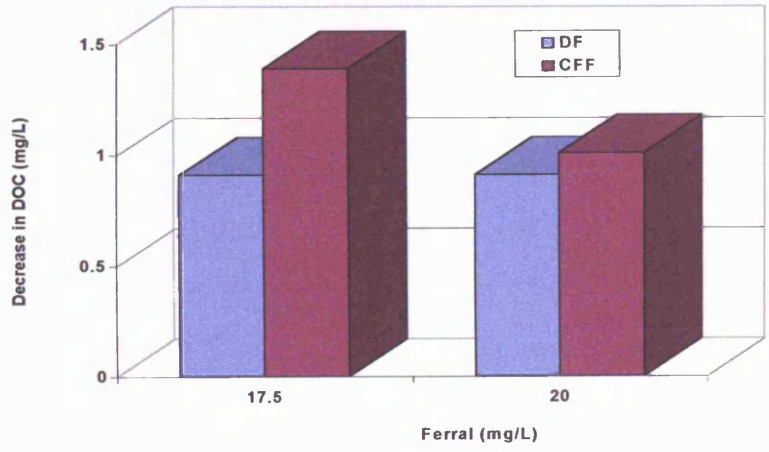

Figure 6.40 Decrease in DOC after treatment with Ferral as a function of dose & mode of operation.

CHAPTER 7

OVERALL CONCLUSIONS

7.0 Overall Conclusions

The broad objective of this research was to investigate the potential for use of CFF in conjunction with the seeds of the tree *M.oleifera* in the treatment of low turbidity waters. The research was conducted in two phases: a laboratory and a field study. The laboratory study consisted of two major investigations. Firstly, the effect of certain hydraulic variables on the treatment process. Secondly, a comparison of *M.oleifera* with two other coagulants, namely alum and chitosan, using a kaolin suspension diluted in deionised water. The field study was conducted at Felindre WTW in South Wales in May-July 1997, using a natural low turbidity water. The study included a comparison between *M.oleifera* and Ferral, and the operation of the plant in the CFF and DF modes. The change in organic content of the raw and final waters was also investigated. The conclusions of these two studies are considered below:

- The increase in removal rates with the smaller compared to the larger media, was considered to be a function of the collector surface area. This was concluded because maintaining the collector surface area in two beds with different media sizes and depths produced filtrate of similar quality. Headloss development rates, however, were consistently higher with the smaller media, due to the reduction in particulate penetration prior to capture and removal.

- Deeper beds, regardless of other hydraulic variables, outperformed shallower beds, when *M.oleifera* seed was used in the laboratory study. This was not the case with alum and chitosan. Three reasons have been postulated for this. Firstly, alum forms a precipitate almost instantaneously on mixing with the raw water, which then enmeshes the particulates very rapidly; chitosan, as is the case with other high molecular weight polymers, attaches at 'one end' to a collector surface, while the 'other end' protrudes out into the flow and is therefore able to capture in-coming particles relatively easily. Whereas for low molecular weight 'patch' forming polymers, such as *M.oleifera*, aggregation and capture only occurs when oppositely charged surfaces come into contact with each other. Thus, in-pore flocculation using *M.oleifera* is a relatively slow process, requiring longer retention times in the bed. Secondly, larger floc such as that formed with alum and chitosan, are more likely to be removed by straining, sedimentation, interception and impaction. Although straining does not lead to optimal use of the bed and the total output is reduced, removal rates are higher due to the autocatalytic nature of particle capture. Thirdly, the weaker attachment bonds formed by

M.oleifera compared to alum and chitosan, leads to greater detachment of floc as the run proceeds. Deeper beds are therefore required to ensure a greater opportunity for reattachment.

◆ Higher filtration rates resulted in a reduction in turbidity removal rates with *M.oleifera* in the laboratory and field studies. In fact filtration rate was found to be the significant hydraulic parameter in terms of turbidity removal. The point at which the shear forces exceed the attachment forces of the particles in the bed, occurs at a lower rate with *M.oleifera* than chitosan; consequently, turbidity breakthrough was more rapid with *M.oleifera*. With chitosan, higher filtration rates reduced surface straining and increased the rate of bed penetration of the particulates. This caused a greater distribution of the removed material and consequently a higher total output before terminal headloss was reached.

◆ A further effect of higher filtration rates was to increase the mixing intensity and reduce the duration of the rapid stage. The effect of this was an increase in turbidity removal rates with alum and chitosan. This was considered to be due the rapid rate of floc formation with these coagulants, therefore requiring flash mixing to ensure their distribution throughout the raw water. Furthermore, excessive periods of rapid mix can cause floc break-up. Thus conditions in the rapid mix stage were closer to optimal for these two coagulants at the higher filtration rates. For *M.oleifera* seed the rapid mix stage was not as critical. The retention time in the bed was of more significance due to the relatively slow process of in-pore flocculation. Accordingly, equal removal was achieved with *M.oleifera* when the retention times in the bed were the same, despite the variation in filtration rates, bed depths and mixing intensities.

◆ There is a trade-off of benefits with higher filtration rates. The main advantage is the reduction in land requirement; but in order to maintain a high quality filtrate with *M.oleifera*, deeper beds are necessary, which in turn require large volumes of backwash water. Furthermore, clean bed headlosses are greater at higher filtration rates, and a greater hydraulic head is required. A solution is to use dual media filter beds, such that deeper beds do not effect such large headlosses. Dual media beds were of particular benefit with chitosan, since longer runs were achieved due to deeper particulate penetration and an increase in the floc holding capacity. As headloss development rates were not generally critical when using *M.oleifera*, but the collector surface area was (for efficient turbidity removal), it was concluded that deeper layers of sand should be used at the expense of the anthracite layer in the dual media beds. The

opposite was found with alum and chitosan where it was considered that deeper layers of anthracite at the expense of the sand layer would be more advantageous.

• The laboratory results were validated by the field study, although only low turbidity waters (2-5 NTU) were examined in the field study. It was found that removal rates were very similar in the two studies, indicating that the kaolin suspension effectively simulated natural raw waters. More importantly, low turbidity waters are more difficult to treat using low molecular weight polymers, therefore successful results with a natural low turbidity water offers great promise for higher turbidity waters (5-75 NTU). There was a significant variation in the optimum dosages for the two studies (optimal doses for the laboratory study ranged between 10-50 mg/L for waters of 5-75 NTU, and for the field study the optimal doses ranged between 50-70 mg/L for waters of 2-5 NTU). It was considered that the organic content of the natural raw water exerted a high coagulant demand. The DOC concentration of the raw water used in the field study was not high, and therefore waters with higher DOC concentrations may require considerably higher doses of coagulant in order to significantly reduce the turbidity.

• Treatment using *M.oleifera* seed suspension caused an increase in the DOC of the final water, due to the presence of extraneous seed material. It was considered that some of the original DOC in the raw water was removed, despite the overall increase in the final water. The isothiocyanate compounds in the final water originated from the suspension, since none were found in the raw water. The significance of the presence of these compounds in the final water is that they contain an antimicrobial agent with potential health implications for use in potable water treatment. Furthermore, organic matter in the final water may provide substrate for microbial growth. A more serious concern is the reaction of the organics with chlorine, forming potentially carcinogenic DBP. However, THM concentrations were similar in the waters treated with *M.oleifera* and Ferral, and were also below the PVC for the UK. Furthermore, concern over THMs in drinking water is a quality issue mainly of the developed world. The major concern in many developing countries is firstly the availability of water, and secondly for the water to be free from pathogens. Low concentrations of potentially harmful compounds that may have a long term chronic effect are of limited significance where coagulants and disinfectants are not always available throughout the year. Whilst it is not suggested that lower standards should be set for such countries, it must be recognised that appropriate treatment means the implementation of the best available solutions.

◆ A comparison of DF and CFF treatment processes indicated that turbidity and bacterial removal rates were comparable, although headloss development rates were different. The higher headloss development rates with CFF were a consequence of floc morphology. For the same mass of particulate matter, larger floc have a lower surface area to volume ratio than smaller floc or particles. Higher surface area causes higher fluid drag and therefore greater headloss development rates. A further effect of pre-formed floc entering the filter bed, is that surface removal can dominate and also lead to higher headloss development rates. The extent to which each of these phenomena operates determines whether DF or CFF causes higher headloss development rates. With *M.oleifera*, the headloss was lower with DF because floc formation is relatively slow, thus the floc entering the bed are relatively small and unlikely to clog the surface layers. Therefore, the effect of the increase in particulate surface area was more important. But since headloss development rates are not generally a problem with low to medium turbidity waters when using *M.oleifera*, CFF would be more advantageous than DF due to the reduction in treatment stages and the consequent reduction in costs.

◆ Bacterial removal rates (both total coliforms and E.coli), using *M.oleifera* and Ferral in DF and CFF, were very high. At the optimum dose and media configurations 100% removal rates were achieved. Although a disinfection stage after filtration is always recommended, the very high removal rates show the great potential for using *M.oleifera* with CFF.

◆ An advantage of using *M.oleifera* is that floc formation will not occur in the rapid mix stage, since the process is relatively slow. Thus, surface removal is reduced and deeper penetration can occur, even at lower rates. A further advantage of *M.oleifera* over other coagulants is the broad dose range which can be used, such that exceeding the optimal dose still effects removal. This indicates a robust treatment process appropriate for developing countries. Additionally, there is a reduction in costs since treatment performance is not affected by pH, and sludge production is reduced due to the fundamental nature of the floc. Potential disadvantages of using *M.oleifera* are that deeper beds are required if the benefits of larger media and higher filtration rates to be utilised. Furthermore, the lengths of the filter runs are reduced considerably with higher turbidity raw waters (35-75 NTU) using CFF. Monitoring raw water turbidity may be necessary since the rate of clogging will increase and the interval between filter washings will decrease as turbidity increases. Additionally, staff would be required at the treatment works throughout the day in order to prepare fresh seed suspensions every 8-12 hours depending on climatic conditions.

◆ Thus the combination of CFF and *M.oleifera* operates more effectively at lower filtration rates (5 m/h) and/or deeper beds preferably of dual media. Though there is a requirement for more beds, the cost of construction and operation of sedimentation and flocculation tanks is eliminated. Furthermore there is a reduction in the required coagulant dose. Potentially higher filtration rates may be used with lower turbidity waters (5-20 NTU), than is the case with conventional treatment. Filtration rates can then be reduced if the raw water turbidity increases.

◆ The choice of coagulant is not only dependent on effectiveness in terms of turbidity removal; issues such as cost, availability, degree of expertise required in its preparation and the dosing regime are important, especially in developing countries. For example, Thyolo WTW in southern Malawi, referred to previously, uses imported alum from South Africa. The supply, however, is not guaranteed throughout the year, partly because many roads become impassable during the rainy season. The use of a local material in such a case would not only reduce the problem of transport but also lead to the employment of local labour for cultivation, harvesting and processing of the coagulant.

◆ The use of *M.oleifera* seed would only be appropriate in tropical and subtropical regions where it can be grown, unless the active proteins can be extracted and reliably stored. Chitosan, a by-product of the shellfish industry, is in plentiful supply near the processing plants. Chitosan preparation requires the use of acid and high intensity mixing, and the optimal dose range is relatively small. It may also be necessary to adjust the pH of the raw water in order to obtain maximum turbidity removal, while maintaining a low headloss development (since chitosan was found to form a precipitate as well as forming floc by the bridging mechanism). Preparation of *M.oleifera* seed suspension is relatively labour intensive, although the level of skill required is not high.

◆ This research is unique and has added significantly to the available knowledge on the use of natural coagulants in conjunction with CFF in the treatment of low turbidity waters. *M.oleifera* seed has not previously been used in the treatment of such waters at pilot scale, nor has the significance of the hydraulic variables been assessed. The positive results, in terms of turbidity and bacterial removal, in the natural raw waters were very encouraging, as was the limited production of THMs in the final water after disinfection. The increase in DOC in the final water is a matter that needs to be considered further. The comparison of effectiveness of

M.oleifera with alum, Ferral and chitosan was also unique, and indicates that its effectiveness is comparable with other established coagulants. The combination of CFF and *M.oleifera* seed in the treatment of low turbidity waters has many advantages to developing countries: it is robust, low cost in terms of chemical requirement and construction, and very effective.

CHAPTER 8

FUTURE WORK

8.0 Future Work

There are some interesting areas for further study/investigation and these are:

- A series of full scale trials using CFF in conjunction with *M.oleifera* seed in the treatment of low turbidity waters in the range of 5-75 NTU in developing countries.

- Investigation into the use of the presscake and the isolated active proteins from the seeds in terms of effectiveness of turbidity, bacteria and DOC removal. The latter would be of particular importance since using whole seed suspensions resulted in an increase in the organic matter of the treated water, which may in turn cause an increase in bacterial numbers and THM formation. The viability of suspensions made-up using the presscake and the active proteins would need to be compared to whole seed suspensions, as the latter became inactive within 8-12 hours (probably due the degradation of the extraneous organic matter from the seeds). Therefore using suspensions where the organic content is reduced or eliminated would potentially extend the viability of the suspension.

- A comparison of the costs involved in the production and use of the three aforementioned suspensions is necessary. Production costs for the presscake and active proteins suspensions would be higher but because smaller volumes of material are required to effect the same treatment, then storage and transport costs may be lower. Additionally, the sludge volume is lower and therefore treatment costs are further reduced. There may be some disadvantages however; the presscake and the active protein suspensions have smaller dose ranges and therefore more accurate dosing equipment may be required. Furthermore, the use of the active proteins may require pH adjustment to optimise floc formation. The costs incurred should be considered.

- Investigation into the effectiveness of colour, iron and manganese removal with *M.oleifera* seed. These parameters are considered important because their concentrations can be relatively high in low turbidity waters, and can be difficult to remove with polyelectrolytes. 'Sweep' floc coagulation induced by metal coagulants is more efficient in capturing the colour compounds than are polymers. Iron and manganese removal usually involves oxidation to form a

precipitate, and a high coagulant dose is often required to ensure coagulation and removal in the clarification stage.

- The results of the bacterial removal rates were very promising, but further research should be conducted into the removal rates of other microorganisms especially pathogenic protozoans such as *Cryptosporidia*. Such pathogens are of significance to health, as they are not deactivated by chlorine.

- Filter bed backwashing procedures should be optimised after the use of *M.oleifera*, in terms of flow rate and volume of water used. This is particularly important with CFF and DF, since relatively large volumes of water are used compared to conventional treatment. Furthermore higher backwash rates are required with polymers than with metal salts due to stronger attachment forces between the particulate and the media surfaces. The use of an air scour may be necessary. Only general principles for optimisation of backwashing can be established in any one study. Specific details would need to be ascertained for each water, since it may be advantageous to leave a certain amount of particulate matter in the bed to aid the ripening process.

- Investigation of the use of *M.oleifera* as a coagulant aid in conjunction with metal coagulants and with weighting agents such as bentonite. These combinations are considered to be effective in the treatment of low turbidity waters.

- Determination of the sensitivity of the treatment process to transient shock loading of flow and turbidity on the filter bed. This is because CFF lacks the buffering capability of a sedimentation stage.

- Investigation of the effectiveness of chitosan in treatment of high and low turbidity natural raw waters, in terms of turbidity, bacteria and DOC removal.

- Investigation of the use of chitosan as a coagulant aid in conjunction with alum, in terms of enhanced removal at high filtration rates and a reduction of the required alum dose.

- Investigation of the effect of varying the pH of the raw water on treatment optimisation using chitosan. Altering the mechanism of particle destabilisation and floc formation may improve floc characteristics and allow deeper penetration of the particles and optimal use of the bed. In addition the volume of backwash water may be reduced.

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APPENDIX 1.

Kaolin Stock Solution

Reagents used:

100 g of kaolin light - Sigma (hydrated aluminium silicate). Particle size 0.1-4 μm
1200 ml of deionised water
8.4 ml of sodium bicarbonate stock solution (65 g of sodium bicarbonate per litre deionised water)

Method:

8.4 ml of sodium bicarbonate solution was added to 1200 ml of water, and then a litre portion was taken from the resulting solution. From this 1 litre a small amount was added to the 100 g of kaolin to make paste. This was mixed until a smooth consistency was reached then the remainder of the 1 litre was added mixing continually. The solution was then placed under a mechanical stirrer (AZTEC Sedimentation Jar Tester) and stirred for 2 hours @ 400 rpm. It was then allowed to settle for 30 minutes and the top 800 ml was decanted off. The remaining sludge at the base was discarded and the 200 ml of deionised water (containing the sodium bicarbonate solution) was added. This was then stirred for a further 1 hour @ 400 rpm and then allowed to settle for 15 minutes before decanting the top 800 ml.

