THE APPLICATION OF *MORINGA OLEIFERA* SEEDS AS A COAGULANT FOR WATER TREATMENT IN DEVELOPING COUNTRIES

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ABSTRACT

THE APPLICATION OF *MORINGA OLEIFERA* SEEDS AS A COAGULANT FOR WATER TREATMENT IN DEVELOPING COUNTRIES

The focus of this study has been on the ability of crushed seed suspensions of the pan-tropical tree *Moringa oleifera* Lam. to clarify turbid waters. Information regarding the coagulation effect of the seeds was first published in 1979. Since then a number of studies have been carried out to establish the effectiveness and nature of the coagulation effect. These are reviewed. The coagulation effect has been attributed to water soluble, low molecular weight cationic proteins contained within the seed kernel. The mode of activity has been proposed to be charge neutralisation via the patch mechanism. In an attempt to further the understanding of coagulation behaviour, four water sources in Malawi and Zimbabwe have been examined. A modified theory of protein coagulation that involves a combined charge neutralisation and protein precipitation mechanism is presented. Further work required to fully determine the mechanism of activity is discussed.

In order to establish whether seed solutions could be considered for use within continuous flow water treatment systems a pilot plant was designed and constructed in Malawi. Details of the design are presented. Studies undertaken over a three-year period demonstrated that, as a coagulant, seed solutions were effective. Subsequent to these trials, an evaluation of the efficacy of using seed solutions on an operational treatment works was undertaken. Factors that might be potential constraints to utilisation at this scale, for example seed processing and dosing solution preparation, were investigated. Although effective as a coagulant, it has been concluded that the use of powdered seed suspensions is not a viable option for large-scale applications. Amongst those constraints identified, problems with the preparation of dosing solutions, the potential for increasing Total Organic Carbon levels in treated water and regulatory issues are considered the

most significant. Alternative routes by which *M.oleifera* could still be utilised on a large scale are discussed.

CONTENTS

LIST OF TABLES

LIST OF FIGURES

CHAPTER 1

INTRODUCTION

While the majority of the developed world can take a plentiful supply of pure, clear water for granted, those in the developing world cannot. According to statistics released by the World Health Organisation (WHO) and other agencies, there are currently 1500 million people in rural, peri-urban and urban areas of developing countries (excluding China) without access to safe water supplies. The figures for those without access to adequate sanitation facilities are even higher. Up to 30 million people die each year from causes that can directly be attributed to the lack of adequate water and sanitation facilities (WHO/WSSC/UNICEF, 1996). If population growth in developing countries continues unabated the problems will become more acute. Rising demands on already over stretched raw water supplies for domestic, industrial and agricultural purposes will inevitably lead to increased chemical and microbiological contamination.

It is generally accepted that the provision of potable water in isolation is not sufficient to address current problems. Whilst the cause and effect relationships of water-related disease are complex, they may be classified into three major categories:

i) Infections spread through water supplies (water-borne diseases) including amoebic dysentery, cholera, typhoid fever, hepatitis A and poliomyelitis.

ii) Infections brought about by the lack of water for personal hygiene (water-washed diseases) including scabies, trachoma and bacillary dysentery.

iii) Infections caused by parasitic worms (water vector-borne diseases) including schistosomiasis and guinea worm.

How these infections may be prevented is dependent on the mode of disease transmission. In the first category, the choice of water supply and the provision of suitable treatment facilities and mechanisms to prevent subsequent contamination, can greatly reduce the risk of infection. This reduction is, therefore, dependent on water quality. In the second category, improvements in

1

personal hygiene are seen as being the key to reducing infection rates. In many rural areas collection of water may involve travelling long distances. In this case the priorities for the limited quantity of water collected will be drinking and cooking. Improvements in personal hygiene can only be brought about if sufficient amounts of water are made available, this implies that it is quantity rather than the quality of water that is important. In the final category, protection of supplies from contamination through adequate sanitation facilities can greatly reduce the risks of infection. In all cases health education plays a vital role.

The provision of a safe water supply is, therefore, only part of what must be an integrated approach to the problems outlined, however, its importance cannot be understated. Against this background the continued development of village, community and urban water supply systems appropriate to the needs and capabilities of developing countries has an important role to play in reducing morbidity and mortality rates that result directly from contaminated water supplies.

When considering source waters for supply, the deciding factors relate inevitably to the economic and technical inputs required in relation to water abstraction and treatment. These in turn are related to the source water quality and quantity. Groundwaters are the preferred source since, in general, the bacteriological quality is good. Dependent on the water quality, chlorination may be the only treatment required. Access to groundwaters may be achieved using simple handpumps or motorised pumps that feed into a distribution system. Shallow wells, unless properly protected, are not recommended due to possible contamination from livestock. In some areas groundwaters can be adversely affected by salt water incursions or an excess of chemical contaminants (e.g. fluorides, iron, arsenic) thereby limiting the extent of their use. In such circumstances or where groundwaters are not easily accessible, water of relatively good quality may be obtained from protected upland catchments (streams and lakes). Dependent on raw water quality, treatment may be basic filtration and/or chlorination.

If none of these sources is available, surface water sources must be utilised. The primary disadvantage of using these sources is the complexity of treatment and concomitant costs arising from the inherent variability in raw water quality. The majority of surface sources in the dry

season do not contain large quantities of suspended solids although they may still have high levels of microbiological contamination. In this instance, treatment may be carried out at reasonably low cost using filtration and/or disinfection. It is during the wet season that the problems arise. As a result of extensive run off from surrounding catchments surface waters may become highly turbid (turbidity is a term used to describe the relative clarity of any given suspension and has been defined as an "optical property that causes light to be scattered and absorbed rather than transmitted in straight lines through a sample" (Hach, 1985), in other words, it is a measure of the light scattering effect of suspended particles. The generally accepted units of measurement are Nephelometric Turbidity Units [NTU]). Raw waters containing high levels of turbidity per se do not, in themselves, represent a major health hazard. It is the high levels of microbiological contamination associated with extremes of turbidity that demand effective treatment be carried out.

Turbidity levels must be reduced in order for such waters to be made potable. The reasons for this are two-fold:

i) All methods of disinfection, for example boiling, chlorination, ozonation, ultra-violet exposure etc., are greatly hindered by the presence of turbidity. Chlorine, for example, may still be relatively effective in the presence of high levels of turbidity, however, the quantities required would be excessive. In cost terms it would be uneconomic to chlorinate highly turbid waters. The process of removing turbidity has the effect of significantly reducing microbiological contaminants closely associated with turbidity causing particulates thereby enhancing any final disinfection process;

ii) There is a general misconception that a clear water is a safe water. The provision of water that is aesthetically pleasing is of prime importance in order to satisfy this perception that clear equals safe. The WHO specifies a guideline value of 5 NTU with a maximum permissible of 25 NTU (to put this into context, drinking water in the U.K. generally has turbidity values less than 0.5 NTU). Surface water turbidities in the wet season frequently exceed 200 NTU and can often run into several thousands for long periods. In such circumstances filtration alone is not a viable option and some form of pre-treatment is required to bring turbidity levels down to those that can be

filtered without compromising filtrate quality or adversely effecting the length of filter runs. The simplest pre-treatment options available are plain sedimentation and roughing filtration. In both cases treatment relies on gravity settlement of sufficient particulates to reduce turbidity to acceptable levels. However, in the extreme circumstances found in most tropical developing countries such simple measures are rarely effective. Although the gross settleable solids may be removed significant quantities of colloidal material will still pass through these systems rendering the filtration process ineffective. The only remaining pre-treatment option is that of chemically assisted sedimentation.

In this process, chemicals such as aluminium and ferric salts are added to raw waters to bind suspended particulates into larger agglomerates that may then be settled out under gravity. The process is one that is now used worldwide and that has been demonstrated to be highly effective. The major drawback for developing countries, aside from the cost and complexity of some of the recommended treatment process units, is the requirement for chemicals. If these are not available in-country then they must be imported often at high cost and involving the use of valuable foreign exchange. In general chemical costs constitute a much higher percentage of the operational budget of a treatment works in developing countries than they do in developed countries. It is this specific, significant aspect of the water supply process that this project is directed at.

The study presented here examines a potential alternative to the use of chemicals within the solid-liquid separation stage of water treatment. For years natural materials have been shown to be capable of operating as for conventional chemical coagulants. Of the many natural materials that have been investigated seeds of the tree *Moringa oleifera* Lam. (*M.oleifera*) have been shown to be one of the most effective. The objectives of this study are, therefore:

i) To determine the effectiveness of the seeds over a range of raw water conditions in order to establish mechanisms of action and;

ii) To evaluate performance at different scales of treatment in order to determine viability and potential limitations to their use.

The following chapter provides an introduction to the chemical treatment of raw waters, the theories behind colloid stability and destabilisation, the mechanisms of action of conventional coagulant materials and a brief resume of previous work on alternative coagulant materials.

Chapter 3 gives an introduction to the tree *M.oleifera*, outlines previous work carried out on the water treatment properties of the seeds and puts the current study in context.

Chapter 4 details laboratory studies carried out on raw waters in Malawi and Zimbabwe and puts forward a theory on the mechanism of action of the seeds based on the laboratory results.

Chapter 5 examines the potential for use of *M.oleifera* within continuous flow systems and details work undertaken at pilot and full scale in Malawi. The results are discussed not only in relation to treatment performance but also to ease of seed material processing and dosing solution preparation.

Chapter 6 provides a summary of the findings in Chapters 4 and 5 and discusses these in the context of the practical application to water treatment facilities in developing countries. Further work necessary for implementation to become a reality is outlined.

CHAPTER 2

CHEMICAL TREATMENT OF TURBID SURFACE WATERS

2.1 INTRODUCTION

As introduced in Chapter 1, the primary objective of chemical treatment within water treatment is the reduction of turbidity thereby enhancing the effectiveness of any tertiary disinfection process and providing an aesthetically pleasing water that satisfies the common perception that clear equals safe. The aim of this chapter is to enable a general understanding of how materials can remain in suspension, the mechanisms by which these particles can be destabilised and removed and how this is achieved in practice in the water industry. It must be noted that the inherent variability in natural raw waters has lead to significant volumes of research being generated on the application of individual chemicals to specific raw water conditions. Although such knowledge assists in the general understanding of mechanisms of action of specific coagulants, a review of all such published research is not considered necessary in order to gain an appreciation of the mechanisms involved.

The first section of this chapter provides an overview of the current state of knowledge on the generally accepted mechanisms involved in particle removal using chemical coagulants. The final two sections of the chapter attempt to put chemical treatment in the context of its application in developing countries and outlines potential alternatives to the use of proprietary chemicals.

2.2 THE ROLE OF CHEMICAL TREATMENT

The majority of the sediment load contained within a surface water will settle out under gravity given sufficient time. However, it is those particles that can remain in suspension indefinitely that chemical treatment is concerned with. These particles generally fit into the size range $0.001\mu m$ - $10\mu m$ and are termed colloids (although some bacteria and viruses may lie outside this size range

they will be included for ease of reference). Table 2.1 gives an indication as to the time required for a particle of given diameter to settle:

Diameter of particle (mm)	Order of size	Total surface area[1]	Time required to settle[2]
10.0	Gravel	3.14 cm^2	0.3 sec
1.0	Coarse sand	31.4 cm^2	3 sec
0.1	Fine sand	314 cm^2	38 sec
0.01	Silt	0.314 m^2	33 min
0.001	Bacteria	3.14 m^2	55 hr
0.0001	Colloidal particles	31.4 m^2	230 days
0.00001	Colloidal particles	0.283 ha	6.3 years
0.000001	Colloidal particles	2.83 ha	63 years min.

Source: Adapted from American Water Works Association (1971)
1. Area for particle of indicated size produced from a particle 10mm in diameter with sp. gravity 2.65
2 Calculations based on sphere with a specific gravity of 2.65 to settle 30cm

Table 2.1 Settling time as a function of particle size

From the table it is clear that in order to produce water of suitable quality within a reasonable time span processes other than plain sedimentation are required. To this end, chemicals are added that are capable of binding suspended particles together to form larger agglomerates, termed floc, which can then be removed. In order to understand how these chemicals operate an initial understanding of the mechanisms by which colloidal particles may remain in suspension is necessary.

2.2.1 Colloid stability

It is generally accepted that the stability of colloidal suspensions is caused by the electrostatic interaction of particle surface charges and van der Waals forces of attraction. All solids in an aqueous phase acquire a net electrostatic charge, termed the primary charge. The primary surface charge of the majority of particles encountered in water treatment is negative. In relatively simple terms the origin of this surface charge may arise from three different sources:

i) Isomorphous substitution within the crystal lattice. Clay minerals provide an example of this mechanism. Clay minerals are primarily layered structures comprising tetrahedral layers (consisting of SiO_4 tetrahedra) cross linked with octahedral layers (composed of oxygen and hydroxyl ions and containing aluminium [Al], magnesium [Mg] or other cations). Isomorphous substitution of Al for Si in the tetrahedral layer or Mg for Al in the octahedral layer can both give rise to a net surface negative charge;

ii) Ionisation of surface chemical groups, for example carboxyl, amino and hydroxyl groups. The nature and size of the charge are dependent on the pH of the aqueous phase. Examples include the charge found on bacteria and hydrous metal oxides;

iii) Preferential adsorption of ions from solution. Specific adsorption arises from hydrogen bonding, covalent bonding or van der Waals bonding, all of which can be assisted by electrostatic attraction. As for (ii), the nature of the particle charge will be dependent on the characteristics of the aqueous phase.

Particulate suspensions themselves do not possess an electrical charge, the result being that the particle primary charge must be counter balanced by ions of opposite charge within the suspension. The result is a high concentration of ions close to the surface of the particle and moving with it, termed the Stern layer, with a movable diffuse layer of ions extending out into the solution to a position of electro-neutrality. The diffuse region is known as the Gouy-Chapman layer, the potential at the plane of shear between the Stern and Gouy-Chapman layers is known as the Zeta potential. The overall potential between the surface of the particle and the point of

electro-neutrality is known as the Nernst potential. Figure 2.1 shows schematically this electric double layer.
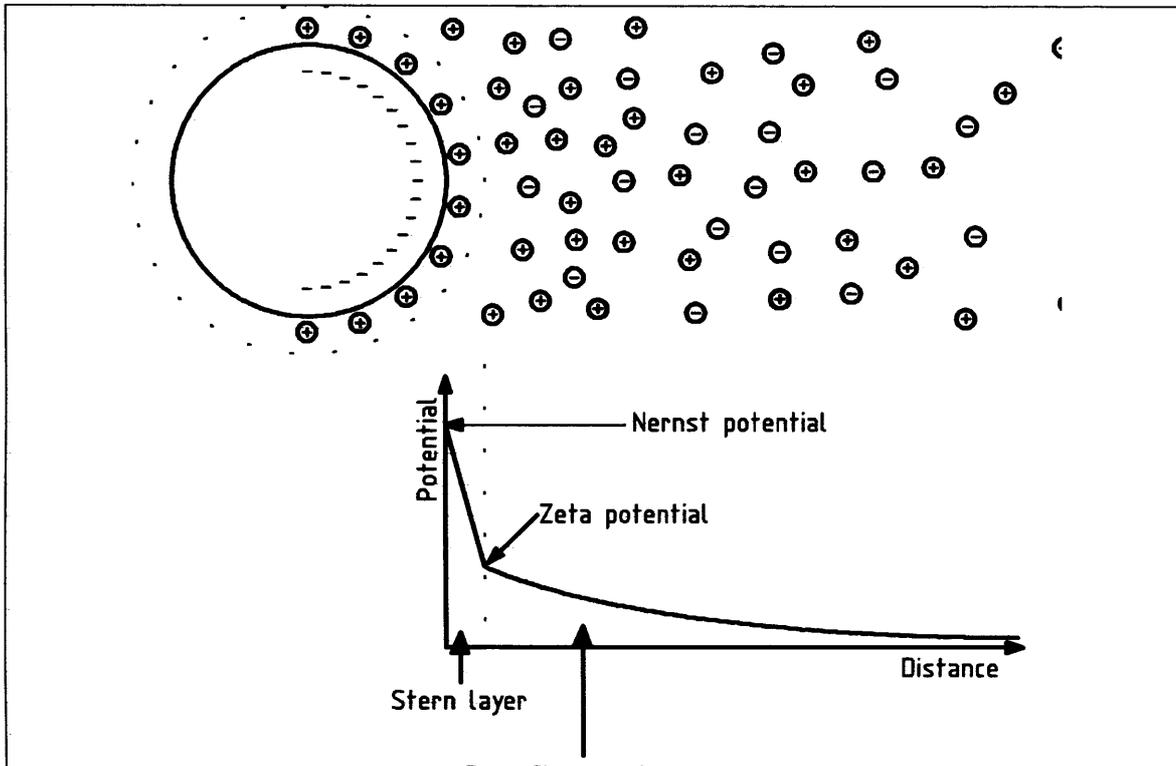
Figure 2.1 The electric double layer surrounding a particle with net negative surface charge in aqueous solution

As two particles of like charge are brought together, the diffuse ion clouds surrounding the particles start to interact. If the zeta potential of both particles is greater than the van der Waals attractive forces then repulsion is the result. If the zeta potential is less than the van der Waals forces then attraction between particles occurs. It is clear, therefore, that in order to remove particles from a stable suspension, these forces of repulsion must be overcome i.e. the particles must be destabilised (for example, Michaels and Morelos, 1955; Black, 1960; Gregory, 1977; Packham and Sheiham, 1977; Dempsey, 1984; Wakeman et al., 1989).

9

2.2.2 Colloid destabilisation

There are two main theories of colloid destabilisation that are of interest in water treatment viz.: reduction of interactive energies and inter-particle bridging.

● **Reduction of interactive energies**

From the theoretical consideration of particle stability outlined in the previous section this would appear to be the most obvious method. The two primary mechanisms by which the electrostatic forces of repulsion may be overcome are:

i) By increasing the ionic strength of the solution surrounding the particle. This has the effect of reducing the zeta potential to a point where the attractive van der Waals forces dominate. This form of destabilisation can only occur with ions of opposite charge and generally obeys an empirical rule known as the Schulze-Hardy rule. This states that higher valence counter ions are more effective at reducing the repulsive forces. Figures given for this effect show a bi-valent ion to be 50-60 times more effective than a monovalent ion, while a trivalent ion is 700-1000 times more effective than a monovalent ion. Figures 2.2 and 2.3 below demonstrate the effect on the Stern and Gouy-Chapman layers upon the introduction of counterions of increasing valence. It must be noted that a similar effect may be noticed upon the addition of precipitates of opposite charge.
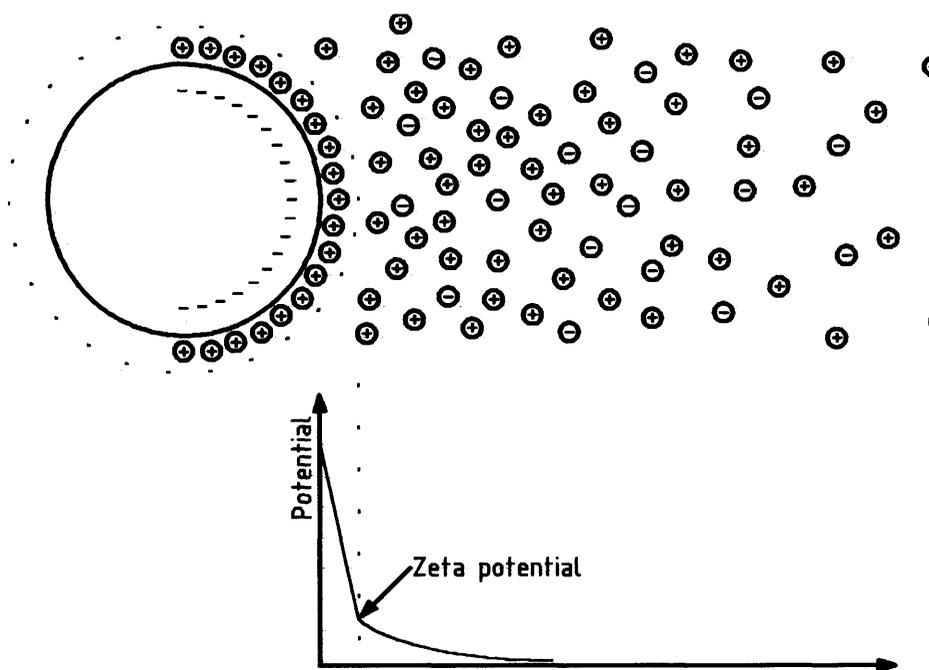
Figure 2.2 Reduction in zeta potential through an increase in solution ionic strength.
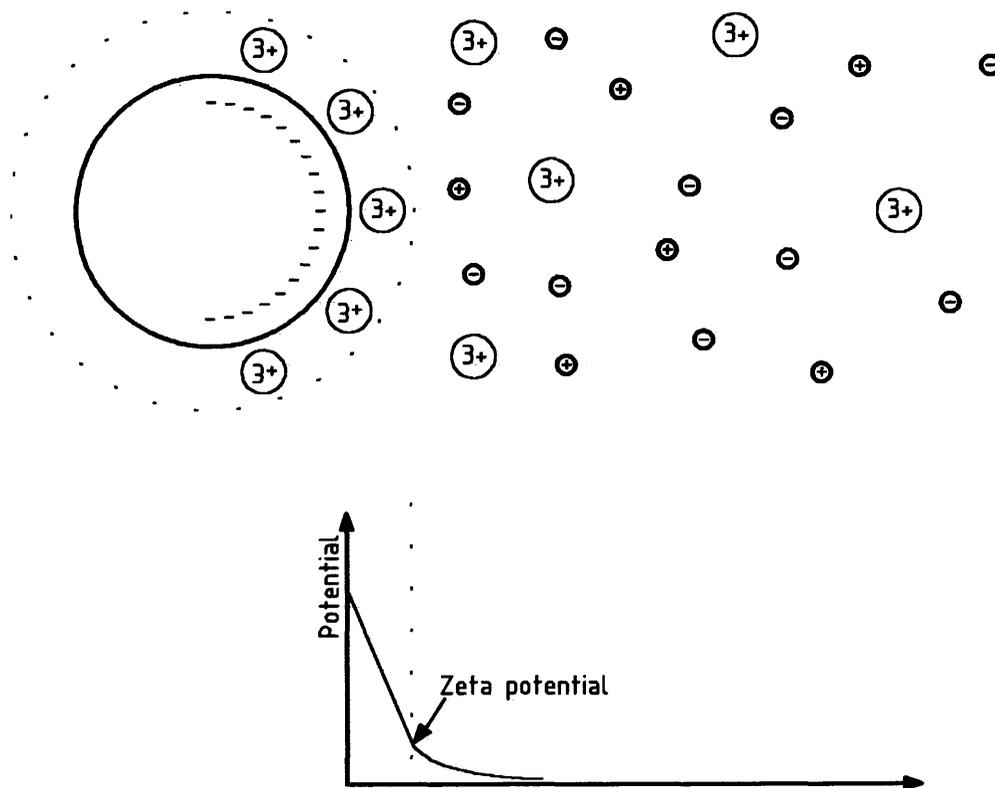
Figure 2.3 Reduction in zeta potential through addition of counterions of higher valence.

In this case charge reversal resulting in particle restabilisation cannot occur since electrostatic forces cannot attract more counterions into the diffuse layer than the magnitude of the primary charge.

ii) By the addition of counterions, or charged precipitates, that are specifically adsorbed on the surface of the particle. The effect of this is to reduce the surface charge enabling particle interaction. As specific adsorption can occur as a result of forces other than electrostatic attraction, e.g. van der Waals bonding, hydrogen bonding or some other specific chemical reaction, charge reversal and hence particle restabilisation can occur if an excess of counterions are specifically adsorbed. It must be noted that subsequent destabilisation of these particles can be brought about by an increase in the ionic strength of the surrounding solution as described in (i) above.

● **Inter-particle bridging**

It has long been recognised that polymers (long chain of repeating molecular units) are capable of destabilising colloid suspensions. The main theory put forward to explain this effect involves that of inter-particle bridging. In this model electric double layer effects play little part in the process. A simplified schematic of the mechanisms involved is shown in Figure 2.4.

(1) POLYMER + COLLOID ⟶ DESTABILISED COLLOID

(2) DESTABILISED COLLOIDS ⟶ FLOC

(3) DESTABILISED COLLOID SECONDARY ADSORBTION ⟶ RESTABILISED COLLOID

(4)
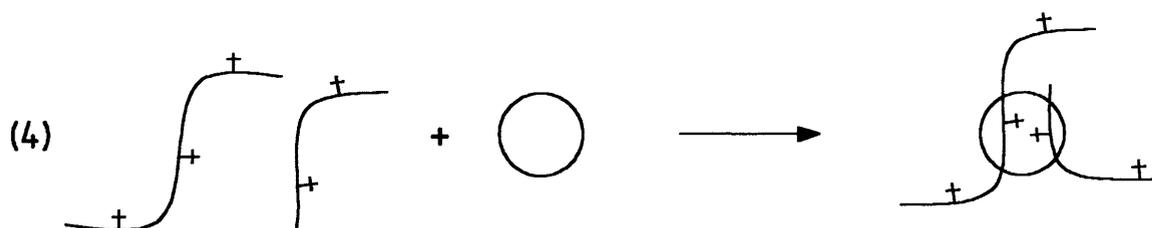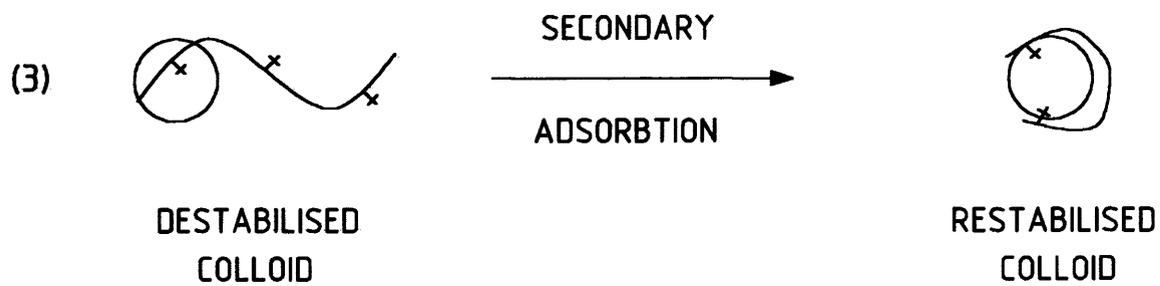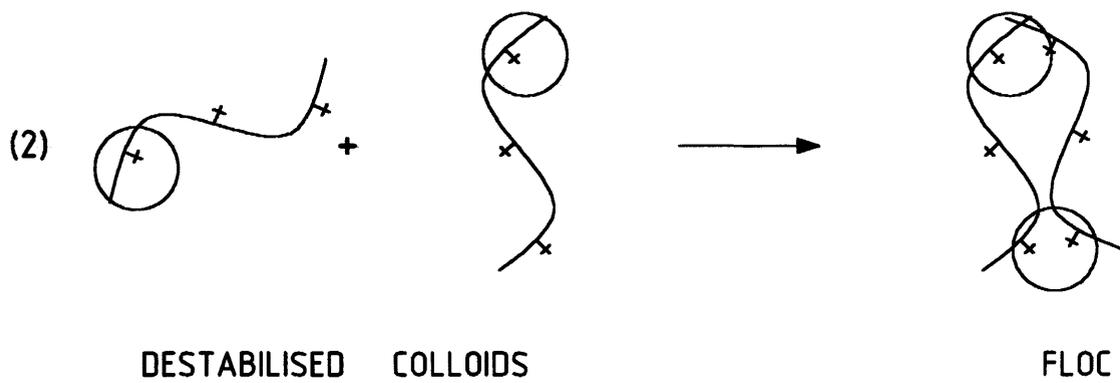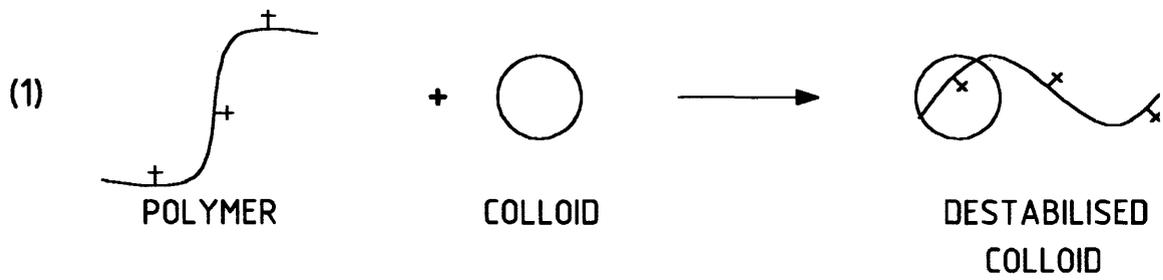
Figure 2.4 Colloid destabilisation through inter-particle bridging

In (1), polymer addition results in adsorption of polymer segment on to one or more sites on the surface of the colloid, the remaining sections of the chain extend out into the solution. Adsorption can result from a number of sources viz.: electrostatic attraction (which can involve suppression of the electric double layer), ion exchange, hydrogen or covalent bonding or van der Waals bonding. One or more of these mechanisms may be involved. In (2), as a result of particle collision the extended chain segments from one destabilised particle are adsorbed on the surface of a second forming a chemical bridge. Bridging between further particles results in the formation of floc. In (3), if no particle collision occurs it is possible for the extended segments of the polymer chain to fold over and be re-adsorbed at other sites on the particle surface. Since all sites are now occupied, segments from other polymer chains are unable to be adsorbed resulting in restabilisation of the particle. A similar effect is observed with an overdose of polymer, as seen in (4). Occupation of all adsorption sites by separate polymer chains results in a stabilised particle as no sites are available for polymer bridges.

The modes of particle destabilisation described above may be considered simplistic, however, they provide the basis for understanding the mechanisms by which chemical coagulants can effect particle removal in water treatment. Which mechanism predominates will be a function of the specific chemical coagulant used and the raw water quality i.e. pH, ionic content, levels of organic matter and the type and concentration of particles to be removed. The practical consideration of particle destabilisation in water treatment, with specific reference to the most commonly used chemical coagulants viz.: metal salts and polyelectrolytes, is addressed in the following section.

2.2.3 Particle destabilisation in water treatment - coagulation and flocculation

The terms coagulation and flocculation have often been used interchangeably in the water industry to describe the destabilisation process and floc formation. La Mer and Healey (1963) proposed the first formal definition of the two terms. Coagulation was defined as the general kinetic process whereby colloidal particles are united (latin 'coagulare' meaning 'to be driven together'). Coagulation being effected by the reduction of the repulsive potential of the electric double layer.

14

The term flocculation (latin 'flocculus' meaning 'a woolly or loosely fibrous structure') was to be restricted to the action of high molecular weight materials bridging between the solid particles to form three dimensional, loose and porous floc. In practice this would imply a process where aggregation by a simple soluble electrolyte would be termed coagulation, the subsequent aggregation of these by a water soluble polymer would be termed flocculation. These definitions do not however, describe the destabilisation and aggregation brought about by aluminium and iron salts, or polyelectrolytes when used as primary coagulants (this will become clear in the following sections on chemical treatment).

A more accepted definition that has been proposed, and one that will be used throughout this study, is that coagulation is the process by which particle repulsion is overcome. Flocculation is the process by which destabilised particles are brought together to form floc (Packham and Sheiham, 1977; Herman, 1983). Flocculation itself may be split into two further sub-processes viz.: perikinetic flocculation and orthokinetic flocculation. The former accounts for floc growth brought about by Brownian motion, i.e. the transport of colloidal particles resulting from bombardment by the random motion of water molecules. The latter involves particle transport and collisions caused by fluid motion and is generally characterised by the mean velocity gradient imparted to the fluid, G value, and the non-dimensional GT value (where T represents the retention time in seconds at this G value) as a criterion to express the extent of the flocculation process (Ives, 1984; Janssens and Buekens, 1987; Tambo, 1991)

2.2.3.1 Coagulation by metal ion salts

Aluminium and iron salts are the most widely used chemical coagulants in the world. The mechanisms by which particulate removal is effected using these coagulants, and the complex chemistry involved in the hydrolysing reactions that occur, have been the subject of extensive studies and are presented in detail elsewhere (Stumm and Morgan, 1962; Packham, 1965; Hanna and Rubin, 1970; Gregory, 1977; Packham and Sheiham, 1977; Rubin and Blocksidge, 1979; Dempsey, 1984; Jiang, 1998). Outlined below is a summary of the mechanisms involved, obtained

15

from the literature previously cited, and an explanation as to the practical use of the chemicals. For clarity the explanation will be limited to the case of the aluminium salts, however, in general the explanation applies equally to iron salts.

It would appear from the previous discussion of the effect of ion valence on the electric double layer (Schulze-Hardy rule), that the presence of the Al^{3+} ion alone could account for the coagulation ability. However, these species do not exist in an aqueous environment. In aqueous solution the aluminium ion is surrounded by six coordinated water molecules forming simple aquo complexes: $Al(H_2O)_6^{3+}$. The high charge on the central aluminium ion causes the O-H groups to polarise resulting in a tendency for the H+ to dissociate:

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

In alkaline waters the liberated acid is neutralised and the equilibrium shifts to the right. In the presence of sufficient alkalinity, hydrolysis continues as shown below:

$$[Al(H_2O)_6]^{3+} \leftrightarrow [Al(H_2O)_5OH]^{2+} \leftrightarrow [Al(H_2O)_4(OH)_2]^+ \leftrightarrow [Al(H_2O)_3(OH)_3](s)$$
$$\leftrightarrow [Al(H_2O)_5(OH)_4]^-$$
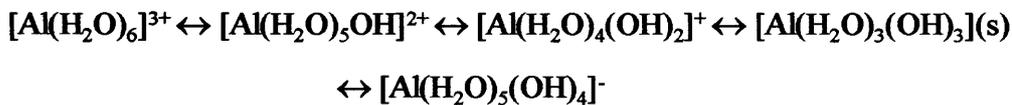
As pH increases, soluble metal hydroxo species are produced up to the point where the uncharged metal hydroxide is formed. Aluminium hydroxide is insoluble in water and produces large gelatinous precipitates. As the pH increases further the precipitate re-dissolves as the aluminate ion is formed. It can be seen that as hydrolysis proceeds from the simple aquo metal complex, the charge on the subsequent ion complexes is reduced. If the action of the complex ions were simply compression of the electric double layer, it would be expected that coagulation effectiveness would be reduced. However, this has been observed not to be the case. It has been demonstrated that these hydroxo metal complexes can be readily adsorbed on a wide variety of surfaces resulting in a reduction in surface charge and particle destabilisation. Excess adsorption can result in charge reversal (a mechanism not possible through direct compression of the double layer) and particle restablisation.

It must be noted that the reaction sequence described is highly simplified. Intermediate polynuclear complex ions have also been shown to exist which may also act as effective coagulants for many types of colloids.

Although it can be shown that the hydroxo metal complexes formed prior to the metal hydroxide can act effectively as coagulants, the practical application of aluminium salts in water treatment utilises the formation of the insoluble metal hydroxide in a process known as sweep flocculation. In this process coagulant is added in such quantities so as to exceed the solubility limit of the hydroxide. The hydroxide is initially precipitated as large numbers of colloidal particles. Perikinetic flocculation occurs quite rapidly increasing the size of the particles. As they grow, colloidal particles in the original suspension become trapped within the floc. Orthokinetic flocculation further increases the size of the floc. These can then be removed by a suitable solid-liquid separation process. The main considerations for optimum performance are the presence of sufficient alkalinity, to enable the hydrolysis reactions to occur up to the point of insolubility, and the maintenance of pH levels for precipitation (for alum pH range 6-8; iron salts pH range 5-8). In order to ensure effective precipitation conditions, raw water quality must be monitored and maintained, if necessary, through suitable chemical addition to control levels of alkalinity and pH. In addition, the production of the metal hydroxide reduces the pH of the treated water and may therefore require adjustment prior to the water entering the distribution system.