The resulting 800 ml of kaolin solution was then tested for stability by the following method. 7 ml of the bicarbonate solution was added to 1 litre of deionised water and placed under the mechanical stirrer and stirred @ 300 rpm. 2-3 ml of the kaolin solution was then added and the mixing velocity reduced to 30 rpm. Formation of floc was checked for in the beaker, especially around the meniscus and just below the surface of the water around the paddle. The solution was then allowed to stand for 15 minute and any settling observed. If no floc formation or settling was observed, then the solution is considered to be stable and suitable for use in making up the model water. There is no shelf life for the kaolin solution, though vigorous stirring must be undertaken to ensure full resuspension.

APPENDIX 2.

Calculation of G and t values in the FWZ in the laboratory pilot rig

G is the velocity gradient	s^{-1}
t is the retention time	s
Q is the volumetric flow rate	2.18×10^{-5} m^3/s
ρ is the density of water	1000 kg/m^3
g is the gravitational constant	9.81 m/s^2
h is the headloss	(m)
μ is the dynamic viscosity of water	$1x\ 10^{-3}$ kg/m s
L is the length of the flocculation tank (the FWZ in this case)	1m
A is the cross sectional area of the flocculation tank	7.85×10^{-3} m^2
D is the diameter of the flocculation tank	0.1 m
f is the coefficient of friction	Dimensionless
Re is the Reynolds number	Dimensionless
v is the velocity of flow through the tank	10 m/h (2.78×10^{-3} m/s)

$$G = \left(\frac{Q\rho gh}{\mu LA}\right)^{1/2} \qquad \textit{Eq. 1}$$

$$h = f\frac{Lv^2}{gD} \qquad \textit{Eq. 2}$$

$$f = \frac{16}{\mathrm{Re}} \qquad \textit{Eq. 3}$$

$$\mathrm{Re} = \frac{\rho Dv}{\mu} \qquad \textit{Eq. 4}$$

$$t = \left(\frac{AL}{Q}\right) \qquad \textit{Eq. 5}$$

Using the above values:

t = 360 s	(from equation 5)
Re = 278	(from equation 4)
f = 0.23	(from equation 3)
h = 4.4×10^{-7} m	(from equation 2)
G = 0.16 s^{-1}	(from equation 1)

Gt = 39.6

References : Camp (1953); Camp & Stein (1943); Munday & Farrar (1983)

APPENDIX 3.

List of all the filter runs undertaken in the field study

Run No.	Coagulant	Dose (mg/L)	Filter Media	Floc-culation Tank	Flow Rate (m/h)	Column	Minimum Turbidity (NTU)	Date
M1	*M.oleifera*	30	S-70 cm & A-50 cm	N	5	1	0.72	14/5/97
M2	*M.oleifera*	40	S-70 cm	N	5	2	0.27	
M3	*M.oleifera*	30	S-70 cm & A-50 cm	N	5	3	0.64	
M4	*M.oleifera*	40	S-70 cm	N	5	4	0.21	
M5	No coagulant	0	S-70 cm & A-50 cm	N	5	1	3.2	19/5/97
M6	*M.oleifera*	50	S-70 cm	N	5	2	0.19	
M7	*M.oleifera*	50	S-70 cm & A-50 cm	N	5	3	0.18	
M8	No coagulant	0	S-70 cm	N	5	4	2.9	
M9	*M.oleifera*	40	S-70 cm & A-50 cm	N	5	1	0.24	20/5/97
M10	*M.oleifera*	30	S-70 cm	N	5	2	0.43	
M11	*M.oleifera*	40	S-70 cm & A-50 cm	N	5	3	0.29	
M12	*M.oleifera*	30	S-70 cm	N	5	4	0.28	
M13	*M.oleifera*	30	S-70 cm & A-50 cm	N	5	1	0.66	21/5/97
M14	*M.oleifera*	40	S-70 cm	N	5	2	0.30	
M15	*M.oleifera*	40	S-70 cm & A-50 cm	N	5	3	0.32	
M16	*M.oleifera*	30	S-70 cm	N	5	4	0.39	
M17	*M.oleifera*	60	S-70 cm & A-50 cm	N	5	1	0.17	22/5/97
M18	*M.oleifera*	50	S-70 cm	N	5	2	0.21	
M19	*M.oleifera*	50	S-70 cm & A-50 cm	N	5	3	0.21	
M20	*M.oleifera*	60	S-70 cm	N	5	4	0.16	
M22	*M.oleifera*	60	S-70 cm & A-50 cm	N	5	1	0.17	28/5/97
M23	*M.oleifera*	60	S-70 cm	N	5	2	0.19	
M24	*M.oleifera*	70	S-70 cm & A-50 cm	N	5	3	0.13	
M25	*M.oleifera*	70	S-70 cm	N	5	4	0.14	
M26	*M.oleifera*	40	S-70 cm	N	10	2	0.28	29/5/97
M27	*M.oleifera*	50	S-70 cm & A-50 cm	N	10	3	0.19	
M28	*M.oleifera*	40	S-70 cm & A-50 cm	N	10	1	0.15	2/6/97
M29	*M.oleifera*	40	S-70 cm	N	10	2	0.20	
M30	*M.oleifera*		S-70 cm	N	10	4	0.19	
M31	*M.oleifera*	40	S-70 cm & A-50 cm	N	10	1	0.15	3/6/97
M32	*M.oleifera*	40	S-70 cm	N	10	2	0.23	
M33	*M.oleifera*	50	S-70 cm & A-50 cm	N	10	3	0.16	
M34	*M.oleifera*	50	S-70 cm	N	10	4	0.12	
M35	*M.oleifera*	60	S-70 cm & A-50 cm	N	10	1	0.21	4/6/97
M36	*M.oleifera*	60	S-70 cm	N	10	2	0.24	
M37	*M.oleifera*	30	S-70 cm & A-50 cm	N	10	3	0.21	
M38	*M.oleifera*	30	S-70 cm	N	10	4	0.25	
M39	*M.oleifera*	30	S-70 cm & A-50 cm	N	10	1	0.21	6/6/97
M40	*M.oleifera*	30	S-70 cm	N	10	2	0.37	
M41	No coagulant	0	S-70 cm & A-50 cm	N	10	3	1.11	
M42	No coagulant	0	S-120 cm	N	10	4	1.05	
M43	*M.oleifera*	60	S-120 cm	N	5	1	0.14	17/6/97
M45	*M.oleifera*	30	S-120 cm	N	5	2	0.49	
M46	*M.oleifera*	40	S-120 cm	N	5	3	0.21	
M47	*M.oleifera*	50	S-120 cm	N	5	4	0.23	
M48	*M.oleifera*	90	S-120 cm	N	5	1	0.11	18/6/97
M49	*M.oleifera*	60	S-120 cm	N	5	2	0.18	

M50	*M.oleifera*	70	S-120 cm	N	5	3	0.16	
M51	*M.oleifera*	80	S-120 cm	N	5	4	0.16	
M52	*M.oleifera*	60	S-70 cm	N	5	1	0.37	25/6/97
M53	*M.oleifera*	90	S-70 cm	N	5	2	0.33	
M54	*M.oleifera*	80	S-70 cm	N	5	3	0.27	
M56	*M.oleifera*	70	S-70 cm	N	5	4	0.26	
M57	*M.oleifera*	60	S-70 cm & A-50 cm	N	5	1	0.33	27 /6/97
M58	*M.oleifera*	90	S-70 cm & A-50 cm	N	5	2	0.25	
M59	*M.oleifera*	80	S-70 cm & A-50 cm	N	5	3	0.19	
M60	*M.oleifera*	70	S-70 cm & A-50 cm	N	5	4	0.23	
M61	*M.oleifera*	60	S-70 cm & A-50 cm	Y	5	1	0.26	2/7/97
M62	*M.oleifera*	60	S-70 cm & A-50 cm	Y	5	2	0.27	
M63	*M.oleifera*	40	S-70 cm & A-50 cm	Y	5	3	0.52	
M64	*M.oleifera*	40	S-70 cm & A-50 cm	Y	5	4	0.48	
M65	*M.oleifera*	50	S-70 cm & A-50 cm	Y	5	1	0.78	3/7/97
M66	*M.oleifera*	50	S-70 cm & A-50 cm	Y	5	2	0.79	
M67	*M.oleifera*	30	S-70 cm & A-50 cm	Y	5	3	0.53	
M68	*M.oleifera*	30	S-70 cm & A-50 cm	Y	5	4	0.55	
M69	No coagulant	0	S-70 cm & A-50 cm	Y	5	1	1.13	4/7/97
M70	No coagulant	0	S-70 cm & A-50 cm	Y	5	2	1.24	
M71	*M.oleifera*	70	S-70 cm & A-50 cm	Y	5	3	0.16	
M72	*M.oleifera*	70	S-70 cm & A-50 cm	Y	5	4	0.23	
M73	*M.oleifera*	70	S-70 cm & A-50 cm	Y	5	1	0.13	12/7/97
M74	*M.oleifera*	70	S-70 cm & A-50 cm	Y	5	2	0.13	
M75	*M.oleifera*	80	S-70 cm & A-50 cm	Y	5	3	0.14	
M76	*M.oleifera*	80	S-70 cm &A-50 cm	Y	5	4	0.13	
M77	*M.oleifera*	50	S-70 cm & A-50 cm	Y	5	1	0.15	14/7/97
M78	*M.oleifera*	50	S-70 cm & A-50 cm	Y	5	2	0.18	
M79	*M.oleifera*	60	S-70 cm & A-50 cm	Y	5	3	0.12	
M80	*M.oleifera*	60	S-70 cm & A-50 cm	Y	5	4	0.12	
M81	*M.oleifera*	40	S-70 cm & A-50 cm	Y	5	1	0.19	17/7/97
M82	*M.oleifera*	40	S-70 cm & A-50 cm	Y	5	2	0.17	
M83	*M.oleifera*	90	S-70 cm & A-50 cm	Y	5	3	0.18	
M84	*M.oleifera*	90	S-70 cm & A-50 cm	Y	5	4	0.17	
M85	*M.oleifera*	60	S-70 cm & A-50 cm	N	5	3	0.15	21/7/97
M86	*M.oleifera*	90	S-70 cm & A-50 cm	N	5	4	0.15	
M87	*M.oleifera*	60	S-70 cm	N	5	3	0.21	23/7/97
M88	*M.oleifera*	90	S-70 cm	N	5	4	0.17	

F1	Ferral	12.5	S-120 cm	N	5	1	0.49	19/6/97
F2	Ferral	22.5	S-120 cm	N	5	2	0.20	
F3	Ferral	20	S-120 cm	N	5	3	0.21	
F4	Ferral	17.5	S-120 cm	N	5	4	0.22	
F5	Ferral	22.5	S-120 cm	N	5	1	0.19	20/6/97
F6	Ferral	12.5	S-120 cm	N	5	2	0.53	
F7	Ferral	17.5	S-120 cm	N	5	3	0.28	
F8	Ferral	20	S-120 cm	N	5	4	0.19	
F9	Ferral	22.5	S-70 cm	N	5	1	0.23	24/6/97
F10	Ferral	12.5	S-70 cm	N	5	2	2.10	
F11	Ferral	17.5	S-70 cm	N	5	3	0.52	
F12	Ferral	20	S-70 cm	N	5	4	0.48	
F13	Ferral	14	S-70 cm & A-50 cm	N	5	1	2.15	28/6/97
F14	Ferral	28	S-70 cm & A-50 cm	N	5	2	0.17	
F15	Ferral	18	S-70 cm & A-50 cm	N	5	3	0.27	
F16	Ferral	23	S-70 cm & A-50 cm	N	5	4	0.23	
F17	Ferral	15	S-70 cm & A-50 cm	Y	5	1	0.71	9/7/97
F18	Ferral	15	S-70 cm & A-50 cm	Y	5	2	0.72	

F19	Ferral	20	S-70 cm & A-50 cm	Y	5	3	0.13	
F20	Ferral	20	S-70 cm & A-50 cm	Y	5	4	0.17	
F21	Ferral	17.5	S-70 cm & A-50 cm	Y	5	1	0.12	10/7/97
F22	Ferral	17.5	S-70 cm & A-50 cm	Y	5	2	0.10	
F23	Ferral	22.5	S-70 cm & A-50 cm	Y	5	3	0.11	
F24	Ferral	22.5	S-70 cm & A-50 cm	Y	5	4	0.15	
F25	Ferral	17.5	S-70 cm & A-50 cm	Y	5	1	0.13	11/7/97
F26	Ferral	17.5	S-70 cm & A-50 cm	Y	5	2	0.14	
F27	Ferral	22.5	S-70 cm & A-50 cm	Y	5	3	0.11	
F28	Ferral	22.5	S-70 cm & A-50 cm	Y	5	4	0.13	
F29	Ferral	15	S-70 cm & A-50 cm	Y	5	1	0.16	17/7/97
F30	Ferral	15	S-70 cm & A-50 cm	Y	5	2	0.17	
F31	Ferral	20	S-70 cm & A-50 cm	Y	5	3	0.17	
F32	Ferral	20	S-70 cm & A-50 cm	Y	5	4	0.15	
F33	Ferral	17.5	S-70 cm & A-50 cm	N	5	1	0.13	21/7/97
F34	Ferral	20	S-70 cm & A-50 cm	N	5	2	0.13	
F35	Ferral	17.5	S-70 cm	N	5	1	0.14	23/7/97
F36	Ferral	20	S-70 cm	N	5	2	0.13	

M1-M88: Runs conducted using *M.oleifera* seed.

F1-F36: Runs conducted using Ferral.

(1) S = Sand size range 0.60-1.18 mm, classified as BS Mesh Size 14/25, and has a UC of 1.3-1.7.

(2) A = anthracite size range 1.2-2.5 mm, classified as BS grade 2 and UC of < 1.5,a hardness of between 2.75-3.25 on the Moh Hardgrove Index, and a specific gravity of between 1.35-1.45.

APPENDIX 4.

Methods of Analyses Used in the Field Study

1. Bacterial analysis

(i) Definitions of microorganisms

The principle of the Membrane Filtration Method is that yellow colonies present on membranes incubated at 37°C are confirmed as coliforms if both acid and gas are produced in Lactose Peptone Water and the Oxidase test is negative. If the same occurs at 44°C the colonies are confirmed as *E.coli* (HMSO, 1984).The bacterial indicators used in this study were total coliforms, *E.coli* and HPC. The total coliforms are defined as aerobic and facultative anaerobic, Gram-negative, non-spore forming, rod shaped bacteria that ferment lactose with the formation of gas within 48 hours at 35°C. This includes certain entrobacteria, klebsiella, citrobacter, *E.coli* and faecal coliforms. The latter two can be further distinguished in the laboratory by growing the isolated colonies on a selective media and incubating at temperatures of 43-45°C The HPC is defined as the number of colonies formed after incubation at 37°C for 1 day and at 22°C for 3 days. The tests for coliforms and *E.coli* are the most important routine bacteriological examinations carried out on drinking water. As these provide the most sensitive method for detecting faecal contamination. The 1991 Water Supply Act states that no coliforms, *E.coli*, faecal coliform (also known as thermotolerant coliforms) should be present in water intended for potable purposes (HMSO, 1983; Hyder Environmental, 1997).

(ii) Sampling procedure and preparation

The sampling procedure for all the microbiological tests was as follows. The tap was left running for 2 minutes, then turned off and flamed for about 90 seconds. The tap was then turned on again and allowed to run for about 30 seconds before the sample was taken. A 50 ml sample was then sent to Hyder Environmental Microbiological Laboratory for analyses within 6 hours.

(iii) Enumeration of heterotrophic bacteria

The sample bottle was inverted several times to ensure an even distribution of the organisms. The cap was removed and the bottle mouth flamed. A series of dilution of the sample were then made by taking 1 ml of sample and adding it to 9 ml of diluent. This was then shaken thoroughly and 1 ml taken from this solution and added to another 9 ml of diluent. Generally a 5 ml sample of treated water was used and 1 ml of raw water was used for incubation at 37°C. A 1 ml sample of treated water and 0.1 ml of raw water was incubated at 22°C. The diluent used was Ringer's solution, made up in the laboratory by adding 2 Ringer's tablets to 1000+/- 10 ml of deionised water. The solution was then autoclaved at 121°C for 15 minutes.

The test volume was pipetted into an empty petri dish using a sterile pipette. Approximately 15-29 ml of molten Yeast Extract Agar was then poured into the petri dish, within 20 minutes of dispensing the sample. The Yeast Extract Agar was prepared in the laboratory from a dehydrated medium. 23 g of Yeast Extract Agar powder

was added to 1000 +/- 10 ml of deionised water. The solution was then mixed thoroughly and autoclaved at 105°C for 30 minutes. The agar was distributed into glass bottles, which were loosely capped and re-autoclaved at 122°C for 15 minutes. The sample and the agar were mixed rapidly but gently in two petri dishes using a circular movement. They were then placed flat on the bench and the solution allowed to solidify. The plates were inverted and one incubated at 37°C for 24 +/- 3 hours and the other at 22°C for 72 +/- 3 hours at 22°C. The plates were examined as soon as they are removed from the incubator.

The number of colonies formed in each plate were counted using a counting device. The plate with the original sample was counted first. If this contained more than 300 colonies, then the colonies from a diluted sample containing between 30 and 300 colonies were counted. If all dilution contained more than 300 colonies, this was recorded as greater than 300 at the highest dilution. The number of colonies, or colony forming units (CFU) represent the number of bacteria present in 1 ml of water sample.

(iv) Enumeration of Coliforms and *E.coli* bacteria using the Membrane Filtration Method

Presumptive estimation of coliform and *E.coli*

The membrane filtration apparatus used to isolate the bacteria from the water was sterilised for 30 minutes before use. The water sample or dilution of the sample were filtered through the apparatus containing a membrane with a nominal pore size of 0.45 μm. This was sufficient to retain all the bacteria present in the water. Two membranes were taken and placed on pads in petri dishes, ensuring no air was trapped between the membrane and the medium. The pads were saturated with Membrane Lauryl Sulphate Broth, a medium suitable for the growth of coliforms and *E.coli.*

The Membrane Lauryl Sulphate Broth was prepared by the addition of 76.2 g Membrane Lauryl Sulphate Broth powder to 1 L of water and mixing thoroughly. The pH was adjusted to 7.6 using 12% HCL and 4% NaOH. The broth was then stored in glass screw-capped bottles and autoclaved at 121°C for 15 minutes.

One membrane was incubated for 4 +/- 0.25 hours at 30°C, followed by 14 +/- 0.5 hours at 37°C. This test gave a presumptive count of the total coliforms present. The second membrane was incubated for 4 +/- 0.25 hours at 30°C followed by 14 +/- 0.5 hours at 44°C to give the presumptive count for the number of *E.coli* present. After incubation the membranes were examined and the number of yellow colonies were counted. This gave the presumptive number of organisms present in the particular volume of water which was filtered.

Confirmation of the results for coliform and *E.coli*

For confirmation purposes the colonies were further subcultured into specific media and again incubated at 37°C to confirm the total coliform count and at 44°C for the *E.coli* count. The confirmation media used was Lactose Peptone Water for the coliforms and Tryptone Water for *E.coli.* The preparation of Lactose Peptone Water consisted of dissolving 10 g of peptone, 5 g of sodium chloride, 10 g of lactose and 2.5 ml of Phenol red into 1 L of deionised water, then adjusting the pH to 7.5 using 12% HCL and 4% NaOH. The Water was then distributed in 6 ml volumes into tubes. The tubes were capped and autoclaved at 110°C for 15 minutes. The

preparation of Tryptone Water consisted of adding 15 g of Tryptone Water powder to 1 L of deionised water and mixing. The pH was adjusted to 7.5 using 12% HCL and 4% NaOH. The water was then distributed in 6 ml volumes into test tubes. These were capped and autoclaved at 121°C for 15 minutes.

A random selection of the yellow colonies which grew in the presumptive media were transferred to tubes containing the confirmation media, each containing an inverted Durham tube to collect any gas formed. These subcultures were grown into two tubes of Lactose Peptone Water and one of Tryptone Water. One tube of Lactose Peptone Water was placed at 37°C for 24 +/- 4 hours. The second Lactose Peptone and the Tryptone water were incubated at 44°C for 24 +/- 4 hours. After incubation Kovac's reagent was added to the Tryptone water. A further test was to streak the inoculum on plates of Yeast Extract Agar and MacConkey agar plates. These were then incubated for 24 +/- 4 hours at 37°C.

A positive result was demonstrated by the production of acid in the Lactose Peptone Water, i.e. the colour of the media turned from red to yellow and gas was formed in the Durham tube. In the Tryptone Water a confirmed result was demonstrated by the presence of turbidity (growth) and the development of a red colour in the amyl alcohol surface layer after the addition of Kovac's reagent. In the MacConkey agar the presence of coliforms was indicated by the presence of circular, convex and smooth surfaced colonies. The colonies should be red but the depth of colour varies and can not be relied upon.

The Yeast Extract Agar was used to perform the Oxidase test. This consisted of placing an absorbent pad into a petri dish and wetting with Oxidase reagent. Then using a sterile plastic loop to remove a colony from the agar plate and smearing it on to the pad. An oxidase positive result was indicated by immediate purple colouration of the pad. Coliforms are oxidase negative organisms and therefore their presence would show no colouration.

2. Dissolved organic carbon (DOC) analysis

(i) Sampling procedure

The water sample was poured into a 40 ml glass stoppered container, ensuring no air was trapped into the container. This was then kept out of direct light until the analysis took place at the Hyder Environmental Laboratory.

(ii) Analysis procedure

The sample was allowed to settle in order to remove the particulate matter. The supernatant was then decanted off. The inorganic fraction was removed from the supernatant sample, prior to analysis, by acidification and purging with nitrogen. The remaining non-purgeable organic carbon fraction was determined by oxidation with potassium persulphate, while subjecting the sample to intense ultraviolet illumination. This process releases carbon dioxide which was detected by an infrared detector at 440 nm. This technique can be used to detect organic carbon concentrations up to 10 mg/L, with a detection limit of 0.5 mg/L.

3. Organics detectable by gas chromatography

(i) Sampling procedure and preparation

The sampling procedure for analysis by gas chromatography mass spectroscopy (GC/MS) consisted of filling a two-litre darkened glass bottle with the sample. This was then kept refrigerated and kept out of direct light until the analysis was undertaken.

(ii) Analysis procedure

The analysis consisted of extracting the organic phase of the sample with dicloromethane. The extract was then reduced in volume by gentle evaporation. The analysis of the extract was by full scan GC/MS at a range of temperatures from 35-300^{o}C. The peaks produced were identified by searching against a library of Mass Spectra. The relative quantities of each compound was estimated with reference to the internal standard D8 Naphthalene.

4. Trihalomethane (THM) analysis

(i) Sampling procedure and preparation.

The water sample was placed in to a sealed container and dosed with sodium hypochlorite (chlorous), at concentration of 1.2 mg/L. This was equivalent to that used in the main works at Felindre WTW. The sample was then stored for a predetermined length of time. For the purposes of this study samples were stored for 1 and 24 hours. The sample was then placed in to a special sample jar which contained sodium thiosulphate crystals. These crystals have the effect of neutralising the sodium hypochlorite and thereby arresting the formation of chlorination by-products.

(ii) Sample analysis

The sample was extracted into pentane. The extract was then analysed by gas chromatography using an electron capture detector (ECD). This method is well suited to compounds which have a high electron affinity such as THMs. The detector is operated by passing the extract over a radioactive beta particle emitter. An electron from the emitter ionises the carrier gas. About 100 secondary electron are produced for each initial beta particle. After further collision, the energy of these electrons is reduced to the thermal level and they are captured by samples molecules. The electron population in the ECD cell is collected periodically by applying a short voltage pulse to the cell electrodes and the resulting current compared with a reference current. the pulse interval is adjusted automatically to keep the cell current constant, even when the electron are being captured by the sample. the change in the pulse rate when a sample enters the ECD is related to the sample concentration.

The methodologies for the bacterial, organic and chlorination by products analyses were supplied by Hyder Consulting (Environmental) Ltd.

APPENDIX 5.

Raw Data from the Field Study

Table A5.1 Total numbers of bacteria & percentage removal using M.oleifera seed and CFF; filter media - 70 cm of sand. (Runs: M18, 20, 52-55 and 88-89).

Determinand	25 June '97					23 July '97			22 May '97		
	Raw water	90 mg/L	80 mg/L	70 mg/L	60 mg/L	Raw water	60 mg/L	90 mg/L	Raw water	50 mg/L	60 mg/L
HPC (1 day at 37°C/ml)	850	220 75%	160 88%	158 88%	202 76%	630	530 16%	270 43%	650	60 91%	60 91%
Total Coliforms /100 ml	300	142 80%	115 84%	98 86%	118 83%	118	5 96%	0 96%	360	31 90%	5 99%
E.coli /100 ml	300	142 80%	115 84%	98 86%	118 83%	106	0 100%	0 100%	360	28 93%	4 99%

Table A5.2 Total numbers of bacteria & percentage removal using M.oleifera seed and CFF; filter media - 120 cm of sand. (Runs: M48-51).

Determinand	18 June '97				
	Raw Water	90 mg/L	80 mg/L	70 mg/L	60 mg/L
HPC (1 day at 37 °C /ml)	100	9 91%	6 94%	<1 100%	8 92%
Total Coliforms /100 ml	64	0 100%	1 98%	0 100%	1 98%
E.coli /100 ml	29	0 100%	0 100%	0 100%	1 98%

Table A5.3 Total numbers of bacteria & percentage removal using M.oleifera seed and CFF; filter media - 70 cm of sand & 50 cm anthracite. (Run: M17, 19, 57- 60, 86 and 87).

Determinand	27 June '97					21 July '97			22 May '97		
	Raw Water	90 mg/L	80 mg/L	70 mg/L	60 mg/L	Raw Water	60 mg/L	90 mg/L	Raw Water	50 mg/L	60 mg/L
HPC (1 day at 37 °C /ml)	330	120 65%	220 34%	300 10%	350 +6%	1010	930 8%	780 23%	1980	60 97%	60 97%
Total Coliforms /100 ml	250	20 92%	9 94%	26 90%	42 79%	40	0 100%	3 99%	650	14 98%	22 97%
E.coli / 100 ml	250	12 95%	6 98%	26 90%	42 83%	10	0 100%	1 90%	360	11 97%	14 96%

Table A5.4 The DOC (mg/L) of the treated water using M.oleifera seed and CFF.

Dose (mg/L)	70 cm Sand			120 cm Sand	70 cm Sand & 50 cm Anthracite		
	M 52 -	M 18 - 20	M 88	M 48 - 60	M 58 - 60	M 17-19	M 85- 86
90	6.69	-	5.56	5.11	5.72	-	5.23
80	6.53	-	5.11	4.69	5.69	-	-
70	6.30	-	-	4.76	5.56	-	-
60	6.06	5.94	-	4.15	5.38	5.51	4.28
50	5.82	5.17	-	-	-	5.16	-
Raw Water	3.56	2.92	-	2.07	2.96	2.92	2.1
Seed	-	-	880	-	-	-	-

Table A5.5 Total numbers of bacteria & percentage removal using Ferral and CFF; Filter media - 70 cm of sand. CFF mode of operation. (Runs: F 9-12, 33 and 34).

Determinand	24 June '97			23 July '97		
	Raw water	20 mg/L	22.5 mg/L	Raw Water	17.5 mg/L	20 mg/L
HPC (1 day at 37 °C /ml)	490	77 84%	16 97%	630	321 49%	241 62%
Total Coliforms /100 ml	750	122 84%	37 95%	118	18 85%	9 92%
E.coli /100 ml	710	113 84%	37 95%	106	18 85%	4 96%

Table A5.6 Total numbers of bacteria & percentage removal using Ferral and CFF; filter media - 120 cm of sand. (Runs: F1-4).

Determinand	19 June '97		
	Raw Water	20 mg/L	17.5 mg/L
HPC (1 day at 37 °C /ml)	101	1 99%	10 90%
Total Coliforms /100 ml	44	0 100%	1 99.5%
E.coli /100 ml	44	0 100%	0 100%

Table A5.7 Total numbers of bacteria & percentage removal using Ferral and CFF; filter media - sand 70 cm and anthracite 50 cm. (Runs: F13-16, 31 and 32).

Determinand	28 June '97			21 July '97		
	Raw water	28 mg/L	17.5 mg/L	Raw Water	17.5 mg/L	20 mg/L
HPC (1 day at 37 °C /ml)	560	179 68%	216 61%	1010	130 87%	140 86%
Total Coliforms /100 ml	>200	1 >99.5%	1 >99.5%	40	0 100%	0 100%
E.coli /100 ml	>200	0 100%	1 >99.5%	10	0 100%	0 100%

Table A5.8 The DOC (mg/L) of the raw & treated water using Ferral and CFF.

	Sand 70 cm		120 cm Sand	Sand 70 cm & Anthracite 50 cm	
Dose (mg/L)	F 9 & 12	F 36	F 3 & 4	F 14 & 15	F 33 & 34
17.5	-	-	1.27	1.27	1.2
20	2.28	1.35	1.24	-	1.09
22.5	2.14	-	-	-	-
28	-	-	-	1.29	-
Raw Water	3.14	-	2.04	3.15	2.1

Table A5.9 Total numbers of bacteria & percentage removal using M.oleifera seed and DF; filter media - 70 cm of sand and 50 cm anthracite. (Runs: M73-86).

Determinand	12 July '97			17 July '97			14 July '97		
	Raw Water	70 mg/L	80 mg/L	Raw Water	40 mg/L	90 mg/L	Raw Water	50 mg/L	60 mg/L
HPC (1 day at 37 °C /ml)	34	21 39%	<1 <97%	950	550 42%	196 79%	690	580 16%	230 67%
Total Coliform /100 ml	39	0 100%	0 100%	52	1 98%	0 100%	31	7 78%	1 97%
E.coli /100 ml	39	0 100%	0 100%	47	0 100%	0 100%	31	3 90%	1 97%

Table A5.10 Total numbers of bacteria & percentage removal after treatment using Ferral and DF; filter media - 70 cm of sand and 50 cm anthracite. (Runs: F 25-30).

Determinand	11 July '97			17 July '97		
	Raw Water	17.5 mg/L	22.5 mg/L	Raw Water	15 mg/L	20 mg/L
HPC (1 day at 37 °C /ml)	760	450 41%	210 72%	950	440 54%	330 65%
Total Coliforms /100 ml	29	0 100%	0 100%	52	1 98%	2 96%
E.coli /100 ml	23	0 100%	0 100%	47	0 100%	1 98%

Table A5.11 The DOC (mg/L) of the treated water using M.oleifera seed and DF; filter media - 70 cm sand & 50 cm anthracite.

Dose (mg/L)	M 74 & 75	M 77 & 79	M 81 & 84
90	-	-	4.50
80	5.31	-	-
70	4.97	-	-
60	-	4.32	-
50	-	4.02	-
40	-	-	2.19
Raw Water	2.43	2.39	2.15

Table A5.12 The DOC (mg/L) of the treated water using Ferral and DF; filter media - 70 cm sand & 50 cm anthracite.

Dose (mg/L)	F 20 & 21
17.5	1.24
20	1.24
Raw Water	2.15

APPENDIX 6 .

The Plate Section

Plate 1. 12 month old *M.oleifera* trees in southern Malawi.

Plate 2. Immature green pods in the foreground and dry pods in the background. Also visible are the highly nutritious green leaves.

Plate 3. Various *M.oleifera* dishes (top left to bottom right): leaf salad, curried pods, fresh leaves in ground nut sauce, fried dry leaf sauce & friend fresh leaves with to tomatoes.

Plate 4. *M.oleifera* seed (right), oil (back) and presscake (front).

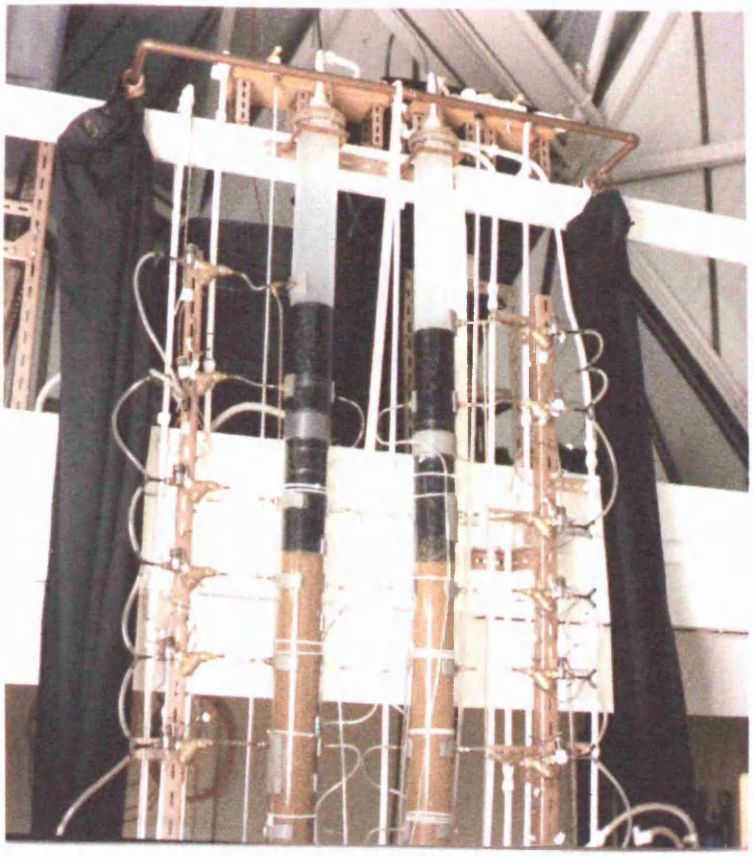

Plate 5. The two columns of the laboratory pilot rig, showing dual media beds of anthracite and sand.