Despite the widespread use of aluminium salts in the water industry, concern has been expressed regarding the potential health risk associated from the long term ingestion of uncomplexed aluminium. The concern relates specifically to a potential link between aluminium and Alzheimer's disease (Martyn *et al.*, 1989). Although a direct link has yet to be proven, this is a factor that must be borne in mind when alternative coagulants are being considered for a particular source.

2.2.3.2 Polyelectrolytes and their applications in water treatment

The concept of particle destabilisation through polymer addition was introduced in section 2.2.2 where it was postulated that the mechanism by which destabilisation was brought about was

17

through polymer adsorption and inter-particle bridging. In recent years a number of investigators have begun to question whether this mechanism can be assumed for all polymers or whether alternative mechanisms exist that depend on polymer charge, charge density and molecular weight. The term polyelectrolyte has been introduced to describe polymers that have electrical charges (ionised sites) along its length. Depending on the nature of the charge, polyelectrolytes may be cationic (positive charge predominates) anionic (negative charge predominates) or nonionic (neutral). Technically speaking the term polyelectrolyte only applies to those polymers that are cationic or anionic, however, the term is frequently used to cover all polymeric coagulants. The mechanisms of action and the practical application of each type of polyelectrolyte will be reviewed separately.

- <u>Cationic polyelectrolytes</u>

The action of cationic polyelectrolytes was originally considered to be one of adsorption, through electrostatic attraction between positively charged groups of the polymer chain and the negatively charged particle surface, and bridging between particles (as described in section 2.2.2). However, this view is changing.

Gregory (1976) and Ghosh and Leu (1986) detail studies that have demonstrated that cationic polyelectrolytes tend to assume a flat configuration on particle surfaces as a combined result of the strong ionic interaction between oppositely charged groups on the particle surface and along the polymer chain, and the strong electrostatic repulsion between charged groups along the chain. This pattern of adsorption, through charge neutralisation, has been termed the patch mechanism. In such a configuration no loops or segments of polyelectrolyte will extend into solution to enable bridge formation. The patch mechanism is shown schematically in Figure 2.5.
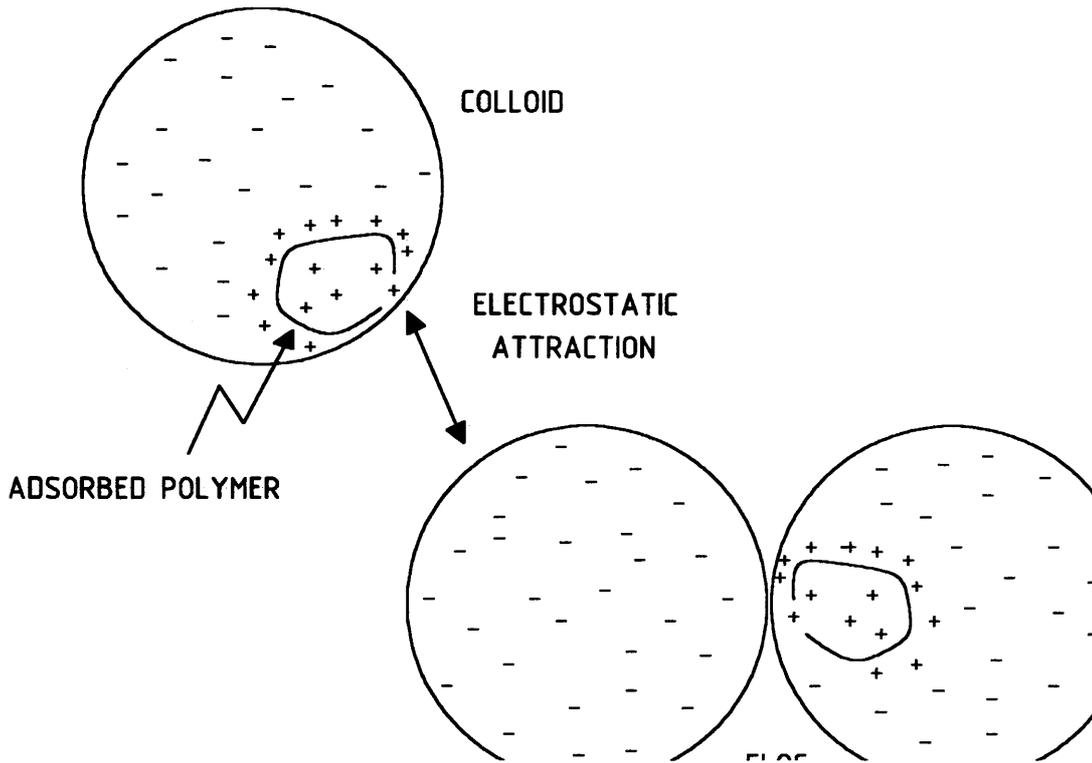
Figure 2.5 Schematic representation of the patch mechanism

The patch mechanism considers the localised adsorption of a polymer or polymeric species on to the surface of a particle thereby inducing a localised region of reduced negative, zero or even positive charge (the nature of the charge will be dependent on the relative size of the polymer and particle charges). Electrostatic attraction between this region of 'positive' charge and the negative site on a second particle leads to the formation of microfloc. Particle collisions induced through fluid agitation result in further agglomeration and the formation floc of increased size. The evidence for such a mechanism comes from a number of studies that have examined the effects of molecular weight and charge density of cationic polyelectrolytes on rates of flocculation. In the majority of cases charge density, not molecular weight, has been shown to be the governing factor, with the amount of positive charge adsorbed at the optimum dose being approximately the same irrespective of molecular weight (Gregory, 1973; Mabire et al., 1984; Ghosh and Leu, 1986; Gregory and Lee, 1990).

It is acknowledged that the patch mechanism cannot explain completely some of the observed behaviour of cationic polyelectrolytes and is postulated that there are two particular instances where the bridging model may still apply. Firstly, where the polyelectrolyte molecule is significantly larger than the particles to be removed. Secondly, when cationic polyelectrolytes are added to concentrated suspensions of colloidal particles. The frequency of particle collisions under such circumstances can lead to the attachment of polyelectrolyte segments on two or more particles, thereby forming a bridge, prior to achieving a flat adsorption configuration (Gregory, 1976).

The effectiveness of cationic polyelectrolytes, due to the opposite nature of their charge to the particles being removed, has lead to an increase in their use in water treatment as primary coagulants i.e. as direct replacements for the metal salt coagulants. Despite their higher cost, their main advantages are the small quantities used coupled with the production of a sludge that is less bulky and easier to dispose of (Montgomery, 1985, Kawamura, 1991).

- Anionic and nonionic polyelectrolytes

The mechanism by which both anionic and nonionic polyelectrolytes operate is generally accepted to be via adsorption and inter-particle bridging. With the effective charge on both the polyelectrolyte and particle being negative, a significant amount of work has been undertaken to determine how such adsorption can take place. Hydrogen bonding and anion exchange with adsorbed anions are possible explanations, however, it has been demonstrated that flocculation using negatively charge polyelectrolytes can only occur in the presence of a suitable electrolyte. It is postulated that the preferential adsorption of cations compresses the electric double layer of the particle thereby reducing the repulsive forces and enabling adsorption of polyelectrolyte segments. In addition, complexing between charged polymer sites and adsorbed cations, particularly with Ca^{2+} ions, results in the formation of strong particle-polymer bonds. It has also been demonstrated that, in the presence of sufficient cations, overdosing can lead to restabilisation with

the particle covered by a 'protective' anionic shell (Ruehrwein and Ward, 1952; Black *et al.*, 1965; Sommerauer *et al.*, 1968; Gregory, 1977).

Although anionic and non-ionic polyelectrolytes can operate as primary coagulants, they are significantly less effective than both the metal salts and cationic polyelectrolytes. Their main use in water treatment is as flocculant aids. Generally added subsequent to the primary coagulant, they are highly effective in forming bridges between previously destabilised particles producing stronger and faster settling floc.

Anionic and nonionic polyelectrolytes, when used as aids in water treatment, are frequently termed 'flocculants' since, within the accepted definition of flocculation described in section 2.2.3, they assist in the bringing together of destabilised particles. Cationic polyelectrolytes and the metal salts are generally termed coagulants since they act to destabilise particles.

- Utilisation of polyelectrolytes in the water industry - health concerns

The efficiency of polyelectrolytes, irrespective of type, in removing turbidity and organics from raw waters is such that their use is becoming increasingly more widespread. However, concern has been raised regarding the long term health effects of their use. Letterman and Pero (1990) conducted a literature and patent review that indicated many of the products used in the US water industry contain, in addition to the polyelectrolyte, measurable amounts of certain contaminants. The contaminants arose primarily from the production process and could be categorised as residual (unreacted) monomers, unreacted chemicals used to form the monomer units, and the products that transform or degrade residual monomer. Of particular concern were those that were produced from the polymerisation or co-polymerisation of acrylamide and epichlorohydrin (both have been shown to be an animal carcinogens and is a known neurotoxin). The potential health problems related to polyelectrolyte use in drinking water treatment has lead to a number of countries refusing permission for their use, for example Switzerland and Japan, and to a number of others establishing stringent limits on application rates, for example France, UK and US.

2.3 APPLICABILITY OF CHEMICAL TREATMENT TO DEVELOPING COUNTRIES

From the previous discussion of the requirement for chemical treatment, it is clear that the high raw water turbidities encountered in the majority of developing countries during the wet season necessitates the use of chemicals. However, although chemicals are in widespread use, particularly in urban water treatment systems, there are two main disadvantages to their use:

i) If manufacture within country is not possible, all chemicals (coagulants, disinfectants and solutions for water quality control e.g. pH, hardness) must be imported, often at high cost and involving the use of valuable foreign exchange. In comparison with many developed countries, the chemical costs involved in water treatment in developing countries form a major element of operational costs with, for example, salaries playing a much smaller part. Coupled with this is the reliance on foreign supplies. Political instability in neighbouring countries can result in the closure of cross border supply routes. Such a situation was observed by the author in Malawi during the time of the civil war in Mozambique. The closure of the southern supply routes meant imported alum from South Africa had to be brought in by a more circuitous route. The result was an increase in cost and a reduction in the quantity supplied;

ii) In order to maintain optimum performance of the coagulation process and hence the quality of the treated water, requires the attention of skilled operators. The general problem of a lack of trained operators is discussed elsewhere (Schulz and Okun, 1984), however, it is clear that without suitably trained personnel the complexity of the treatment process coupled with the potentially hazardous nature of many of the chemicals involved, make chemical treatment the least attractive and least appropriate treatment option particularly for small community water supplies.

The potential health risks associated with the use of aluminium sulphate and synthetic polyelectrolytes are, as indicated, a major concern in the developed world. However, it has been argued that in developing countries the lower life expectancy reduces the potential effects of any long term exposure. Although such an argument may have certain validity, such double standards should have no place in the provision of a safe water supply for communities in developing countries.

22

2.4 ALTERNATIVES TO CHEMICAL TREATMENT - NATURAL COAGULANTS

An alternative to the use of chemicals in developing countries that is gaining in interest is the use of natural coagulants. Although a wide variety of natural materials, ranging from 'clarifying' clays to the deacetylated exoskeletons of arthropods (chitosan), have been shown to be effective primary coagulants and coagulant aids, the discussion here will restrict itself to those of plant origin.

The use of plant materials to clarify turbid surface waters is not a new concept. Sanskrit writings in India dating from several centuries BC make reference to seeds of the tree *Strychnos potatorum* as a clarifier, Peruvian texts from the 16th and 17th centuries detail the use by sailors of powdered, roasted grains of *Zea mays* as a means of settling impurities. Chilean folklore texts from the 19th century refer to water clarification using the sap from the 'tuna' cactus, *Opuntia fiscus indica* (Jahn, 1981).

Although primarily used as flocculant aids, a number of plant extracts have been applied on a large scale. For example, sodium alginate (extracted from seaweed) has been used by a number of water authorities in the UK, starch derivatives (marketed under the trade name Wisprofloc) have been used in Germany and two cactus extracts (marketed under the trade names Tunafloc A and Tunafloc B) are currently being employed in Peru (Packham, 1967; Schulz and Okun, 1983).

Of all the plant materials that have been investigated over the years, seeds from trees of the family *Moringaceae* have been shown to be some of the most effective particularly when applied as primary coagulants (Jahn, 1981). The study being presented here focuses on coagulant extracts from seeds of probably the most well known tree in the family viz.: *Moringa oleifera* Lam. (*M.oleifera*). Although the coagulating properties of seeds from other members of the family have been demonstrated, most notably those from *Moringa stenopetala* Bak.f.cufod, the primary advantages of *M.oleifera* are its rapid growth rate and widespread cultivation throughout many developing countries.

Chapter 3 that follows provides a brief introduction to the tree, outlines its many current uses, details previous work on the coagulation properties of the seeds and puts into context the study presented here.

CHAPTER 3

MORINGA OLEIFERA Lam.

3.1 INTRODUCTION

The focus of this project as a whole is the application of seeds of *M.oleifera* as a natural coagulant for the treatment of surface waters in developing countries. However, *M.oleifera* is a truly multipurpose tree and as such its intrinsic value as a multiprovider will affect and potentially enhance its acceptance as a tree crop and hence increase the possibility of implementing the use of the seeds within water treatment practices. The multipurpose nature of the tree and the potential impacts of this on implementation within water treatment practices are discussed in more detail in Chapter 6. The following chapter, therefore, provides a general introduction to the tree and the many products and uses that may be obtained from it, a section that details activities that have already been carried out on the utilisation of the seeds within water treatment practices and a final section that defines the context of the study being presented in relation to these previous investigations.

3.2 GENERAL INFORMATION

The *Moringaceae* is a single genus family with 14 known species. Of these *Moringa oleifera* Lam. (syns. *Moringa pterygosperma* Gaertn.) is the most widely known and utilised species. A native of the sub-Himalayan regions of northwest India, *Moringa oleifera* (*M.oleifera*) is now indigenous to many countries in Africa, Arabia, South East Asia, the Pacific and Caribbean Islands and South America (Verdcourt, 1985). Commonly known as the 'horse-radish' tree (arising from the taste of a condiment prepared from the roots) or 'drumstick' tree (arising from the shape of the pods), *M.oleifera* has a host of other country specific vernacular names, an indication of the significance of the tree around the world.

25

As a result of the ubiquitous nature of the tree, an immense amount of botanical literature has been produced relating to its description and ecology. Only a brief summary will be presented here (for more detailed information the reader is directed to the selected references). The tree ranges in height from 5 - 12m with an open, umbrella shaped crown, straight trunk (10-30cm thick) with corky, whitish bark. The tree produces a tuberous tap root which helps explain its observed tolerance to drought conditions. The evergreen or deciduous foliage (depending on climate) has leaflets 1-2 cm in diameter, the flowers are pleasantly fragranced, white or cream coloured and are borne profusely in axilliary drooping panicles 10-25 cm long. The fruits (pods) are initially light green, slim and tender eventually becoming dark green and firm up to 120cm long depending on the variety. Fully mature, dried seeds are round or triangular shaped, the kernel surrounded by a lightly wooded shell with three papery wings (Morton, 1991; Booth *et al.*, 1988; Verdcourt, 1985; Ram, 1994; Ramachandran, 1980).

Originally considered a tree of hot, semi-arid regions (annual rainfall 250 - 1500mm) it has also been found to be well adapted to hot, humid, wet conditions with annual rainfall in excess of 3000mm (Ram, 1994). The tree was originally considered to be suitable only for lowland cultivation at altitudes less than 600m. However, the adaptability of the tree has been demonstrated by the discovery of natural stands in protected microclimates at altitudes of 1200m in Mexico (Jahn, 1986) and, recently observed by the author, in excess of 2000m in Zimbabwe.

M.oleifera can also grow in a variety of soil conditions. Although preferring well drained sandy or loamy soils, heavier clay soils will be tolerated although water logging should be avoided (Ram, 1994). The tree is reported to be tolerant to light frosts and can be established in slightly alkaline soils up to pH 9 (Valia *et al*, 1993).

As stated in the introduction, *M.oleifera* is a truly multipurpose tree with almost all parts of the tree utilised. Table 3.1 provides details of the principal uses.

Tree part and usage	Comment	Relevant literature
Roots - Production of a condiment similar to horseradish	Use gave rise to one of the more common vernacular names for *M.oleifera* - the horseradish tree.	Morton,1991
Leaves - Human nutrition	Contain significant quantities of calcium, protein (complete amino acid complement), iron, and vitamins A, B and C and are considered to offer great potential for those who are nutritionally at risk.	Ramachandran *et al.*,1980; D'souza and Kulkarni, 1993; Ram,1994
Leaves - Animal nutrition	Recent studies indicate the leaves to be an acceptable fodder crop for ruminants, pigs and an important browse crop for goats and sheep.	Makkar and Becker, 1996; Makkar and Becker, 1996; ECHO, 1995; Morton, 1991
Leaves - Growth hormones	The fast growing nature of the tree has lead to investigations into the potential utilisation of 'growth hormones' from the tree to promote growth in other plant species. Nodulation of *Vigna mungo* L. has been shown to increase vigorously with the application of an ethanol extract from *M.oleifera* leaves. A commercial enterprise in Nicaragua is currently utilising an ethanol extract on a large scale to promote growth in *Jatropha curcas*.	Bose, 1980; Makkar and Becker, 1996
Flowers - Human nutrition	The flowers, which must be cooked, are rich in potassium and calcium.	Busson,1965; Ram,1994
Seeds - Human nutrition	Utilised either as a green 'pea', in their immature state, or fried, in their mature state, possessing a peanut like flavour. Concern has been expressed that mutagenic compounds, arising from the thermal degradation of seed glucosinolates, may be formed during heating.	Jahn, 1989; Villasenor *et al.*, 1989
Immature green pods - Human nutrition	Probably the most valued and widely used of all the tree parts. The pods, generally prepared in a similar fashion to green beans and having a slight asparagus taste, contain all the essential amino acids. Primarily used around the world by the Asian population use by other peoples is increasing.	Ramachandran *et al.*, 1980

Seeds - Source of vegetable oil	Interest in the oil extracted from *M.oleifera*, known commercially as 'Ben' or 'Behen' oil, has existed for well over a century. The commercial name originates from the high levels of behenic acid in the oil. In addition the oil contains high levels of oleic, palmitic, stearic, eicosenoic and arachidic acids. The fatty acid composition is similar to that of olive oil. Seed kernels contain between 25 and 40% oil. Reported to have been previously used as a watch lubricant, an enfleurage in the cosmetic industry and as a lamp oil, it is now considered eminently suitable for edible purposes. The presscake obtained following oil extraction may be applied as a fertiliser. Its use as an animal feed is not recommended as it contains an alkaloid and a saponin.	Anon,1904; TEI, 1996; Jamieson, 1939; Morton 1991; NRI (1993); Ferrao & Ferrao (1970); Dahot & Memon (1985); TEI (1995); Raval and Toliwal, 1996; Booth and Wickens, 1988
Whole tree - Live fencing	Used as a live fence or windbreak. Normally grown from cuttings for this purpose the tree is quick to establish itself providing an effective barrier within a few months of planting.	
Whole tree - Medicinal purposes	Almost all parts of the tree have been utilised within traditional medicine. In recent years interest has grown in these medicinal properties and studies have been carried out on the chemistry and pharmacological applications of different parts of the tree.	Morton, 1991; Caceres *et al.* 1991; Caceres *et al.* 1992; Caceres and Lopez, 1991; Pal *et al.* 1995
Whole tree - Pulp and paper	The wood provides a pulp that is considered suitable for newsprint, wrapping, printing and writing papers and for viscose rayon grade pulp for textiles and cellophane.	Guha and Negi, 1965

Table 3.1 Principal uses and products of *M.oleifera*

A more detailed list of other, lesser uses may be found elsewhere (Booth and Wickens, 1988). It is clear from the table that the tree has enormous potential as a 'new' cash crop for a variety of purposes. The extent to which this potential may be realised within the context of the application of the seeds to water treatment practices is discussed further in chapter 6.

3.3 *M. OLEIFERA* IN WATER TREATMENT

The traditional use of the *M.oleifera* seeds for domestic household water treatment has been limited to the rural areas of parts of the Sudan. Having collected their water, powdered seeds, placed in a small cloth bag to which a thread is attached, would be swirled around in the turbid water. The flocculated solids would then be allowed to settle prior to boiling and subsequent consumption (Jahn, 1981).

3.3.1 Clarification studies and isolation of the active fraction

Since discovery of the traditional use of the seeds in the early 1970's, a number of studies have been carried out to evaluate the performance of the seeds in clarifying turbid raw waters and to determine the mechanisms of coagulant action. Although a lot of work has been undertaken on performance evaluation, it is difficult to cross correlate results from the different studies. Individual studies have either been conducted on very specific raw waters (Jahn and Dirar, 1979; Jahn, 1981; Jahn, 1984; Jahn ,1986; Muyibi and Okuofu, 1995) or synthetic test waters involving the use of either kaolin or bentonite clays to provide artificial levels of turbidity (Bhole and Shrivastava, 1983; Folkard *et al.* 1986; McConnachie, 1993; Ndabigengesere and Narasiah, 1996). In many cases, particularly those involved with the testing of specific raw waters, many water quality and test parameters are unstated making it difficult to fully interpret the results. However, the general findings of the many studies may be summarised:

i) For high raw water turbidities (generally considered >150 NTU) seed solutions are highly effective giving residual turbidity levels similar to or, in some cases, superior to those obtained using alum;

ii) For low to medium raw water turbidities (<150 NTU) the performance of seed solutions is significantly more variable with alum treatment far superior to that obtained using the seeds. This variability in performance is clearly a factor of the mechanism by which seed solutions are able to interact with suspended solids and is considered to be a function of raw water characteristics and the type and concentration of particulate material in suspension;

iii) Treatment performance is maintained irrespective of raw water pH and application has no effect on treated water pH.

In an attempt to understand the mechanisms by which seed solutions are able to clarify water, several investigations have been undertaken to isolate and characterise the active fraction contained within the seed kernels. It was initially postulated that the active materials were contained within the N-free fraction of the seed kernels i.e. the carbohydrate fraction (Jahn and Dirar, 1979). However, a study undertaken by Bart *et al.* (1982) identified 6 water soluble basic polypeptides (proteins or protein sub-units) with molecular weights in the range 6,000-16,000 daltons to be responsible for the observed activity. Subsequent investigations have analysed the isolated proteins using polyacrylamide gel electrophoresis techniques and have indicated that a number of the proteins are in fact hetero-dimers i.e. unique polypeptides varying in sequence by only a few amino acids, linked by covalent di-sulphide bonds. The significance of this fact in relation to their application to water treatment has yet to be determined. To date only two of the isolated proteins have been sequenced i.e. the position and nature of the amino acids has been determined. Comparison of the sequences obtained to known sequences stored in the European Molecular Biological Laboratory data bank have shown no significant homology. The proteins are not, therefore, considered to belong to any known protein family and as such new and unique (Gassenschmidt *et al.* 1995; BFE, 1997)

Several investigations have provided further general information on the protein characteristics viz.: confirmation of the molecular weight range and confirmation of the highly basic nature of the proteins with iso-electric points (that point at which the proteins carry a net neutral charge) determined to be between pH 10 and 11. The iso-electric points of the proteins provide the first indication as to a potential mechanism of action. At pH values below their iso-electric points (raw water pH values are generally between 6 and 8.5) the proteins carry a net positive charge. This, coupled with the fact that proteins are long chain molecules consisting of sequences of amino acids, has lead to the conclusion that the proteins act in a similar manner to synthetic cationic

polyelectrolytes i.e. either through the interparticle bridging or patch mechanisms detailed in Chapter 2 (Tauscher, 1994; Gassenschmidt *et al.* 1995; Ndabigengesere *et al.*, 1995).

An attempt to explain the reduced performance of the seeds at low turbidity, as indicated earlier in this section, has been given based on the assumed cationic polyelectrolyte mechanisms of action. The inter-particle bridging and patch mechanisms rely on particle collisions for effective floc formation. Low turbidity waters generally contain low levels of suspended particulates. Under such conditions, the opportunities for inter-particle interaction are greatly reduced thereby resulting in reduced floc formation and an adverse effect on treatment performance. This is reasonable explanation for the general behaviour of cationic polyelectrolytes on low turbidity waters, however, it does not explain the variability in performance achieved in studies carried out on different raw water samples at similar turbidity levels.

3.3.2 Treatment performance in relation to the microbiological content of raw waters

In Chapter 1 it was stated that the primary function of chemical treatment is to reduce raw water turbidity to such levels that enable effective disinfection of the treated water. The physical removal of floc formed during the chemical treatment process has the additional benefit of significantly reducing the microbiological content due to the close association of bacteria, viruses and protozoa to the inorganic turbidity causing particles. Several of the studies undertaken to evaluate the effectiveness of *M.oleifera* in clarifying natural raw waters also determined bacterial removals. In the majority of cases 90-99% reductions in total bacterial counts within two hours of treatment were recorded. However, in all cases it was also reported that regrowth of bacteria occurred to at or above original levels within a 12 hour period following treatment. Similar results were observed using alum as the coagulant although regrowth did not occur to the same extent. It has been postulated, therefore, that the addition of whole seed material to effect clarification has the disadvantage of supplying a suitable substrate that promotes bacterial regrowth (Jahn and Dirar, 1979; Jahn, 1984; Jahn, 1986). A subsequent study by Madsen *et al.* (1987) to determine whether the removal of bacteria by *M.oleifera* was strain specific proved inconclusive.

The reductions in bacterial concentration observed during the study by Jahn and Dirar (1979) led to an investigation to determine whether or not the seeds contained an antimicrobial agent. Although the premise for the investigation may have been mistaken (bacterial removals known to be a function of the turbidity removal process) the study did reveal the seeds to contain a potent antimicrobial compound, identified as the glucosidic mustard oil 4 (α-L-rhamnosyloxy) benzyl isothiocyanate. However, further studies have concluded that, over the range of seed doses generally applied in water treatment, the quantity of the antimicrobial compound present would not be sufficient to have any significant effect (Eilert et al., 1981; Dayrit et al., 1990).

In common with all coagulants used in water treatment, M.oleifera seed treatment can significantly reduce microbiological contamination, however, in order for the treated water to be termed potable a final disinfection process is still recommended.

3.3.3 Toxicity

A number of investigations have been undertaken to assess the risks to human health associated with the use of M.oleifera in water treatment. Barth et al. (1982) carried out a study to determine acute toxicity. Peroral applications of ground seed suspended in olive oil to laboratory rats at concentrations of 5g/kg showed no toxic effects. In subsequent studies by Berger et al. (1984) using similar procedures, no sub-acute toxic effects could be observed in rats receiving doses of 50 and 500mg/kg per day over a six week period. Similarly, no chronic effects were observed in rats dose with 1.5 or 15 g/kg per day over the same period. Those effects that were observed were considered to be caused by the olive oil vehicle used for dose application as similar effects were seen in the control samples receiving solely olive oil.

A more recent study by Grant et al. (1995) contradicts these first studies. In an investigation to determine the potential toxic effects of a variety of seed lectins it was found that ".....M.oleifera also had a high oral toxicity" but concluded that the toxicity was not due to the lectins but some other unknown substance. A criticism of the study is that no definition of toxicity is provided. In the investigation, rats were fed ground seed powder as part of an iosenergetic diet. In addition to

recording mortality rates, the effects on food intake and weight gain or loss were also determined. In the case of M.oleifera no deaths were recorded but a reduced food intake (19g/day as compared to the control 30g/day) and marginal weight loss (0.1g/day as compared to the control of a weight gain of 11g/day) were observed. It is argued that does not indicate a toxic effect. It is possible that the rats did not like the taste of the diet that included *M.oleifera*, the seeds have been reported to be very bitter when eaten raw. The results provided by Barth *et al.* (1982) and Berger *et al.* (1984) are considered to be more realistic as the rats received the seed irrespective of whether they liked it or not.

Grabow *et al.* (1985) carried out a study to determine the toxic and mutanegenic effects of water coagulated with *M.oleifera*. Toxic effects were observed on fish (guppies - *Poecilia reticulata*), protozoa (*Tetrahymena pyriformis*) and bacteria (*Eschericia coli*). The toxic effect observed was considered to be due to the anti-microbial compound 4 (α-L-rhamnosyloxy) benzyl isothiocyanate discussed in section 3.3.2. No mutanegenic effects were observed in the Ames *Salmonella* mutanegenicity assays with doses up to 1000mg/l of *M.oleifera*. The Ames *Salmonella* mutanegenicity assay provides indication as to possible carcinogenic effects. The authors concluded that "In view of complex associations of the toxin with organic matter, instability and the mechanism of action, it would not appear to constitute a human health risk....in the concentrations generally involved in the utilisation of the seeds for nutritional, medicinal or water purification processes".

3.3.4 *M.oleifera* product utilisation in associated water treatment applications

• **Seed shells as an activated carbon**

The use of the seed kernel as coagulant material requires the removal of the shell. The utilisation of the shell as a potential by-product was the focus of a study by Pollard *et al.* (1995). The study demonstrated that carbonisation of the shells under nitrogen gas followed by steam activation at 850°C produced a highly microporous carbon with adsorptive properties similar to commercial

activated carbons. Further work to reduce the complexity of the carbonisation process making it more appropriate to developing countries is being undertaken (McConnachie *et al.*, 1996).

- *M.oleifera* seeds for water softening

A study carried out by Muyibi and Evison (1995) using a synthetic hard water (distilled water spiked with calcium chloride), a surface water and two groundwater sources demonstrated that seed solutions could significantly reduce hardness levels. The whole seed doses required to achieve these reductions ranged from 950 to 2400mg/l. Comparing these doses to those commonly applied for clarification purposes i.e. 100 to 250 mg/l for high turbidity waters, and one has to question the viability of the treatment. In addition no comment is made regarding the introduction of such high levels of organic material and the quantities and constituents of the organic material that may be left in suspension. On a positive note, the interaction between the proteins, assuming the proteins are responsible for the hardness reduction, and the magnesium and calcium ions primarily responsible for hard waters may indicate a potential use for the proteins in the specific recovery or removal of other metals. This is an area that requires further work.

3.4 THE CURRENT STUDY IN CONTEXT

It is clear from the previous sections that a significant amount of work has already been done on many aspects of the application of *M.oleifera* seeds as an alternative to the chemical treatment of surface waters. However, there are significant gaps in the current state of knowledge regarding the mechanisms of protein activity and the practical application of seed treatment. The study objectives and the context within which they were formed may be summarised:

- **To further current knowledge on the mechanisms of action of the active coagulant fraction of the seeds.**

Although a basic understanding of the mechanisms of activity has been established i.e. that activity is via the bridging or patch mechanism, no attempt has been made to fully explain the variable

performance of seed solutions at low to medium levels of turbidity. In an attempt to expand on current knowledge, this study has focused on the examination of seed solution performance on 4 surface water sources from separate locations in Malawi and Zimbabwe. By evaluating performance under differing seasonal conditions and by monitoring a range of raw water quality parameters it was considered that a more in depth knowledge of the mechanisms of activity would be gained. The results of this study are detailed and discussed in Chapter 4.

- **To evaluate treatment performance on continuous flow systems and to determine the feasibility of such application as related to seed processing.**

The primary objective of the majority of studies that have been undertaken has been the application of seed treatment to batch household water treatment practices. Although a potentially viable option, implementation at this scale is highly complex task and one that is preferably carried out in-country. As such it was considered that the utilisation of the seeds within community or urban water treatment systems offered the greatest potential for acceptance and implementation provided that the feasibility of such an application could be demonstrated. A comprehensive study at pilot and full scale was planned in order to evaluate:

i) whether treatment performance observed in batch studies could be maintained in continuous flow systems - floc settlement under quiescent batch conditions does not necessarily infer that settlement will be achieved with fluid motion;

ii) whether any significant modifications i.e. increased flocculation times, extended settlement retention times etc., would be required to existing treatment works to enable seed treatment to operate effectively - any such major capital alterations may negate possible savings made in the utilisation of a potentially cheaper coagulant material;

iii) whether treatment performance would be adversely affected by extreme climatic conditions prevalent in many developing countries and

iv) whether problems with seed processing and seed solution preparation would severely limit the scale of application - preparation of a few grams of material for the batch treatment of, say, 40

35

litres is not a difficult task, however, processing of several tonnes of material may prove to be highly complex.

The pilot and full scale trials presented in Chapter 5 and the discussion presented in Chapter 6 based primarily on these trials represent the first and most rigorous attempt to determine the feasibility of applying *M.oleifera* as an alternative chemical coagulant within continuous flow systems.

CHAPTER 4

LABORATORY STUDIES TO DETERMINE *M.OLEIFERA* TREATMENT PERFORMANCE AND MECHANISM OF ACTION ON A RANGE OF SURFACE WATER SOURCES

4.1 INTRODUCTION

The preparation of a test water that can be used to model 'real' conditions is almost impossible as all natural raw waters vary in both chemical constituents and solids load concentration and load type. The use of kaolin or montmorillonite clay suspensions have been and are still used to model real waters, however, their use is limited primarily to the establishment of comparative performance of a range of coagulant materials under very specific conditions, the laboratory evaluation of treatment processes and, to a lesser extent, the 'microscopic' evaluation of coagulant mechanisms of action (for example, Ruehrwein and Ward, 1952; Black *et al*, 1965; McCooke and West, 1978; Gregory, 1988). The complex interrelationships that exist between coagulant and the constituents of the water to be treated mean that model waters cannot be used to predict dose response nor treatment performance on specific raw waters. As such the two studies presented here focus solely on the application of *M.oleifera* to natural surface waters. The argument that, because all raw waters vary in quality, such a study will only give an indication as to effectiveness on the specific raw water tested is valid, however, by choosing a number of different sources and by investigating them over a range of conditions an attempt has been made to provide sufficient data to enable conclusions as to how raw water quality affects treatment performance to be drawn. It was considered that this information combined with detailed observations of floc formation rates, floc size and shape and settling behaviour would enable a better understanding of the mechanisms of action of the active proteins contained within the seeds to be gained.

The first study, carried out in Malawi over a 5 month period, was designed to cover both dry and wet season conditions thereby enabling a variety of raw water qualities to be investigated. Three

rivers with significantly different catchment areas and flow characteristics were chosen to provide a cross section of the type of raw water conditions that *M.oleifera* would be expected to perform on.

Subsequent to these studies the opportunity arose to carry out a series of experiments on a eutrophic water, a type of water not previously investigated in any study using *M.oleifera*. The source water concerned was Lake Chivero, a reservoir that feeds the Morton Jaffray Water Treatment Works serving Harare, the capital city of Zimbabwe. Eutrophic waters are generally characterised by low turbidity, low suspended solids and high levels of organic materials. Such characteristics were in complete contrast to those of open flowing river courses experienced in Malawi and provided an opportunity to extend the knowledge available on factors that could affect the performance of *M.oleifera*.

The two studies were carried out under very different conditions and hence, for clarity, are presented separately in sections 4.2 and 4.3 respectively. A general discussion that attempts to relate observations made during both studies to potential mechanisms of protein activity is presented in section 4.4. Section 5 presents a summary of the conclusions from the studies.

4.2 PERFORMANCE OF *M.OLEIFERA* ON RIVER WATERS IN MALAWI

4.2.1 Sample sites

The main criteria for choice of sampling site were that the rivers and catchment areas should be as different from each other as possible and that the travelling distance to the site from the laboratory in Blantyre should be kept to a minimum. Following discussions with the appropriate authorities the sites chosen were at the Thyolo and Mwanza Water Treatment Works (under the control of the Ministry of Works and Supplies Water Department) and at the Walkers Ferry Treatment Works (under the control of Blantyre Water Board [BWB]). Sampling at the Thyolo and Mwanza sites was carried out by the author. BWB agreed to collect and deliver samples from the Walkers Ferry site once a week since the site could only be reasonably accessed by a 4-wheel drive vehicle.

Figure 4.1 shows the locations of the sites. The following paragraphs provide a brief description of the sampling sites.

Figure 4.1 Field study sampling sites

i) Thyolo

Thyolo is a fairly large district and main market centre situated 30 kms south south east of Blantyre in the Thyolo highlands. At an elevation of approximately 900m the surrounding landscape exhibits considerable but uniform fluctuations in elevation. Thyolo itself is the centre of the main tea growing region in Malawi. The urban centre of the town is supplied by water from a conventional treatment works, installed in the 1960's, situated on the river Nswadzi approximately 3 km from the town centre. The river has its headwaters in the Thyolo mountain forest reserve and is fed by a large number of small tributaries that draw water from a considerable catchment area. As a result, during the wet season the river is frequently subjected to flash floods producing raw water turbidities that can exceed 6000 NTU.

ii) Mwanza

Mwanza is a district centre situated approximately 60kms west north west of Blantyre. The surrounding area is relatively sparsely populated with the main land use being subsistence farming. Recent years have seen the growth of coffee estates although only to a limited extent. The urban centre is supplied by water from two conventional treatment works situated on the Mwanza river. The river drains a hilly catchment area of approximately 87 km2 which receives an annual rainfall of 1100 - 1200mm. The response of the river during the wet season is similar to that at Thyolo with flash floods occurring frequently, however, the nature of the land use in Mwanza i.e. less intensively farmed, means that river turbidities rarely achieve those experienced at Thyolo.

iii) Walkers Ferry

The Walkers Ferry treatment plant is located on the Shire river approximately 40 km northwest of Blantyre. The treated water from the plant is pumped to Blantyre to supplement that produced by the main BWB works on the river Mudi in Blantyre itself. The Shire River is the main outlet from Lake Malawi and eventually joins up the Zambezi River. The river is large and relatively slow

moving with the result that, although floods are a regular occurrence during the wet season, the high turbidities experienced at Thyolo and Mwanza are rarely encountered.

4.2.2 Experimental procedure

- **Sample collection**

40 litre samples were collected from each water source once a week. Turbidity analysis of samples was carried out on-site in order to determine the extent of settlement that occurred during transport.

- **Coagulant solution preparation**

Seed supplies for the study were collected from two mature *M.oleifera* trees growing outside the main hall of the University of Malawi in Zomba approximately 80km from Blantyre. Care was taken to ensure that all seeds were taken from pods that were completely dry. Individual seed suspensions were prepared for each jar within a series of tests by crushing the required amount of seed in a pestle and mortar, then diluting to the required concentration using distilled water. Seed suspensions were normally prepared at a 1:100 weight to volume ratio of coagulant to water (although not strictly a concentration for ease of presentation throughout this and subsequent sections this will be referred to as a 1% concentration). In hindsight this method of seed preparation was time consuming and did not take into account possible variations in individual seed protein content. For jar tests carried out subsequent to this study, stock concentrations of seed were prepared from which the required dose was taken. This method requires less preparation time and reduces the effect of variations in seed protein content.

Alum solutions were prepared at a 1% concentration by dissolving the required amount of commercial grade aluminium sulphate (obtained from BWB) in distilled water.

- **Jar test procedures and water quality analyses**

Investigations of treatment performance were undertaken using the conventional jar test. The jar test has a long history of use in the water industry and is generally accepted as being the most convenient method of determining optimum dose for a particular raw water under specific

41

hydraulic conditions. The standard laboratory apparatus consists of 4 or 6 stirrers connected to a single variable speed motor. Utilising sample volumes of between 500ml and 2 litres, the apparatus should allow for simultaneous coagulant addition to all samples. In the practical evaluation of optimum dose for a specific treatment works, it is recommended that the prevailing hydraulic conditions in the works i.e. mixing intensities and durations and settling times, should be utilised within the test. However, with respect to the sole determination of coagulant effectiveness there are no standard procedures although a number of investigators have made procedural recommendations (Dentel, 1991; Kawamura, 1991).

In the absence of a standard laboratory jar test device, investigations in this study were carried out using a portable 4 stirrer apparatus (Aztec Environmental Limited) and 1 litre sample volumes. Jar test parameters were as used in previous studies at Leicester University (Sutherland, 1986):

Rapid mix at 200 rpm for 2 minutes for coagulant dispersion;

Slow mix at 20 rpm for 20 minutes to enable flocculation;

Quiescent settling for 2 hours.

Prior to each set of experiments, the following basic water quality parameters were monitored: Turbidity (Hach Portalab Turbidimeter, model 16800, turbidity expressed as nephelometric turbidity units [NTU]); pH (Jenway portable pH meter); Conductivity (Jenway portable combined conductivity/temperature meter, conductivity expressed as $\mu S/cm$); Alkalinity (Palintest tablet method, expressed as mg/l $CaCO_3$); Total, Calcium and Magnesium hardness (Palintest tablet method, expressed as mg/l $CaCO_3$).

Treatment performance was established on the basis of residual turbidity following the 2 hour quiescent settling period. Samples for turbidity analysis were taken from the centre of the jar 2 cm below the water surface.

It is acknowledged that water temperature can affect coagulant and treatment performance (Kawamura, 1991), however, given that no control could be exerted over this parameter, a detailed investigation of temperature effects was not included in the study.

4.2.3 Results

For ease of presentation the raw water turbidities encountered at the three sample sites have been classified as low, medium or high. The classification used viz.: low - up to 50 NTU; medium - 50 to 150 NTU; high - > 150 NTU, is based on the author's observations of typical seasonal turbidity values experienced at the Thyolo site during this and subsequent field studies.

Table 4.1 presents the range of raw water characteristics experienced at the three sites over the period of the study.

	Thyolo	Mwanza	Walkers Ferry
Turbidity NTU	13 - 1450	24 - 1800	5.5 - 180
pH	7.4 - 7.9	7.3 - 8.1	8.0 - 8.4
Conductivity μS/cm	57 - 155	83 - 159	159 - 273
Alkalinity mg/l as $CaCO_3$	15 - 70	30 - 70	50 - 110
Total hardness mg/l as $CaCO_3$	15 - 55	25 - 60	45 – 75
Calcium hardness mg/l as $CaCO_3$	15 - 50	25 - 60	45 – 70
Magnesium hardness mg/l as $CaCO_3$	0 - 10	0 - 10	0 – 20

Table 4.1 Raw water characteristics of the three sources investigated

- **Low Turbidity (< 50 NTU)**

Figure 4.2 shows results obtained from two low turbidity samples from Thyolo following treatment with *M.oleifera*. In both cases floc formation was evident, however, the floc formed were small, compact and light resulting in large numbers still in suspension even after the two hour settling period. Figure 4.3 shows a comparative test carried out with alum. The figure demonstrates clearly the advantage of the heavier aluminium hydroxide floc with turbidity removals significantly greater than those obtained for *M.oleifera*. As turbidity increases, an improved performance from *M.oleifera* is observed. Figure 4.4 shows a comparative

alum/*M.oleifera* test carried out at a slightly higher initial turbidity. Although alum still outperforms *M.oleifera* the improved treatment performance of *M.oleifera* can be clearly seen.

Figure 4.2 Thyolo - *M.oleifera* low turbidity dose response
(Initial turbidity of sample as indicated in legend)

Figure 4.3 Thyolo - comparative low turbidity dose response
Initial turbidity 18 NTU

Figure 4.4 Thyolo - Improved *M.oleifera* performance at low turbidity
Initial turbidity 25 NTU
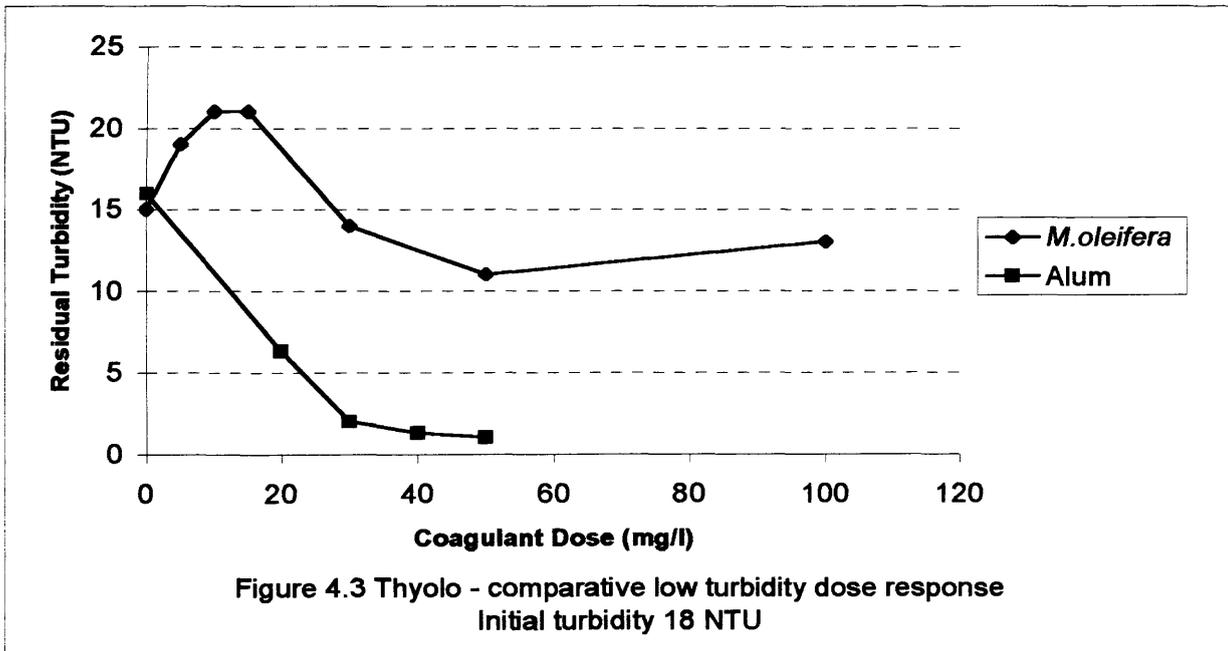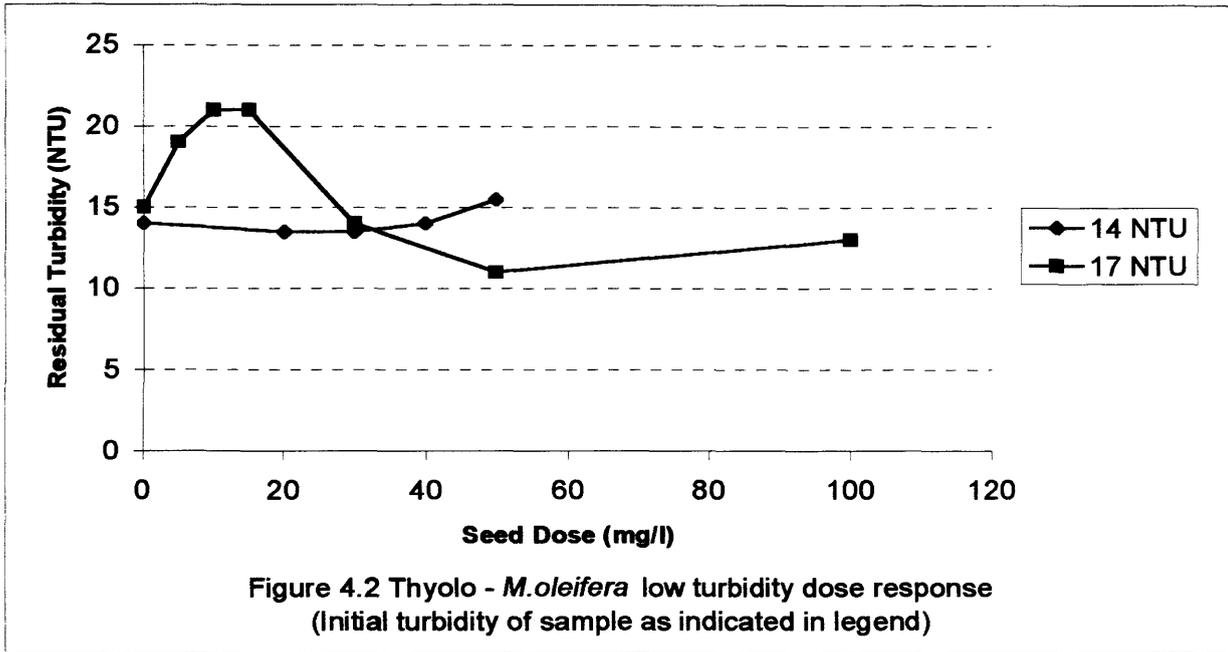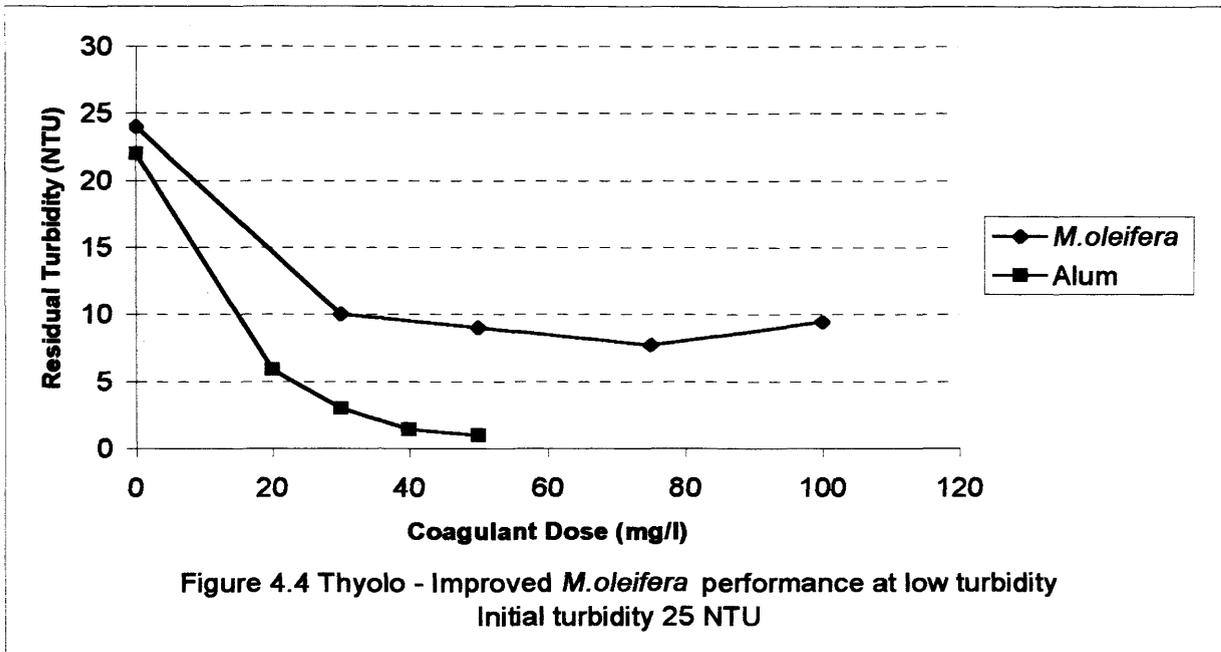
Figure 4.5 shows *M.oleifera* treatment performance on three separate low turbidity samples from the Mwanza sampling site. As for the Thyolo samples treatment performance improves with increasing turbidity, however, in contrast to the Thyolo samples, residual turbidities are far higher for an equivalent initial turbidity. As for the low turbidity samples encountered at Thyolo, floc formation was evident at all doses, however, the flocs formed were small and light showing little tendency to settle out. Indeed, it can be seen that for the initial turbidity of 24 NTU, as a result of seed addition turbidities subsequent to treatment have increased over the range of seed doses examined. It must be noted that the turbidity increase is not the result of the addition of solids from the seed solution, rather the change in the nature of the particles in the water from discrete to flocculated changes the refractive behaviour of the light passing through the water. Turbidity measurement, as used in this study, based on the refractive behaviour of particulates is a function of the particulate characteristics. A change in particulate characteristics can give distorted readings, such as in this case where a rise in turbidity is observed with no addition of material. The results must therefore be taken as an indication of minimal settlement.

45
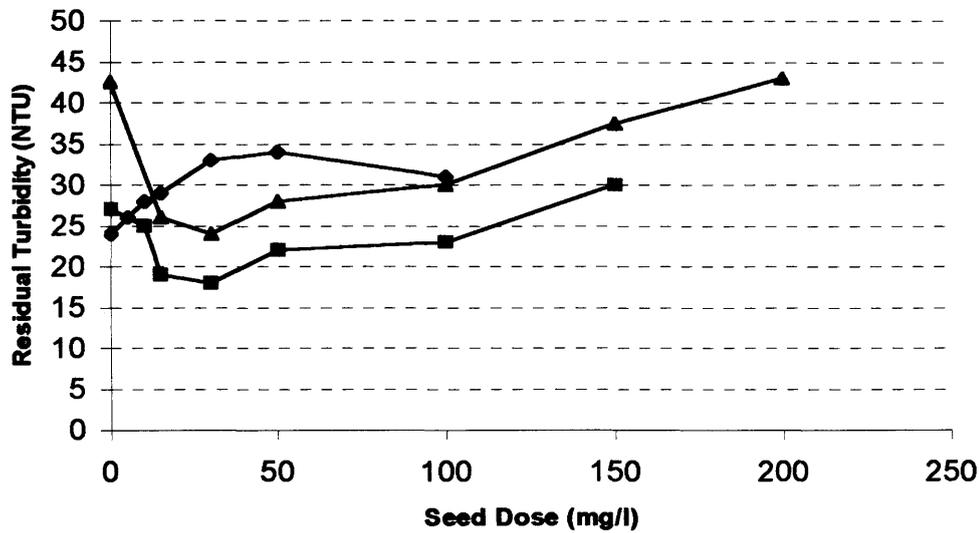
Figure 4.5 Mwanza - *M.oleifera* low turbidity dose response
(Initial turbidity of sample as indicated in legend)

In contrast to both the Thyolo and Mwanza sites the results and observations provided by the Shire river samples from Walkers Ferry demonstrate a clear effect of how variations in source water characteristics can affect coagulant performance. Figure 4.6 shows results obtained from two low turbidity samples. It can be seen that even at the low initial turbidities encountered viz.: 8 and 13 NTU, effective treatment can be achieved. The floc that formed were, in contrast to Thyolo and Mwanza, large, woolly and fast settling with complete clarification being obtained within the two hour settling period. A further observation was that with increasing dose floc size decreased while the number of floc increased. Figure 4.7 provides comparative data between alum and *M.oleifera* on a low turbidity sample. Although alum still outperforms *M.oleifera* the difference is less significant than that shown for Thyolo in figure 4.4.

Figure 4.6 Walkers Ferry - *M.oleifera* low turbidity dose response
(Initial turbidity of sample as indicated in legend)

Figure 4.7 Walkers Ferry - Comparative low turbidity dose response
Initial turbidity 18 NTU
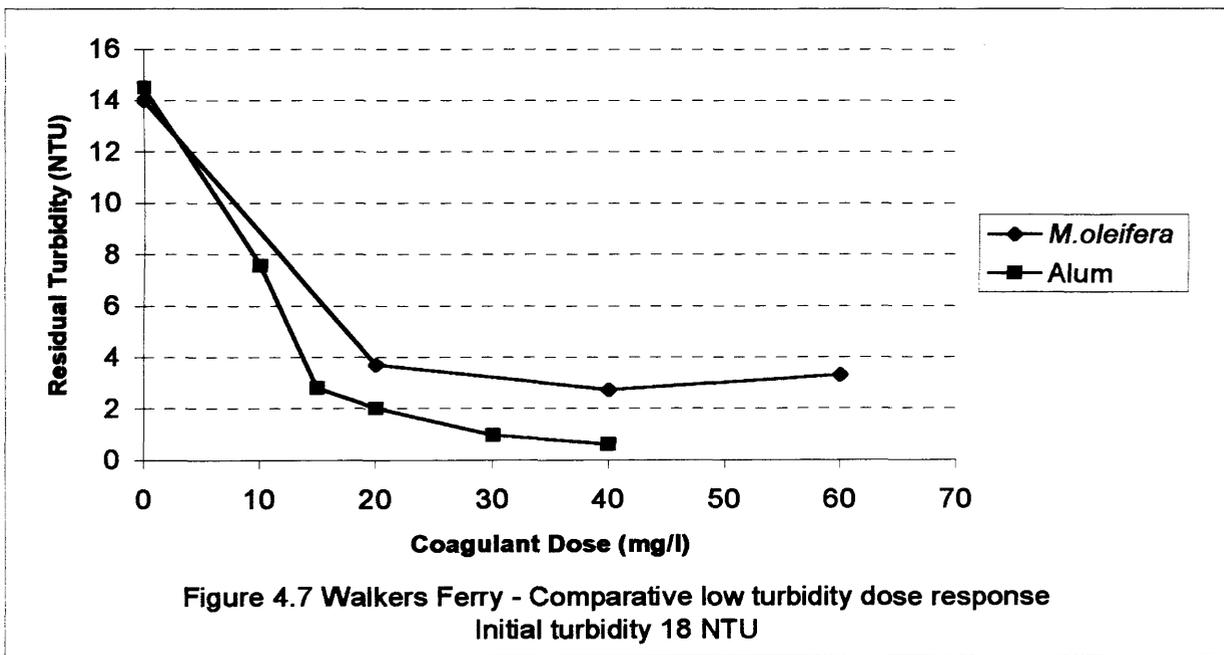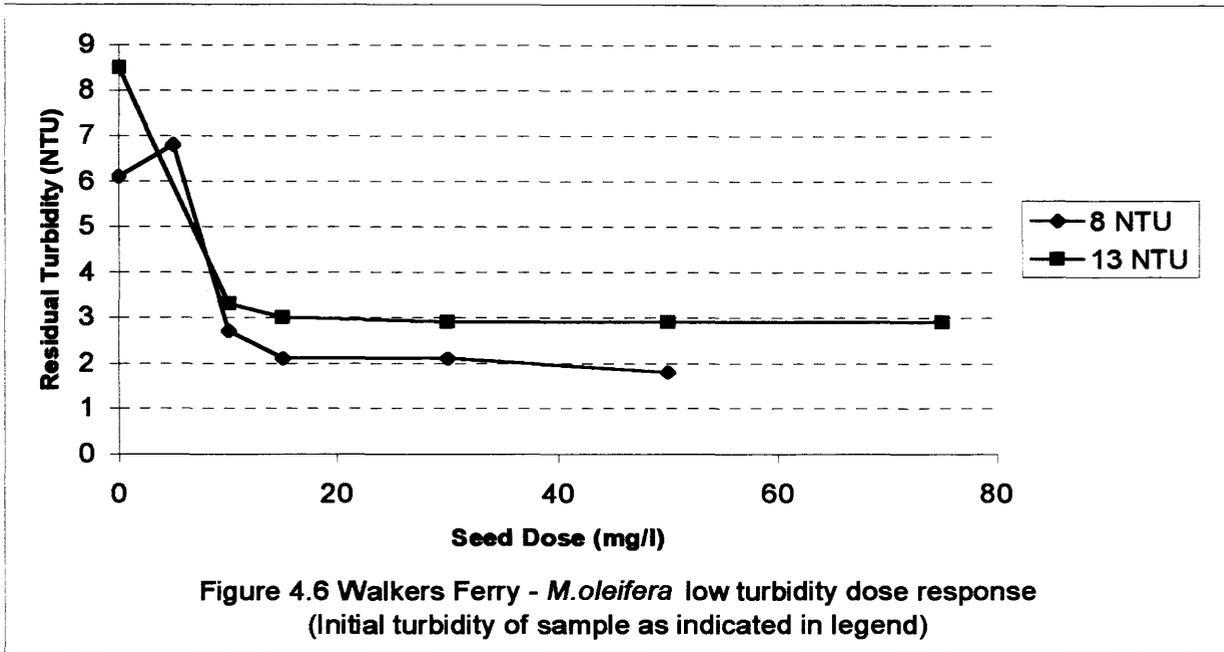
- **Medium Turbidity (50 - 150 NTU)**

Figure 4.8 shows results obtained from the treatment of a sample of medium turbidity from the Thyolo site. Two important points to note are firstly, the equivalent treatment performance

achieved by M.oleifera, and secondly, the sharp increase in residual turbidity at alum doses above the optimum. This is considered to be due to a combination of insufficient natural alkalinity to enable effective formation of the insoluble hydroxide and a reduced buffering capacity of the raw water resulting in a drop in pH to below that necessary for hydroxide formation (as discussed in Chapter 2). The absence of this effect in other tests on the Thyolo source using alum is considered to be due to a temporary change in the chemical quality of the river water around the time of sampling. The fact that no effect is observed with M.oleifera demonstrates clearly one of the primary advantages of M.oleifera treatment.

No samples from the Mwanza sampling site were received during the study period that fitted the medium turbidity classification.

Figure 4.9 shows comparative alum/M.oleifera results obtained from a Walkers Ferry sample. Equivalent performance is achieved across the range of doses examined. In addition floc size and settling characteristics were observed to be similar.

Figure 4.8 Thyolo - Comparative medium turbidity dose response
Initial turbidity 145 NTU

Figure 4.9 Walkers Ferry - Comparative medium turbidity dose response
Initial turbidity 150 NTU
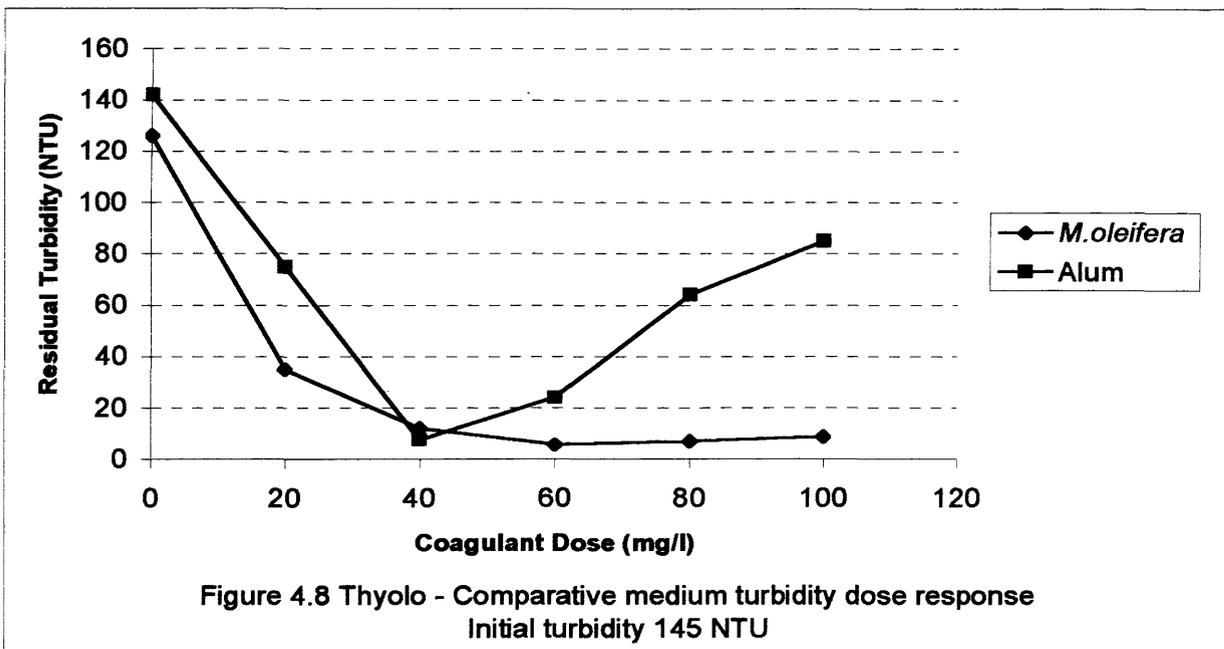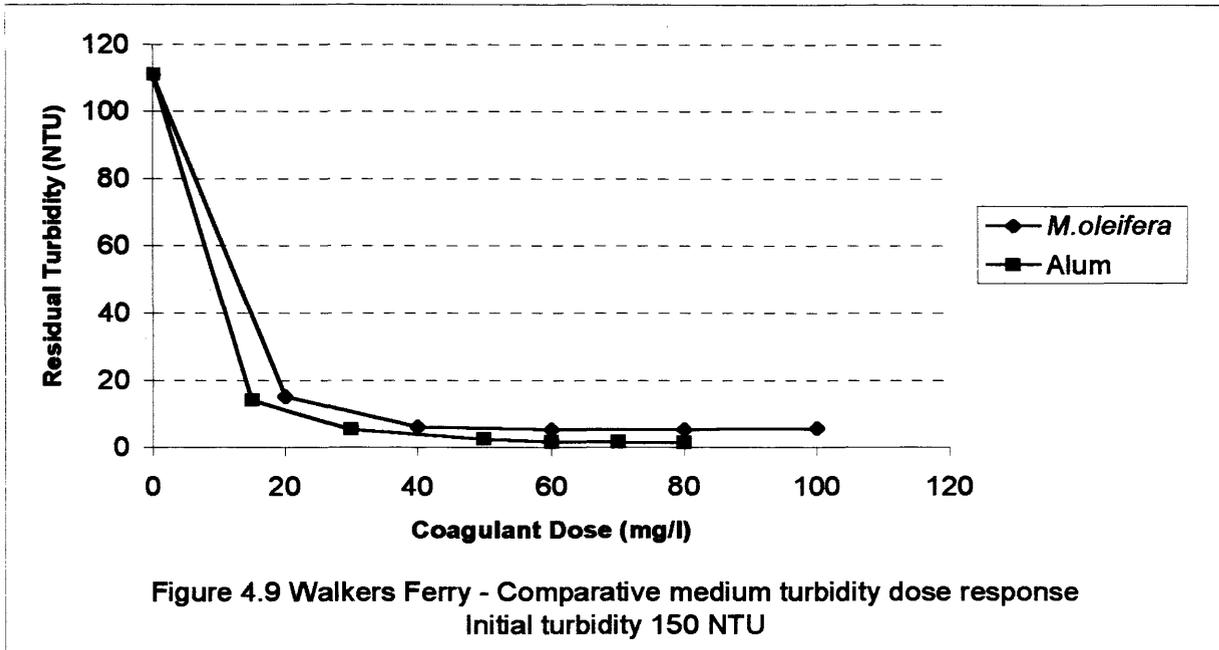
- **High Turbidity (> 150NTU)**

Figures 4.10, 4.11 and 4.12 show comparative alum/*M.oleifera* performance data from Thyolo and Mwanza. It can be seen that equivalent performance is achieved over the range of doses examined. In addition, Figure 4.11 shows a similar alum dose response to that observed on the medium turbidity sample from Thyolo with residual turbidities increasing sharply above the optimum dose. Again this is considered to be due to insufficient alkalinity resulting in incomplete hydroxide formation at the higher doses. Confirmation of this fact may be obtained by considering, at a very basic level, the stoichiometric hydrolysis of aluminium ions:

$$Al^{3+} + 3H_2O \Leftrightarrow Al(OH)_{3(s)} + 3H^+$$
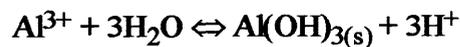
The overall reaction with water would indicate that each mole of trivalent ion should produce 1 mole of the metal hydroxide and 3 moles of hydrogen ion if the reaction goes to completion. On this basis it is estimated that 1mg of alum will consume approximately 0.5mg of alkalinity (Montgomery, 1985). The sample shown in Figure 4.11 had a raw water alkalinity of 30mg/l. At the optimum dose for alum of 40mg/l, approximately 20mg of alkalinity will be consumed. This is less than the raw water value so it may be assumed that the reaction goes to completion. At a

49

dose of 60mg/l approximately 30mg of alkalinity will be consumed, as such it would be questionable whether the hydrolysis reaction will go to completion. A dose of 80mg/l will potentially result in partial hydrolysis formation, hence the turbidity reduction observed, however the products formed are clearly less effective than those formed at the optimum dose. As for the Thyolo sample, *M.oleifera* performance is unaffected by the lower alkalinity levels.