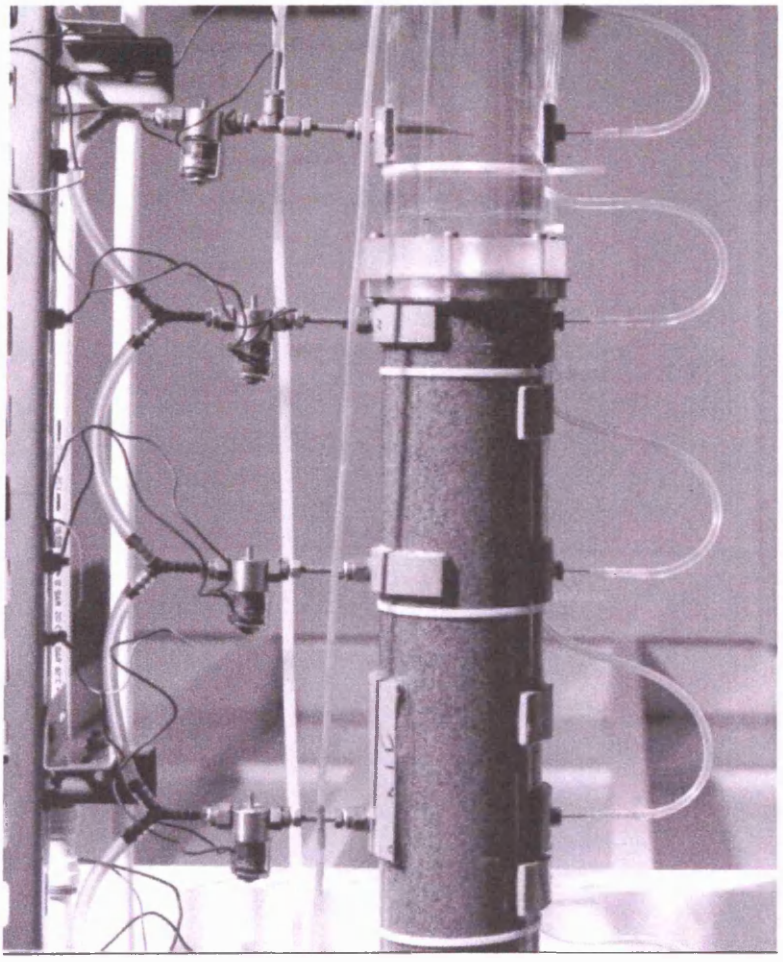

Plate 6. Filter column showing headloss measurement ports on the left side consisting of tubes entering the sand bed and solenoid coil valves. Turbidity measurement ports on the right side consisting of narrow tubes entering the bed attached to plastic tubing for sample draw-off.

Plate 7. The HACH turbidity meter and Gelex Secondary Standards used for calibration.

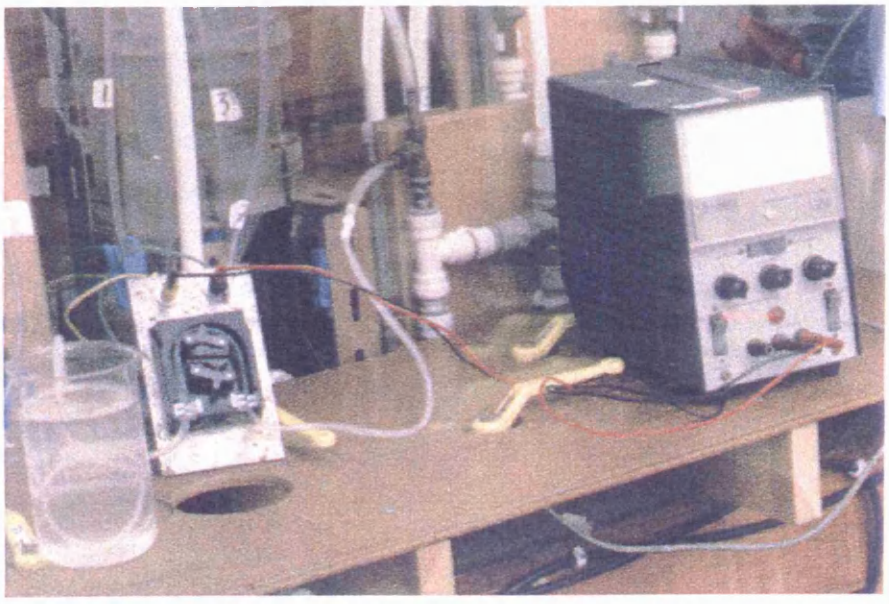

Plate 8. The seed suspension (left), peristaltic dosing pump (middle) and the power supply (right).

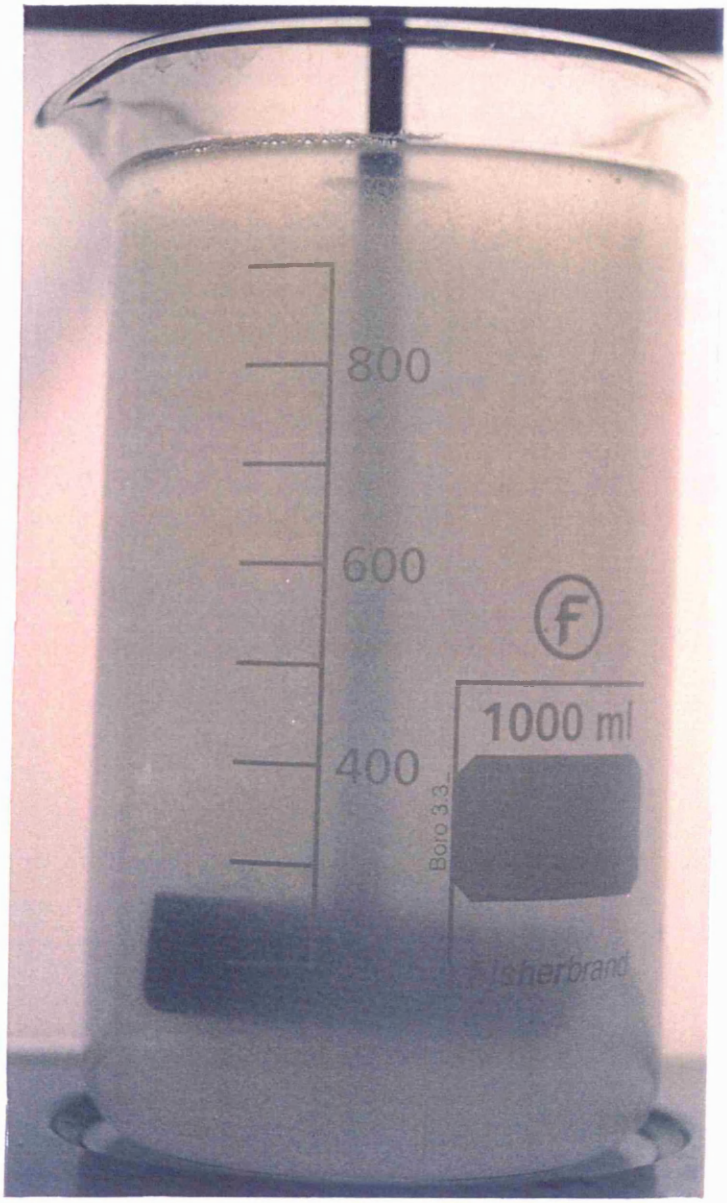

Plate 9. *M.oleifera* floc after 2 minutes mixing at 300 rpm and 15 minutes at 30 rpm.

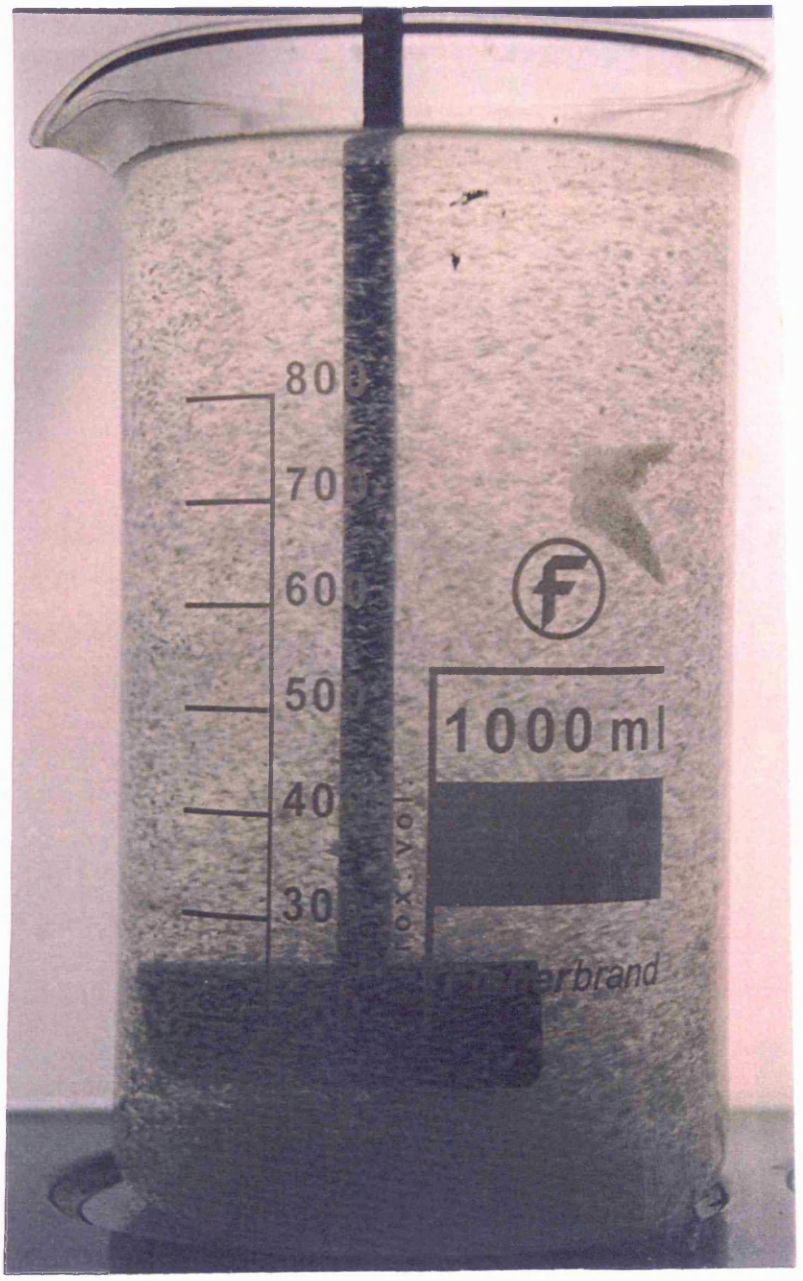

Plate 10. Chitosan floc after 2 minutes mixing at 300 rpm and 15 minutes at 30 rpm.

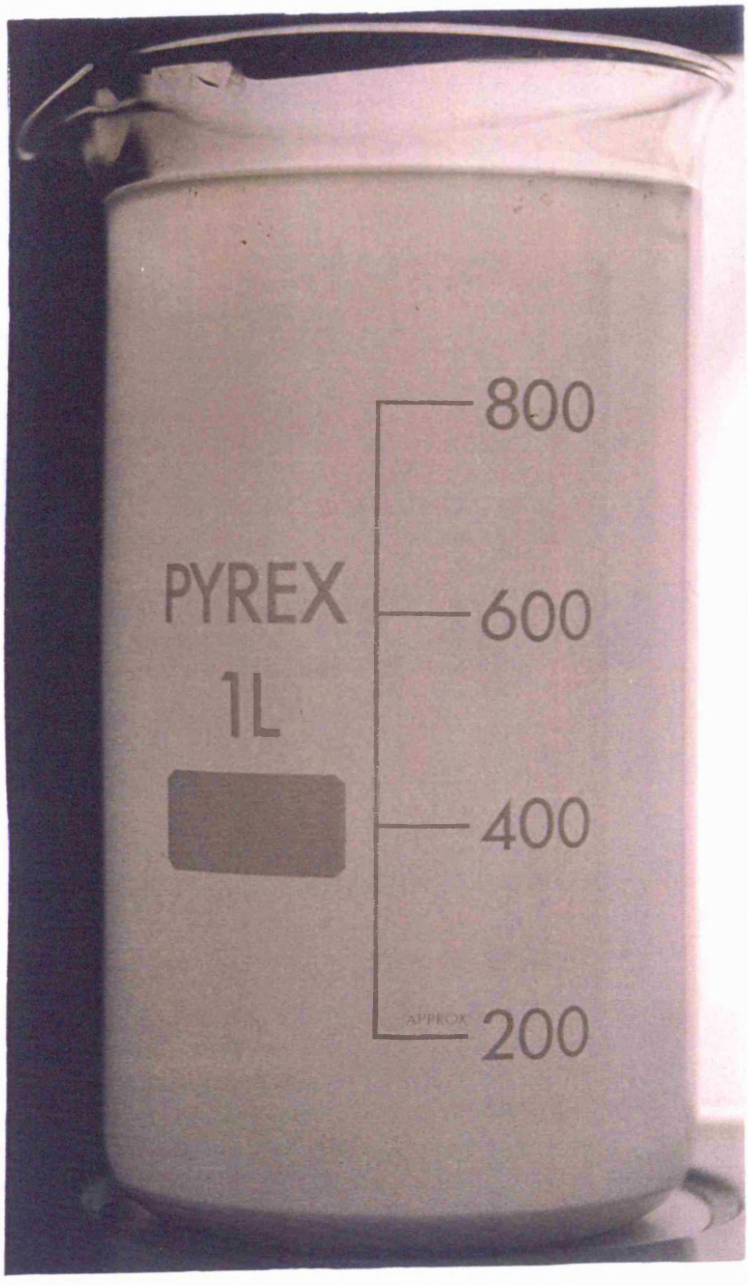

Plate 11. Alum floc after 2 minutes mixing at 300 rpm and 15 minutes at 30 rpm.

Plate 12. Aerial view of the Upper & Lower Lliew Reservoirs. (Source: Dwr Cymru Welsh Water)

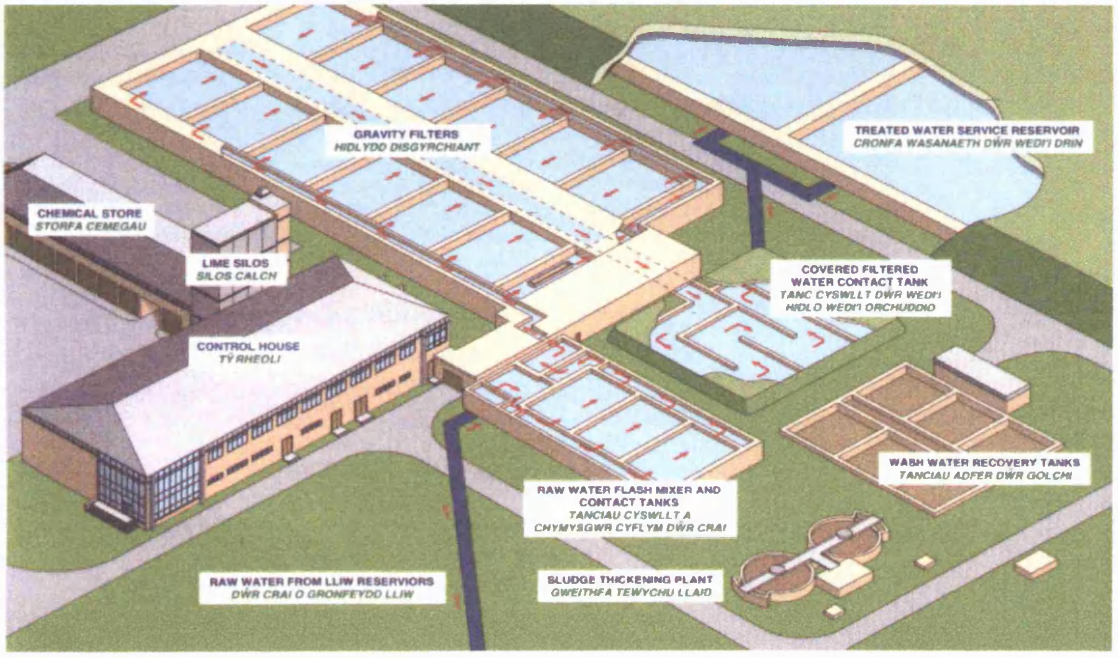

Plate 13. Schematic of the Felindre WTW. (Source: Dwr Cymru Welsh Water)

Plate 14. The 4 columns of the pilot plant at Felindre WTW.

APPENDIX 7.

Publications Emanating from this Research

1. G.K. Folkard, J.P. Sutherland & R.S. Al-Khalili. (1995) "A naturally occurring cationic protein for coagulation of raw waters". *Chemi Oggi*, Vol. 14. No. 11/12. pp 36-40. ISSN 0392-839X (Biocides Today supplement).