Figure 4.10 Thyolo - Comparative high turbidity dose response
Initial turbidity 1400 NTU

Figure 4.11 Mwanza - Comparative high turbidity dose response
Initial turbidity 510 NTU

Figure 4.12 Mwanza - Comparative high turbidity dose response
Initial turbidity 1800 NTU
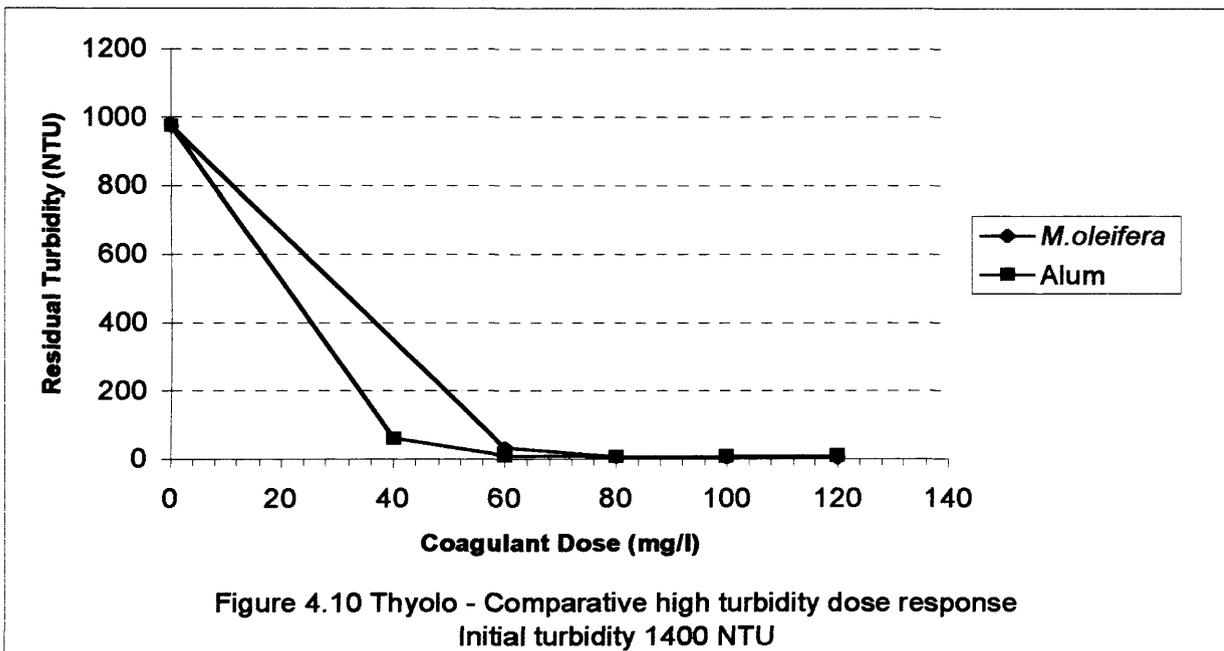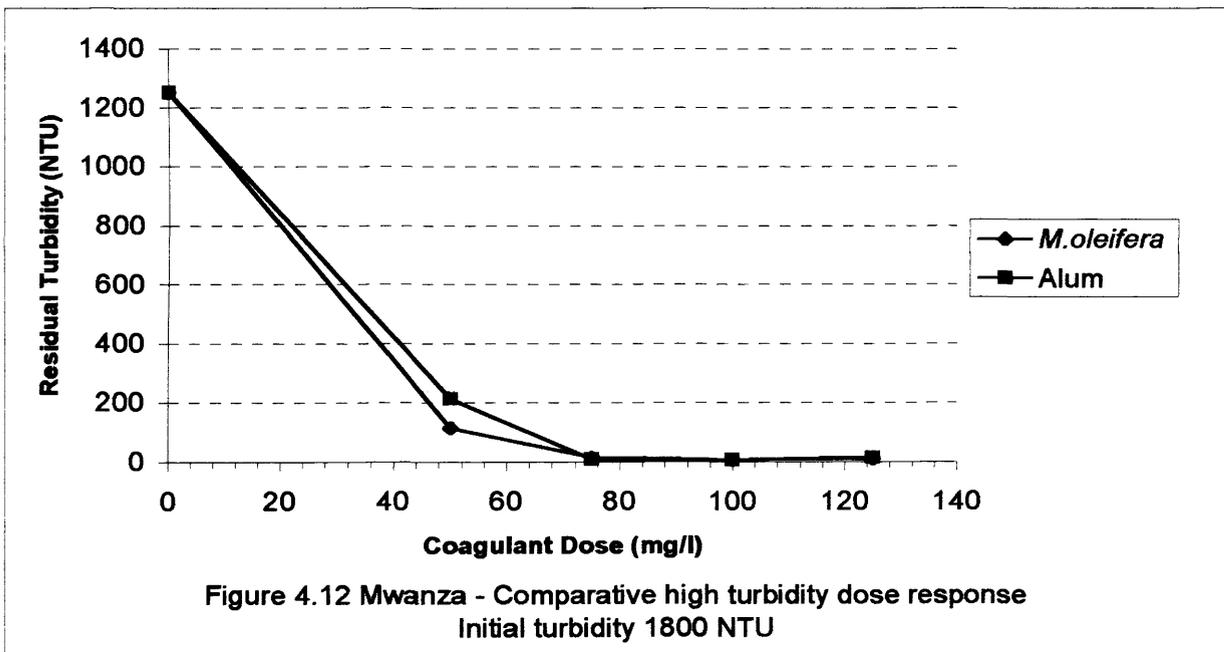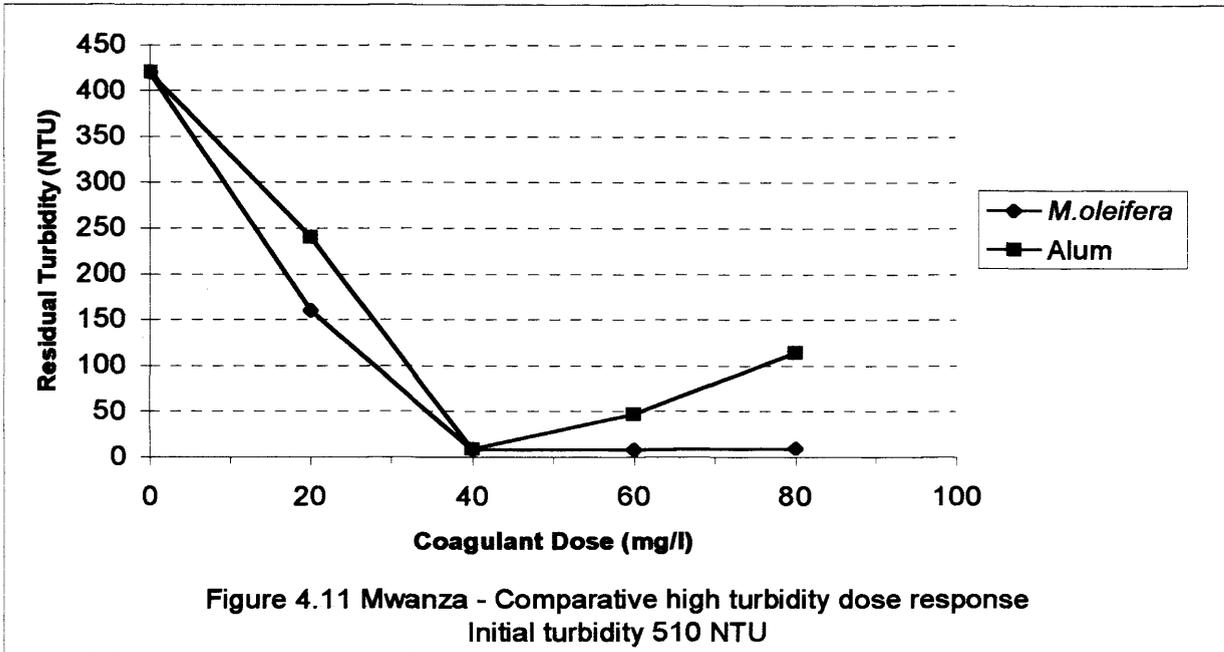
4.2.4 Discussion

The results presented demonstrate clearly that over the range of raw water sources examined *M.oleifera* can be used as an effective treatment at medium or high turbidities. However, the results demonstrate the significant variability of treatment performance at low turbidities. Leaving the results from Walkers Ferry aside for the moment, the results from Thyolo and Mwanza show a high degree of similarity. Floc formation does not appear to be the problem. In each case sufficient floc were seen to be formed that should enable full clarification to be achieved. The fact that the process appears to be ineffective is a result of the inability of the formed floc to settle. The floc were seen to be small, almost granular in shape and obviously light. In addition, the size of the floc was seen to decrease while the numbers increased as the dose of coagulant increased. In comparison, the Walkers Ferry floc characteristics were markedly different. Even at very low turbidities *M.oleifera* treatment produced effective floc formation with large fast settling floc evident. The only similarity in behaviour between Walkers Ferry and the Thyolo and Mwanza sources was the reduction in floc size and increase in floc numbers as coagulant dose was increased.

The differences in floc characteristics observed are clearly a function of raw water quality, particle type and concentration and the mechanisms of protein activity. These aspects are discussed further in section 4.4.

4.3 PERFORMANCE OF *M.OLEIFERA* ON A EUTROPHIC WATER IN ZIMBABWE

The raw water that serves the Morton Jaffray (MJ) Water Treatment Works is inherently difficult to treat with a low suspended solids concentration, high organic content and algal blooms that can occur throughout the year. Over the years the water has become increasingly eutrophic as a result of phosphate and nitrate additions from fertiliser run-off and effluent discharges. In addition, the lack of significant rainfall over a 6 to 7 year period prior to this study has prevented the reversal of this eutrophic state. The water entering MJ is currently treated with aluminium sulphate (alum) as a coagulant with activated silica as a weighting agent. Although treatment is effective a major

problem exists with the alum sludge produced during treatment. The sludge is voluminous with a high water content. This prevents adequate settling and compaction within the two sludge lagoons currently being used to treat the sludge. As a result high concentrations of alum sludge are being disposed of to the local watercourse causing extensive problems to the farmers downstream.

4.3.1 Experimental procedure

Samples were collected from the raw water intake at the MJ works prior to any form of treatment. Standard laboratory jar tests were carried out in the environmental laboratory of Mr.J.McKendrick, retired Chief Water Quality Chemist of Harare City Council. The equipment used was a Baird and Tatlock five jar stirrer with 500ml samples. The age of the jar test apparatus was such that the test parameters listed below are nominal, difficulty being experienced in maintaining a steady stirring speed particularly during the flocculation stage. The rotational speed indicated was obtained by counting the number of revolutions in a given time period and hence are subject to experimental error. Jar test parameters were as follows; rapid mix for 2 minutes (approx.100rpm) for coagulant dispersion followed by slow mix for 15 minutes (approx. 30rpm +/- 20rpm) to allow flocculation. A 1 hour quiescent settling period followed prior to sample extraction for turbidity and other analyses. Samples for turbidity analysis were taken from 2 cm below the water surface. Observations were made of floc formation rates, floc size, settleability and floc strength.

M.oleifera seed powder obtained from Malawi was used throughout the tests. Solutions were prepared in tap water at a 1% concentration.

In order to provide comparative data, the study incorporated treatability tests using locally produced granular alum (obtained from and used by MJ) and liquid Ferric Aluminium Sulphate (FAS) (Clinty Chemicals Ltd, Northern Ireland). FAS was included in the study as the manufacturers claimed that it was more effective than alum in the treatment of eutrophic waters.

Alum solutions were prepared in tap water at a 1% concentration. FAS was used undiluted as supplied. No quantitative data can be given regarding the dose of FAS utilised as information

regarding constituents was not forthcoming from the supplier. It was not possible with the equipment available to measure precisely the applied dose of FAS.

4.3.2 Results

Raw water turbidities were consistently in the range 2.0 - 3.5 NTU throughout the period of the study.

Tests carried out using *M.oleifera* in isolation were disappointing. Over the dose range examined, 15 to 80mg/l, floc formation was evident, however, it was observed to be slow requiring the full 15 minutes flocculation time to produce floc of significant size. On standing these floc grew producing large 'woolly' structures but did not settle well. Effective clearance was still not achieved even after 4 hours settling. At doses of 20,25,30,35 and 40mg/l a residual turbidity of 1.0 NTU was obtained following this 4 hour settling period, however, large quantities of floc were still evident in the bottom half of the sample jars.

An optimum alum dose of 80mg/l was determined giving a residual turbidity after settlement of 0.4 NTU. Floc formation was rapid with visible floc after the rapid mix phase. Floc continued to grow during the flocculation period producing a large 'woolly' structure. Settlement occurred rapidly with maximum clearance being achieved after 20 minutes. Some light floc was still evident in suspension at the end of the settlement period.

Jar tests showed an optimum dose for FAS to be 3 drops from a 10ml pipette of the undiluted solution producing a settled water turbidity of 0.3 NTU. Floc formation and settling were rapid with little or no floc left in suspension following settlement.

The poor performance of *M.oleifera* in isolation lead to an alternative approach being adopted. The addition of particulate material to the raw water was seen as a means of improving flocculation and settling performance. The use of weighting agents is an accepted treatment process, the MJ works uses activated silica in combination with alum for this very purpose. It had been hoped to use activated silica within these trials to provide comparative performance data to that obtained with alum, however, it was not possible to determine the concentration of the

activated silica suspension used at MJ, despite intensive questioning of the operators, nor the method by which the suspensions were prepared. An alternative weighting agent was therefore sought. Bentonite clay has been previously used as a weighting agent (Montgomery, 1985) and was therefore selected for use in combination with *M.oleifera*. A supplier of imported bentonite (locally mined sources were unavailable) was located and samples of three different types of bentonite obtained. Following preliminary testing, a sodium bentonite, designated bentonite 1 by the supplier was found to be the most effective. Following initial sequence addition testing, the clay (made up as a nominal 1% solution) was added to the test water at the end of the rapid mix phase following *M.oleifera* addition. Under this treatment regime a dose of 40mg/l *M.oleifera* and 80mg/l bentonite was found to give the best performance. It must be noted that the timing and location of clay addition within the treatment train subsequent to *M.oleifera* addition were not optimised due to insufficient time. Under the treatment conditions established, floc formation was observed to be rapid with visible floc appearing at the end of the rapid mix phase. Floc growth continued during the slow mix and settling phases. In comparative tests, settling of the *M.oleifera*/bentonite 1 (*M.oleifera*/B1) floc occurred at a rate similar to that of alum but slower to that for FAS. At the end of the settling period small light floc were still observed within the supernatant of the *M.oleifera*/B1 sample.

Although residual turbidity is generally regarded as the primary parameter measured to determine treatment effectiveness, with a raw water turbidity in the range 2.0 - 3.5 NTU (c.f. WHO guideline value for potable water of 5 NTU), residual turbidity becomes a less effective and, to an extent, a less important measure. Thus, a series of tests were carried out using the optimum conditions established for the 3 coagulant combinations to determine the effect of treatment on other water quality parameters. Following settlement, samples were filtered through Whatman no.1 filter paper to simulate the filtration process (Kawamura, 1991). The following analyses were carried out: pH, conductivity, total alkalinity, iron, manganese, turbidity, total dissolved solids (TDS) and colour. The results are presented in the Table 4.2. All analyses, other than those indicated, were carried out after filtration.

	Raw water	M.oleifera/B1	Alum	FAS
pH	8.1 (pf)	8.1	7.9	7.3
Alkalinity (mg/l as CaCO₃)	172 (pf)	176	148	96
Conductivity (μS/cm)	573 (pf)	582	484	578
Colour (Hazen)	25 (pf)	< 5 (pf)	< 5 (pf)	< 5 (pf)
Turbidity (NTU)	2.5 (pf)	1.0 (pf) 0.75	0.325 (pf) 0.25	0.225 (pf) 0.15
Iron (mg/l)	0.26 (pf)	NIL	NIL	TRACE
Manganese (mg/l)	0.04 (pf)	NIL	TRACE	0.07
TDS	482.0	466.0	422.4	453.6

pf - analyses done prior to filtration

Table 4.2 The effect of treatment on final water quality

The table indicates little difference in final water quality with the following exceptions:

i)   Treatment using M.oleifera does not affect pH. The results from both the metal salts indicate how their action has the effect of reducing pH and alkalinity values;

ii)   A possible benefit of M.oleifera may be seen through enhanced iron and manganese removal. This is covered in more detail in chapter 5;

iii)   Turbidity removal with M.oleifera is not as good as for either of the metal salts despite the bentonite addition. Settling times were observed to be significantly less than for FAS and marginally less than for alum. It is possible that enhanced performance may be achieved through optimisation of timing of clay addition or the use of an alternative weighting agent.

The floc characteristics of M.oleifera are significantly different to those obtained on the Thyolo and Mwanza sources in Malawi. Some similarities exist between the floc formed on the Walkers Ferry source and those on MJ viz.: larger size and the floc growth that continued to occur during the settling phase. An attempt to explain the observed differences and similarities in floc

characteristics in relation to the mechanisms of protein activity and the effects of raw water quality is presented in the following section.

4.4 DISCUSSION

As stated in the introduction to this chapter, the primary objectives of the study were to evaluate treatment performance on a range of raw water sources under seasonal variations in raw water quality and, from the results and observations made, attempt to explain performance variability in terms of the mechanisms of protein activity and the effects of raw water quality. It could be argued at this point that a complete understanding of the mechanisms of action is not absolutely necessary when considering the practical application of seed solutions. Treatability studies, such as those presented here, could, and should, be undertaken in order to determine whether or not a coagulant material was suitable for treating a particular water. However, it can also be argued that a better understanding of the mechanisms of activity could lead to the developments of methods to improve treatment performance. For example, if it could be shown that specific ions enhanced floc formation and settlement at low turbidities, then improved performance could potentially be achieved through the addition of an additional chemical component at some point in the treatment train.

It is acknowledged that the study presented is limited in terms of being able to define absolutely the mechanisms of protein activity, however, analysis of the results and observations, in the light of recently obtained data from a separate investigation, does allow an interpretation that provides at least the basis for a discussion. It must be noted that concepts will be introduced into this section that have arisen from a recent investigation into the potential commercialisation of *M.oleifera* seed proteins as a coagulant and, as such, they must be considered strictly confidential. With respect to the initial objectives, the results presented in the previous sections may be summarised:

- At high turbidities *M.oleifera* seed solutions give equivalent and in some cases superior performance, as measured by residual turbidity, to that obtained for alum. The dependence of

alum treatment on raw water alkalinity and pH values has been demonstrated, as has the independent nature of seed treatment;

- At low to medium turbidity levels treatment performance is significantly more variable and is clearly dependent on the characteristics of individual raw waters. Such variations were not observed with alum treatment.

Both findings confirm those of previous studies on *M.oleifera*, outlined in Chapter 3, however, perhaps of more significance are the observations made on floc characteristics at low to medium turbidity for the different source waters. To reiterate, for the Thyolo and Mwanza sources formed floc were small and with a granular appearance whilst for both the Walkers Ferry and MJ sources formed floc were larger and 'woolly' in appearance and continued to increase in size following the end of the flocculation period. With the exception of Walkers Ferry, floc formed for the other sources at low turbidity were light and slow settling. In addition, for all sources distinct dose related effects could be observed: where settlement did occur, a wide dose range existed over which effective performance could be achieved; as dose increased above what may be considered the optimum i.e. the lowest dose at which effective performance could be achieved, floc size decreased whilst floc numbers increased.

In the following section the current state of knowledge on the assumed mechanisms of activity for the proteins will be restated and discussed in the context of the results and findings of this and subsequent studies. An attempt will then be made to relate the potential mechanisms of action to the three main questions that have arisen from this study viz.: i) Why does there exist such a wide dose range over which equivalent performance may be achieved and how are floc characteristics influenced by an increase in dose? ii) What influences floc structure at low turbidity? iii) What governs the ability of formed floc to settle? The final section details observations relevant to the practical application of *M.oleifera* to water treatment systems and the potential advantages over proprietary chemical coagulants.

4.4.1 Mechanisms of protein activity and their effects on floc characteristics

In Chapter 2 it was postulated that two potential mechanisms of action viz.: adsorption and interparticle bridging and charge neutralisation via patch adsorption, could explain the ability of synthetic cationic polyelectrolytes to flocculate colloidal suspensions. Which mechanism predominated depended on the concentration of suspended particles and the molecular weight and charge of the polyelectrolyte.

With respect to the proteins from *M.oleifera* consideration has to be given not only to molecular weight and charge, but also to the amphoteric nature of the proteins i.e. although cationic ionisable groups predominate the proteins will also contain amino acid residues that ionise to give negatively charged sites, the extent and nature of the ionisation being dependent on the pH of the solution. With the iso-electric point of the low molecular weight proteins assumed to be responsible for coagulation having been determined to be between pH 10-11 (Tauscher, 1994; Gassenschmidt *et al.* 1995), it is clear that at the pH levels commonly encountered in water treatment (pH 6-8), an overall positive charge will be exhibited. However, this does not preclude the existence of negatively charged sites.

In addition recent work undertaken by the author in collaboration with the Campden and Chorleywood Food Research Association (C&CFRA) to examine the techniques that might be used to extract and process the active fractions on a commercial scale, has produced results that further complicate the understanding of coagulation activity. Using defatted seed kernel powder a range of solvents were examined with a view to maximising protein extraction. Extractions in dilute hydrochloric acid (HCl) at pH 1 and ammonium bicarbonate (NH_4HCO_3) at pH 8.2 proved to be the most effective not only in terms of quantity of material but extraction also limited the recovery of what may be termed 'contaminant' proteins i.e. those proteins not considered to be involved in coagulation process (it should be noted that for reasons of confidentiality, figures relating to solvent strengths, quantities of material extracted etc., shall not be mentioned here, it is the observations that have been made in relation to the techniques that have been employed that are of particular relevance). Extraction in tap water was only marginally less effective than that in

HCl and NH₄HCO₃, the primary difference being that all proteins present in the powdered seed were extracted to a certain degree.

In order to concentrate the extracted proteins with a view to the eventual production of a dried product, various precipitation techniques were evaluated. Whilst testing the original HCl extracts for coagulation effectiveness it was observed that upon addition of the extract to tap water with no particulates present (kaolin clay suspensions have, to date, formed the basis of determining the extent of coagulation activity) a white precipitate formed. As the flocculation stage progressed the precipitates increased in size and eventually settled out. It is known that proteins have a tendency to precipitate at their isoelectric points. Why this should happen is the result of some complex chemistry, however, in relatively simple terms, proteins can remain in aqueous solution at pH values less than their isoelectric points as a result of dissociation of the amino acid groups and residues and the formation of a hydration shell, i.e. water molecules closely associated with the protein structure. At their isoelectric pH values the proteins achieve electroneutrality. The ionic state of the amino acids at the isoelectric point is called a zwitterion or dipolar ion. In the state of neutrality each zwitterion acts as a dipole and there is electrostatic attraction between neighbouring ions that tends to orient them and so induce systematic packing among them. Such packing increases the proximity of amino hydrogens and carboxyl oxygens leading to the formation of hydrogen bonds. The combined electrostatic attraction and hydrogen bonding increases systematic packing thereby driving water molecules away from the protein surface and inducing aggregation and precipitation (Taylor, 1981).

Clearly the pH of tap water, pH 7.9 in this case, is some way from the isoelectric point of pH 10-11 determined for the active proteins. Why then should a precipitate be forming? At the moment this is one question that cannot be answered, however, in utilising this precipitation phenomena as a means of concentrating protein suspensions, it has been shown that proteins extracted in HCl will precipitate at pH values between 2 and 3, much less than the determined isoelectric point, upon the addition of sodium hydroxide (NaOH) (C&CFRA, 1998). It must be noted that prior to the addition of NaOH, the HCl extract solutions were themselves clear, however, precipitation

has been observed in extract solutions at higher pH values, for example, in NH_4HCO_3 solutions at pH 8.2. The precipitates were significantly smaller than those observed in HCl extracts following NaOH addition, although the rate at which they appeared was similar.

It is clear from the above that there exists a complex relationship between the extracted proteins and their aqueous environment. An understanding of this relationship is beyond the scope of this study and is unlikely to become clearer until the three dimensional structure of the proteins has been determined (Whitelam, 1998). However, the existence of a precipitation mechanism must now be acknowledged in any attempt to explain the mechanisms by which seed solutions can effect coagulation.

Whilst further information is required to determine the extent to which precipitation is involved in the coagulation process there is evidence from this and other studies that indicates that a combination of mechanisms exists. The inter-particle bridging model, originally proposed to explain the activity of polyelectrolytes, is probably least favoured for application in this case. The reasons for this are two-fold. Firstly, it can be inferred from the low molecular weight (estimated to be 13,000 daltons) of the active proteins that they will be significantly smaller than the particulates to be removed. This size relationship may be illustrated by taking haemoglobin as an example of a low molecular weight protein. Haemoglobin has a molecular weight of 68,000 daltons that corresponds to an estimated size of $4m\mu$ (Taylor, 1981). In comparison, the average colloid diameter is approximately $500m\mu$ (Stumm, 1977). It is clear that unless significant cross linking existed between the active proteins thereby effectively increasing their molecular weight, and there is no current evidence to suggest that this occurs, it is unlikely that, following adsorption of a segment of the protein on a particle surface, sufficient segments will be left in solution to span the inter-particle gap thus forming a bridge.

The second reason for discounting the bridging model is that it is now generally accepted that inter-particle bridging is a less significant factor in the activity of cationic polyelectrolytes and that the primary mechanism is one of charge neutralisation through a patch wise adsorption on particulate surfaces (a more detailed discussion follows). At high particle concentrations the

bridging model may still apply, since inter-particle collisions may occur prior to full adsorption of the polyelectrolyte onto the particle surface, however such a situation can only be envisaged for high turbidity raw waters and those instances where the particles to be removed are smaller than the polyelectrolyte molecules (Gregory,J. 1976; Mabire *et al.* 1984; Ghosh and Leu, 1986).

As indicated above, the destabilisation mechanism of cationic polyelectrolytes towards negatively charged particles is widely accepted as being one of charge neutralisation brought about through a patch wise adsorption on particle surfaces. Such adsorption induces localised regions of reduced negative charge or the formation of positively charged sites that are able to interact with negatively charged sites on other particles. Given this scenario, what evidence is there to suggest that patch adsorption plays any role in protein activity? The effect of seed solution addition on the zeta potential of a synthetic test water containing kaolin has been examined (Ndabigengesere *et al.*, 1995). The results, reproduced in Figure 4.13, demonstrated that as seed dose was increased the negative zeta potential decreased, optimum removal being achieved at that dose that corresponded to the zero point of charge (ZPC) of the particle.

Figure 4.13 Residual turbidity and zeta potential of kaolin suspension as a function of seed dose
Coagulant: M.oleifera whole seed including shell at a 5% solution concentration
Adapted from Ndabigengesere et al. (1995)
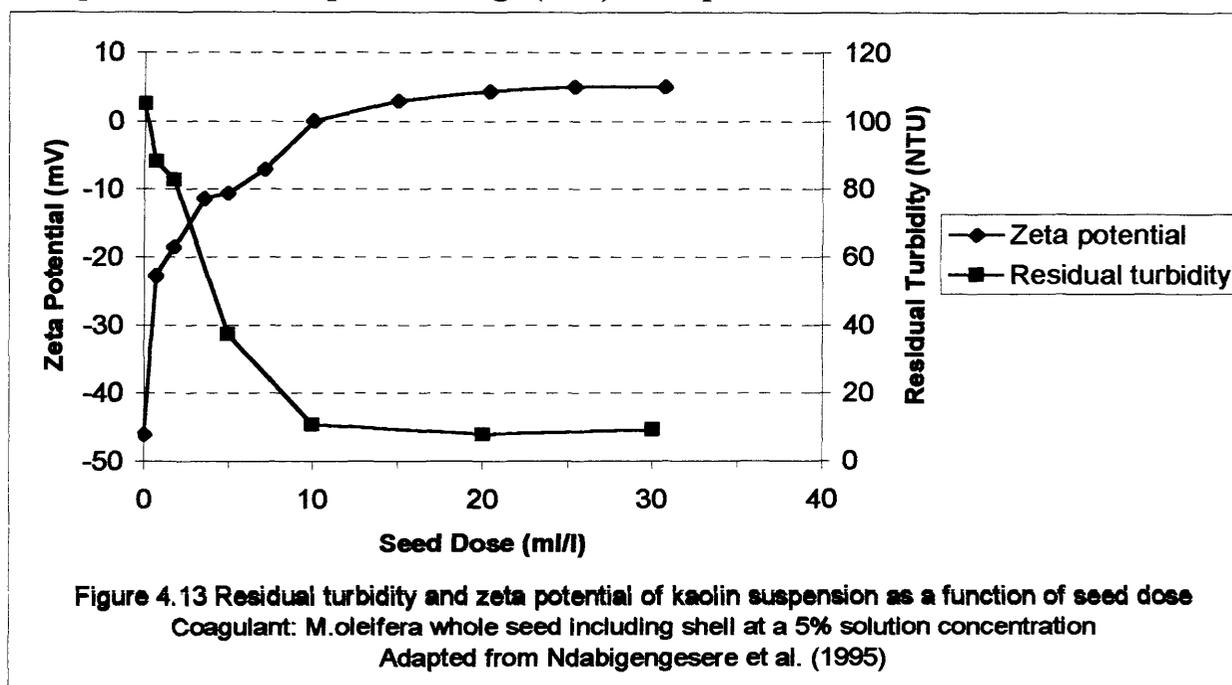
Increasing the dose above the optimum induces a positive zeta potential on the particle surface, the plateau region indicating complete coverage of all surface sites available for adsorption. Leaving aside the overdose situation for the moment, the reduction in zeta potential that leads to effective floc formation is clearly a function of the charge neutralisation behaviour of the cationic proteins. Further evidence for this fact may be gained from results obtained in this study. Figure 4.14 shows the relationship obtained between optimum dose and raw water turbidity for the three Malawi sources.

Figure 4.14 Relationship between raw water turbidity and optimum dose as a function of test site
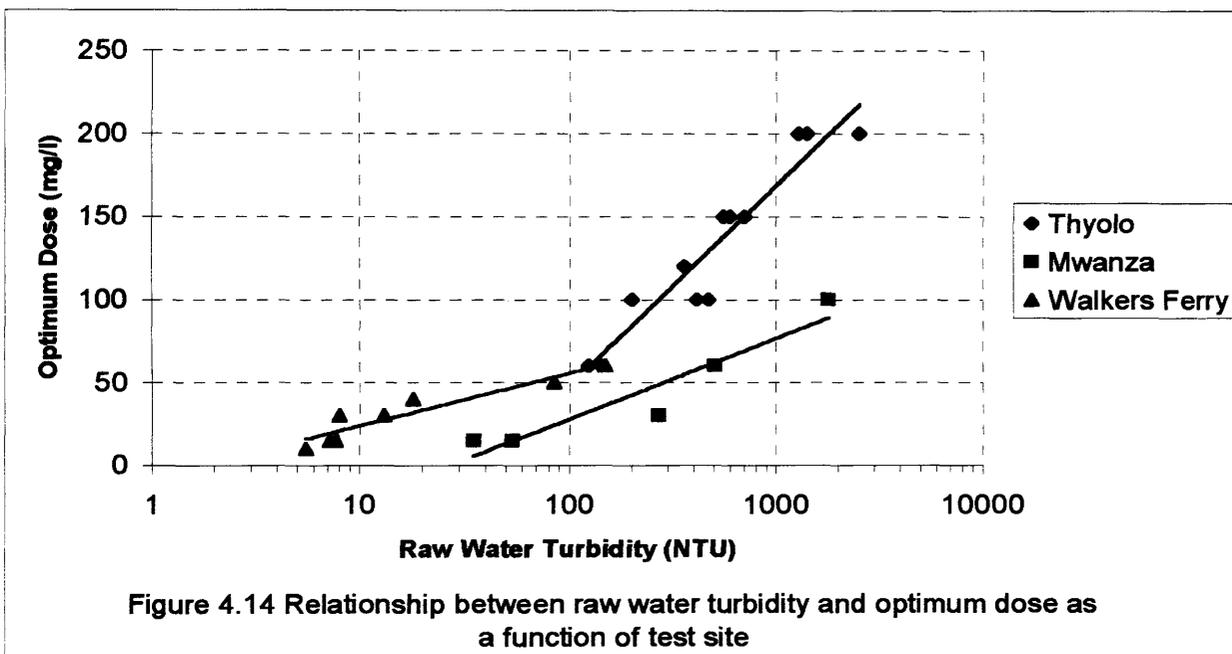
In analysing the data for inclusion in the figure the following assumptions / omissions have been made: i) results presented for the different source waters demonstrate that effective performance may be achieved over a wide dose range. The optimum dose indicated in the figure is the lowest dose required to achieve such performance; ii) for the Thyolo and Mwanza sources, settlement at low turbidities was problematic. With effective performance being determined by residual turbidity, those samples that showed minimal settlement, despite effective floc formation, have been omitted. With these factors in mind, it is clear that for each source there would appear to be

a stoichiometric relationship optimum dose and initial turbidity. Such relationships would be expected if charge neutralisation was the predominant mechanism (Stumm and O'Melia, 1968; O'Melia, 1969). The fact that a specific relationship exists for each source would also be expected since each water will have its own range of water quality parameters and will contain particulate material of differing charge neutralisation requirement dependent on the catchment of the source. Returning to Figure 4.13, according to the theoretical considerations of charge neutralisation it would be expected that charge reversal, as shown by the positive zeta potential, would result in particle restabilisation, inter-particle repulsion and an increase in residual turbidity. This is clearly not the case as turbidity removal continues to occur despite the reversal in particle charge. It must be assumed, therefore, that charge neutralisation is not the sole mechanism of action. It is possible that the observed effect may be explained by the protein precipitation phenomena outlined previously. Of particular interest is the observation of colloidal precipitates in solution at pH values similar to those of raw waters i.e. NH_4HCO_3 at pH 8.2. It is, therefore, postulated that activity is brought about by a combination of charge neutralisation and protein precipitation with the former being the dominant mechanism. At doses above the optimum excess protein not involved in the charge neutralisation process precipitates out. As the precipitates form aggregation, either through electrostatic attraction between neighbouring dipoles or through hydrogen bonding, may result in particulates being trapped within the growing precipitates (c.f. sweep flocculation using alum). The dual, and potentially competitive, mechanisms of charge neutralisation and protein precipitation would help explain not only the results of Ndabigengesere et al. (1995), but also the observations made on the Malawi raw waters that an increase in dose leads to greater numbers of floc of reduced size irrespective of the initial turbidity of the raw water.

One of the main findings of the studies in Malawi and Zimbabwe was the difference in floc structure produced on the four separate sources. It is not the mechanisms of action per se that can account for the observed differences. By consideration of the raw water quality of the different sources it is postulated that it is the presence of natural organic materials (NOM) that is of

significance. The Thyolo and Mwanza source waters are small, shallow and fast flowing, even during the dry seasons, and are unlikely to offer suitable conditions for the decomposition of organic matter. The Walkers Ferry source is a large deep river that outlets from Lake Malawi. As such, it is likely to have a higher NOM content than both Thyolo and Mwanza. The MJ source is a large impounded reservoir. The regular occurrence of algal blooms is an indication of the presence of high levels of organic matter (Montgomery, 1985; Randtke, 1988). It is generally accepted that the vast majority of NOM present in water results from the decomposition of animal and plant tissues and the exudates of microorganisms. Although other sources of organic materials also exist, NOM predominates. The mechanisms by which decomposition results in the formation of NOM are complex and have been detailed elsewhere (Montgomery, 1985; Randtke, 1988; Gregor et al, 1997), however, NOM is considered to take two major forms viz.: humic acid and fulvic acid. Humic acids are generally characterised as high molecular weight compounds (up to 200,000) of low charge density and may exist either as colloidal particulates or as soluble compounds depending on the pH of the water. Fulvic acids are low molecular weight soluble compounds with comparatively higher charge densities. A number of mechanisms have been proposed to describe how these compounds may be removed from suspension, however, of relevance to this discussion is charge neutralisation.

It is clear from the previous discussion of the behaviour of M.oleifera proteins that charge neutralisation and complexing of both humic and fulvic acids can be envisaged, however, it is considered that it is the interaction between the proteins and the high molecular weight humic acids that has a major influence on the observed floc characteristics. In this instance it is postulated that the humic acid compounds act in a similar manner to synthetic anionic polyelectrolytes bridging between destabilised particles or, potentially, protein precipitates thereby forming the large, woolly structures observed. A similar argument could be put forward for the larger floc formed in the treatment of the Walkers Ferry source. For the Thyolo and Mwanza sources, the absence of such 'bridging' components provide what may be considered 'ideal' charge neutralisation conditions that results in the formation of closely packed, granular floc.

The variations in settling behaviour observed throughout the study is considered to be primarily due to the nature and concentration of the particles to be removed. An increase in the specific gravity or the concentration of particles will result in the formation of heavier, faster settling floc.

4.4.2 Potential advantages of *M.oleifera* seed treatment

In addition to treatment performance being independent of raw water pH (thereby reducing the potential costs of treatment) and the wide dose range over which effective performance may be maintained (indicative of the robustness of treatment), it became clear during both studies that a further advantage of *M.oleifera* application over that of the chemical coagulants was in the sludge produced following treatment. Both the aluminium and iron salts have a tendency to produce large volumes of sludge. This occurs through the formation of large quantities of insoluble metal hydroxides during treatment and the inclusion of water within the hydroxide floc structure (Montgomery, 1985). The result is a voluminous sludge that can present problems in dewatering and subsequent disposal. As discussed in the previous section *M.oleifera* seed addition, in the absence of high levels of natural organic material, produces floc that are generally smaller and more compact. The proximity of the particles to each other within the floc structure, arising from the proposed mechanism of activity, precludes the inclusion of water and results in a closely packed sludge. It is considered that such a sludge would pose less of a technical problem in terms of dewatering and subsequent disposal. A further benefit is that, unlike the metal hydroxide sludges, the added material contained in the *M.oleifera* sludge is bio-degradeable. The potential for use of a dewatered slurry or the dry solids as a fertiliser or soil conditioner would be greatly enhanced. No studies have yet been carried out to establish this potential.

4.5 CONCLUSION

In light of recent findings, an analysis of the results obtained in this study has lead to the development of a new theory that describes that coagulation mechanism of the active proteins viz.: charge neutralisation coupled with protein precipitation. Further experimental evidence is

required to confirm this hypothesis, however, should protein precipitation be found to be a significant factor it may, in the future, be possible to enhance the precipitation process thereby providing a natural material that could operate as effectively as the metal salt coagulants over a broader range of raw water conditions.

The investigation presented has demonstrated that *M.oleifera* seed solutions can be used effectively to treat a variety of raw water sources under different conditions. It is argued that the variable performance of seed solutions on some low to medium turbidity waters is not necessarily a major drawback to their potential use as replacements for proprietary chemicals in water treatment systems. In general, the majority of chemical consumption occurs during the wet season when turbidity levels are high. As this study has demonstrated, seed solutions can provide equivalent and in some cases superior performance to their chemical counterparts under such conditions. The use of the seed solely during the wet season could potentially result in significant savings in chemical costs.

The batch studies reported were carried out under controlled laboratory conditions. Although these studies are able to provide a certain amount of information regarding both treatment performance and the mechanisms by which performance is effected, success on this scale does not automatically infer that use on a continuous flow system under operational conditions would produce similar results. The determination of seed solution performance under continuous flow conditions and the viability of treatment in terms of seed processing and seed solution preparation is the subject of the following chapter.

CHAPTER 5

EXPERIMENTAL DETERMINATION OF TREATMENT VIABILITY - PILOT SCALE AND FULL SCALE STUDIES

5.1 INTRODUCTION

In Chapter 4 the laboratory investigations carried out on the three raw water sources in Malawi demonstrated that treatment using *M.oleifera* was feasible particularly in respect of raw waters containing medium to high turbidities. Although problems were encountered in treating low turbidity waters, the successes achieved at medium and high turbidities merited continued investigation. The justification for this stems from the fact that the majority of chemical coagulant consumption occurs during the wet season when raw water turbidities are generally in the medium to high categories (as defined in Chapter 4). It may be argued that even if *M.oleifera* were adopted as coagulant solely during this period the cost savings would be considerable.

The work presented in this chapter extends the laboratory studies reported through scaling up the investigation to enable treatment effectiveness on continuous flow systems to be determined. It had originally been considered that this could be most effectively achieved by utilising one of the existing treatment works in Malawi. The works would be monitored in operation using the regular coagulant; this would subsequently be replaced by *M.oleifera* seed solution. However, two problems were encountered. Firstly, at the time the scaled up investigations were proposed, sufficient seed supplies for continued use over a long period of time could not be guaranteed. Secondly, the Ministry of Works and Supplies Water Department of the Malawi Government, without whom full scale trials could not be carried out, were not convinced as to the treatment effectiveness of the seeds and to the ease with which seed solutions could be prepared under operational conditions. In order to overcome both these problems a compromise was reached whereby initial testing would be carried out on a purpose built pilot plant. If pilot scale trials

proved successful, permission to operate an existing rural water treatment works would be forthcoming. The primary objectives of both the pilot plant and full scale trials were:

i) To evaluate the practicability of the process in terms of seed processing, solution preparation and solution dosing;

ii) To confirm the effectiveness of the treatment process on continuous flow systems and to determine potential limitations to use that a) may require treatment works modifications and b) may arise from severe climatic conditions prevalent in developing countries;

iii) To compare the effectiveness of the proposed treatment to alternatives currently available. In this instance alum was selected to provide data for comparison due to its worldwide use in treatment works.

Although four separate water sources had been investigated during the laboratory studies, Thyolo was established as the location for the pilot and full scale trials primarily for the reasons of ease of access to river intake and site topography, inherent problems of high raw water turbidities during the wet season and the close proximity of the site to Blantyre keeping travel time to the site to a minimum and allowing the use of back up technical support available in Blantyre. Figure 5.1 shows the location of the test site.
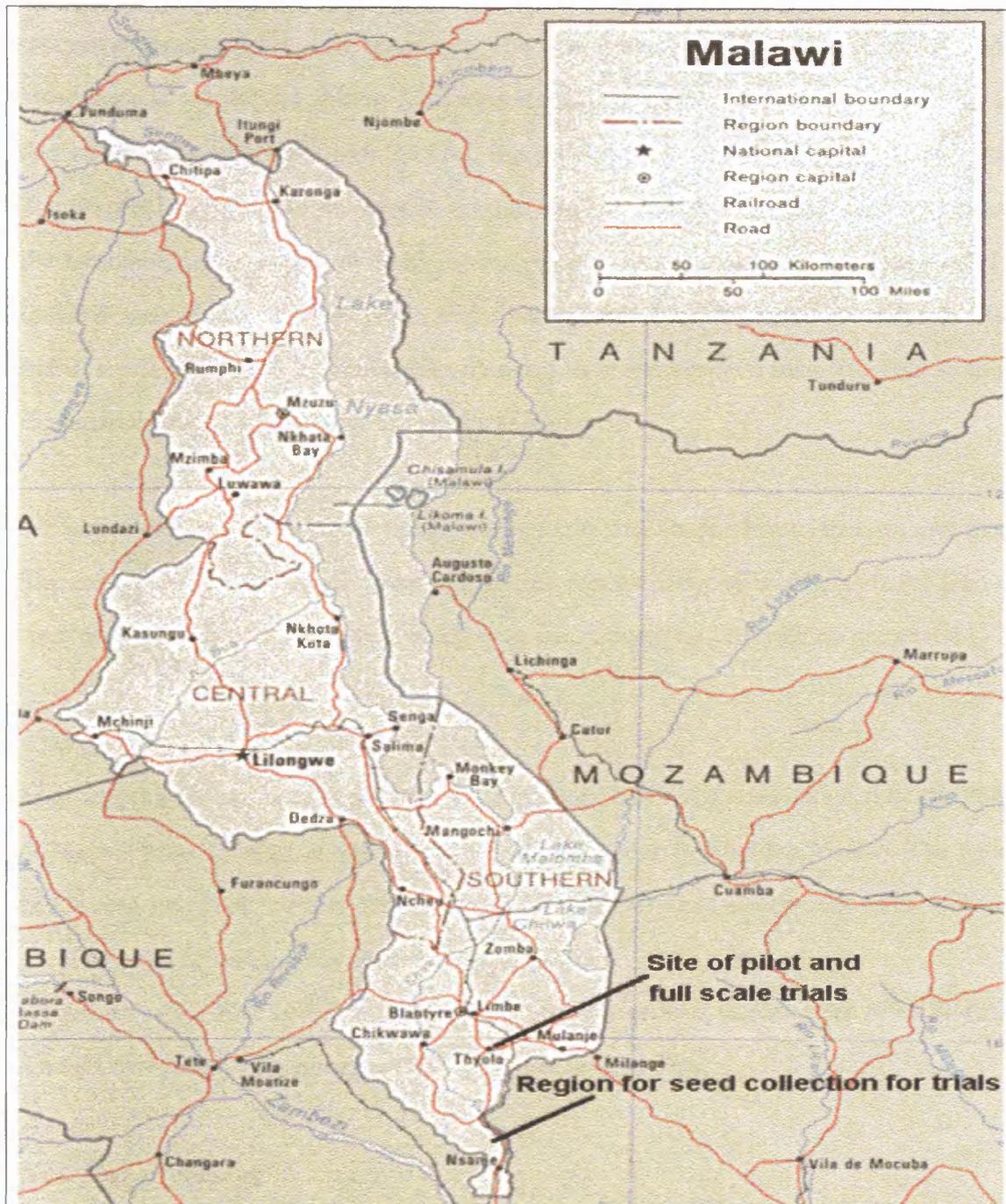
Figure 5.1 Location of test site and seed collection region

The pilot scale trials were carried out over a three year period, all the trials were planned to extend over the transition between the wet and dry seasons in Malawi to enable a wide range of

raw water characteristics to be examined. The work presented in this chapter is a compilation of investigations carried out during four separate field studies over a three year period and was undertaken in collaboration with the Civil Engineering Department of the Polytechnic of Malawi. The chapter is divided into three main sections. The first examines problems involved in scaling up the treatment with respect to seed processing and solution preparation. The second section outlines the design of the pilot scale works and the modifications required following commissioning and discusses the results obtained from experimental runs. The final section details the full scale trials and discusses the results in context.

5.2 SEED SUPPLIES, PROCESSING TECHNIQUES AND SUSPENSION PREPARATION AND STABILITY

The following sections details observations and results obtained during both the pilot and full scale trials relating to the problems involved in seed processing and solution preparation.

5.2.1 Seed supplies

Seed supplies for use in both the pilot scale and full scale trials were obtained by the Polytechnic of Malawi from villagers in and around the Nsanje region in southern Malawi. The region from which seed was obtained is shown in Figure 5.1 (location map also for test site). Villagers were given payment in cash per kilogram of seed collected.

It must be noted that sufficient seed supplies for the various studies were often limited due to drought conditions experienced during two of the field studies.

5.2.2 Seed processing

Seed solutions throughout the laboratory studies had been prepared by crushing the required amount of seed with a pestle and mortar prior to the addition of water to the required dilution. The volume of seed to be utilised during both the pilot and full scale trials required the development of an alternative method of processing. In order to maintain the appropriate technology theme that runs throughout this work, a method was developed using local skills, local

equipment and traditional grinding processes. The method developed to obtain the seed powder involved removal of the shells by grinding the seeds between a stone and a hard, flat surface, the shells then being separated from the kernels by traditional winnowing techniques. The seed kernels were then pounded using a traditional maize pestle and mortar and sieved through a traditional maize sieve of approximate aperture 0.8mm. Retained seed particles were returned for further pounding. Seed processing is illustrated in Figures 5.2, 5.3 and 5.4. All seed for the pilot and full scale trials was processed by local women from Thyolo.
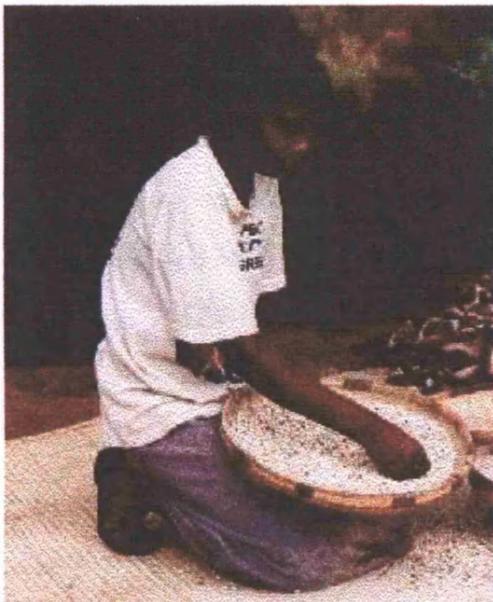
Figure 5.2 Winnowing of seed to remove shell fragments
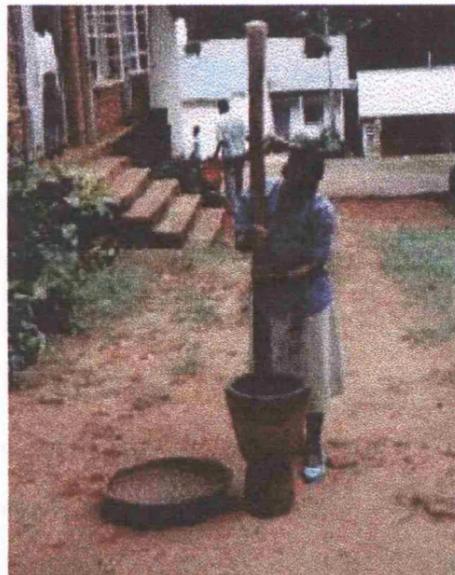
Figure 5.3 Crushing of seed kernels using traditional pestle and mortar

Figure 5.4 Sieving crushed seed kernel powder

The full scale trials provided an opportunity for an evaluation of the efficiency of the processing technique with large quantities of seed being required for the trials. A total of 1200 kg was collected by the beginning of the field study. The seed was stored at the Polytechnic before being taken to Thyolo for processing. The seeds were processed as per the method described above with the exception that following initial crushing and sieving of the seed kernels, material retained on the sieve was returned for crushing a second time. This process was subsequently repeated a third time. Material from each successive crushing and sieving were kept separate from each other. Subsequent jar tests showed material from the third crushing to have little or no activity, this is considered to be due to excess oil produced during this final stage. Recovery rates during each stage of processing are summarised in Table 5.1. The actual figures for seed kernel material recovery were, unfortunately, not recorded due to a breakdown in communication. The 780 kg of seed kernel material available for processing was estimated assuming the husk to be on average 25% of the whole seed weight. An additional 10% was included for wastage during the husk removal and winnowing processes. From observations made during the processing and from the fact that husk material from the same batch sent to Edinburgh University for activated carbon trials was found to contain significant quantities of kernel material (McConnachie, 1995) it is considered that this wastage figure is on the low side.

	kg	% of seed kernel material	% of total seed weight collected
Seed collected	1200		
Seed kernel material (estimated)	780		65
Powdered seed - Total	727	94	61
- Active	472	61	39
- Inactive	255	33	22

Table 5.1 Recovery rates following seed processing

As the figures stand it can be seen that the process of crushing and sieving the seed kernel material is carried out with minimal wastage, in this case 6%. However, of concern is the fact that if this figure is added to that of the inactive powdered seed (which may be considered wasted) then the overall efficiency of the process drops from 94% to 61%. In terms of overall recovery from the original quantity of seed collected the recovery rate of active seed powder material is 39%. In practice this would mean that in order to produce 1 tonne of active seed powder, 2.54 tonnes of whole seed would be required. If the process were 100% efficient (assuming a 25% deduction arising from the seed husk) 1.3 tonnes of whole seed material would be required.

Although the processing technique used provided sufficient seed for the field trials, if the seed powder was the form to be adopted for use in large urban treatment plant then alternative mechanical methods of processing would have to be used to improve recovery rates and sustain commercial viability.

5.2.3 Seed storage

No definitive studies have been conducted with respect to storage methods and conditions of whole seed and/or seed processed seed powder. In Malawi the dry whole seed and processed seed powder were stored in a covered, dry area for periods up to 6 months with no sign of degradation

or pest attack. Powdered seed samples have been stored in sealed containers and used in the laboratory at Leicester University for over a year with no measurable loss in activity.

5.2.4 Seed harvesting and processing - effects on coagulation efficiency

In the initial stages of developing the method for processing the seeds, sieving following crushing had been carried out through a no.16 sieve of aperture 1mm. Standard jar tests (as described previously) carried out on the seed powder produced using this method gave excellent clarification but at doses higher than anticipated. Subsequent re-sieving of the powder through a traditional maize sieve (aperture approx. 0.8mm) produced a more effective end product. Figure 5.5 demonstrates the effect of seed particle size on optimum dose to give equivalent turbidity reductions. It can be seen that decreasing the effective particle size reduces the optimum dose required from 100 mg/l to 50 mg/l. The active fraction of the seed kernel is water soluble. Increasing the surface area available from which protein can be leached or solubilised increases the volume of active material produced thereby reducing the dose, in mg/l of kernel material, required.

Seed performance was also found to be affected by the stage at which harvesting took place and the method by which the seeds were allowed to dry. It was discovered that during the initial stages of seed collection from the villagers in Nsanje that, with payment being offered, some seed pods were harvested prior to being fully sun dried on the tree. The seeds were subsequently dried in the sun. Jar tests carried out following processing demonstrated a reduction in efficiency. Figure 5.6 shows the results obtained, optimum dose for the force dried seeds being of the order of 200 mg/l as compared to 50 mg/l achieved using seeds which had been allowed to dry fully on the tree. This reduced effectiveness is considered to be due to the immaturity of the seeds resulting in a reduced protein content. Thereafter, instructions were given to the Forestry workers to refuse acceptance of those seeds that appeared to be immature as a means of quality control. The difference between the two is easily seen, the immature seeds being smaller and the husk significantly thinner and lighter in colour than the mature dry seeds.

Figure 5.5 Effect of seed particle size on turbidity removal
Initial turbidity 400 NTU

Figure 5.6 Effect of maturity and drying on turbidity removal
Initial turbidities - tree dried 400 NTU; sun dried 510 NTU
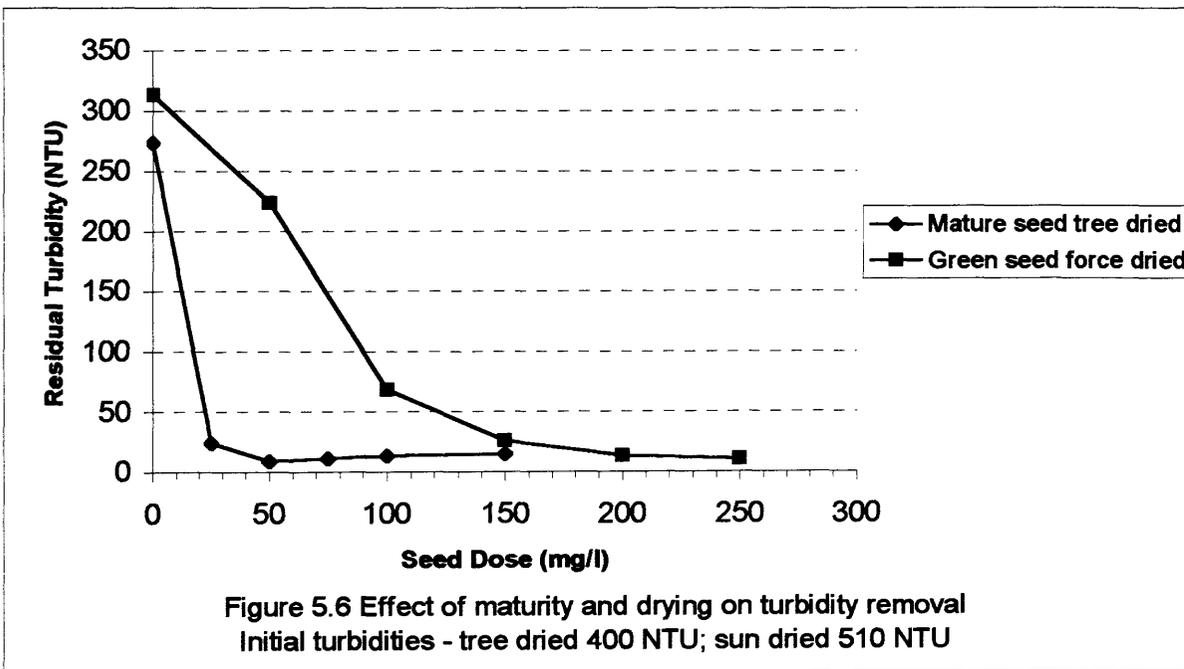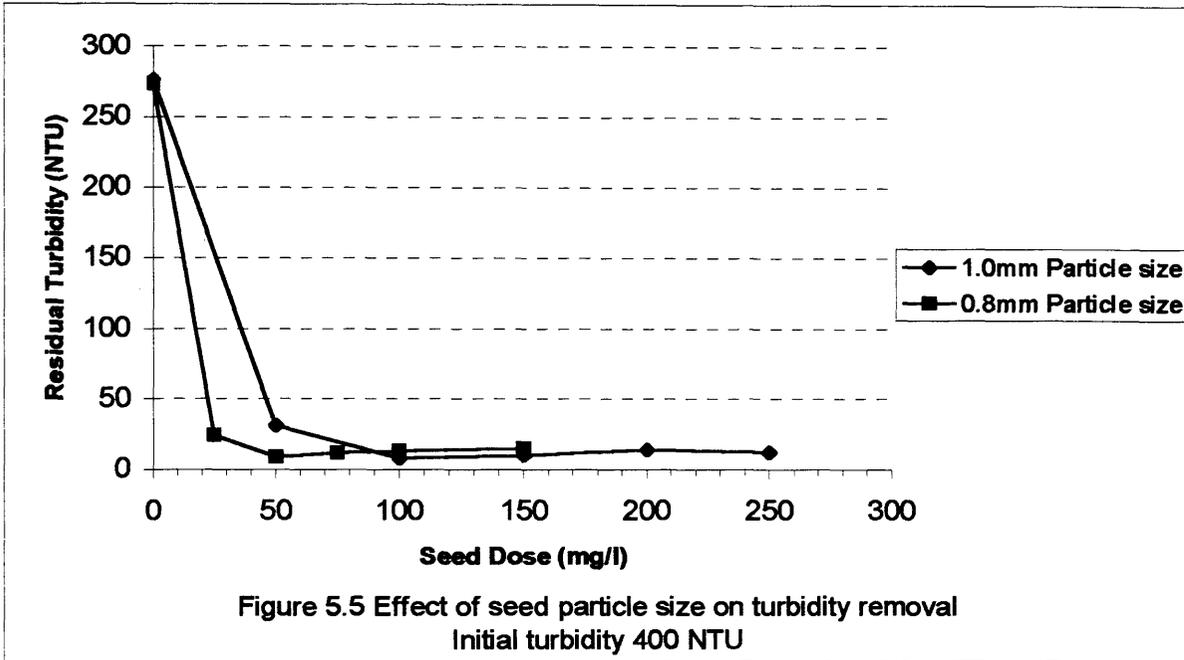
5.2.5 Seed solution preparation

Preparation of seed solutions for the pilot plant studies initially involved the addition of seed powder to the required amount of water to achieve the desired solution concentration. However, this method produced suspensions of reduced activity. On adding to the water it was observed that the powder tended to clump together rather than producing a fine suspension. It is considered that the presence of the seed oil prevents full contact between the seed and water. In order to overcome this a small quantity of water was added to the powdered seed and mixed to a smooth paste. This was then gradually diluted with continuous mixing to the required concentration.

Further problems were experienced in the preparation of large volumes of seed solution for the full scale trials. Initially preparation followed that for the pilot plant i.e. dilution of a seed paste to the required concentration. However, it was found that seed particulate material in the prepared suspension did not settle to the base of the coagulant tank as would have been anticipated. Instead a foam sludge layer formed on the surface. Although performance of the seed was not affected, operationally this could pose problems in determining the actual level of the coagulant in the tank and the requirement for subsequent cleaning of the tanks prior to the make up of further coagulant solutions. The reason for the layer forming at the surface is considered to be due to a combination of trapped air and the high concentration of seed oil present. This effect had not been experienced during the pilot plant trials when settlement of remaining seed particles occurred rapidly. In the short term the problem was overcome by using two tanks for coagulant preparation. Suspensions prepared in one tank were siphoned to the second via a fine mesh sieve to remove the residual seed particles. In the context of the trials the procedure worked well, however, the process cannot be considered a long term solution to the problem. A long term solution is currently being pursued and is outlined in the following section.

5.2.6 Use of *M.oleifera* oil free presscake as coagulant

The problems outlined in the previous sections with respect to seed processing and seed suspension preparation are considered primarily to be a function of the presence of the oil

contained within the seed kernel. The seed kernel contains on average 40% by weight of oil. In a previous study Ademiluyi and Eze (1990) used defatted *M.oleifera* seed in studies to determine the potential for *M.oleifera* as a sludge conditioner. The results obtained showed an improved performance over the non-defatted seed. No information is available on studies to show whether similar performance enhancement may be obtained for potable water treatment.

The study presented here was carried out in collaboration with colleagues from the Natural Resources Institute, Chatham, Kent. In contrast to the study by Ademiluyi and Eze (1990) in which oil extraction was performed using petroleum ether as the extracting solvent, this study was carried out using a physical extraction process.

Seed kernels for the study were obtained from Malawi. Extraction of the oil was achieved using the following procedure. Seed kernels were pre-treated prior to physical extraction of the oil. This comprised roasting followed by milling to a fine paste. The moisture content of the paste was then raised to approximately 18% by kneading it into warm water. The resulting paste was then subjected to extraction using a modified meat mincer. The seed cake produced following oil extraction was oven dried to a solid cake. A small quantity was then ground to produce granules 2-3 mm in diameter. An analysis of the composition of the seed kernel prior to extraction and of the resulting presscake is given in Table 5.2.

Fraction	Composition %	
	Seed kernel	Seed cake
Moisture	4.58	9.85
Crude protein	36.94	46.15
Ether extract	39.72	17.39
Crude fibre	3.63	3.58
Ash/Minerals	3.61	4.5
Nitrogen free extract (by difference)	11.52	18.53

Table 5.2 Composition of seed kernel and presscake

It can be seen from the table that the oil has been extracted at an efficiency of 56%. This is lower than would be achieved from a dedicated oil expeller where efficiencies of between 70 and 80% would be expected (Donkor, 1993). It must be noted that sufficient quantities of seed were not available for the process to be optimised. The analysis also indicates the preservation of protein content through the process.

Determination of the coagulant activity of the presscake was carried out using standard jar test procedures outlined in Chapter 4. The efficiency of the presscake was compared to that of hand crushed whole seed kernels. A model raw water was used for the investigation consisting kaolin clay in deionised water. An ionic background was introduced in the form of sodium bicarbonate to produce a final alkalinity of 350 mg/l as $CaCO_3$. Final pH was in the range 8.0-8.5. Kaolin clay suspension was added to give an initial turbidity of 300 NTU. The use of a high turbidity was considered necessary to produce an effective comparison. Both seed suspensions were prepared at a 1% concentration using the standard test water without the kaolin. Presscake suspensions were prepared by initially making a paste. This was then diluted to the required concentration and used immediately. A similar procedure was used for the hand crushed seed solution.

Figure 5.7 shows the results obtained and demonstrates equivalent performance indicating the presscake to have retained its coagulation properties. Both seed solutions show an optimum dose of 20 mg/l, above the optimum residual turbidities are maintained at a marginally increased value. The figure also shows that repeated tests using the presscake produced little variation in final water quality indicating the homogeneous nature of the presscake.

The preliminary experiments have highlighted a potential problem. It had been expected that by removing the oil and effectively concentrating the protein material, the optimum dose would be lower than for the whole seed. This is evidently not the case. There are two possible reasons for this. Firstly, it is possible that the heat treatment of the seed kernels prior to extraction has resulted in some denaturing of the protein material thereby reducing effectiveness. Secondly, the hard granular nature of the presscake may require that more extensive mixing than was conducted is required to ensure complete solubilisation of the protein material. Both these aspects requires

further investigation, however, the significant result is that the potential exists for two valuable products, oil and coagulant, to be gained from a single resource. The implications of these findings with respect to implementation as a whole are enormous and are discussed further in Chapter 6.

Figure 5.7 Treatment performance of presscake obtained following oil extraction as compared to hand crushed whole seed; Initial turbidity 300 NTU
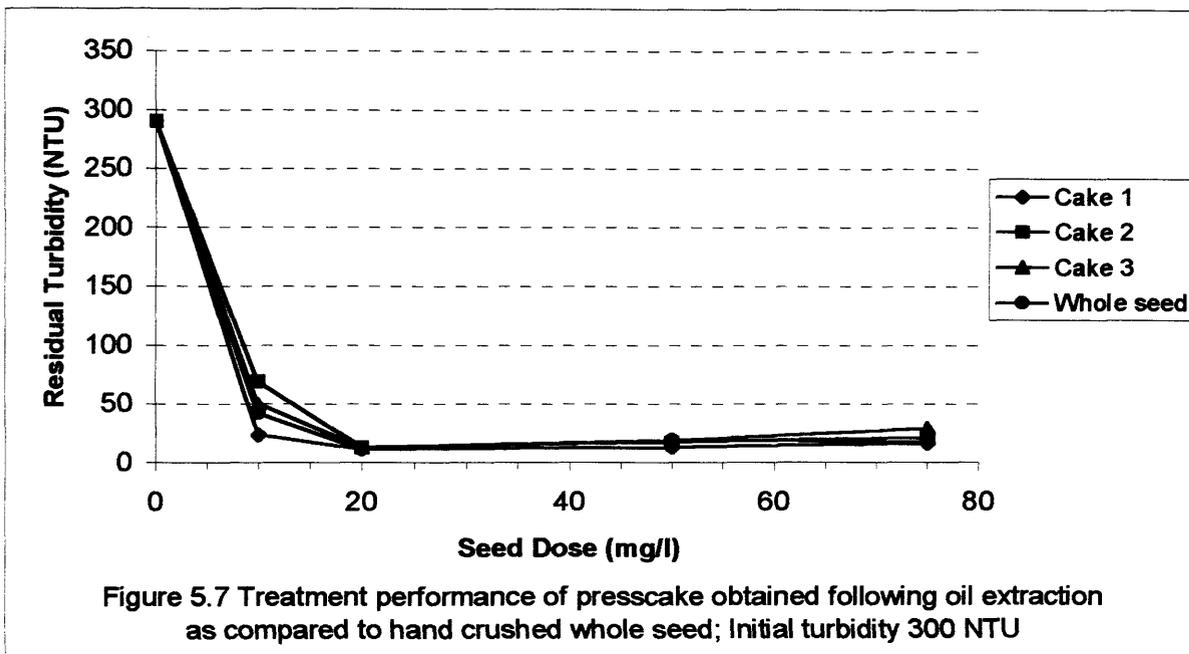
With respect to the operational problems of seed processing and seed suspension preparation that the study was addressing, it is envisaged that use of the presscake would have significant benefits. Firstly, the solid form of the presscake would make it more suitable for mechanical grinding processes. In the whole seed form, commercial maize flour producers in Malawi who were approached during the full scale trials with a view to processing of seed powder, refused. In their opinion the high oil content would result in their machinery becoming fouled. The reduced oil content coupled with the solid nature of the presscake may make it more acceptable for processing. In addition, the problems of sieving the ground presscake would be less than that for the whole seed powder thereby enabling a finer end product to be achieved. The second benefit is envisaged to be a reduction in the time and effort required for coagulant solution preparation.

With the whole seed powder considerable time and effort was required in making up the seed paste prior to dilution to the required concentration. Although it has not been tried in the quantities required for the full scale trials, the laboratory experiments indicate that solution preparation may follow that conventionally used for granular alum i.e. the required amount of coagulant material is added to water and stirred continuously. This is an area that requires further work but the initial signs are encouraging.

One problem that the presscake does not overcome is that of a residual seed particle sludge that remains in the coagulant solution tank following suspension preparation. It is possible that this material could be removed and have potential use as a fertiliser by-product. However, the extent to which this would complicate operations in large treatment plant is unknown.

5.2.7 Stability of seed suspensions

It has been suggested in a number of studies (Jahn, 1981; Jahn, 1986) that seed solutions should be prepared fresh on the day of testing, indeed all tests carried out by the author have been done following this procedure. The reasons why this procedure should be adopted have, as yet, not been scientifically justified. It would indicate, however, that a potential problem of *M.oleifera* utilisation exists with the possible degradation of seed suspensions over time leading to a reduction in coagulation efficiency. Considering the climatic conditions under which it is envisaged that suspensions of *M.oleifera* will be used i.e. hot tropical climates, the potential for accelerated degradation must be taken into account.

In the case of the proteins from *M.oleifera* responsible for coagulation, degradation is likely to occur as a result of the presence of proteases. Proteases are enzymes that effectively digest proteins into smaller lengths. These enzymes may be present in the seed itself or may arise externally within aqueous solutions from the presence of bacteria that may release proteases as part of their natural growth and death processes (Taylor, 1972).

The evidence to date on the extent to which degradation occurs is contradictory. A study carried out on seed samples from Malawi by the paper division of Allied Colloids Ltd. on behalf of

Leicester University indicated a rapid decline in cationic content over a 5 hour period. At 24 hours hydrogen sulphide (H_2S) gas was evident, indicating a break in the protein sulphur bonds (Allied Colloids, 1991). However, the experiences of the author provide contrary evidence. An investigation was carried out whereby seed samples prepared in tap water were incubated at $37^{o}C$ over a period of 4 weeks. Activity was determined throughout the period by standard jar tests using kaolin suspensions. No reduction in activity was observed. The seed sample remained clear, indicating no bacterial growth, and the formation of H_2S was not evident. The contrast in results is considered to be due to either the fact that degradation in the first study was not the result of protease activity, or that the conditions in the second study were such that the protease was not activated. Although some proteases can become activated over a broad range of conditions others require very specific conditions in terms of pH and salt concentration (Taylor, 1972).

The extent to which protease activity will affect coagulant activity must also be considered within the context of the proposed mechanisms by which the proteins effect coagulation. It was hypothesised in Chapter 4 that a dual mechanism involving charge neutralisation and protein precipitation exists. If, for example, protease activity solely effected a cleavage of the di-sulphide bonds between hetero-dimers, as could be the case in the Allied Colloid's study, the result would be the formation of two polypeptides. If both polypeptides maintained the characteristics of the original dimer, in terms of charge and precipitation behaviour, then it is possible that there would be no significant effect on activity. However, if cleavage resulted in a change of polypeptide characteristics, then a significant effect may be observed.

There also exists the possibility that degradation may occur during seed storage. If, under favourable conditions during storage, the seeds have begun to germinate it is likely that the proteases will have been activated. The proteins responsible for the coagulation effectiveness of *M.oleifera* have been postulated to be storage proteins. If this is indeed the case then the function of the proteins will be to provide a readily convertible energy source during germination and initial growth phase. The action of the protease will be to break down the protein into a form that may be utilised for energy conversion. If seeds that have begun to germinate are used for coagulation

there could be an accelerated rate of degradation due to previously activated protease in the germination phase.

The potentially complex interaction between proteases and the active proteins is not an area that has been investigated in any depth but is one that does need to be addressed. However, from a practical point of view, an important aspect that must be taken into consideration is the length of time over which seed suspensions are left during use. In both the pilot and full scale trials presented later in this chapter the maximum length of time between fresh solution preparations was 8 hours. No apparent reduction in effectiveness was observed during this time, which provides further evidence to counter the Allied Colloid findings. If, in subsequent investigations, it is found that degradation occurs but the effect does not become appreciable for, say, 24 hours then it can be argued that from an operational point of view no problem is posed since it is unlikely suspensions will be kept for that length of time. If it is found that degradation occurs in a shorter period of time, for example less than 12 hours, then either methods must be found to reduce or halt the effect, or operational practices may have to be altered to ensure that seed suspensions are used up within a specified time period.

From the above it is clear that there are potential problems with seed suspension stability. Further investigations are required to determine the extent of degradation, the mechanisms behind such degradation processes, the conditions under which such degradation may occur, the extent to which degradation could effect the coagulation process and, if required, methods by which the degradation process may be reduced or halted.

5.3 PILOT SCALE STUDIES

5.3.1 Introduction

As presented in Chapter 3, the majority of previous studies carried out to determine the treatment performance of *M.oleifera* have been on a batch scale irrespective of whether the studies have been laboratory or field based. One exception to this is a study carried out in Burundi (Kaser *et al.*, 1989). In this study a small scale treatment unit was designed and constructed in a rural area

by staff and students at the University of Burundi. The works comprised pre-settling tank, coagulant addition, hydraulic flocculation in bamboo pipes, horizontal sedimentation, rapid gravity filtration and slow sand filtration and was designed to be operated for 8 hours a day producing on average $2m^3$ of treated water in this period (i.e. flow rate of approximately 0.25 m^3/hour). One of the problems with the location of the works was that no power was available to conduct jar testing for dose optimisation, or to allow accurate turbidity measurements to be taken. Thus both operations were carried out by simple visual observation. Results from the study demonstrated the treatment using *M.oleifera* on this works was effective with turbidities in the storage tank maintained below the WHO limit of 5 NTU. However, there are a number of limitations of this study:

i) No data with respect to the operation of the works without coagulant is supplied. It could be argued that at the raw water turbidities encountered, the three filters in series may have reduced the turbidity of the treated water to that obtained during the test runs without the requirement for coagulant. The utilisation of slow sand filters alone to treat raw waters containing less than 50 NTU is a well established treatment procedure (Graham, 1988). It would be reasonable to assume that pre-filtration, such as that provided in this works, could extend the operating range of the slow sand filters to include the medium turbidities encountered in this study.

ii) No information is provided on the nature of the floc that formed, whether settlement was evident or on the extent to which floc were carried over to the pre-filter. The primary aim of chemical addition within a treatment train is to reduce the solids loading on the filters thereby increasing performance both in respect of final water quality and the volume treated prior to filter cleaning.

iii) The trials were discontinued during periods of extremely high turbidities in order to economise on quantities of coagulant used. It would have been useful to have investigated whether treatment performance could still have been maintained under these extreme conditions. An operational situation could be envisaged whereby, following high consumer

84

demand, treatment would have to be continued even during periods of high turbidity in order to maintain sufficient supply.

The pilot scale trials reported here extend significantly the preliminary studies carried out in Burundi. In addition to the main goals outlined in section 5.1, the trials aimed to:

i)   Determine the extent to which filter loading could be reduced over a wide range of raw water turbidities;

ii)   Determine factors in plant design that would affect coagulant performance

The following sections deal with the design of the pilot plant and the subsequent modifications made following commissioning. The final section presents results of the pilot plant studies using *M.oleifera*.