2. G.K. Folkard, J.P. Sutherland & R.S. Al-Khalili. (1995) "Natural coagulants - a sustainable approach". In: 'Sustainability of Water and Sanitation Systems' (Ed.) J. Pickford. Intermediate Technology Publication. ISBN 0 906055 46 6.

3. G.K. Folkard, R.S. Al-Khalili & J.P. Sutherland (1996) "Contact flocculation filtration using a natural polyelectrolyte for the treatment of low turbidity surface water in developing countries". In: 'Chemical Water and Wastewater Treatment IV' (Eds.) H.H. Hahn, E. Hoffmann & H. Ødegaard. Springer - Verlag Berlin Heidelberg New York. ISBN 3-540-61624-1.

4. R.S. Al-Khalili, G.K. Folkard & J.P. Sutherland (1998) "Filtration with a natural coagulant". In: 'Water and Sanitation for All: Partnerships and Innovations' (Ed.) J. Pickford. Intermediate Technology Publication, London. ISBN 185339 444 0.

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A naturally occurring cationic protein for coagulation of raw waters

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ABSTRACT

Suspensions of crushed seed of the M. oleifera tree in water yield low molecular weight, highly basic, water soluble, cationic proteins. Their use as primary coagulants within pilot and full-scale water treatment works in Malawi is reviewed. The water to be treated is derived from a river that exhibits high suspended solids, particularly in the rainy season.

Laboratory studies are reported on a novel combination of M. oleifera seed coagulant within a contact flocculation filter (CFF). CFF is seen as a relatively simple, single stage treatment for low turbidity waters, prevalent in the dry season. Such waters are inherently difficult to treat using the conventional treatment sequence as described in the pilot and full-scale studies.

INTRODUCTION

In developing countries, river water drawn for human consumption and general household use can be highly turbid particularly in the rainy season. River silt is churned into suspension and run off from fields and other surfaces carries solid material, bacteria and other micro-organisms into the river. It is of paramount importance to remove as much of this suspended matter as possible prior to a disinfection stage and subsequent consumption. This can generally only be achieved by the addition of coagulants to the raw water within a controlled treatment sequence. In many developing countries proprietary chemical coagulants, such as aluminium sulphate (alum) and synthetic polyelectrolytes are either not available locally or are imported drawing on scant resources of foreign exchange.

A viable alternative is the use of crushed seed of the pantropical tree, *Moringa oleifera* Lam. (*M. oleifera*), as a natural coagulant. The seed pods are allowed to dry naturally on the tree prior to harvesting. The seeds may be easily shelled, crushed and sieved using traditional techniques employed for the production of maize flour. Suspensions of crushed seed in water yield low molecular weight (approximately 6000 daltons), highly basic iso-electric point pH 11) water soluble proteins that act in a similar manner to that of synthetic cationic polyelectrolytes [1]. The low molecular weight of the active proteins indicate that charge neutralisation and floc formation are brought about by the patch mechanism as opposed to the bridging mechanism. Although the mechanism of action is, as yet not fully understood, it is considered that the formation of compact, dense floc that are observed even at high turbidities confirms this hypothesis of action of the active constituents.

The *M. oleifera* tree is a native of Northern India which now grows widely throughout the tropics. It may be propagated from seeds or cuttings, even in poor soils, requiring minimal horticultural attention and is able to survive long periods of drought. It grows rapidly - growth of up to 4 metres in height, flowering and fruiting were all observed within one year during trials in Southern Malawi. The tree is used for many purposes and gives many products [3].

Mature seed contains 40% by weight of oil. The fatty acid profile shows oleic acid at 73% confirming that the oil is similar to olive oil and thus of high market value. The cation protein profile of the presscake (solids residue remaining after oil extraction) is homologous to that of the whole seed. Thus in the future, presscake coagulant may be derived as a by-product of commercially viable oil extraction. For the developing world both commodities are in demand.

M. OLEIFERA COAGULANT WITHIN A CONVENTIONAL TREATMENT TRAIN

M. oleifera seed solution has been shown to be an effective primary coagulant for the treatment of river waters exhibiting relatively high levels of suspended solids. Such waters may be characterised by turbidity values in excess of 100 NTU (Nephelometric Turbidity Units). Studies conducted at pilot and full-scale in Malawi using a conventional treatment sequence comprising coagulation flocculation, sedimentation and rapid sand filtration are summarised.

River water treatment at pilot scale

It is now regarded as axiomatic that both water and wastewater technology for developing countries must be no more complex than strictly necessary, be robust and cheap to install and maintain. A protective treatment works was designed founded on this philosophy. The pilot plant was constructed adjacent to an existing rural water treatment works and is shown schematically in Figure 1. River water is pumped at 24 m³/d to a header/mixer tank where the *M. oleifera* seed solution is dosed. An 18 minute flocculation period is provided within the gravel packed columns prior to sedimentation. A rapid gravity filter removes residual floc carried over in the outlet flow from the sedimentation stage. The

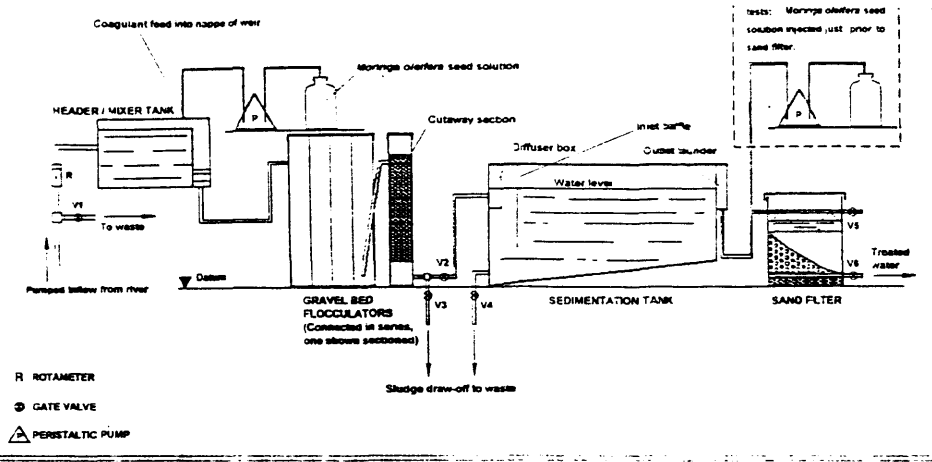


Figure 1 - Schematic diagram of pilot scale water treatment works

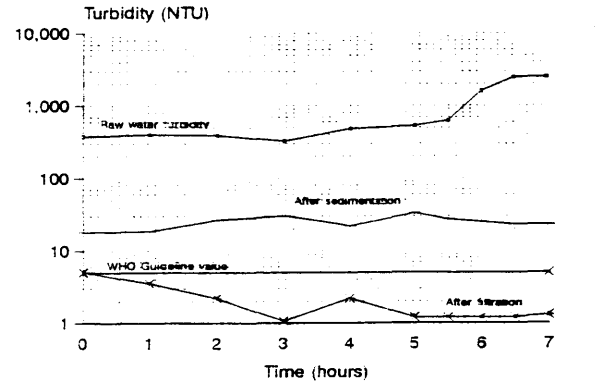


Figure 2 - Pilot plant performance under conditions of high turbidity.
M. oleifera dose averaged 150 mg/l

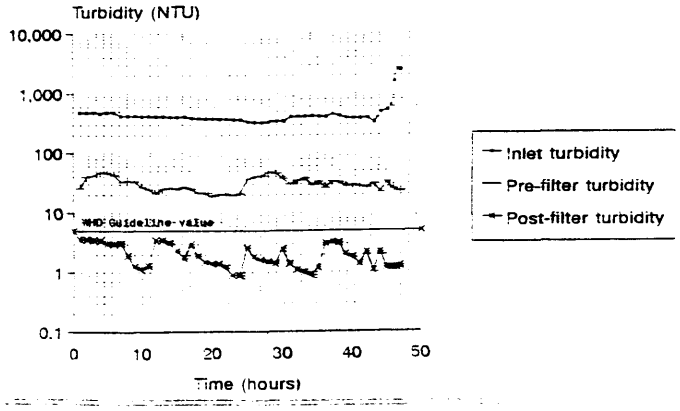


Figure 3 - Composite of treatment data from the pilot plant studies

system was successfully commissioned during the rainy season with the source river exhibiting turbidity levels in excess of 400 NTU throughout the study period. Figure 2 shows the results of one test conducted over an 8 hour period with the full treatment sequence in operation. *M. oleifera* seed was dosed at 200 mg l^{-1}. Raw water turbidity of 1000 NTU was reduced to below 10 NTU by coagulation/ sedimentation. As the sand filter "worked in" the final outlet turbidity was below 1 NTU and still improving until the run was terminated by a regional power failure. Presumptive coliform reductions were in the order of 96%. Figure 3 is a composite sequence of all runs conducted. It can be seen that solids removal within the plant was consistently above 90% following a gravel bed flocculation stage and plain horizontal flow sedimentation. Subsequent rapid gravity sand filtration gave a final, treated water turbidity generally well below 5 NTU. *M. oleifera* seed dose ranged from 75-250 mg l^{-1} depending on the initial raw water turbidity (4).

River water treatment at full-scale

During the following rainy season, the main rural treatment works was operated using *M. oleifera* solution as coagulant. The works comprise upflow contact clarifiers followed by rapid gravity filters and chlorinator. The clarifiers are in a state of disrepair with the impeller drives and chemical feed pumps inoperative. Alum solution is introduced into the incoming flow of 60 $m^3 h^{-1}$ via a simple declining rate, gravity feed tank. Comparable treatment performance with alum was achieved. Inlet turbidities of 270-380 NTU were consistently reduced to below 4 NTU. This was the first time that *M. oleifera* had been successfully used as a primary coagulant at such a scale with the treated water entering supply (5). The considerable quantity of *M. oleifera* seed required for the full scale trials was purchased from enthusiastic villagers in the region. It was viewed as a temporary, yet very welcome source of additional income in what is a poor rural community. The tree is widely cultivated in this region of Malawi, being highly prized as a source of fresh

final pH correction) for the works were imported from South Africa at an annual imported equivalent cost of US $ 39,000 (March 1993). It was estimated that if the water utility established and maintained a plantation of *M. oleifera* trees for oil production/presscake coagulant, a net operating profit could be achieved.

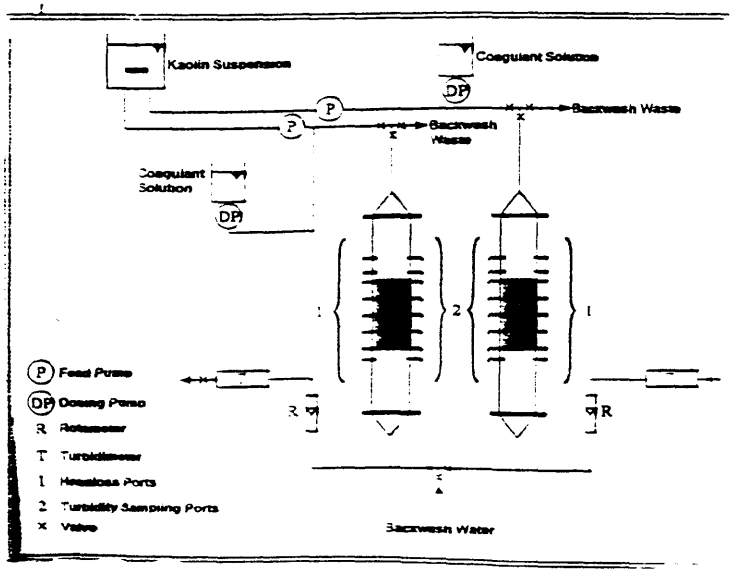
M. OLEIFERA COAGULANT WITHIN A CONTACT FLOCCULATION FILTER

Low turbidity waters are inherently difficult to treat using the conventional treatment sequence of coagulation - flocculation - sedimentation - filtration treatment processes due primarily to the relatively low concentration of suspended particles. The use of contact flocculation filtration (CFF), a process whereby coagulant is introduced directly to the raw water inflow immediately prior to the filter inlet, is under investigation as a single stage treatment alternative. Flocculation and deposition occur entirely within the sand bed.

The objectives of this current study are two-fold. To determine the viability of CFF as a single stage treatment process appropriate for water treatment in developing countries and to extend the range of raw waters that may be treated by *M. oleifera* as a primary coagulant. In order to establish performance characteristics laboratory studies were carried out using a model raw water consisting of Kaolin clay in a deionised water base. The use of such a model water allows for direct comparisons between experimental runs without variations in raw water quality that may be encountered when using a natural water. Subsequent field trials carried out in Southern Malawi were conducted to establish the efficacy of treatment on a low turbidity surface water and provide comparative data.

The laboratory experimental rig consists of two perspex columns of internal diameter 100 mm which may be operated in parallel in upflow or downflow mode from the same header tank and is shown schematically in Figure 4. Diametrically opposed connections for piezometric headloss measurements and turbidity sample extraction are located at intervals along the columns. Headloss measurements were made via pressure transducers linked to a computer to facilitate data acquisition, display and analysis. Turbidity sampling ports consist of stainless steel tube extending 3 mm into the sand bed. Roller clamps on the sample draw off tubes allow continuous turbidity measurement at low flow rates such that minimal disturbances are transmitted to the flow regime within the filter columns. Residual turbidity was measured using a Hach Ratio Turbidimeter. Prior to each experimental run the filter was backwashed to a final washwater turbidity of <2 NTU. Kaolin stock solutions were prepared using a standard procedure. 200g samples of Kaolin (BDH, Grade light, particle size 0.4-1 µm) were mixed to a paste and gradually diluted with deionised water, containing 450 mg l^{-1} sodium bicarbonate, to a final volume of 1 litre. Samples were then rapid mixed for 2 hours at 400 rpm, allowed to settle for 30 minutes and the top 800 ml decanted. This was then made up to 1 litre again with deionised water and mixed for a further 1 hour at 400 rpm. Following settlement for 15 minutes the top 800 ml was decanted and used as a stock solution for dilution to the required turbidity. *M. oleifera* seed solutions were prepared using a previously established method (6). The seeds were de-husked and the kernels crushed using a pestle and mortar. The resulting powder was made into a paste and diluted to give a 1% solution concentration for dosing purposes.

Figure 4 - Schematic of experimental rig shown in downflow mode



Extraction of the active fraction of the seed kernel occurs more readily in the presence of an ionic background. Thus in order to ensure repeatability of experimental results, solution preparation was carried out with deionised water containing sodium bicarbonate at a concentration of 110 mg l^{-1}. Experimental runs were terminated when treated water turbidity exceeded the World Health Organization guideline value of 5 NTU (7) or headloss exceeded 240 cm (8,9). Three initial turbidities were examined viz. 20, 35 and 50 NTU, at filtration rates of 5, 10 and 20 m m^{-2} h^{-1}. A single media bed viz. filter sand of effective size 0.6 mm and uniformity coefficient 1.21 to a depth of 700 mm was used throughout the laboratory study. A summary of the main results is presented in Table

The optimum dose for the combination of run parameters investigated was determined by carrying out a series of dose response runs extending to whichever termination criteria was first reached. Two distinct dose responses

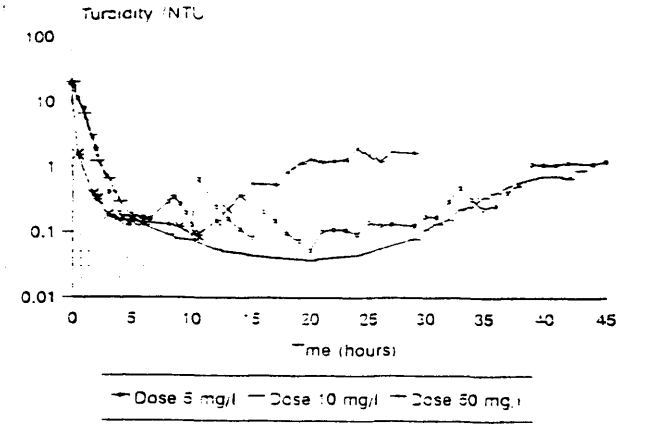


Figure 5 - Dose response at a filtration rate of 5 m/h and initial turbidity 20 NTU

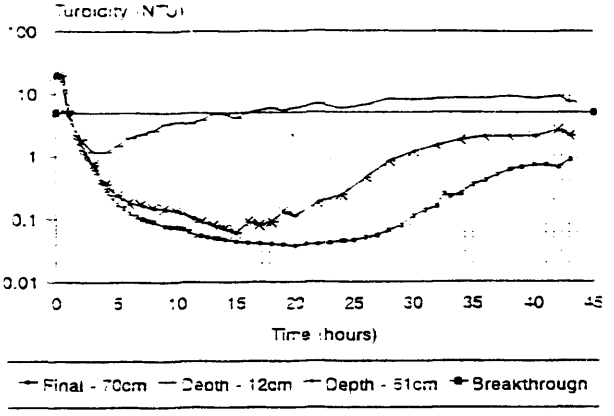


Figure 6 - Residual turbidity as a function of bed depth at a filtration rate of 5 m/h and initial turbidity 20 NTU

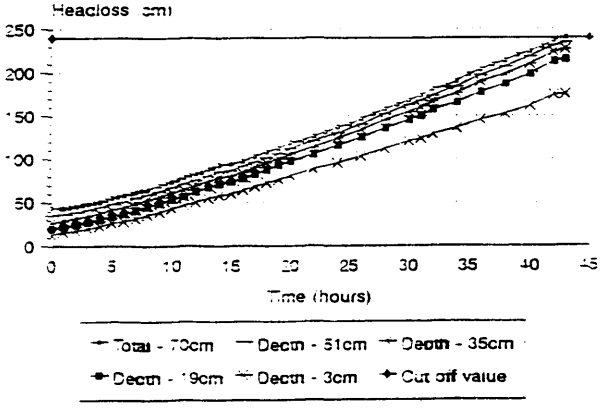


Figure 7 - Headloss development as a function of bed depth at a filtration rate of 5 m/h and initial turbidity 20 NTU

were observed. Figure 5 provides an example for an initial turbidity of 20 NTU and filtration rate of 5 m h^{-1}. Increasing the dose from 5 mg l^{-1} to 10 mg l^{-1} improves filtrate quality with no significant reduction in run time. Further increasing the dose to 50 mg l^{-1} produces a reduced filtrate quality with a significantly shorter run time. It is considered that particulate charge reversal resulting from coagulant overdose, coupled with the adsorption of excess "free" protein on the bed media causes particle - media repulsion accounting for the reduced performance at this high dose. Table 1 also shows that for the same initial turbidity, as the filtration rate is increased, a higher optimum dose is required to maintain performance. In order to maintain this filtrate quality a rise in the inter-pore shear forces must be accompanied by an increase in the strength of the attachment forces. The patch mechanism describes the full adsorption of polyelectrolyte onto a region of opposite charge. In the case of cationic polyelectrolytes, the adsorption is onto the surface of the particle to be destabilised.