5.3.2 Pilot plant design

It is now regarded as axiomatic that both water and wastewater treatment technology for developing countries must be no more complex than strictly necessary, be robust and cheap to install and maintain. In order to assess the efficacy of using *M.oleifera* within such systems, a prototype treatment works was designed at pilot scale based on this philosophy.

Design criteria for individual 'pilot scale' unit processes are generally not available. Reference can be found in the literature to design and to the suitability of certain unit processes for 'small treatment works'. However, when one considers that a 'small' works can be one that treats 5000m^3/day (Schulz and Okun, 1984), this puts into context the problems associated with obtaining specific design information for a works that may treat, for example, 20 m^3/day. For the majority of unit processes within a conventional treatment train this does not pose a serious problem and design criteria used for large works can be adapted. However, the design of 'small scale' sedimentation tanks poses difficulties. The problem associated with the scaling down of full size sedimentation tanks is that floc size will remain constant irrespective of the size of treatment works. Although it is possible to adjust the physical characteristics of the floc by the addition of

appropriate flocculant aids, in the context of this study where it is the effectiveness of the coagulant that is being investigated, the addition of aids would be inappropriate.

Within the context of the overall aims of the pilot scale studies, the design of the pilot plant was subject to three main criteria:

i) The works should follow, as close as possible, the treatment train utilised in conventional treatment works operating with chemical coagulants viz.: rapid mix - for coagulant dispersion, slow mix - to enable floc formation, solid/liquid separation - by sedimentation and/or filtration;

ii) The treatment processes should function effectively without the need for external power i.e. all processes to be carried out hydraulically. The reasons for this are two fold. Firstly, from an experimental point of view, any process that required electrical power would be subject to frequent power failures during the wet season resulting in significant down times during continuous experimental runs. Secondly, the use of the seeds is seen as being technology appropriate to developing countries. It is logical then that the effectiveness of the treatment should be established on a works that is itself appropriate i.e. technology with low capital, operation and maintenance costs and requiring a minimum of technical knowledge for its operation;

iii) The limited space available on the chosen site for the pilot plant called for the design of compact units.

Space restriction was also the main criteria for the selection of flow rate. As plain horizontal sedimentation was envisaged as the primary method of solid - liquid separation, a flow rate of 1 m^3/hour was chosen thereby keeping the size of the sedimentation tank to within acceptable limits.

The following section outlines the rationale used to determine the selection of unit processes and their design characteristics based on the 1 m^3/hour design flow rate.

5.3.2.1 Intake

The location of the intake on the Nsuwadzi River required that the water be pumped to the pilot works inlet. A diesel engine (Lister Petter type AA1) was chosen as the prime mover for the

86

pump (Mono) to allow continued operation of the works during periods of electrical power failure. A coarse screen on the pump intake pipe prevented large debris from being sucked in.

5.3.2.2 Pre-treatment, dosing and rapid mix

Extremely high raw water turbidities (>1000 NTU) are experienced periodically in the river and some form of pre-treatment had to be provided. A header tank with a 15 minute retention time was provided to remove the gross settleable solids. The header tank also acts to balance the flow within the system as all subsequent processes operate by gravity flow.

The coagulation process involves the addition of chemicals to bring about particle destabilisation prior to flocculation. In particular, with metal salt coagulants the process occurs rapidly with the formation of the hydrolytic products and subsequent particle destabilisation occurring in less than 1 second (Kawamura, 1991). Thus, complete and effective mixing of coagulant with raw water must be effected instantaneously. In contrast, for cationic polyelectrolytes, such as *M.oleifera*, colloid adsorption is a much slower process with the result that instantaneous mixing is not critical. It is the time of mixing that is more significant. One of the aims of the pilot plant trials was to compare performance using alum and *M.oleifera* and hence a compromise had to be reached between the instantaneous mixing requirements of alum and the less critical requirements of *M.oleifera*. The turbulence created by the flow over a 20° V- notch weir was chosen as the method by which initial intense agitation of the coagulant with raw water could be achieved. By ensuring that turbulent flow existed in the pipe work following the header tank, a further 11 seconds of intense mixing was provided at a G value of 468 s^{-1} (value obtained by calculation using headloss within the pipe as the energy input into the system − P in equation 5.1 detailed in the next section).

Coagulant solution was dosed into the nappe of the 'V'-notch weir via a peristaltic pump operating off mains converted D.C. voltage. A battery back up was available for use during periods of mains power failure. Although solution feeders that operate without electrical power are available, it was decided that, for experimental purposes, peristaltic pumps would provide a more accurate method of dose control.

5.3.2.3 Flocculator design

Flocculation is the process whereby electrolytically destabilised particles undergo gentle agitation allowing particle collisions such that agglomeration will result in the formation of floc that may be removed by settlement and filtration. Agitation should be of such intensity that particle collisions and aggregation can occur but not so high that formed floc are broken due to the imposition of excessive shear forces. The design of flocculators is based primarily on the required velocity gradient, G (s^{-1}), and retention time, t (s). It must be noted however, that the use of the G value as a design parameter may not be entirely valid but has been adopted mainly due to the ease of calculation (Liu *et al*, 2000). Flocculators come in two forms, those that require an external power source to rotate mechanical stirrers to provide the agitation and those that rely on gravity flow termed hydraulic flocculators. The discussion of alternative forms of flocculator presented here will be limited to the latter type.

For all flocculators, the velocity gradient, G, may be calculated from the following basic equation developed by Camp and Stein (1943):

$$G = (P/\mu V)^{0.5} \dots\dots\dots(5.1)$$

In equation 5.1, P represents the power required to provide the turbulence for mixing. In flocculators that operate under gravity flow, the power requirement is a function of the headloss i.e. $P = Q\rho gh$. Hence, for hydraulic flocculation equation 5.1 may be re-written:

$$G = (Q\rho gh/\mu V)^{0.5} \dots\dots\dots(5.2)$$

Where

$\quad G$ = velocity gradient (s^{-1})

$\quad Q$ = flow rate (m^3/s)

$\quad \rho$ = density of water (kg/m^3)

$\quad g$ = gravitational constant (9.81 m/s^2)

$\quad h$ = head loss (m)

$\quad \mu$ = dynamic viscosity (kg/ms)

$\quad V$ = volume of unit (m^3)

By controlling the headloss it is possible to control the intensity of mixing. Over the years a variety of hydraulic flocculators have been developed utilising various methods of energy transfer to provide fluid motion. The most widely used type is the baffled channel flocculator. Mixing is accomplished by reversing the flow of water through channels formed by baffles. The major advantages of such systems are their ease of construction and reduced risk of short-circuiting, a major problem with many mechanical flocculators (Schulz and Okun, 1984). The primary disadvantage with respect to the design being considered was that for the flow rate to be maintained such that adequate headloss could be achieved in the channel required the baffles to be placed very close together. It was envisaged that this would cause problems in the construction of the units and cleaning of the units during operation. Also, the water under consideration may require different flocculation conditions dependent on raw water quality. For large treatment works this problem may be overcome by adding or removing baffles as required, thereby altering the velocity and headloss in the channel. However, at pilot scale this would not only have been difficult to achieve in terms of construction but would be a time consuming practical operation.

Hydraulic jet-action flocculators were a second alternative considered at the initial design stage. In these the jet action of the influent flow into a chamber is used to impart a rotational movement to the water. They are currently extensively used in China and Latin America (Schultz and Okun, 1984). Although effective, the main disadvantages were considered to be the difficulty of construction and the reported high degree of short-circuiting. The short time span available for the field studies did not warrant the risk of designing and constructing flocculators that might, at a later date, require extensive modifications to ensure effective performance.

A final alternative that was considered and eventually selected was gravel bed flocculation (GBF). As the name suggests GBF employs packed gravel beds to divert the flow allowing continuous re-contacts of floc and destabilised particles. Previous studies have demonstrated the effectiveness, both experimentally and operationally, of the process (Kardile, J.N. 1978; Bhole and Potdukhe, 1983; Schulz and Okun, 1984; Deshpande *et al.*, 1986;) and their use has been recommended primarily for small treatment plant. The advantages of GBF are that the units are compact, simple

to construct, easy to clean, are subject to a minimal risk of short circuiting and can be flexible in terms of required velocity gradient. A further advantage that has been reported is the ability of the system to store floc within the interstices of the gravel or to settle floc on top of the gravel. No figures have been presented to indicate the extent of in-bed deposition, however, the end result is a reduced solids load on subsequent sedimentation and filtration stages.

The design of the units was based on the following equations assuming laminar flow across the bed (Schulz and Okun, 1984):

$$G = (Q\rho gh/\mu\alpha V)^{0.5} \quad\quad\quad (5.3)$$

$$h = \frac{f(1-\alpha)Lv^2}{\theta\alpha^3 dg} \quad\quad\quad (5.4)$$

$$f = \frac{150(1-\alpha)}{R_N} + 1.75 \quad\quad\quad (5.5)$$

$$R_N = \frac{dv\rho}{\mu} \quad\quad\quad (5.6)$$

Where

G = velocity gradient (s^{-1})

Q = flow rate (m^3/s)

ρ = density of water (kg/m^3)

g = gravitational constant (9.81 m/s^2)

h = head loss (m)

μ = dynamic viscosity (kg/ms)

V = volume of unit (m^3)

α = porosity (≈ 0.4)

f = friction factor

L = depth of gravel bed (m)

θ = shape factor (≈ 0.8)

R_N = Reynolds number

d = average size of gravel (m)

v = face velocity (m/s) = Q/A   where A = x-sectional area (m^2)

From equations 5.3 to 5.6 it can be seen that the velocity gradients induced in the gravel bed are a function of headloss, which itself is a function of (i) the size and shape of the gravel, (ii) the rate of flow, (iii) the cross sectional area of the bed.

90

By sequential substitution of the above equations, a single relationship can be obtained for G:

$$G = \sqrt{\frac{[150(1-\alpha)A\mu + 1.75Q\rho d](1-\alpha)Q^2}{\theta d^2 \mu \alpha^4 A^3}} \quad \ldots\ldots\ldots(5.7)$$

Knowing the design flow rate $(Q = 2.78 \times 10^{-4} \text{ m}^3/\text{s})$, water density and dynamic viscosity and taking the assumed values for bed porosity $(\alpha=0.4)$ and gravel shape factor $(\theta=0.8)$ and assuming a square unit section $(A=D^2)$ and an average gravel diameter of 0.015m, the equation above reduces to:

$$G = \sqrt{\frac{0.894D^2 + 0.07}{D^6}} \quad \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots(5.8)$$

Using an iterative process a value of D is assumed from which G may then be calculated. The value of D may then be increased or decreased depending on the value of G obtained and the calculation repeated. It has been claimed in previous studies that a 5 minute retention time within a GBF produced equivalent performance to that obtained from a 15 minute jar test flocculation period (Wagner, 1982; Schulz and Okun, 1984). It was therefore decided to base the design on G values used during standard jar tests carried out in the laboratory studies reported in Chapter 4. Data supplied with the Aztec jar test apparatus used during the study indicated that a G value of approximately 9.9 s^{-1} would be expected at a paddle rotation speed of 20 rpm. Substituting this value for G into equation 5.8 provides a value for **D = 0.35m**.

The depth of bed required may be determined by considering the retention time within the bed for effective flocculation. Although previous studies had indicated a shorter flocculation time than used in standard jar tests was required for GBF, it was decided to err on the conservative side and provide for an extended flocculation period, in this case providing a 10 minute retention time. The retention time is given by the following equation:

$$T = \frac{AL}{Q} \quad \ldots\ldots\ldots(5.9)$$

Where:

T = retention time (s)

A = face area (m²)

L = bed depth (m)

Q = flow rate (m³/s)

Substituting into equation 5.9 the known values of T, A and Q provides a bed depth L = 1.35m.

A sheet metal grating was provided to support the gravel. In summary the grating consisted of 1,369 ports of diameter 5mm at a spacing of 8mm. Total headloss due to the grating was 0.02m with a velocity gradient through each individual port of $22s^{-1}$. Although higher than in the bed itself, this value of G was not considered high enough to cause damage to the formed floc.

To ensure easy removal and replacement of the gravel as and when required one side of the flocculator was bolted to the others functioning as a full depth, removable panel.

5.3.2.4 Sedimentation tank design

A variety of different methods may be employed to separate formed floc from the treated water. Horizontal plain sedimentation was chosen for the pilot plant due to its simplicity of construction, ease of operation and tolerance to shock loading. It also offered the possibility of increasing throughput by inserting high rate tube settlers at a later date. As previously noted, one of the major disadvantages of plain sedimentation for small scale plant, such as the one proposed, is that of scaling down the design. Even if the hydraulic flow characteristics have been properly established, it is difficult adjust the physical characteristics of the floc (Kawamura, 1991). One technique that may be employed is the use of flocculation (floc) aids. However, in the context of this work investigating the sole use and performance of primary coagulants, floc aids were not considered appropriate. Despite this drawback the advantages of plain sedimentation were considered to outweigh the difficulties posed in design.

A review of the literature available also highlighted the difficulty in obtaining design information for small sedimentation tanks. In general, the initial design of a sedimentation tank is based on the

expected settling velocities of the flocculated particles to be removed, given in its idealised form by equation 5.10:

$$v_s = \frac{Q}{A} \quad \text{.................(5.10)}$$

Where

v_S = settling velocity (m/s)

Q = rate of flow (m^3/s)

A = surface area of tank (m^2)

The ratio Q/A is commonly known as the surface loading or overflow rate expressed in m/h. Knowing the settling velocity of the particles to be removed and knowing the design flow rate the physical basin size, in terms of surface area, may be established. However, at the time the design was being considered information relating to the settling velocity of flocculated particles at the Thyolo site was unavailable. The jar test studies on the Thyolo water, as reported in Chapter 4, considered solely the effectiveness of the treatment in terms of turbidity reduction. Although observations were made on floc structure and setting ability, no discrete settling measurements were taken. The design was therefore based initially on a required retention time to ensure complete settlement. As a 2 hour quiescent settling period had proved to be adequate for the jar tests carried out previously, it was decided to design conservatively and allow a 4 hour settling period as the behaviour of *M. oleifera* floc under dynamic conditions was unknown. At a flow rate of 1 m^3/hr this required a settling basin of total volume 4 m^3. The physical shape of the tank was determined using the suggested guidelines of Smethurst (1979) of a length to width ratio of 3:1. Using a length of 3m and width 1m from these guidelines and knowing the volume required gives a tank depth of 1.33m. For a surface area of 3m^2 and flow rate 24 m^3/day equation 5.9 gives an overflow rate of 8 m/day. Although this is lower than would generally be expected, an overflow rate of 20 - 30 m/day is given by Schultz and Okun (1984) for 'small' works, Smethurst (1979) recommends 9 m/day for works operating under very bad conditions without coagulant aids being used.

The inlet to the tank consisted of a diffuser box and full width baffle placed 0.3m from the inlet to a depth of 1.5m below the water surface. Both the diffuser box and baffle were designed to ensure and even distribution of the inlet flow over the full width of the tank. The outlet launder consisted of a full width rectangular weir plate.

In order to assist cleaning of the tank the base was sloped at an approximate 5° angle from the outlet to the inlet.

5.3.3 Construction and installation

All the units were fabricated in sheet steel of gauge SWG16. The header/mixing tank and GBF unit were fabricated in the Civil Engineering Department at the Polytechnic. Fabrication of the sedimentation tank was subcontracted out to a local firm.

In order to support the weight of water in the sedimentation tank, and the weight of water and gravel in the GBF unit, concrete foundations were provided. In addition, a brick plinth and working platform were constructed for the header tank and associated dosing apparatus.

Shelters were constructed over the header and sedimentation tanks to prevent possible flooding of the units during heavy rain. In the case of the sedimentation tank the roof shelter was augmented by side screens to provide shade in an attempt to reduce hydraulic 'short circuiting' occurring as a result of solar heating.

5.3.4 Operation

Referring to the schematic shown in Figure 5.8, the raw water is pumped via a 2" diameter pipe to the header tank. Flow rates were measured and controlled using the rotameter (R) and valve V1 located immediately prior to the header tank. Valve V1 also acted as a pressure release safety valve. Coagulant was dosed approximately 3 cm above the nappe of the weir discharge prior to the header tank exit. The dose of coagulant was controlled using variable speed peristaltic pumps supplied from DC power packs operating off mains electricity. A battery back up system was available for use in the event of mains power failure.

Following rapid mix the treated water enters the GBF unit. Two valves, V2 and V3, prior to the sedimentation tank allow the GBF unit to be emptied and cleaned without affecting levels in the sedimentation tank. Valve V4 drains the sedimentation tank. Cleaning of all units was effected using a hose pipe and mains water supplied from the Thyolo Treatment Works.
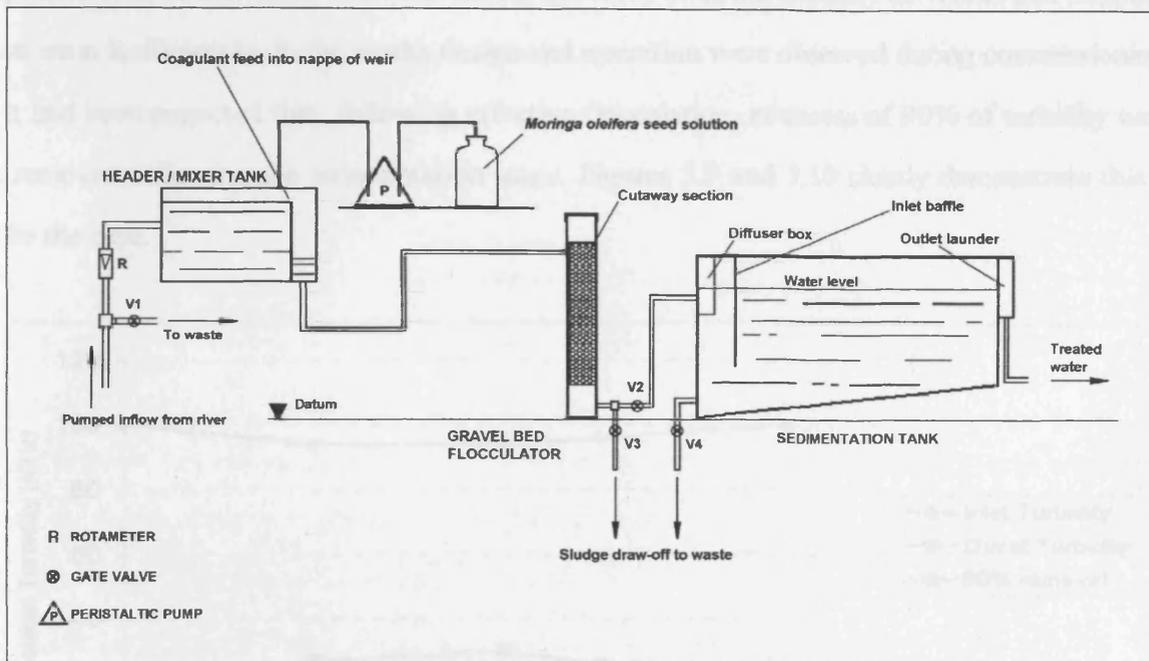
Figure 5.8 Schematic diagram of initial pilot plant layout

5.3.5 Water quality analyses

The reduction in turbidity was the main parameter used to determine the effectiveness of the treatment process. Turbidities were measured using a Hach Ratio XR Turbidimeter. In addition to turbidity, basic raw and treated water characteristics viz.: temperature, alkalinity, hardness and conductivity, were also monitored. It was not possible for pH to be measured due to damaged equipment.

95

5.3.6 Commissioning and modifications

The primary aims during commissioning were to establish the hydraulic flow characteristics of the works and to optimise the procedures for operation. Commissioning was carried out using aluminium sulphate (alum) as the coagulant. The alum used was a commercial granular grade imported from South Africa and obtained for the study from the Ministry of Works and Supplies. Two main inefficiencies in the works design and operation were observed during commissioning:

i) It had been expected that, following effective flocculation, in excess of 90% of turbidity would be removed following the sedimentation stage. Figures 5.9 and 5.10 clearly demonstrate this not to be the case.

Figure 5.9 Initial commissioning of pilot works using alum
Alum dose 65 mg/l

Figure 5.10 Initial commisioning of pilot works using alum
Alum dose - 80 mg/l for 0-23 hours; 50 mg/l for 23-32 hours
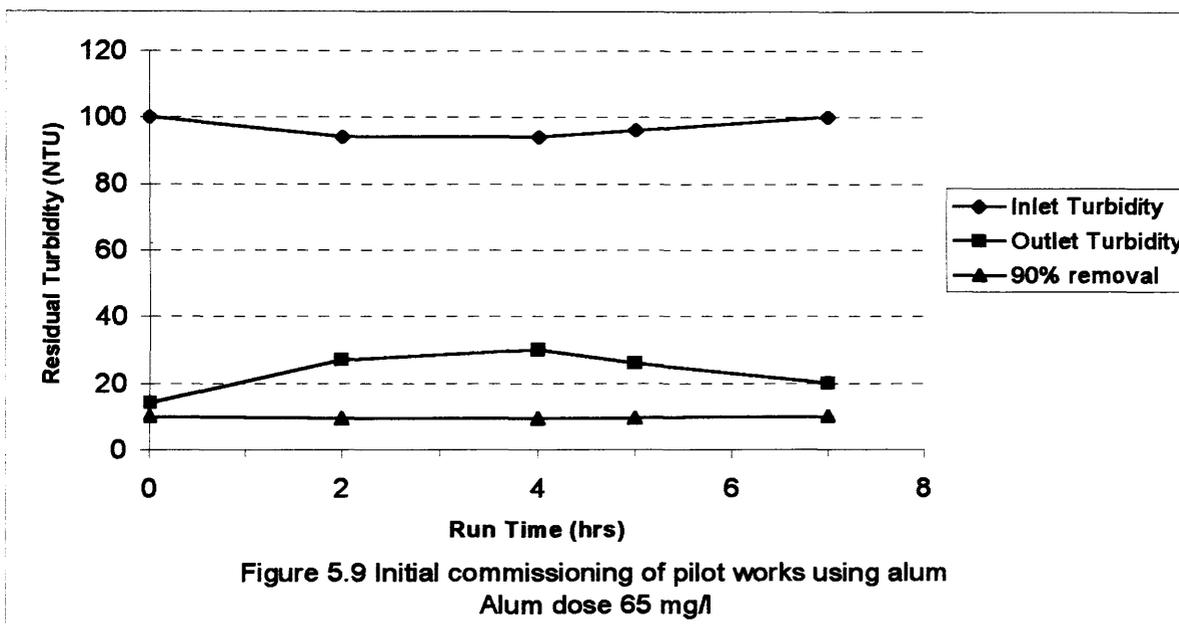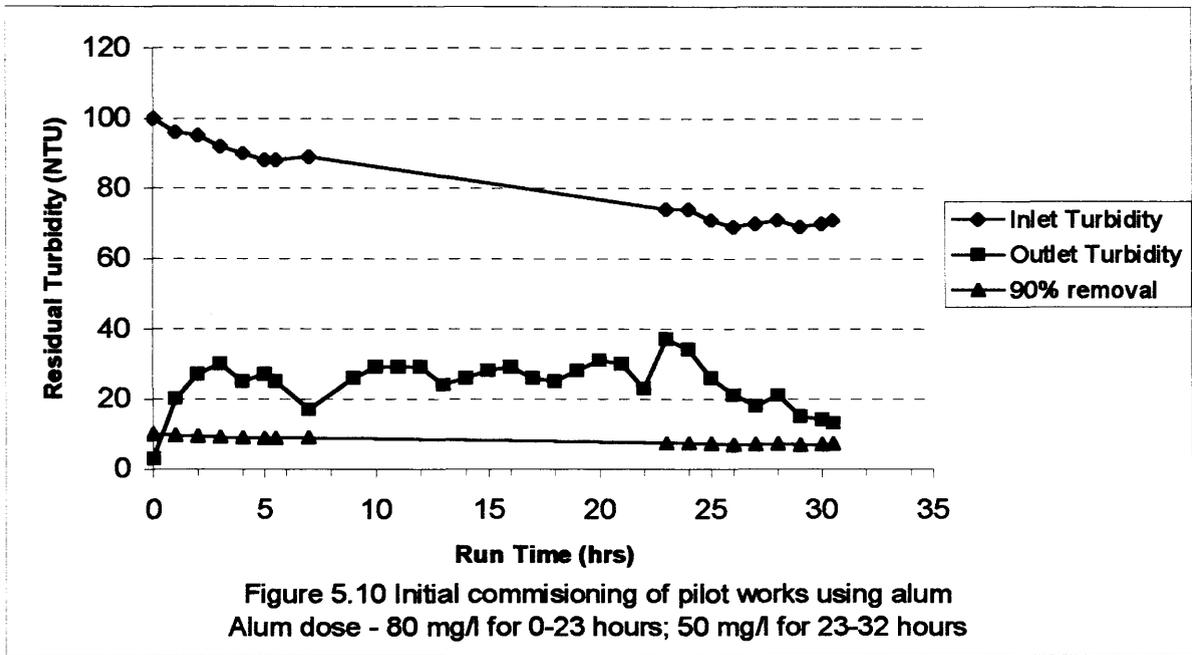
In a review of the design calculations relating to flocculator design, two factors were identified as being the cause of the incomplete flocculation observed. Firstly, an error was found in the original equation used to calculate the retention time. Equation 5.9 calculates retention time based on the total volume of the GBF unit, however, the equation neglects the fact that with the addition of the gravel the volume of the unit is effectively reduced. By inserting the porosity of the bed media into the equation a more realistic retention time is obtained. Equation 5.9 now becomes:

$$T = \frac{\alpha A L}{Q} \quad ....(5.11)$$

Using a value of α = 0.4 reduces the original design retention time of 10 minutes to 4 minutes. The second factor related to gravel size. Although rounded river gravel with an average diameter of 15mm had been requested, gravel with an average diameter of 50mm was delivered. There was insufficient time during the original study to find an alternative source. By inserting this figure into equation 5.7 and using the other known parameters a velocity gradient, G value, of 4.1 s⁻¹ is obtained. The combined factors of a reduced retention time and a lower G value were considered to be responsible for the inefficient operation of the GBF unit. In order to overcome this problem 4 further GBF units were constructed using the same design principles as for the first. With the

same design parameters as used previously, with the exception of an increased average gravel diameter, 4 units were obtained with the following characteristics: face area of square section 0.32m x 0.32m; bed depth 1.5m; average G value throughout the bed of 12 s^{-1}; nominal retention time 4 minutes (nominal total retention time 16 minutes). Reducing the face area increases the velocity of the water in the bed thereby increasing the velocity gradient G. The slightly higher design G value in the new flocculators was chosen in order to provide in effect tapered flocculation (Schulz and Okun, 1984), G values reducing from 12 s^{-1} to 4 s^{-1} in the final original flocculator. A total retention time of approximately 20 minutes was provided. Figures 5.11 and 5.12 show examples of runs carried out during commissioning of the new GBF units. Comparison with Figures 5.9 and 5.10 shows a significant improvement in final water clarity and a lower dose requirement.

Figure 5.11 Commissioning of new flocculators using alum
Alum dose 65 mg/l

Figure 5.12 Commissioning of new flocculators using alum
Alum dose 65 mg/l
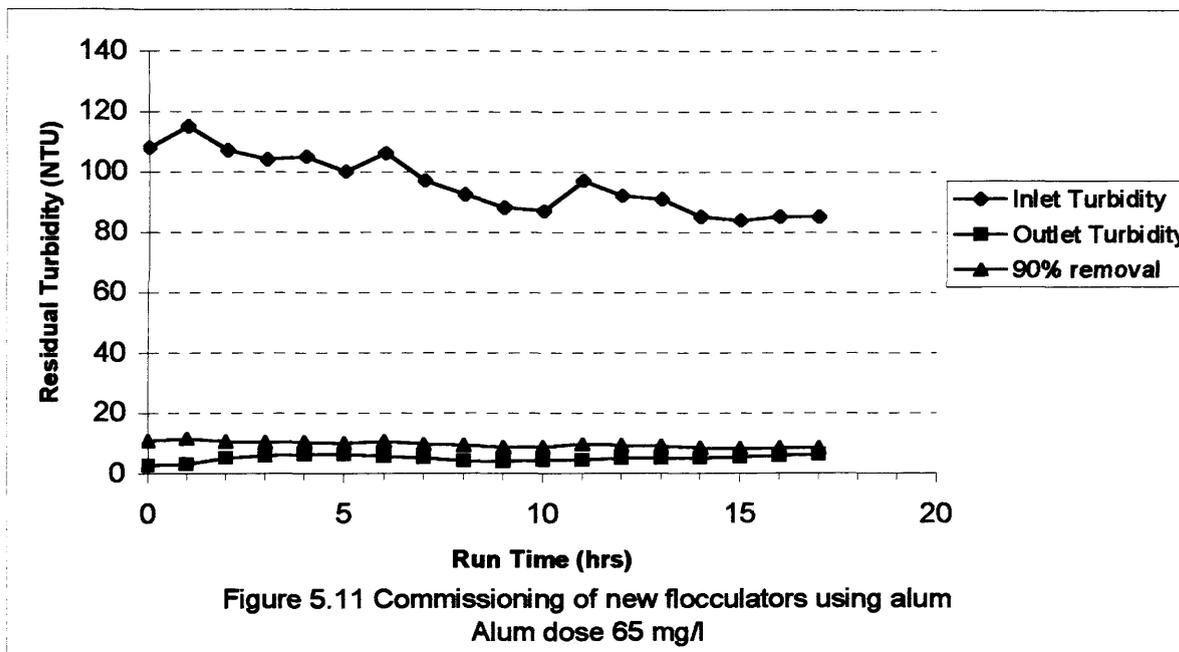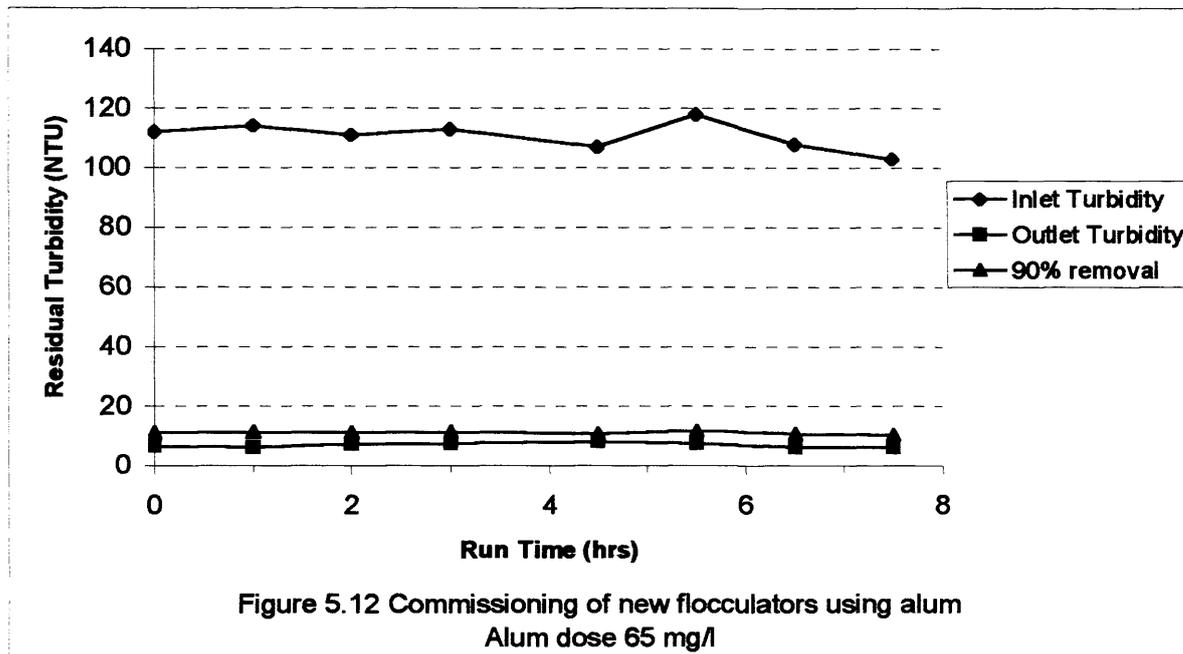
ii) The second inefficiency is one that is frequently observed in treatment works operating in tropical climates and involves the formation of temperature induced density currents within the settling tanks. The effect may be caused by either a rise in the temperature of the influent over that of the bulk of the water or, conversely, a reduction in the temperature of the influent. In the first case, the tendency is for the warmer influent to rise above the bulk of the water and short-circuit across the surface of the tank to the outlet launders. In the second case, the tendency is for the influent to submerge under the bulk of the water and short-circuit across the base of the tank before rising to the outlet launders. In both cases the short-circuits can reduce the retention time in the tank to a fraction of the nominal retention time. It has been reported that a $0.5^{\circ}C$ difference in temperature can cause density currents to occur (Montgomery, 1985; Hahn, 1990; Kawamura, 1991). In the case of the pilot plant in this study the former effect was more noticeable. In general the sedimentation tank was left full after the end of each experimental run with the result that during the night the water in the tank cooled. By the time a new run began on the following day (runs normally commenced at around 10:00 a.m.) the temperature of the water in the river had begun to rise. As the day progressed the river water temperatures continued to increase. This was

99

compounded by the additional heating effect of the water passing through the pipe work and the metal flocculators, both of which were fully exposed to the sun. The overall result is shown schematically in Figure 5.13. As the run progresses the influent to the sedimentation tank quite rapidly rises to the surface and short-circuits to the outlet weir. Figures 5.14 and 5.15 provide examples of the effect on outlet turbidity of the short-circuit for both alum and *M.oleifera*. The rise in the temperature differential is followed closely by a rise in outlet turbidity. The effect can be seen to be more pronounced for *M.oleifera*. This is considered to be due to the higher initial turbidity of the run and the lighter floc produced by *M.oleifera*. Additional density currents were also observed in the sedimentation tank arising from heating of the sides of the tank. The result of this was to bring to the surface settling floc from along the heated edge. This flow was observed to combine with the surface short-circuit current.

Inlet baffle

From flocculators

Inlet baffle

From flocculators

Inlet baffle

From flocculators
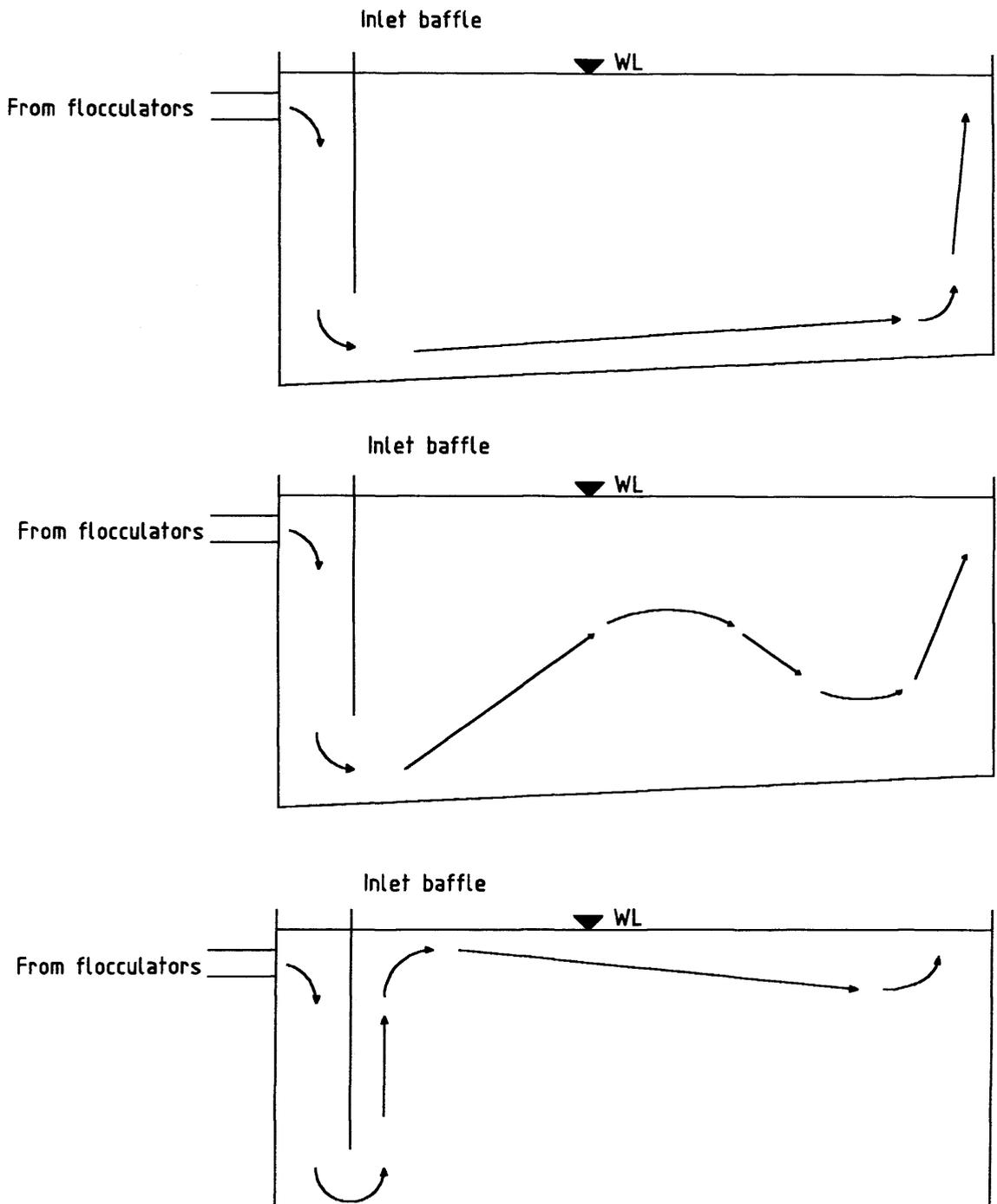
Figure 5.13 Schematic representation of the formation of temperature induced density currents within the sedimentation tank.

There is little that can be done to overcome density currents once they have been established (Kawamura, 1991). It has been suggested that the use of long, narrow (length to width ratios in

the order of 6:1) and shallow sedimentation basins can help to reduce the overall effects of temperature density currents (Kawamura, 1991).
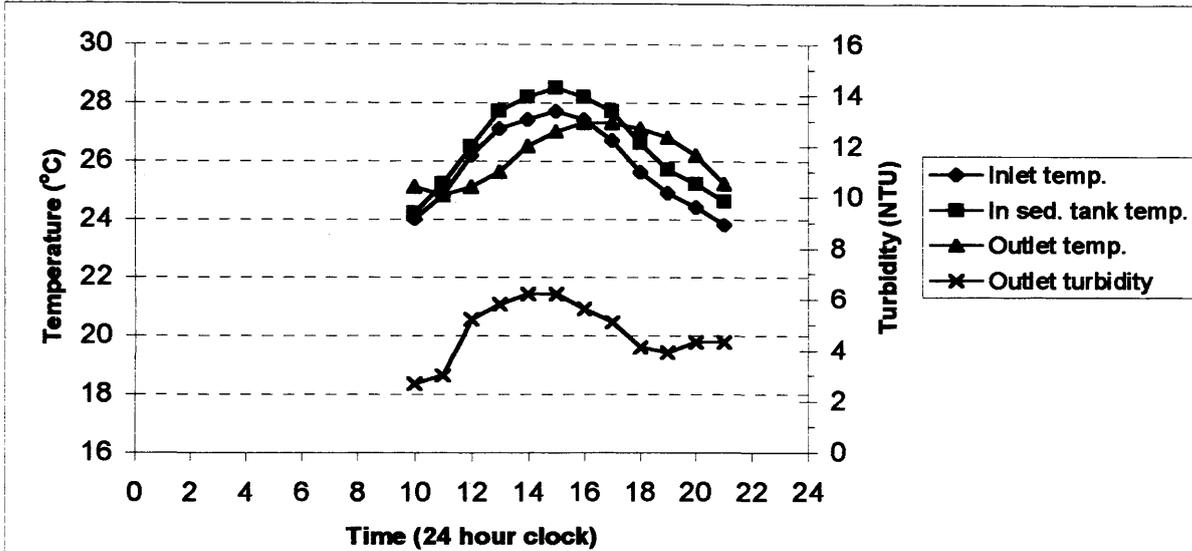
Figure 5.14 Effect of temperature induced density currents on treatment performance
Run carried out using alum as coagulant
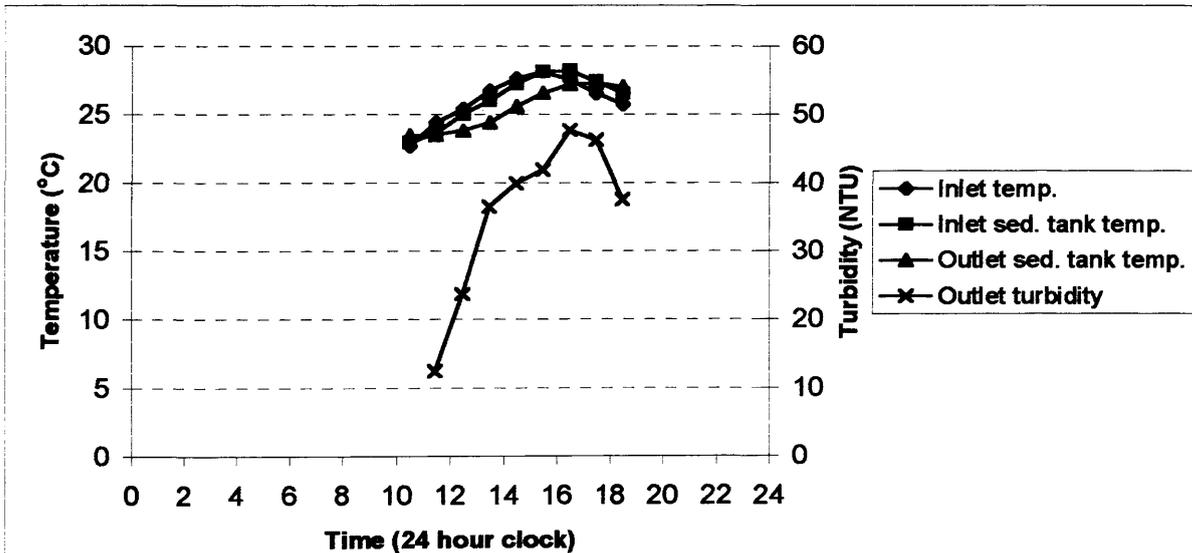
Figure 5.15 Effect of temperature induced density currents on treatment performance
Run carried out using *M.oleifera* as coagulant

In an attempt to reduce temperature effects in this case, side shade screens were provided for both the flocculators and sedimentation tank. For the former the aim was to reduce any additional heating through the metal sides of the flocculators. For the latter the aim was to reduce edge density currents. Both remedies were observed to be relatively successful although density currents arising from the general increase in river water temperature still occurred.

Two further modifications were made following initial commissioning. Firstly, the diffuser box at the inlet to the sedimentation tank was removed. It had been observed that rather than directing the flow across the full width of the diffuser baffle it was creating a jet action that forced the influent across and down over approximately one-third of the baffle width. Removal of the box resulted in a more even flow distribution. Secondly, a rapid gravity sand filter was placed at the outlet to the sedimentation tank in order to provide an indication of final water clarity removing residual floc carry over from the sedimentation tank. The filter was constructed from an oil drum filled with sand (Effective Size 0.62mm and Uniformity Coefficient 1.67) to a depth of 0.7m. A temporary backwash system was installed using river water as the wash water. This arrangement proved effective provided the river water turbidity was less than 100 NTU. During periods of heavy rain when the river was in flood, the sand was removed and washed by hand. A schematic of the works following the modifications is presented in Figure 5.16, whilst Figures 5.17 and 5.18 show details of the modified pilot plant.
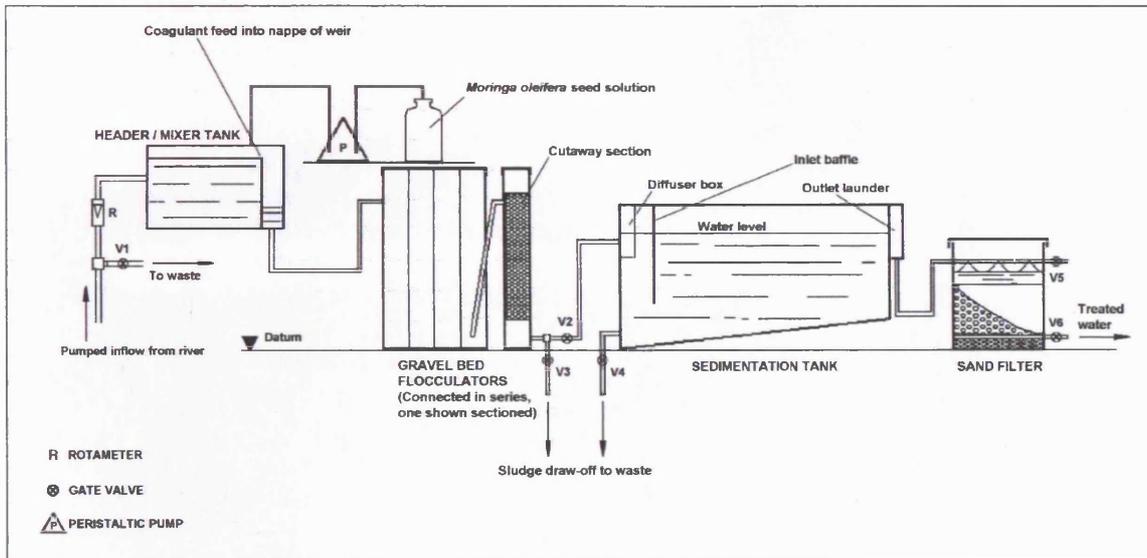
Figure 5.16 Schematic diagram of modified pilot plant layout
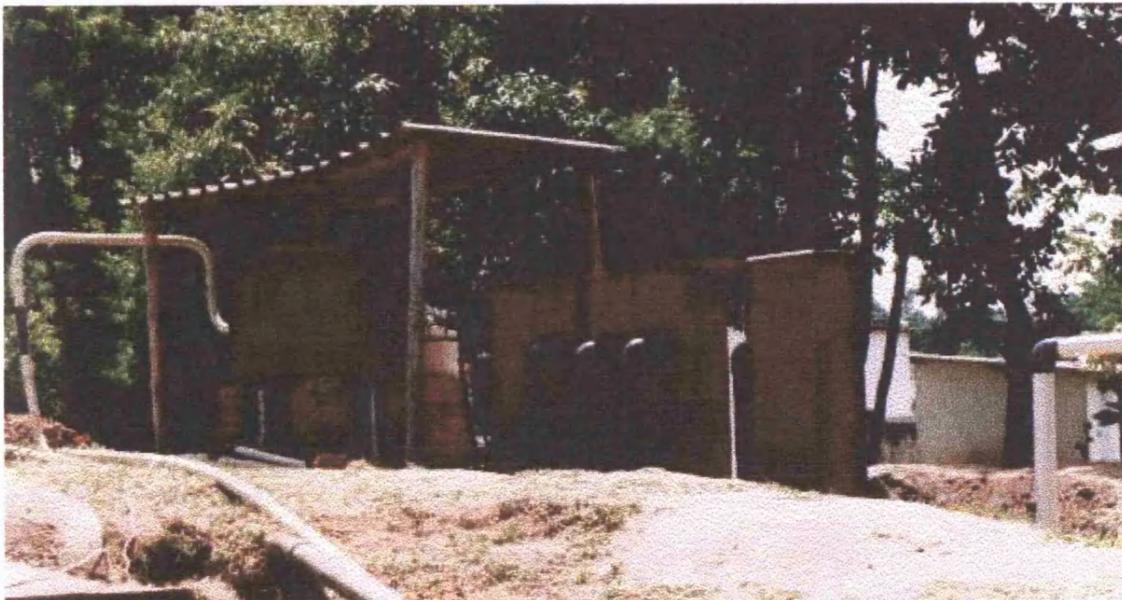
Figure 5.17 Photograph showing header tank and modified flocculation units

Figure 5.18 Photograph showing sedimentation tank and rapid gravity filter

5.3.7 Experimental procedure

Prior to the start of each run a jar test was carried out to determine the optimum dose. If the quality of the raw water changed during the run period, further jar tests were carried out to ensure that the optimum dose of coagulant was maintained.

During the initial stages of commissioning, the flocculators were cleaned prior to the start of each run. It was observed that with this procedure it took longer for floc to be produced than if the beds had not been cleaned. The flocculators had a run in time similar to that experienced with sand filters. With all subsequent experimental runs the flocculators were only cleaned if they had been subjected to extreme loading such as those experienced during flood conditions.

The sedimentation tank was emptied and cleaned at least once a week. As experimental runs were not necessarily carried out on every day there was a danger that by leaving the tank un-cleaned anaerobic decomposition may have ensued within the accumulated sludge layer.

5.3.8 Results and discussion

The results presented were obtained from a series of experimental runs carried out between 1991 and 1994 and are not necessarily presented in chronological order.

This section is divided into 4 sub-sections. The first and second consider results obtained from treatment studies on high and medium raw water turbidities respectively. At no time during the studies did turbidity levels fall below 50NTU thereby enabling the treatment of low turbidity to be examined. The third section describes a significant finding relating to the removal of iron and manganese. The final section deals with the design of the pilot works and considers modifications to improve performance, particularly with respect to *M.oleifera*.

5.3.8.1 High turbidity

Figures 5.19 - 5.22 show examples of results obtained from individual runs. Note that, for clarity, all figures show residual turbidity plotted on a log scale on the Y axis. The 90% removal line is included as a 'marker' to demonstrate the effectiveness of the sedimentation process. The figures clearly demonstrate the effectiveness and consistency of the treatment with in excess of 90% solids being removed following flocculation and sedimentation. Outlet turbidities from the filter were consistently below the WHO guideline value for potable water of 5 NTU.

Figure 5.19 *M.oleifera* pilot works run
Dose 100 mg/l

Figure 5.20 *M.oleifera* pilot works run
Dose 100 mg/l

Figure 5.21 *M.oleifera* pilot works run
Dose 100 mg/l

Figure 5.22 *M.oleifera* pilot works run
Dose 100 mg/l
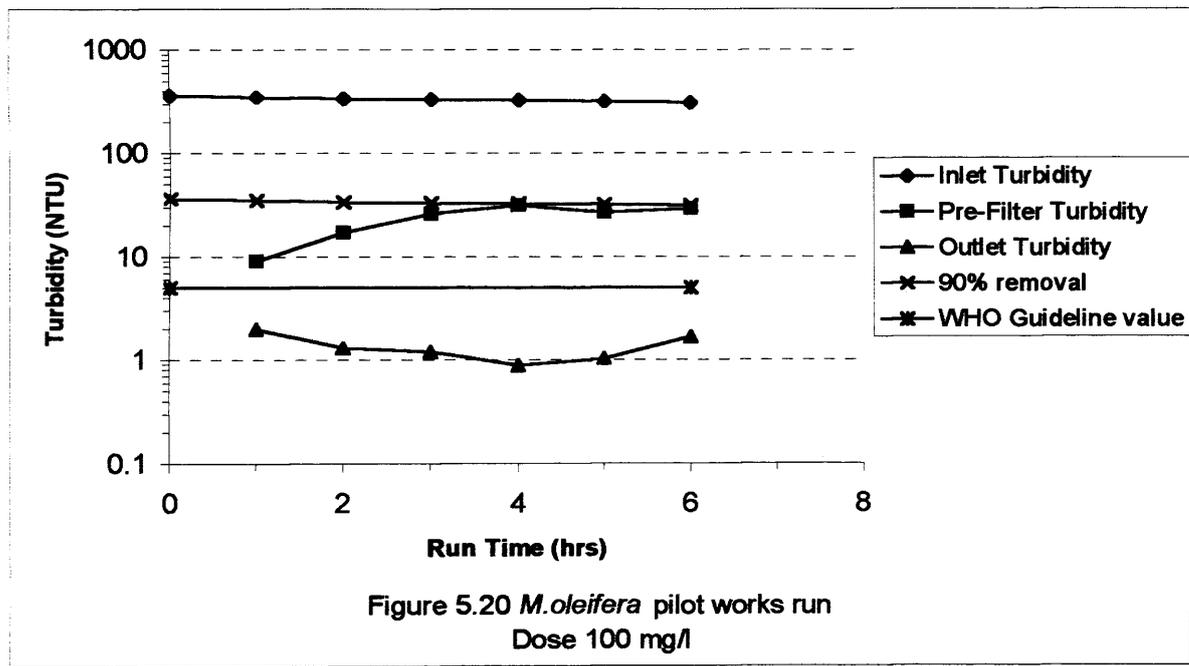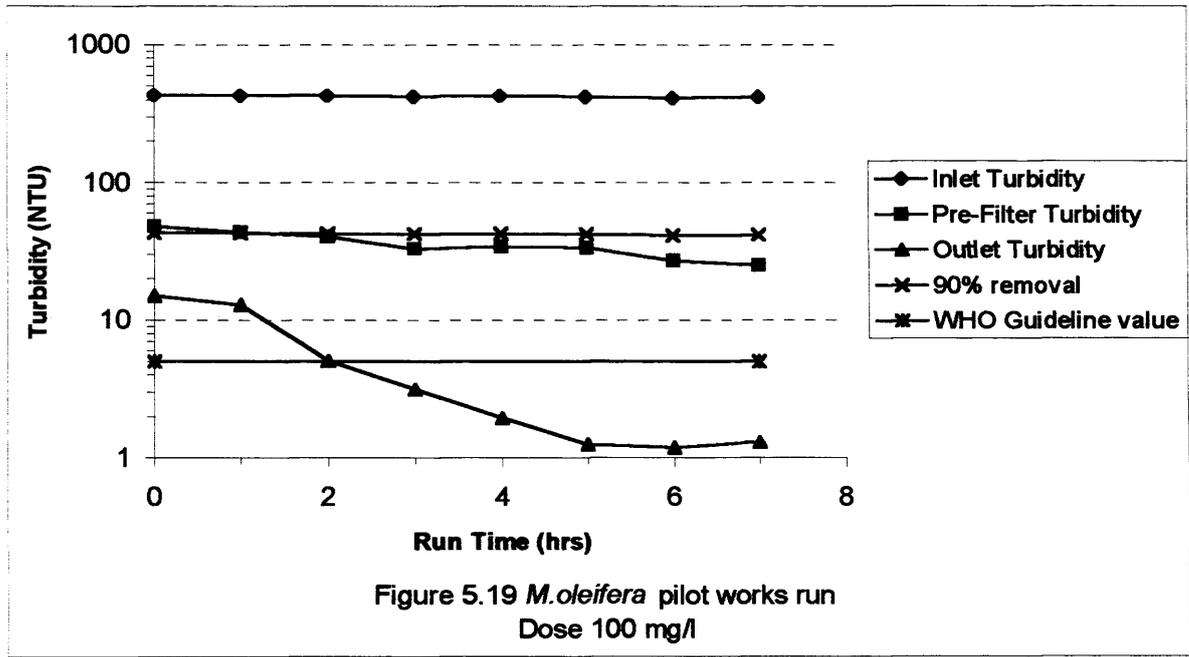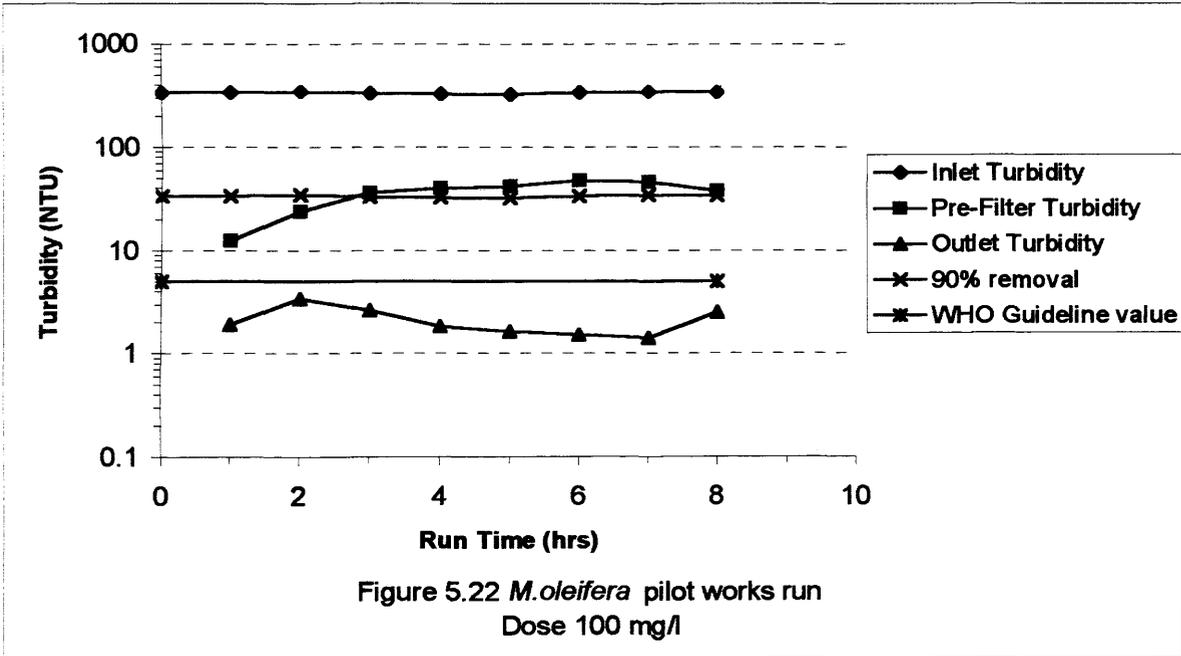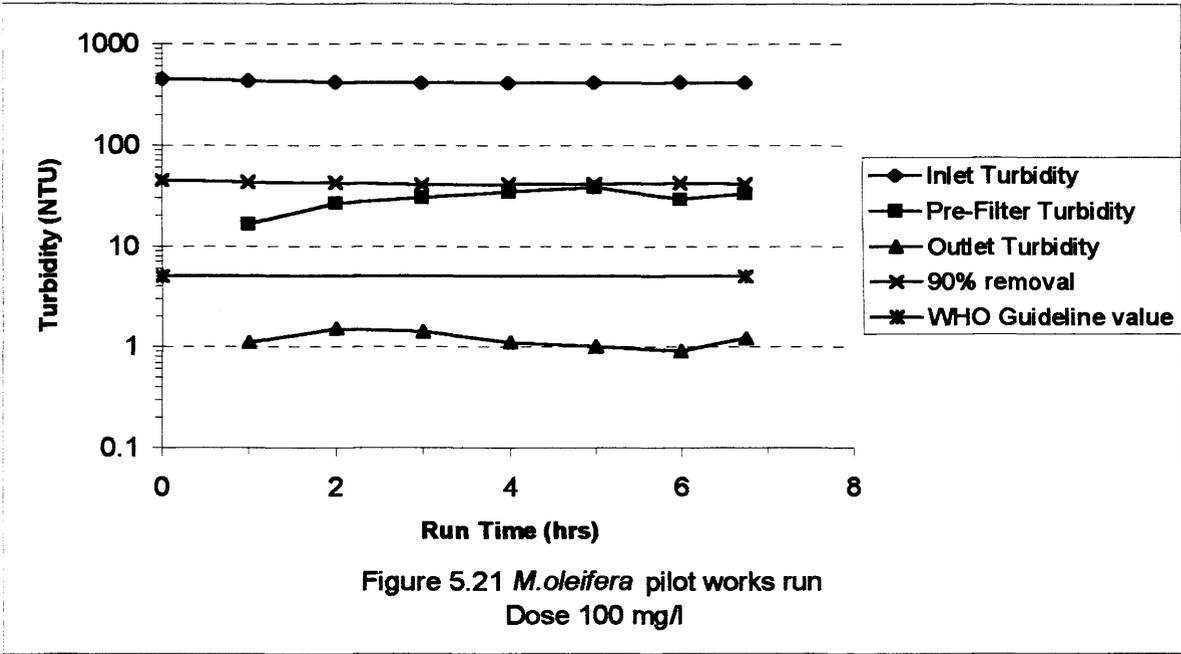
The variation in final water quality produced after filtration arose depending on whether backwashing or hand cleaning had been carried out prior to commencement of the run. Most filters require a 'working in' period before reaching optimum performance. The filters were

backwashed when either outlet turbidities began to rise significantly, indicating turbidity breakthrough, or the head above the filter bed could not be maintained at the same level, indicating a position of excess headloss. This procedure was carried out irrespective of the progress of a run.

It can be seen that the majority of the experimental runs were carried out over a period of 6 - 9 hours. With a nominal retention time of 4.25 hours in the whole system, steady state was normally reached at around 3 hours (an indication of flow short circuiting) providing between 3 and 6 hours over which performance data could be obtained. It was considered that this length of run time was sufficient. Several runs were carried out for longer periods of time with operation of the works during night hours being performed by 2 workers from the Thyolo main works. Although full training was given in seed solution preparation and works operation, it was found that on a number of occasions the results were not as expected and difficulty was experienced in determining the reasons for the discrepancies. Figure 5.23 provides an example of an overnight run.

Figure 5.23 *M.oleifera* overnight pilot works run
Dose 150 mg/l
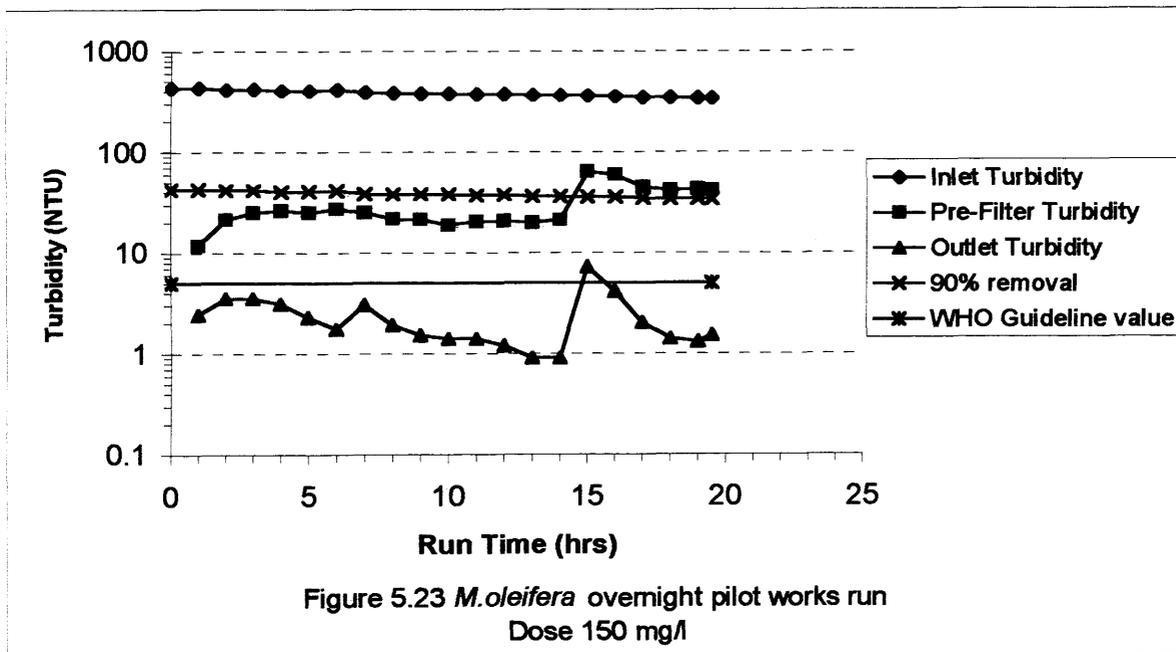
Up to 15 hours the works can be seen to be operating effectively, as the filter 'works in' turbidity levels can be seen to be dropping. At 16 hours a sudden rise in outlet turbidity from both the sedimentation tank and filter can be observed. In this case it was discovered later that the seed solution had not been checked and had run out at approximately 12 hours. The works was allowed to run for 30 minutes with no coagulant being dosed, the resulting 'slug' of untreated water passing through the works. The fact that outlet turbidities did not rise to the level of the raw water is considered to be due to a dilution effect with treated water already in the system and particle entrapment within floc already formed in the system. In this instance the mistake was admitted to and, as a consequence, the variation in results could be explained. In other cases no error was admitted, and was unlikely to be without the author resorting to intensive interrogation procedures! As a consequence the discussion and conclusions of the study are based primarily on the shorter runs carried out by the author.

One criticism of the previous pilot scale trials carried out in Burundi (Kaser *et al.*, 1989) was the limited data available on the treatment of extremely turbid water. Although treatment effectiveness has been demonstrated at 300 - 400 NTU in this study, raw water turbidity levels in excess of 6000NTU have been experienced at this site. Figures 5.24 and 5.25 present runs during which initial turbidities rose above 1000 NTU with no deterioration in treated water quality. Unfortunately, in both cases the high turbidities were combined with flash floods. Their severity caused the runs to be terminated with the pump inlet pipe and the pump itself in danger of being swept away. The pump was subsequently re-sited further up the slope from the inlet.

Figure 5.26 shows a composite of runs carried out. The data points have been taken from individual runs that were carried out allowing 3 hours for each run to reach steady state. In this format the figure clearly demonstrates the consistency and effectiveness of the treatment process.
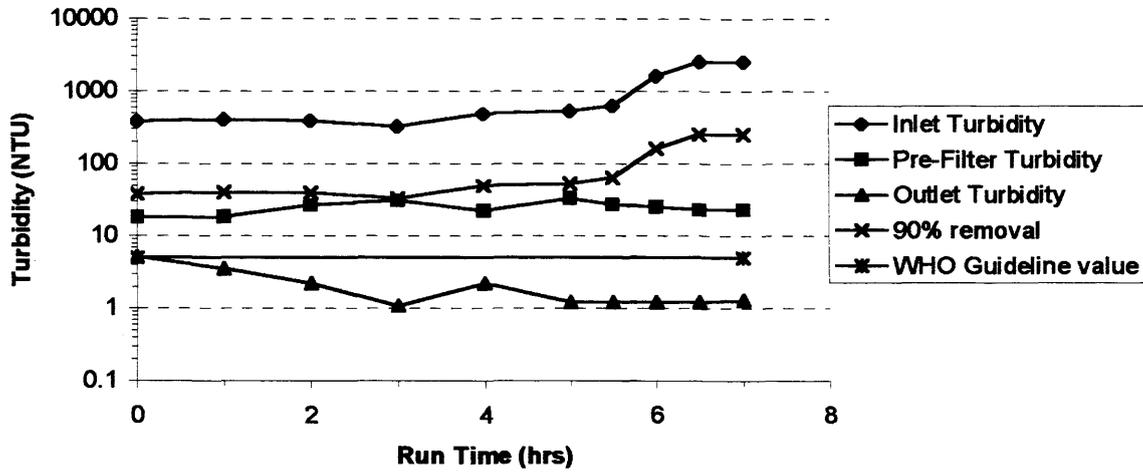
Figure 5.24 Effect of turbidity increase above 1000NTU on *M.oleifera*
treatment performance
Dosing regime - 0-0.5hrs 100mg/l; 0.5-4hrs 65mg/l; 4-5.5hrs 100mg/l;
5.5-6hrs 150mg/l; 6-7hrs 200mg/l
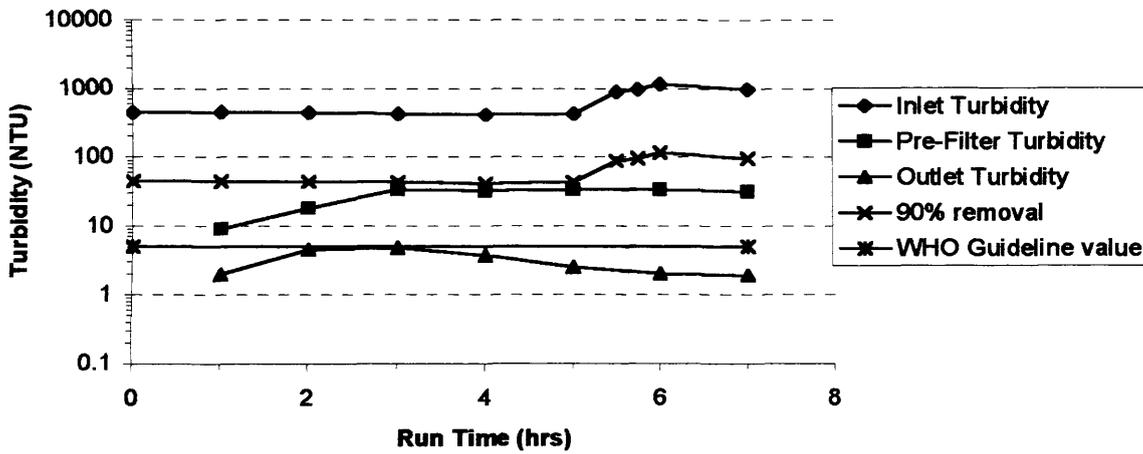
Figure 5.25 Effect of turbidity increase above 1000NTU on *M.oleifera*
treatment performance
Dosing regime - 0-5.5hrs 100mg/l; 5.5-5.75hrs 150mg/l; 5.75-7hrs 200mg/l

Figure 5.26 Composite of all *M.oleifera* pilot works runs
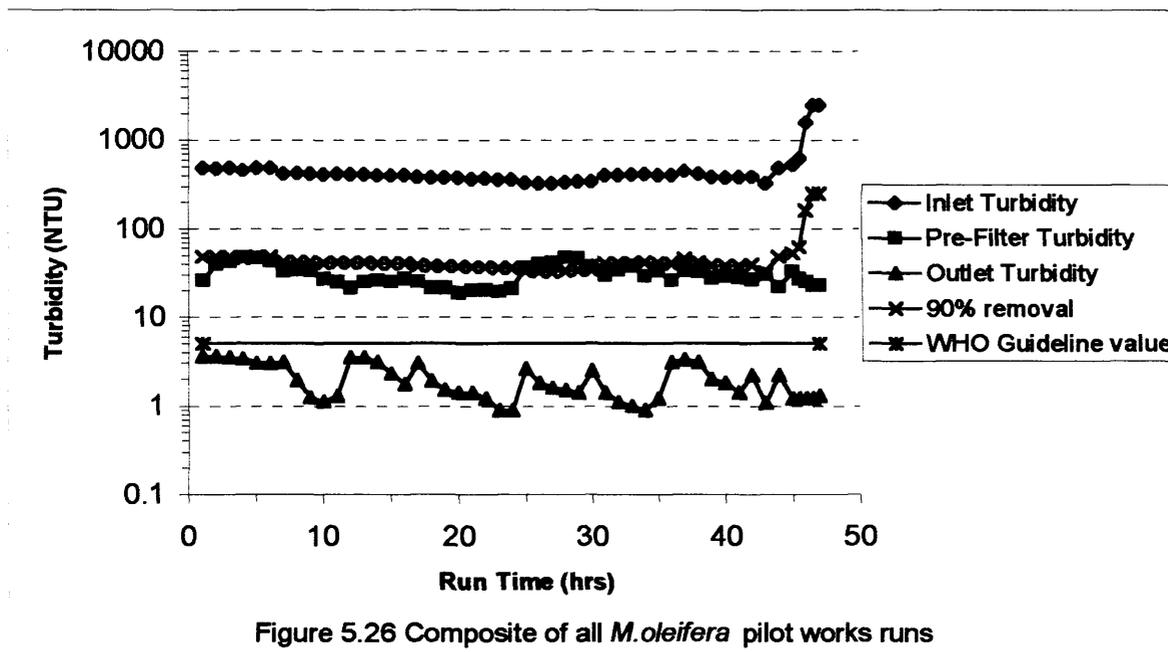
5.3.8.2 Medium turbidity

Figure 5.27 provides an example of one of the runs that was carried out with raw water of a medium turbidity. It can be seen that over the period of the run treated water quality from both sedimentation tank and filter improve up to hour 8 and then stabilise. Comparing this result with those obtained previously for alum (Figures 5.11 and 5.12), demonstrates the reduced effectiveness of *M.oleifera* on the low to medium turbidity waters experienced at Thyolo. For alum, removal within the sedimentation tank can be seen to be in excess of 90%, for *M.oleifera* this figure is only marginally above 50%. Although floc formation was evident with *M.oleifera* the floc formed were small and light with a significantly lower settling velocity. In addition, the low density of the floc made them more susceptible to the short-circuiting density currents that occurred due to temperature differentials between the influent and the existing water in the system (discussed in section 5.3.6). However, it can be seen that following filtration a final water turbidity well below the WHO guideline value of 5 NTU is obtained.