It may be envisaged that at the lower filtration rates only one site requires polyelectrolyte adsorption for strong attachment. As the shear forces increase at the higher filtration rates more than one site on the particle may require polyelectrolyte adsorption to ensure attachment.

Table 1 - Summary of the major laboratory CFF results

Turbidity (NTU)	Filtration rate ($m\,h^{-1}$)	Dose ($mg\,l^{-1}$)	Run time (hours)	Reason for termination	Total headloss (cm/m^2)	Backwashes per 1000 m^3
20	20	25	4.5	breakthrough	90.0	11.2
	10	20	25.5	headloss	255.0	4.0
	5	10	43.0	headloss	215.0	4.7
35	20	50	1.5	breakthrough	30.0	33.3
	10	25	5.5	breakthrough	55.0	18.2
	5	15	31.0	breakthrough	155.0	6.5
50	20	25	0.83	breakthrough	16.6	58.8
	10	25	3.58	breakthrough	35.8	22.7
	5	25	14.48	breakthrough	72.2	12.7

Investigations using zeta potential measurements to examine this dose effect are planned.

Figures 6 and 7 show data obtained from a run at a turbidity of 20 NTU and filtration rate 5 m h^{-1} and serve as an example of typical trends that were observed for turbidity removal and headloss development throughout the study. Figure 6 shows clearly the working in stage, working stage and a breakthrough stage which are characteristic of rapid filtration. Additionally it can be seen that the majority of the removal occurs in the top third of the bed.

CONCLUSIONS

The laboratory work to date has demonstrated that CFF in conjunction with a natural polyelectrolyte can be used effectively as a single stage treatment for low turbidity waters at filtration rates up to

10 m h^{-1}.

The main advantages to developing countries of adopting CFF as a single stage treatment process may be summarised:

i. Filtration rates far in excess of those for slow sand filtration significantly reduce the land area required to produce equivalent volumes of treated water;
ii. Unlike direct filtration, CFF does not require a flocculation stage thereby reducing initial construction costs and treatment complexity;
iii. The use of *M. oleifera* as a natural coagulant will reduce foreign exchange expenditure and reliance on imported proprietary chemical coagulants and offer an alternative cash crop to subsistence farmers. Treatment advantages include maintained performance over a wide pH range.

ACKNOWLEDGEMENTS

The authors gratefully acknowledge the continued financial support of this work provided by the Overseas Development Administration of the British Government and that of the Commission of the European Communities.

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SUSTAINABILITY OF WATER AND SANITATION SYSTEMS

Natural coagulants — a sustainable approach

Geoff Folkard, John Sutherland and Reya Al-Khalili, UK

THE M. OLEIFERA TREE is a native of Northern India which now grows widely throughout the tropics. English vernacular names include 'drumstick' (shape of the pods) and 'horseradish' (taste of the roots). It may be propagated from seeds or cuttings, even in poor soils, requiring minimal horticultural attention and is able to survive long periods of drought. It grows rapidly - growth of up to 4 metres in height, flowering and fruiting were all observed within one year during trials near Nsanje, Southern Malawi. Extended and multiple harvests in a single year are evident in many parts of the world. The many products and numerous uses of the tree are given:

Vegetable

- Green pods, leaves, flowers and roasted seeds are highly nutritious.

Oil

- Seeds contain 40% vegetable oil by weight - may be used for cooking, soap manufacture, cosmetics base and as a lamp fuel.

Coagulant

- Seeds used traditionally for 'household treatment' in the Sudan and Kenya.
- Successfully used at pilot and full scale water treatment works in Malawi.
- Presscake remaining after oil extraction effective as a coagulant - bench scale testing.
- Potential for use as an aid to primary sedimentation in wastewater treatment - bench scale testing.

Other uses

- All parts of the plant are used in a variety of traditional medicines.
- Presscake as a soil conditioner/fertiliser and potentially as an animal/poultry feed supplement.
- Green leaves as a fertiliser - mulch.
- Powdered seed used in ointment to treat common bacterial skin infections.
- Grown as live fences and windbreaks.
- Fuelwood source following coppicing.
- Agroforestry within an intercropping system - also providing semi-shade.
- Wood pulp for paper making industry.
- Planted for specific protective and soil melioration functions.
- Planted as an attractive ornamental tree.

M. oleifera is a multipurpose tree of enormous potential. Multipurpose trees are cultivated and managed in such a way that they foster sustainable land use with more than one product and/or function. They are particularly significant in agroforestry systems, performing specific, protective and stabilising functions.

Water treatment

River water drawn for human consumption and general household use can be highly turbid particularly in the rainy season. River silt is churned into suspension and run off from fields and other surfaces carries solid material, bacteria and other micro-organisms into the river. It is of paramount importance to remove as much of this suspended matter as possible prior to a disinfection stage and subsequent consumption. This can generally only be achieved by the addition of coagulants to the raw water within a controlled treatment sequence. In many developing countries proprietary chemical coagulants, such as alum and synthetic polyelectrolytes are either not available locally or are imported using foreign exchange.

A viable alternative is the use of crushed seed of *M. oleifera* as a natural coagulant. The seed pods are allowed to dry naturally on the tree prior to harvesting. The seeds are easily shelled, crushed and sieved using traditional techniques employed for the production of maize flour. The crushed seed powder, when mixed with water, yields water soluble proteins that possess a net positive charge. Dosing solutions are generally prepared as 1-3% solutions. The solution acts as a natural cationic polyelectrolyte during treatment. (Sutherland et al, 1990)

River water treatment at pilot scale

It is now regarded as axiomatic that both water and wastewater technology for developing countries must be no more complex than strictly necessary, be robust and cheap to install and maintain. A prototype treatment works was designed founded on this philosophy. The pilot plant was constructed within the grounds of the Thyolo Water Treatment Works, the works being controlled by the Ministry of Works and Supplies Water Department of the Malawi Government. The system was successfully commissioned during the 1992 rainy season with the source river exhibiting turbidity levels in excess of 400 NTU throughout the study period. Solids removal within the plant was consistently above 90% following a gravel bed flocculation stage and plain

horizontal flow sedimentation. Subsequent rapid gravity sand filtration gave a final, treated water turbidity generally well below 5 NTU. *M. oleifera* seed dose ranged from 75-250mg/1 depending on the initial raw water turbidity. (Folkard et al,1993)

River water treatment at full scale

In February 1994, the main Thyolo works was operated using *M. oleifera* solution as coagulant. The works comprise upflow contact clarifiers followed by rapid gravity filters and chlorinator. The clarifiers are in a state of disrepair with the impeller drives and chemical feed pumps inoperative. Alum solution is introduced into the incoming flow of 60 cubic metres per hour by simple gravity feed. Comparable treatment performance with alum was achieved. Inlet turbidities of 270-380 NTU were consistently reduced to below 4 NTU. This was the first time that *M. oleifera* had been successfully used as a primary coagulant at such a scale with the treated water entering supply. (Sutherland et al, 1994)

M. oleifera seed for the full scale trials was purchased from enthusiastic villagers in the Nsanje region. The tree is widely cultivated in this area, being highly prized as a source of fresh, green vegetable.

Alum and soda ash (for final pH correction) for the Thyolo works were imported from South Africa at an annual imported equivalent cost of £26,000 (March 1993). It was estimated that if the water utility established and maintained a plantation of *M. oleifera* trees for oil production, presscake coagulant, a net operating profit would be achieved.

Treatment of eutrophic water

Treatment studies have recently been conducted at bench scale on a eutrophic lake water serving the main treatment works to Harare, Zimbabwe. (Sutherland et al, 1995) The impounded water contains much light organic matter in suspension due to high algal growth and exhibits relatively low turbidity throughout the year. As such, the water is problematic to treat consuming significant quantities of alum (as primary coagulant) and activated silica (as weighting agent). Alum floc carry over from the clarifiers causes "filter blinding" and the sludge from the clarifiers is voluminous , difficult to dewater and presents pollution problems on discharge to the receiving water. *M.oleifera* in combination with sodium bentonite as weighting agent produced a final water quality equivalent to that produced using the conventional chemical coagulants. The sludge was significantly more compact and represents a potentially useful output as a soil conditioner/ fertiliser.

M.oleifera as vegetable/oil source

M.oleifera pods are an important commercial vegetable crop throughout India. They are also exported fresh under refrigeration and in cans to countries with sizeable Indian communities.

The leaves have outstanding nutritional qualities - amongst the best of all the perennial tropical vegetables. The protein content is 27% and significant quantities of calcium, iron, phosphorous and vitamins A and C are also present. A particular advantage to people nutritionally at risk is that leaves can be harvested during the dry season when no other fresh vegetables are available.

M. oleifera seed contains 40% by weight of oil. The fatty acid profile shows oleic acid at 73% confirming that the oil is similar to olive oil and thus of high quality and high market value. Laboratory tests at Leicester confirm that the presscake remaining after oil extraction still contains the active constituents effecting coagulation. Coagulant may be regarded as a by product of viable oil extraction. (Sutherland, 1993)

Edible oils are an essential component of human nutritional requirements. In developing countries, the production and marketing of edible oils is usually dominated by a few large scale urban-based companies. Rural supplies of the finished product are erratic with increased prices due to additional transport costs.

The Intermediate Technology Development Group (ITDG) in Zimbabwe have successfully introduced technology appropriate for small scale decentralised rural processing of edible oils. A recent evaluation of the 17 oil mills established to constitute this ITDG project concluded (Sunga and Whitby, 1995):

- the oil mills are commercially viable returning an average of 51% on investment with profits of 21% on sales
- a typical mill employs 10 permanent and 3 temporary workers
- ready cash markets for oil seed crops had been created
- lower cost oil of significantly higher quality is now available in the rural areas.
- alternative edible oil seeds such as soya, cotton seed and *moringa* should be investigated
- the market for and sales of presscake are important for the overall viability and profitability of the mills....new outlets are required

Conclusions

The products of the *Moringa oleifera* tree are underexploited resources. Realisation of their potential will contribute towards sustainability in the rural areas, addressing major issues of current concern:

- **water quality and health**
- **food and nutrition**
- **employment opportunities**
- **income generation**

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Contact Flocculation Filtration Using a Natural Polyelectrolyte for the Treatment of Low Turbidity Surface Water in Developing Countries

G. K. Folkard, R. Al-Khalili, and J. P. Sutherland

Abstract

The use of contact flocculation filtration (CFF), a process whereby coagulant is dosed immediately prior to the filter inlet with subsequent flocculation and deposition occurring within the filter bed, is considered to be a viable single stage treatment option for the treatment of low turbidity waters in developing countries. The process is being investigated using a natural cationic polyelectrolyte obtained from seeds of the pantropical tree *Moringa oleifera* Lam.

Laboratory and field studies have demonstrated the seeds to be particularly effective in the treatment of raw waters less than 35 NTU at filtration rates of 5 and 10 m/h. Increasing raw water turbidity and filtration rate reduces filtrate quality and treated volume. It is envisaged that a combination of increased sand media size and filter bed depth will extend the range of initial turbidities that may be effectively treated at higher filtration rates.

1. Introduction

Low turbidity waters are inherently difficult to treat using the conventional treatment sequence of coagulation – flocculation – sedimentation – filtration treatment processes due primarily to the relatively low concentration of suspended particles. Alternatives that may be considered include slow sand filtration (SSF) and direct filtration (DF). SSF has the disadvantage of requiring extensive land area/capital costs to provide significant quantities of treated water. DF demands a flocculation stage involving capital expenditure and generally an external power source for effective floc generation. The use of contact flocculation filtration (CFF), a process whereby coagulant is introduced directly to the raw water inflow immediately prior to the filter inlet, is considered to be a viable single stage treatment alternative. With CFF flocculation and deposition occur entirely within the sand bed.

The main disadvantage with CFF, particularly for developing countries, is the requirement for coagulant addition to ensure optimum performance. In an attempt to address the problems associated with chemical coagulant usage in developing countries, the process is being investigated using a natural polyelectrolyte, specifically seeds of the pantropical tree *Moringa oleifera* Lam (*M.oleifera*). Crushed

seed suspensions yield low molecular weight (approximately 6000 daltons), highly basic (iso-electric point pH 11) water soluble proteins that act similarly to synthetic cationic polyelectrolytes [1]. Pilot plant and full scale trials carried out in southern Malawi have demonstrated the seeds to be as effective as aluminium sulphate for the treatment of raw waters with turbidities greater than 100 NTU [2]. The seeds have shown reduced effectiveness at low turbidities for some raw waters. Although floc formation is evident, the flocs formed are small, compact and light resulting in significantly reduced settling velocities. This is considered to be a function of the mechanism of coagulation and flocculation involved. The low molecular weight of the active proteins indicates that charge neutralisation and floc formation are brought about by the patch mechanism as opposed to the bridging mechanism [3]. Although the mechanism of action has yet to be fully understood, it is considered that the formation of compact, dense floc that may be observed even at high turbidities offers some confirmation.

The objectives of this study may, therefore, be considered two-fold. To determine the viability of CFF as a single stage treatment process appropriate for water treatment in developing countries and to extend the range of raw waters that may be treated by *M.oleifera* as a primary coagulant. In order to establish performance characteristics laboratory studies were carried out using a model raw water consisting of Kaolin clay in deionised water. The use of such a model water allows for direct comparisons between experimental runs without variations in raw water quality that may be encountered when using a natural water. Subsequent field trials carried out in southern Malawi were used to establish the efficacy of treatment on a low turbidity surface water and provide comparative data to that obtained in the laboratory.

2. Laboratory Studies

2.1 Experimental Procedure

The laboratory experimental rig consists of two perspex columns of internal diameter 100 mm which may be operated in parallel in upflow or downflow mode from the same header tank, or in series, for example as an upflow-downflow filtration system. The experimental rig, set up in conventional downflow mode, is shown schematically in Figure 1. Push fit fittings allow for rapid conversion to upflow mode. Diametrically opposed connections for piezometric headloss measurements and turbidity sample extraction are located at intervals along the columns. Headloss measurements were made via pressure transducers linked to a computer to facilitate data acquisition, display and analysis. Turbidity sampling ports consist of stainless steel tubes extending 5 mm into the sand bed. Roller clamps on the sample draw off tubes allow continuous turbidity measurement at low flow rates such that minimal disturbances are transmitted to the flow regime within the filter columns. Residual turbidity was measured using a Hach Ratio Turbidimeter. Prior to each experimental run the filter was backwashed to a final turbidity below

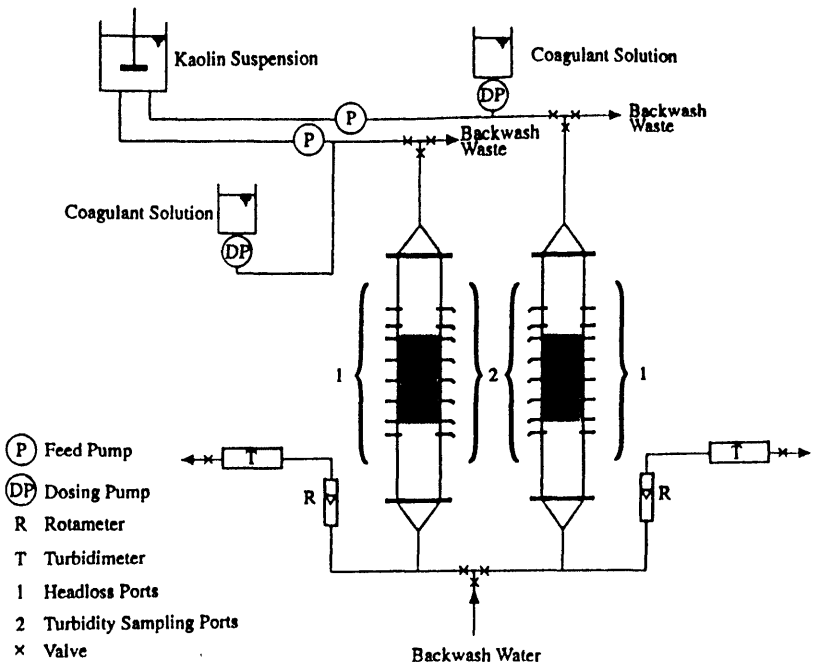

Fig. 1. Schematic of experimental rig shown in downflow mode

Kaolin stock solutions were prepared using the following procedure. 200 g samples of Kaolin (BDH, Grade light, particle size 0.4–1 μm) were mixed to a paste and gradually diluted with deionised water, containing 450 mg/l sodium bicarbonate, to a final volume of 1 litre. Samples were then rapid mixed for 2 hours at 400 rpm, allowed to settle for 30 minutes following which the top 800 ml was decanted. This was then made up to 1 litre again with deionised water and mixed for a further 1 hour at 400 rpm. Following settlement for 15 minutes the top 800 ml was decanted and used as a stock solution for dilution to the required turbidity.

M.oleifera seed solutions were prepared using a previously established method [4]. The seeds were de-husked and the kernels crushed in a pestle and mortar. The resulting powder was made into a paste and diluted to give a 1 % solution concentration. Extraction of the active fraction of the seed kernel occurs more readily in the presence of an ionic background hence, in order to ensure repeatability of experimental results solution preparation was carried out with deionised water containing sodium bicarbonate at a concentration of 110 mg/l.

Experimental runs were terminated when treated water turbidity exceeded the World Health Organization guideline value of 5 NTU [5] or headloss exceeded 240 cm [6, 7].

Three initial turbidities were examined viz.: 20, 35 and 50 NTU, at filtration rates of 5, 10 and 20 m/h. A single media bed viz.: filter sand of effective size 0.6 mm and uniformity coefficient 1.21 to a depth of 700 mm was used throughout the laboratory study. A summary of the main results is presented in Table 1.

Table 1. Summary of the main laboratory CFF results

Turbidity NTU	Filtration rate m/h	Dose mg/l	Run time h	Reason for termination	Total output m^3/m^2	Backwashes per 1000 m
20	20	25	4.5	Breakthrough	90.0	11.2
	10	20	25.5	Headloss	255.0	3.95
	5	10	43.0	Headloss	215.0	4.7
35	20	50	1.5	Breakthrough	30.0	33.3
	10	25	5.5	Breakthrough	55.0	18.2
	5	15	31.0	Breakthrough	155.0	6.5
50	20	25	0.83	Breakthrough	16.6	58.8
	10	25	3.58	Breakthrough	35.8	22.7
	5	25	15.83	Breakthrough	72.2	12.7

Optimum dose for the combination of run parameters investigated was determined by carrying out a series of full length dose response runs. Two distinct dose responses were observed. Figure 2 provides an example for an initial turbidity of 20 NTU and filtration rate 5 m/h. Increasing the dose from 5 mg/l to 10 mg/l improves filtrate quality with no significant reduction in run time. Further increasing the dose to 50 mg/l produces a reduced filtrate quality with a significantly shorter run time. It is considered that particulate charge reversal, resulting from coagulant overdose, coupled with the adsorption of excess 'free' protein on the bed media causing particle-media repulsion, would account for the reduced performance at high dose. Table 1 also shows that for the same initial turbidity, as filtration rate is increased a higher optimum dose is required to achieve performance. A rise in the inter-pore shear forces must be countered by an increase in the strength of the attachment forces. The patch mechanism describes the full adsorption of polyelectrolyte onto a region of opposite charge, in the case of cationic polyelectrolytes, on the surface of the particle to be destabilised. It may be envisaged that at the lower filtration rates only one site requires polyelectrolyte adsorption for strong attachment. As the shear forces increase at the higher filtration rates more than one site on the particle may require polyelectrolyte adsorption to ensure attachment. Investigations using zeta potential measurements to examine this dose effect are planned.

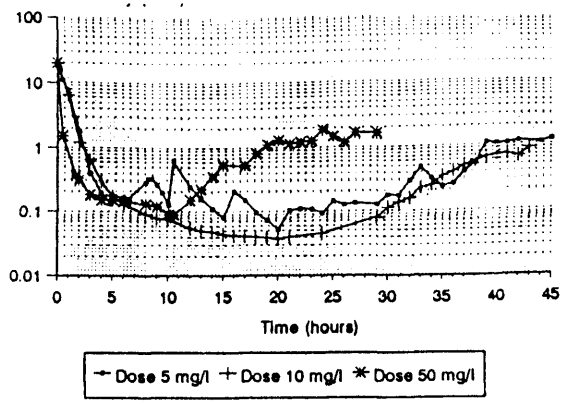

Fig. 2. Dose response; filtration rate 5 m/h and initial turbidity 20 NTU

Figures 3 and 4 show data obtained from a run at a turbidity of 20 NTU and filtration rate 5 m/h and provide an example of the trends that were observed for turbidity removal and headloss development throughout the study. Figure 3 shows clearly the working in stage, working stage and breakthrough stage which are characteristic of rapid filtration [6]. Additionally it can be seen that the majority of the removal occurs in the top third of the bed. During the initial stages of the run the majority of headloss is developed in the top 3 cm of the bed, as the run progresses the bed to a depth of 19 cm becomes active. This can be seen from the increased rate of headloss development over that of the top 3 cm. At run termination, no significant headloss development has occurred in the bottom two thirds of the bed. The minimal bed penetration is considered to be due to strong particle to particle and particle to media attachment forces under the hydraulic conditions of the run. The effects of increasing filtration rate and solids loading on these forces are considered in the next section.

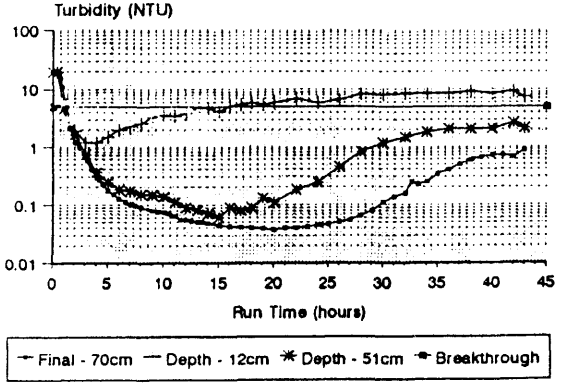

Fig. 3. Residual turbidity as a function of bed depth at a filtration rate of 5 m/h and initial turbidity 20 NTU

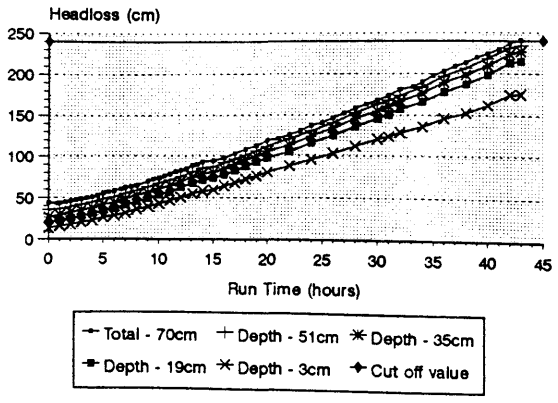

Fig. 4. Headloss development as a function of bed depth at a filtration rate of 5 m/h and initial turbidity 20 NTU

Figures 5 and 6 show the effect of increasing filtration rate and initial turbidity on filtrate quality and output. The output is expressed as that volume of water passing through a unit area of the filter and provides a more effective measure of run length when comparing experimental runs at different filtration rates [6]. With reference to Figures 5 and 6 and Table 1 it can be seen that, in general, an increase in both filtration rate and initial turbidity reduces filtrate quality and output.

Increasing the filtration rate increases the rate at which material is deposited, as shown by the shorter working in stage, however, this has the additional effect of increasing the interpore shear forces. If the shear forces exceed the attachment forces, the result will be a higher degree of breakoff from the surface of the filter bed media [8, 9]. The net result is that material is removed from suspension at a reduced level, as indicated by the reduction in filtrate quality. A similar situation may be considered for an increase in initial turbidity. Increasing the solids load would promote more rapid deposition with a subsequent rise in interpore shear

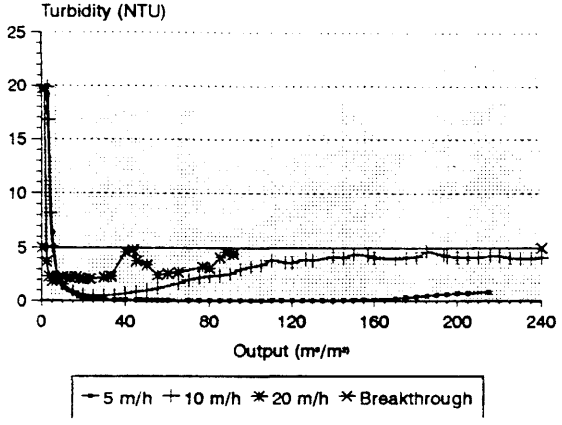

Fig. 5. Effect of increasing filtration rate on turbidity removal and output; initial turbidity 20 NTU

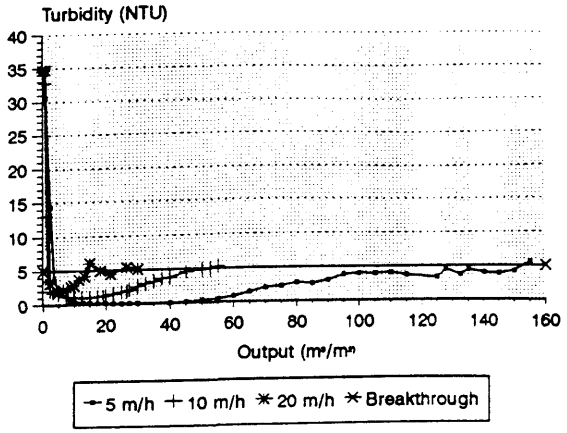

Fig. 6. Effect of incrasing filtration rate on turbidity removal and output; initial turbidity 35 NTU

forces potentially a higher rate of breakoff. Both these aspects would appear to be confirmed by the run carried out at 10 m/h and a turbidity of 20 NTU. In comparison with the run at 5 m/h at the same turbidity, the working in stage is reduced as a result of increased deposition, however, the working stage is significantly shorter. Following breakthrough a balance between breakoff and deposition results in an almost constant filtrate quality. In addition, it was observed that turbidity breakthrough occurred with significant quantities of floc appearing in the filtrate. This would tend to support the theory that floc breakoff, and hence the forces involved in the attachment mechanism, is a major factor in controlling the length of run and filtrate quality. However, with no knowledge of the previous history of the floc, i.e. whether it has been formed as a result of breakoff or as a direct result of inter-pore flocculation, it is difficult to come to any definite conclusions.

3. Field Trials

Field trials of the CFF process using *M.oleifera* were carried out at Thyolo in southern Malawi during the transition period from the wet to dry season in August 1994. A scaled up version of the filter columns used in the laboratory studies was utilised. Column diameter was 279 mm with a bed depth 700 mm. Headloss and turbidity ports were provided at the same depths previously used. Headloss was measured using a differential pressure transducer. Filter sand of effective size 0.6 mm but with no uniformity coefficient given, was purchased from Blantyre Water Board, Blantyre, Malawi. A summary of the main results is presented in Table 2.

Inlet turbidities remained within the range 20–30 NTU throughout the period of the study. The main findings of the study may be summarised as follows.

Figures 7 and 8 provide an example of turbidity removal and headloss development at a filtration rate of 5 m/h. As for the laboratory studies the majority of

Table 2. Summary of the main field trial CFF results

Filtration rate m/h	Average inlet turbidity NTU	Dose mg/l	Run time h	Reason for termination	Total output m^3/m^2	Backwashes per 1000 m
5	20.4	17	31	Headloss	155	6.2
5	23.6	21	38	Headloss	190	5.0
10	28.4	25	19	Headloss	190	5.5
10	20.5	22	25	Headloss	250	4.2
20	27.9	29.5	7.5	Headloss	150	7.0

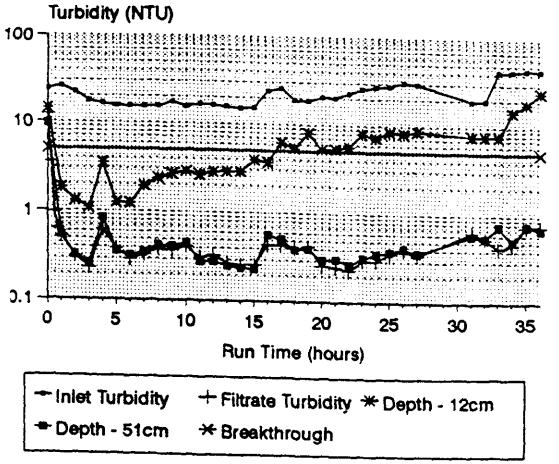

Fig. 7. Field trials: Run carried out at a filtration rate of 5 m/h; seed dose 16.5 mg/l

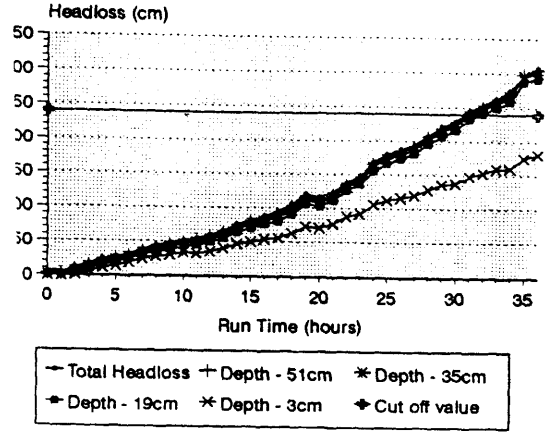

Fig. 8. Field trials: Headloss development at a filtration rate of 5 m/h; seed dose 16.5 mg/l

removal occurs in the top third of the bed, however, the increased efficiency of the bed from 3 to 19 cm would indicate an improvement in bed penetration.

The effect of increasing filtration rate is shown in Figure 9. Significant differences between laboratory and field results may be observed. For comparative purposes residual turbidity is plotted against output. Increasing the filtration rate from 5 m/h to 10 m/h produces only a marginal reduction in filtrate quality with output remaining the same. The reason for the deterioration and subsequent improvement of filtrate quality at 10 m/h is unknown, however, this was observed on more than one occasion. At a filtration rate of 20 m/h output and filtrate quality were significantly improved over that obtained in the laboratory. As inlet turbidity remained approximately the same for all runs, it is considered that the improved performance is a result of an increase in the strength of inter-particle and particle to bed media attachment forces, brought about by the presence of cations in the natural raw water, coupled with a more poly-disperse particle size range.

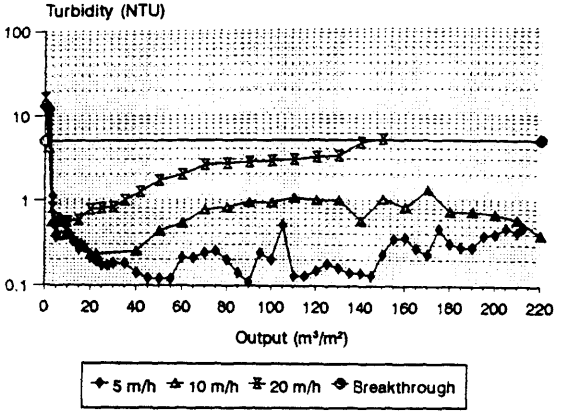

Fig. 9. Field trials: Residual turbidity as a function of filtration rate

4. Discussion

The laboratory and field trials have demonstrated that CFF in conjunction with a natural polyelectrolyte can be used effectively as a single stage treatment for low turbidity waters at filtration rates less than 10 m/h. At higher filtration rates, filtrate quality and treated volume are significantly reduced. In addition as initial turbidity increases a similar effect is obtained. In both cases there is a significant body of evidence to suggest that a move to a larger bed media size would improve performance (e.g. [6, 9]). A larger media size results in lower pore velocities, at equivalent filtration rates, and hence lower shear forces enhancing particle to media attachment. The lower pore velocities result in reduced headloss effects and it has been reported that deeper bed penetration occurs, approaching what may be

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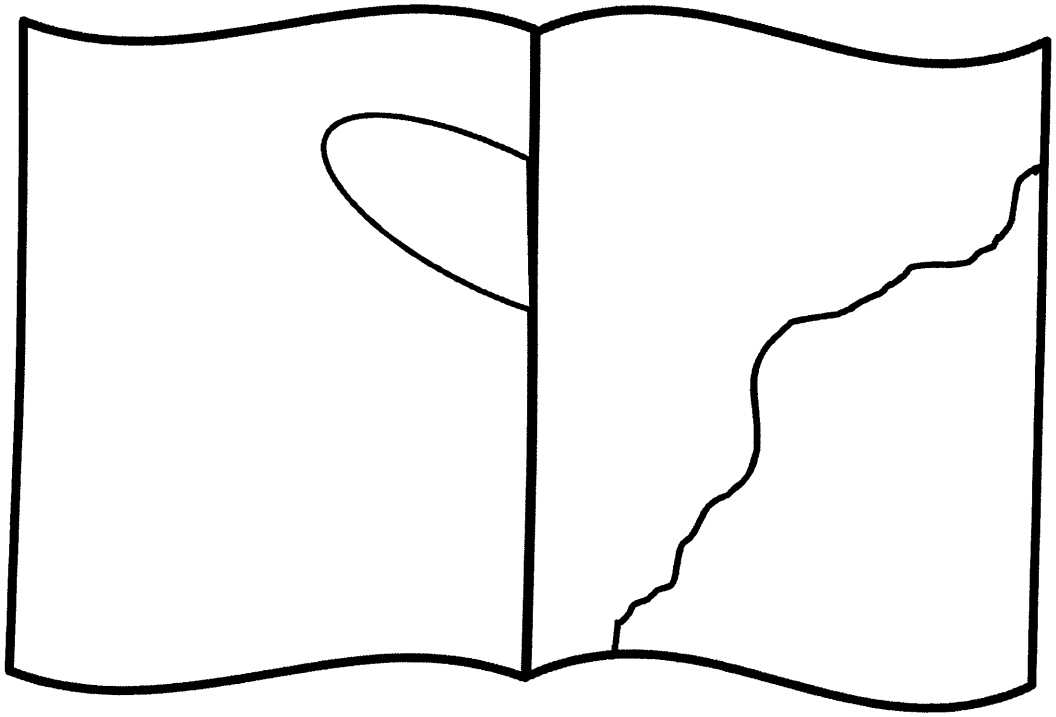

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bed depths to ensure performance, the disadvantage of this being an increase in the volume of backwash water required [9].

Preliminary studies using the laboratory experimental system outlined previously have indicated that by increasing the size of the bed media alone improved performance may be achieved at higher filtration rates and initial raw water conditions, however, sufficient data are not yet available for firm conclusions to be drawn.

The main advantages to developing countries of such a combined treatment system may be summarised as follows:

i) Filtration rates far in excess of those for slow sand filtration significantly reduce the land area required for producing the same volume of treated water;
ii) Unlike direct filtration, CFF does not require a flocculation stage thereby reducing initial construction costs;
iii) The use of *M.oleifera* as a natural coagulant may potentially reduce foreign exchange expenditure and reliance on imported proprietary chemical coagulants and offer an alternative cash crop to subsistence farmers. Treatment advantages include operation over a wide pH and dose range.

With respect to SSF, a potential disadvantage of CFF is a requirement for a separate disinfection stage following treatment. Studies are planned to determine the extent to which microorganisms are removed.

5. Conclusions

The CFF process utilising a natural polyelectrolyte as coagulant has been shown to be highly effective at reducing turbidity levels at filtration rates less than 10 m/h. Increasing bed media size and depth are considered means by which performance at higher filtration rates and initial turbidities may be improved. The use of a natural coagulant within such a treatment system is considered to offer significant advantages over proprietary chemical coagulants particularly for developing countries.

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Filtration with a natural coagulant

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RIVER WATER DRAWN for human consumption in many developing countries can be highly turbid particularly in the wet season. Such waters can be treated effectively using conventional treatment systems incorporating coagulation, flocculation, solid-liquid separation and disinfection. Low turbidity waters, as may be experienced during the dry season, are inherently difficult to treat due, primarily, to the relatively low concentration of suspended particles. Under such circumstances the use of a conventional treatment train can result in the inefficient use of excessive coagulant to provide effective treatment. Alternatives that may be considered for the treatment of low turbidity waters include slow sand filtration (SSF) and direct filtration (DF). SSF provides effective treatment at low filtration rates (generally 0.2 to 0.4m/h). As such, SSF has the disadvantage of requiring an extensive land area to provide significant quantities of treated water. DF involves the formation of floc prior to high rate filtration (generally up to 15m/h). The primary disadvantage of DF is the requirement for a flocculation stage, and backwashing facilities can involve significant construction and maintenance costs. The use of contact flocculation filtration (CFF), a process whereby coagulant is introduced directly to the raw water inflow immediately prior to the filter inlet, is considered to be a viable single stage treatment alternative. With CFF flocculation and deposition occur entirely within the sand bed. A flocculation stage is not required, consequently there is a reduction in construction and operational costs over DF, suggesting that such a process may be a viable option for developing countries.

The CFF process is under investigation at Leicester University in the UK using material of plant origin as a coagulant. The natural coagulant is obtained from the seeds of the pantropical tree *Moringa oleifera* Lam (*M.oleifera*). This tree is native to Northern India, Pakistan and Afghanistan, but is now widely cultivated throughout the tropics. It can be grown from seeds or cuttings in poor soils with minimal horticultural attention and watering. The tree can grow up to 4 metres and commence fruiting within the first year (Folkard *et al*, 1993). It is a multipurpose provider, with the tree, seeds, flowers, seed oil and leaves used variously (Folkard *et al*, 1995). For water treatment applications the seed pods are allowed to dry naturally on the tree prior to harvesting. The seeds are then removed from the shells, crushed into a fine powder and sieved. When mixed with water, the seed powder yields low molecular weight, highly basic proteins which act in a similar manner to that of synthetic cationic polyelectrolytes.

Laboratory studies

Laboratory studies were conducted on model low turbidity waters using laboratory-scale sand filters in a CFF configuration. The rig consists of two perspex columns of internal diameter 100mm. Diametrically opposed connections for piezometric headloss measurement and turbidity sample extraction are located at intervals along the column to enable monitoring of the removal process within the filter bed. The filter medium used was silica sand. In order to establish performance characteristics laboratory studies were carried out using a model raw water consisting of Kaolin clay in deionised water. The use of such a model water allows for direct comparisons between experimental runs without variations in raw water quality that may be encountered when using a natural water (McCooke and West 1978). *M.oleifera* seed solutions were prepared using a previously established method and dosed as a 1 per cent solution (Folkard *et al*, 1996). The effectiveness of the filtration process was measured in terms of final water turbidity, headloss development and volume of treated water produced. Experimental runs were terminated when treated water turbidity exceeded the World Health Organisation guideline value for potable water of 5 NTU (WHO, 1993) or headloss exceeded 2.4m (Adin and Rebhun, 1974).

Results

Five raw water turbidities were examined viz.: 10, 20, 35, 50 and 75 NTU, at filtration rates of 5, 10 and 20 m/hour. The bed depths considered were 0.7 and 1.2 metres. Two sand grain sizes were compared for effectiveness as filter media. The smaller classified as British Standard (BS) mesh size 16/30 (nominal size range 0.50-1.00mm), the larger as BS mesh size 10/18 (nominal size range 0.85-1.70mm). The optimum dose was determined by conducting a series of filter runs to termination. A summary of the main results is presented in Table 1.

The effect of sand grain size

Increasing the size of the sand grains used as the filter media had two major effects. Firstly a reduction in filtrate quality and secondly a reduction in the rate of headloss development across the filter bed. Figures 1 and 2 show turbidity removal and headloss development with the two sand sizes at an initial turbidity of 50 NTU. In both cases turbidity breakthrough was the terminating factor for the filter run. Consequently the filter bed with the smaller sand size was more effective than the larger sand, producing

Table 1. Summary of the main results obtained from the laboratory scale filter

Initial Turbidity (NTU)	Sand Size Range (mm)	Sand Bed Depth (cm)	Filtration Rate (m/hour)	Seed Dose mg/L	Total Output (m^3/m^2) *
75	0.50-1.00	70	10	5	20
75	0.50-1.00	70	5	25	70
75	0.50-1.00	120	10	50	40
75	0.50-1.00	120	5	25	78
75	0.85-1.70	70	10	50	10
75	0.85-1.70	120	10	25	45
75	0.85-1.70	120	5	25	72
50	0.50-1.00	70	20	35	17
50	0.50-1.00	70	10	25	54
50	0.50-1.00	70	5	25	46
50	0.50-1.00	120	10	25	72
50	0.85-1.70	70	20	25	25
50	0.85-1.70	70	10	25	33
50	0.85-1.70	120	10	25	33
35	0.50-1.00	70	20	25	55
35	0.50-1.00	70	10	25	80
35	0.50-1.00	70	5	15	150
35	0.50-1.00	120	10	25	80
35	0.85-1.70	70	10	15	50
35	0.85-1.70	70	5	10	154
35	0.85-1.70	120	10	25	80
20	0.50-1.00	70	20	25	10
20	0.50-1.00	70	10	20	134
20	0.50-1.00	70	5	10	145
20	0.85-1.70	70	20	20	129
20	0.85-1.70	70	10	20	134
10	0.50-1.00	70	20	10	116
10	0.50-1.00	70	10	10	224
10	0.50-1.00	70	5	15	242
10	0.85-1.70	70	5	15	331

* Total output for an equivalent filter of surface area $1m^2$

54 m^3/m^2 as compared to 33 m^3/m^2. Note that the output is expressed as that volume of water passing through a unit area of the filter and provides a more effective comparison of experimental runs at different filtration rates (Adin and Rebhun, 1974).

However, for an initial turbidity of 10 NTU (Figures 3 and 4), the reduction in filtrate quality produced from the filter with the larger sand grain size was less significant. The reduction in headloss development was of greater significance, since this was the termination factor for the filter run. The bed with the smaller sand grain size produced 242 m^3/m^2 as compared to 331 m^3/m^2 from the bed with the larger grain size - an increase of 37 per cent of useful throughput.

The effect of higher filtration rates

Figure 5 shows the effect of increasing filtration rate on filtrate quality and output for experimental runs carried out at an initial turbidity of 35 NTU. With reference to Figure 5 and Table 1 it can be seen that in general an increase

Increasing the filtration rate increases the rate at which material is deposited, as shown by the shorter working-in stage; however, this has the additional effect of increasing the interpore shear forces. If the shear forces exceed the attachment forces the result will be a higher degree of breakoff from the surface of the filter bed media. The net result is that material is removed from suspension at a reduced level, as indicated by the reduction in filtrate quality.

The effect of increasing bed depth

From Figure 6 and Table 1 it can be seen that increasing the bed depth from 0.7 to 1.2m improves filter performance in terms of filtrate quality and volume output. It was found that the reduced performance (in terms of filtrate quality) of the filter bed of larger sand size could be counteracted by using deeper beds, whilst still maintaining the benefit of lower headloss development. Figure 6 demonstrates this phenomenon. One filter comprises a 1.2m deep bed of the larger sand grains. The other comprises the smaller grains to a bed depth of 0.7m. Both filters were operated at 10 m/h with an inlet turbidity of 35 NTU and similar turbidity reduction is evident. This is considered to be due to the filter media providing an approximately equivalent surface area for floc attachment in both filter beds. The potential benefits from increasing bed depth viz. reduced headloss, must be balanced against an increased requirement for backwash water.

Conclusions

The CFF process utilizing a natural polyelectrolyte as coagulant has been shown to be highly effective in the treatment of low turbidity raw waters at filtration rates less than 10m/h. The study is still continuing and as yet no firm conclusions can be drawn. However, results do indicate that alterations in filter configuration, through changes in sand grain size and filter bed depth, can enhance performance depending on initial raw water conditions. Current work is examining the potential benefits of dual media beds in the CFF system. The use of a natural coagulant within such a treatment system is considered to offer significant advantages over proprietary chemical coagulants particularly for developing countries.

Acknowledgements

The authors gratefully acknowledge the financial support of the Overseas Development Administration of the British Government and the European Union.

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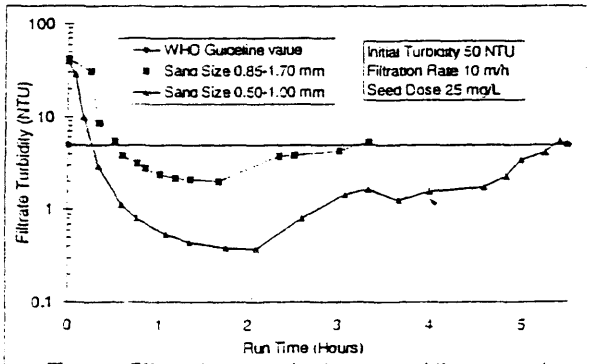

Figure 1 Effect of sand grain size on turbidity removal

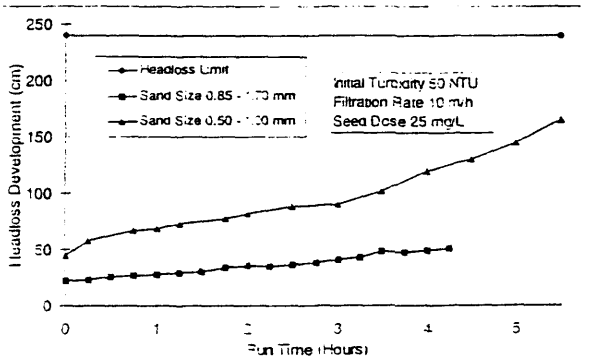

Figure 2 Effect of sand grain size on headloss development

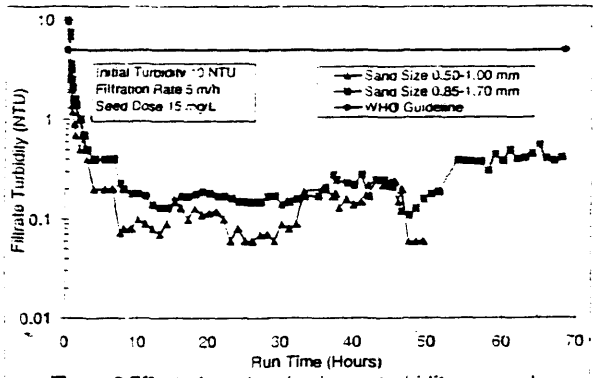

Figure 3 Effect of sand grain size on turbidity removal

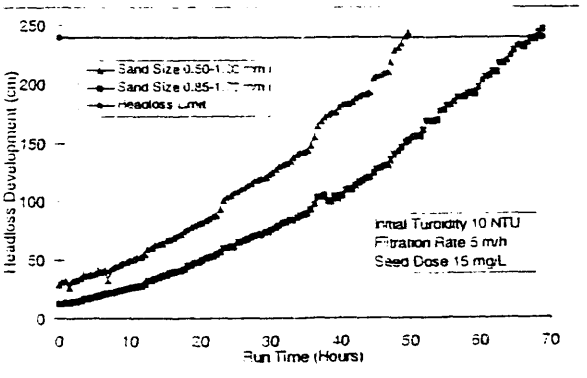

Figure 4 Effect of sand grain size on headloss development

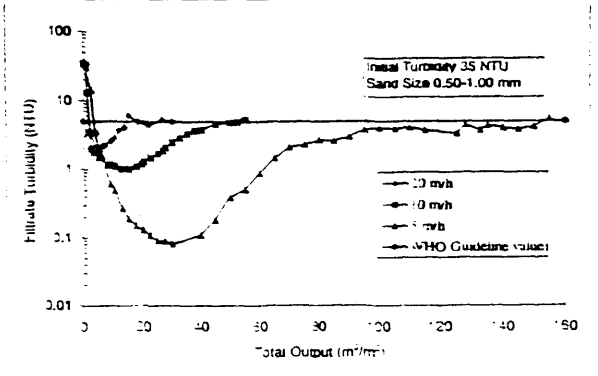

Figure 5 Effect of filtration rate on turbidity removal

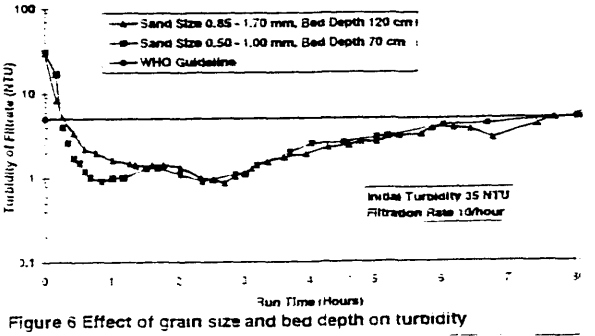

Figure 6 Effect of grain size and bed depth on turbidity