112

Figure 5.27 *M.oleifera* pilot works run in the medium turbidity range
Dose 60mg/l
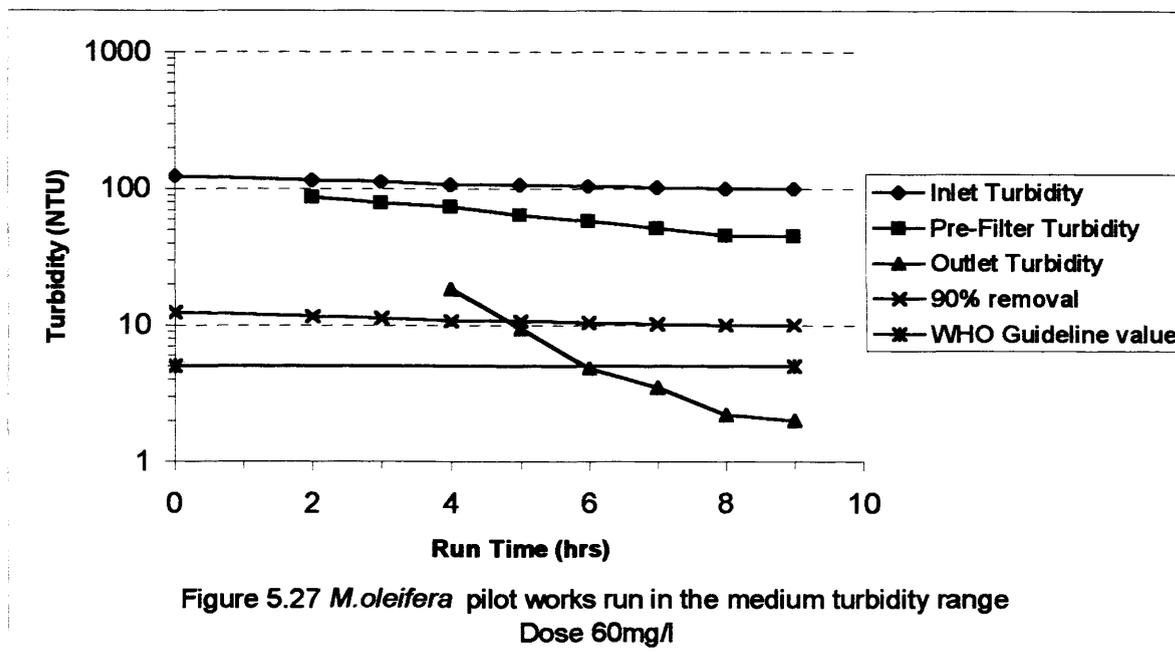
5.3.8.3 Iron and manganese removal

It was observed during commissioning of the additional flocculators that following runs with *M.oleifera*, if the flocculators remained unused for a short period of time black slimy 'floc' deposits formed on the face of the gravel near the water surface. During an experimental run samples of the raw and treated water were taken and a full chemical analysis carried out by Blantyre Water Board in Blantyre. The relevant results are shown in Table 5.2. It must be noted that although a similar chemical analysis was not carried out for alum, such deposits were not observed following alum treatment.

	Raw water	Treated water	% Removal
Iron (as Fe^{2+}) mg/l	0.71	0.13	82
Manganese (as Mn^{2+}) mg/l	0.16	Nil	100

Table 5.3 Iron and manganese removal

The results presented in Table 5.3 were backed up by the investigations carried out on the use of *M.oleifera* to treat the raw water serving the Morton Jaffray Water Treatment Works in

Zimbabwe (detailed in Chapter 4). In this case both iron and manganese concentrations were reduced to below detectable levels from initial values of 0.26 mg/l and 0.04 mg/l respectively. Taking into account both sets of results, it is assumed that the deposits observed during the Malawi studies were manganous oxides formed subsequent to the removal of manganese during the treatment process, however, as no direct analysis of the deposits was possible this assumption could not be confirmed.

Without further work it is not possible for conclusions to be made on the potential mechanisms by which removal has taken place. However, it is known that both iron and manganese can form organic complexes in raw waters with natural organic materials (Montgomery, 1985) and, as such, one possibility is that removal occurs as a result of the formation of protein-iron and protein-manganese complexes. If these protein complexes were subsequently involved in floc formation, then floc settlement would result in the concomitant removal of the iron and manganese. Although neither iron or manganese constitute an acute or chronic health risk, their removal is required to prevent possible taste and odour problems and to reduce the risk of staining. These results, coupled with those obtained by Muyibi and Evison (1995) on the use of *M.oleifera* to soften hard waters (detailed in Chapter 3), indicate a propensity for *M.oleifera* to form organo-metallic complexes. The potential, therefore, exists for *M.oleifera* to be used to remove specific metal ions from solution, for example, the removal of heavy metals from wastewater discharges. This is an area that deserves further consideration.

5.3.8.4 Pilot plant design - the effect on *M.oleifera* treatment effectiveness and potential modifications to enhance performance

Following the modifications instigated after commissioning, the pilot works itself performed with excellent overall efficiency. However, it was clear that certain of the unit processes had disadvantages where treatment using *M.oleifera* seed solutions was concerned. This section highlights the problems involved and suggests potential modifications to the pilot plant itself that may be utilised to overcome the problems. The implications of the observations made during the

pilot plant trials to the practical application of *M.oleifera* to existing treatment facilities will be discussed further in Chapter 6.

Gravel bed flocculation, as a process, has been shown to be highly effective producing floc of adequate size and density for removal within the sedimentation stage. The ability of the flocculators to store formed floc on the surface of and within the interstices of the gravel was observed, however, whether or not this is beneficial, particularly for *M.oleifera*, must be questioned. During periods of high turbidity this would be advantageous through reduced loading on the sedimentation tank. At lower turbidities this could be a disadvantage. Continued floc aggregation tends to occur once the floc enter the sedimentation stage resulting in larger floc and faster settlement. Reducing the load on the sedimentation stage, through floc storage, lowers the number of floc available for inter-floc contact and aggregation. Increasing the velocity in the gravel bed would potentially reduce the tendency for the lighter floc, formed by *M.oleifera* during periods of lower turbidity, to settle on or in between the gravel. This would increase the number of floc entering the sedimentation tank and thus benefiting by flocculation settling. At high turbidities *M.oleifera* floc tend to form at a higher rate and is denser. It is considered that settlement in the gravel bed would still occur despite the higher bed velocities reducing the potential shock load on the sedimentation stage. With respect to the pilot plant this is an aspect that could be investigated in the future by replacing the gravel currently in the beds with gravel of a smaller diameter.

Of primary concern is the general poor settling performance of *M.oleifera* floc formed at low to medium turbidities in the sedimentation tank. At a high turbidity of around 400 NTU the tank proved to be extremely effective with floc removal in excess of 90% despite the problems with temperature induced density currents. At a medium turbidity of 100 NTU the efficiency of the tank reduced to 50% for *M.oleifera* floc while maintaining a 90% efficiency for alum. A review of the literature subsequent to the work presented indicated that by changing the shape of the tank enhanced performance could have been achieved. Kawamura (1991) reports that by increasing the tank length and reducing tank width and depth can reduce the effects of density currents and

improve flow stability. Suggested guideline values are length to width ratio of 6:1 with a minimum of 4:1; width to water depth ratio of 3:1 with a maximum of 6:1 and a length to water depth ratio of a minimum of 15:1. In addition, the flow characteristics may be estimated using the Reynolds (Re) and Froude (Fr) numbers:

$$Re = \frac{\rho v R}{\mu} \dots\dots\dots\dots\dots(5.10)$$

$$Fr = \frac{v^2}{gR} \dots\dots\dots\dots\dots(5.11)$$

Where

        v = flow velocity (m/s)

        R = hydraulic radius = area of flow passage (m^2) / wetted perimeter (m)

        ρ = density (1000 kg/m^3)

        g = gravitational constant (9.81 m/s^2)

        μ = dynamic viscosity (1.01 x 10^{-3} kg/ms)

Kawamura (1991) suggests that for flow stability Re < 2000 and Fr > 10^{-5}. With the current tank design Re = 75 which is well within the limit, however, Fr = 1.2×10^{-8} well below the suggested value indicating a high degree of flow instability. It is clear that from equations 5.10 and 5.11 that a decrease in the width and depth of the tank will increase the flow velocity and reduce the hydraulic radius with a resultant increase in the Reynold and Froude numbers. However, applying this design philosophy to the design of a pilot scale works does not necessarily result in ideal conditions being achieved. The following example, under the same flow rate and retention time conditions used in the actual design, demonstrates the problem. The parameters are as follows:

Flow rate: 2.78×10^{-4} m^3/s (1m^3/hr)

Retention time: 4 hours

Volume of tank required: 4m^3

Dimension ratios, as suggested by Kawamura (1991) and noted previously, are given using the following nomenclature Length - L; Width - W; Depth - D:

$$L/W = 6:1; \quad W/D = 3:1$$

116

Rearranging gives:

$$L = 6W; \quad W = 3D \text{ and therefore } L = 18D$$

If the tank volume (V) required is $4m^3$ then:

$$V = 4 = L \times W \times D = 18D \times 3D \times D = 54D^3$$

Which gives:

$$D = 0.42m$$

And hence:

$$W = 1.26m$$

$$L = 7.56m$$

The L / D ratio works out at 18 which is > 15 and therefore acceptable.

Knowing the dimensions of the tank and the flow rate, the Reynolds and Froude numbers may be calculated using equations 5.10 and 5.11:

$$Re = 131 < 2000 \text{ therefore acceptable}$$

$$Fr = 1.1 \times 10^{-7} < 1 \times 10^{-5} \text{ therefore NOT acceptable}$$

An alternative tack is to compute the cross sectional area needed to give the Froude number required. For this example the minimum W / D ratio of 3:1 will be used   i.e. $W = 3D$.

From equation 5.11:

$$Fr = \frac{v^2}{gR}$$

Where:

$$v = \text{flow velocity (m/s)} = Q / A = Q / WD = Q / 3D^2 = 9.27 \times 10^{-5} / D^2$$

$$R = \text{hydraulic radius} = \frac{WD}{(2D+W)} = 0.6\,D$$

Substituting these into equation 5.11 gives:

$$Fr = \frac{1.46 \times 10^{-9}}{D^5}$$

Using the minimum Froude number, 1×10^{-5}, as suggested by Kawamura gives:

$$D = 0.17m \text{ and } W = 0.51m$$

The required retention time is 4 hours and based on a volume required of 4m^3 gives a tank length of:

$$L = 46m$$

This gives an L / W ratio of approximately 90:1. Kawamura does not give a maximum L / W ratio, however, from a practical point of view, for the size of treatment works under consideration, a tank length of 46m would not be acceptable. The example presented does not offer a modification option, however, it serves to illustrate that there is no simple route to the design of horizontal flow sedimentation tanks at pilot scale.

An alternative approach that enables modifications to be made to settlement tanks in order to enhance settlement efficiency is the installation, within the existing sedimentation tank, of tube settler modules. Tube settler modules are generally up-flow units and consist of a bank of tubes 5cm in diameter (or alternatively 5cm x 5cm square section) inclined at an angle of 60° to the horizontal. From considerations of tube geometry, floc contained within the flow need only settle a nominal distance of 9cm as compared to the full depth of the existing tank. Settled floc slide down the inclined tubes to the tank bottom for subsequent removal. Detention times within such modules ranges from 5 to 20 minutes depending on the type of module and the surface loading rate. The surface loading may be calculated from the area of the sedimentation tank covered by the units and is generally in the range 5-9m/h depending on floc type and the prevailing climatic conditions (lower surface loading rates are recommended for cold climates). The main advantage of these units is that the settling efficiency of such high rate modules is equivalent to a conventional sedimentation tank with a designed detention time of 3-4 hours and a surface loading rate of 1.3 m/h (Montgomery, 1985; Kawamura, 1991). Whether or not the installation of such units in the system used in this study could have been practically accomplished, in relation to 'required' design considerations (e.g. the front one quarter of the length of the tank should be left as open space to ensure acceptable inlet conditions to the modules), or would have increased the settling efficiency of *M.oleifera* floc is difficult to determine since the design of such units requires detailed knowledge of the settling velocities of formed floc. Unfortunately this knowledge is not

available on the specific raw water examined and as such a detailed example of the design calculations involved in modifying a settlement tank to include tube settler modules will not be pursued here.

5.3.9 Conclusion

The pilot scale trials have demonstrated that effective and consistent treatment of high turbidity raw waters can be achieved using *M.oleifera* within continuous flow systems. The use of seed solutions in a pilot plant designed with an appropriate technology philosophy viz.: low cost, low maintenance and minimal operator knowledge requirement, has also demonstrated the robustness of the treatment. Although problems with floc settlement were evident in the treatment of medium to low turbidity water this is considered primarily a function of the specific raw water being treated and the characteristics of the sedimentation tank used in the study. However, the problems highlighted cannot be ignored and it is clear that the practical application of *M.oleifera* to existing water treatment facilities will depend on both the raw water to be treated and the unit processes within the existing treatment train. This aspect will be discussed further in Chapter 6.

The observation that, despite limited floc removal at low to medium turbidity during the sedimentation stage, the filter maintained a high final water quality indicates the use of *M.oleifera* within direct or in-line filtration systems to be a viable option and one worth further investigation.

5.4 FULL SCALE TRIALS

5.4.1 Introduction

Following on from the success of the pilot scale studies permission was granted by the Ministry of Works and Supplies Water Department to carry out full scale trials on the main works at Thyolo. The planned trials were the first time *M.oleifera* had been used at this scale with the treated water entering the supply.

The main works at Thyolo consists of two upflow contact (reactor) clarifiers, one circular the other rectangular, followed by rapid gravity filters. Figure 5.28 shows the general layout of the main works.
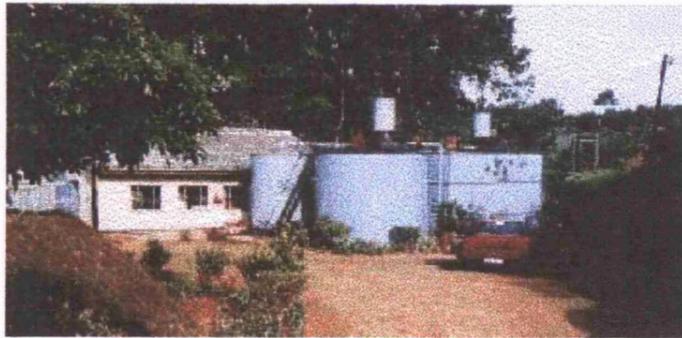
Figure 5.28 General layout of the main works at Thyolo

The reactor clarifiers are similar in design to the one shown in Figure 5.29. In standard operation, the water enters the lower central chamber where chemical addition, rapid mixing and preliminary flocculation occur, the water being kept in motion by a single drive impeller unit. The water then passes to the secondary mixing zone where flocculation continues. From here the flow is directed down over the hood of the primary mixing chamber. The decrease in upflow velocity promotes a combination of floc blanket formation and settling with the clarified water passing up and over the collecting launder. The downward motion of the water over the hood combined with the circular motion of the water in the primary reaction zone results in a high percentage of the settling floc being returned to the primary mixing chamber thus promoting initial floc formation. The downward flow also sweeps settled floc from the base of the hood to the sludge concentrator. Regular removal of sludge from the concentrator ensures free flow throughout the system.
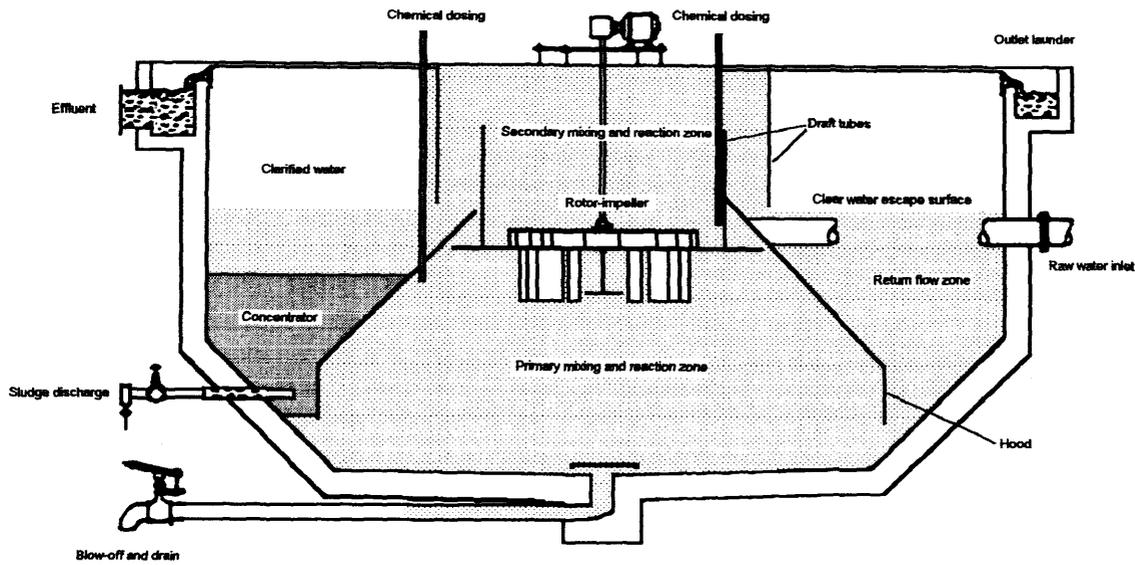
Figure 5.29 Reactor clarifier of the type used at Thyolo

However, since installation in the late 1970's both tanks have fallen into a state of disrepair. The tanks no longer have operational impellers to ensure efficient mixing and floc formation, both of which are vital to ensure optimum works operation. In addition, dosing of the coagulant does not occur in the main tank as shown in the figure but into a V-notch weir prior to the inlet to the main tank. Figure 5.30 shows the circular clarifier and the extent of disrepair. The frequency and efficiency of the desludging process must also be questioned. Prior to the works being utilised for the comparative *M.oleifera* seed and alum trials the tanks were drained in order for them to be cleaned. It was found that the sludge at the base of the tank had built up to a depth of approximately 1.5m completely closing off the primary mixing chamber from the rest of the tank. The result of this is that no recirculation flow occurred. Instead of the water passing down over the hood and back to the primary reactor, it was diverted to the outside wall of the clarifying zone and passing straight up to the outlet launder. The absence of the impeller results in inefficient mixing and a disruption to the original design flow pattern. As a result, although some settlement was evident the floc carry over was excessive.

121
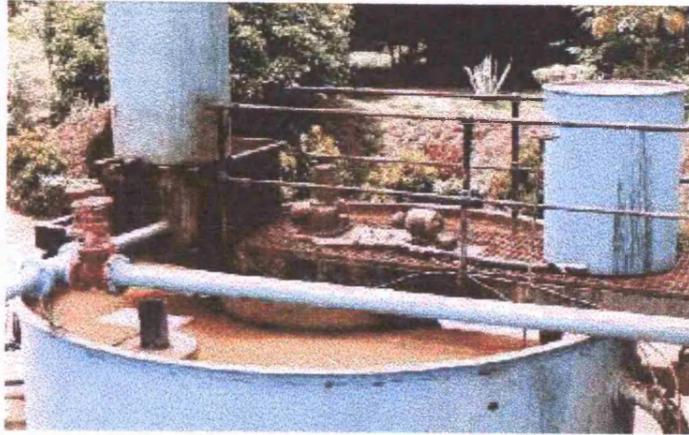
Figure 5.30 Circular clarifier – note the lack of
impeller motor and drip feed of coagulant into 'v'-
notch weir

The original design flow rate of the combined works is unknown. Currently one pump is used to deliver water to the works from the river. The flow through the whole works, according to the Ministry of Works, is either $60m^3/hr$ or $42m^3/hr$ depending on which of the two available pumps is operating. However, there is a discrepancy between these figures and those determined in previous investigations which gave the flow rates for the same pumps to be $44m^3/hr$ and $32m^3/hr$ respectively. With no flow rate monitoring station available it was decided to operate the works assuming the Ministry of Works and Supplies figures. The doses calculated and shown on the figures must therefore be considered to be nominal.

5.4.2 Modifications to the main works

As already stated, dosing of coagulant was previously carried out via gravity feed from solution tanks above the V-notch weir. In order to provide more accurate control of the dose a centrifugal dosing pump was exported to Malawi and installed. Coagulant dosing rates were monitored using a rotameter, alteration of flow rates being effected by a close control needle valve. The dose was injected via a non-return valve into the inlet rising main prior to the flow splitting to the two clarifiers. The application of coagulant into the rising main has the added advantage of providing

122

more effective initial mixing of the coagulant with the raw water promoting more effective floc formation.

5.4.3 Trials procedure

The problems involved with preparation of seed solutions for the full scale trials have been discussed previously (see section 5.2.5) and will not be repeated here.

The relatively low capacity of the dosing pumps combined with a higher dose requirement than alum meant that seed solutions had to be prepared at a 4% concentration. In order to prevent clogging of the pump and rotameter with residual seed particles, seed solutions were sieved following dilution of seed paste to the required concentration. Alum solutions were prepared at a concentration of 3%. Care was taken to ensure the alum had completely dissolved prior to use.

Samples for turbidity analyses were taken from the inlet weir box, clarifier outlet launder and filter outlet prior to the chlorine contact tank.

5.4.4 Results

Dose determination using standard jar tests was carried out prior to commencement of each run, however, as stated earlier the applied dose is nominal due to uncertainty of the flow rate.

Figure 5.31 shows a run carried out using *M.oleifera* at a flow rate of 42 m^3/hr and dose 100 mg/l. The initial high filtrate turbidities arose from previous incorrect use by the operators of the filters. It had been standard practice at the works to run the filters with no head of water being maintained above the sand bed. This procedure can result in considerable short circuiting with only a part of the bed being used effectively for filtration. This was corrected resulting in subsequent improvement to filtrate quality. The practice of keeping a head of water above the bed was subsequently maintained until the end of the trials. From the figure it can be seen that approximately 80% removal is achieved within the clarifiers with filtrate quality following adjustment of filter operation being maintained below the WHO guideline value of 5 NTU.

Figures 5.32 and 5.33 show runs carried out using *M.oleifera* at a flow rate of 60 m³/hr with doses of 75 and 100 mg/l respectively. The difference in clarifier outlet turbidities seen in figure 33 is considered to be a thermal effect producing higher floc carry over rates in the smaller circular clarifier. Filtrate turbidities are maintained below the WHO guideline value.

Figure 5.34 shows a comparative run carried out using alum at a flow rate of 60 m³/hr with a dose of 50 mg/l. As with the *M.oleifera* runs, removal rates within the clarifiers are approximately 80%, however, an improved filtrate quality consistently below 1 NTU can be seen. This is considered to be due to the filters having 'run in' to a greater extent than during the *M.oleifera* trials.

Figure 5.31 Main works operating at a flow rate of 42m³/hr with *M.oleifera* seed solution dosed at 100mg/l
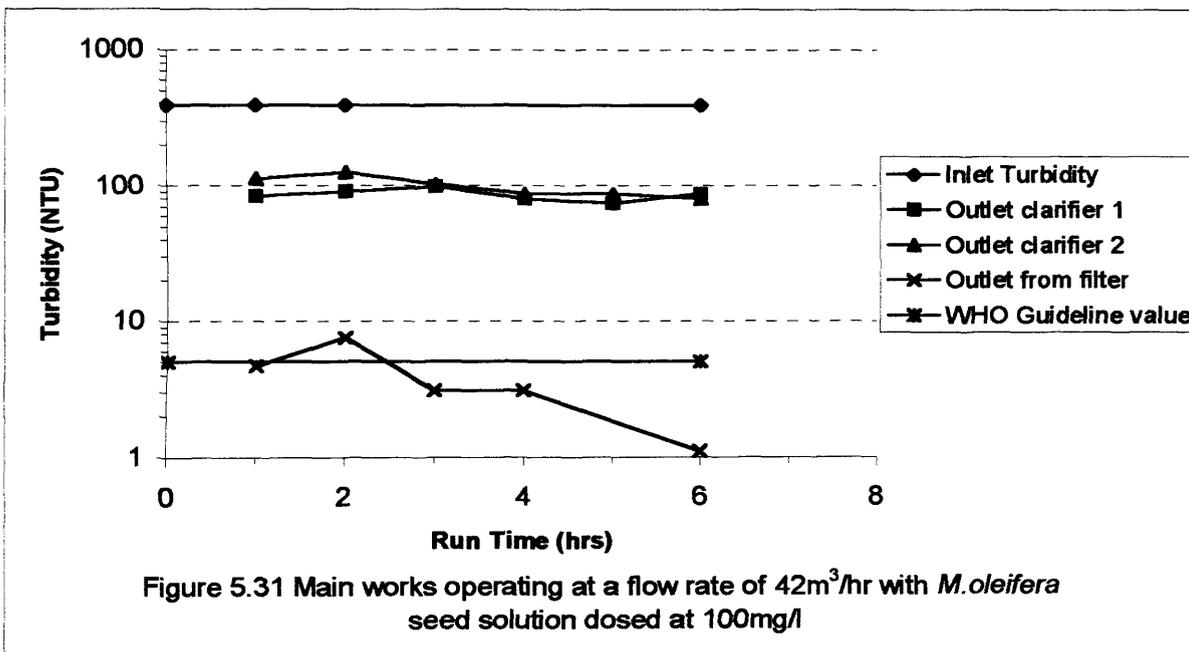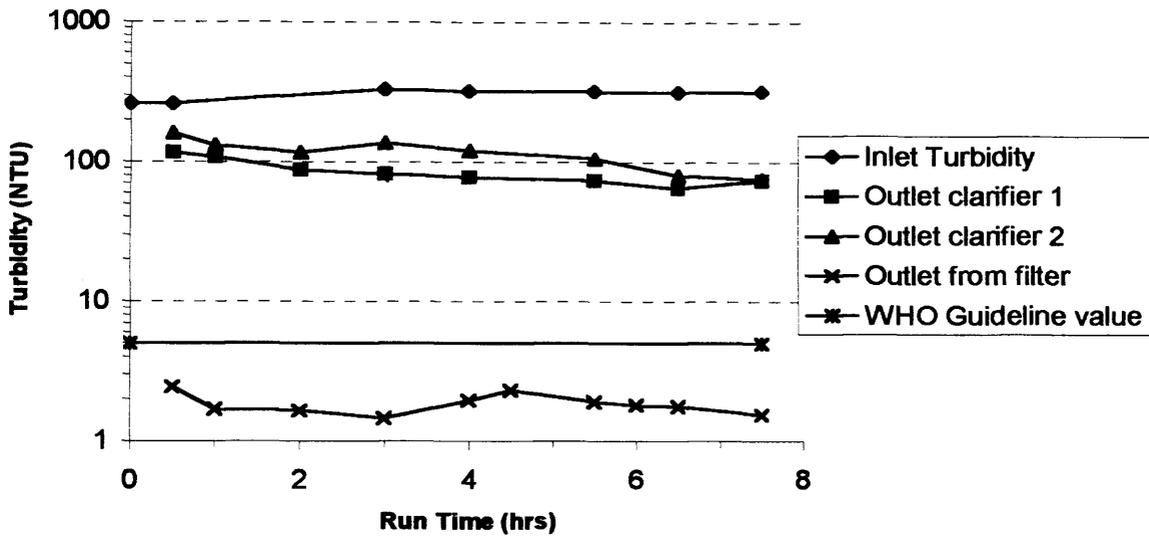
Figure 5.32 Main works operating at a flow rate of 60m³/hr with *M.oleifera* seed solution dosed at 75mg/l
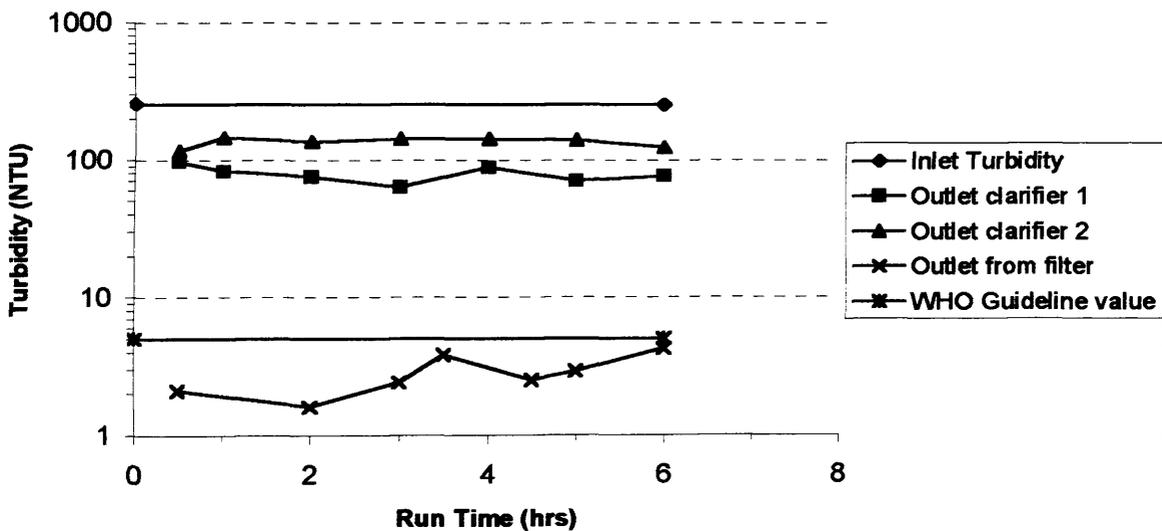
Figure 5.33 Main works operating at a flow rate of 60m³/hr with *M.oleifera* seed solution dosed at 100mg/l

Figure 5.34 Main works operating at a flow rate of 60m³/hr with alum solution dosed at 50mg/l
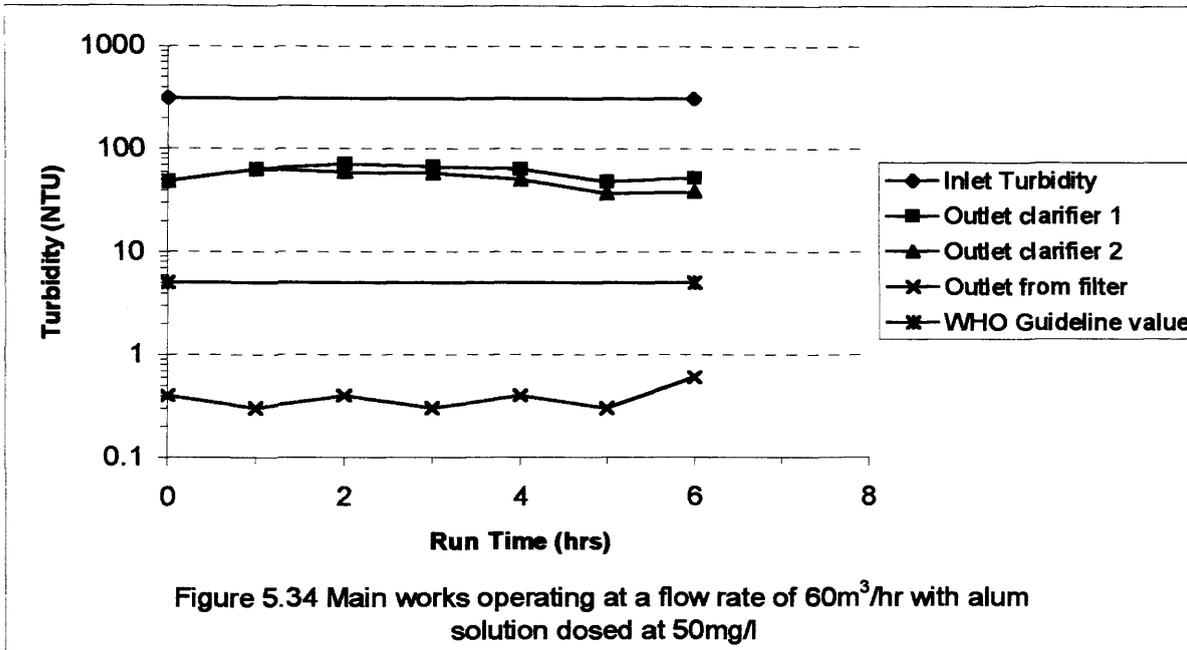
5.4.5 Discussion

The full scale trials are the first of its kind to have demonstrated that, as a coagulant, seed solutions of *M.oleifera* can effectively be used within large scale continuous treatment systems. The success is, perhaps, more significant considering the dilapidated state of the works on which they were carried out, thus providing an indication of the robustness of the treatment process. However, the trials must be viewed within the limitations and context of the study:

• As with all studies carried out on specific source waters, the results obtained should be taken as being indicative of performance capability. Success on one source water, albeit a problematic source, does not imply that similar success would be achieved on other source waters;

• As a result of continued rain turbidity levels during the trials were maintained at or around the 400 NTU mark. The laboratory and pilot plant studies carried out on the same source water both demonstrated a reduced effectiveness at lower turbidities. Further study at the Thyolo main works would be necessary to investigate comparative performance at lower turbidities;

126

- Efforts were made to optimise the treatment process. The installation of a dosing pump, thereby enabling coagulant to be dosed into the rising mains, undoubtedly improved the overall performance of the works irrespective of the coagulant. It was subsequently learnt that about 4 months after the end of the trials the pump broke down. The pump could not be repaired with the result that the old system of dosing via a drip feed was reinstated. Whether or not *M.oleifera* would operate as effectively as alum under such a regime, which is one from the experience of the author that will continue, is unknown.

The trials have also highlighted problems with the use of *M.oleifera* seed powder within the context of large scale treatment. In addition, aspects of operator skill that may affect the use of *M.oleifera* must be considered if acceptability of the treatment process and implementation are to be effectively achieved. These aspects are discussed in more detail in Chapter 6.

5.5 CONCLUSIONS

As a concluding remark it is considered important to restate the findings of the pilot and full scale trials that, as a coagulant, seed solutions of *M.oleifera* can effectively be used as a full or partial replacement for proprietary chemical coagulants in the treatment of surface waters in continuous flow water treatment systems. Treatability studies on individual raw waters will provide an indication as to whether use of seed solutions is limited solely to treatment at high turbidity or whether the treatment can be effected irrespective of the season. Identified problems of scale up relating to seed processing and dosing solution preparation may be solved by the further development of techniques and protocols.

CHAPTER 6

GENERAL DISCUSSION ON THE APPLICATION OF *M.OLEIFERA* TO WATER TREATMENT PRACTICES IN DEVELOPING COUNTRIES

6.1 INTRODUCTION

The laboratory, pilot scale and full scale trials detailed in Chapters 4 and 5 have demonstrated that, as a coagulant, seed solutions of *M.oleifera* can effectively be used as a full or partial replacement for proprietary chemical coagulants in the treatment of surface waters. The studies have highlighted areas in which problems still exist and that need to be addressed. Specific aspects pertain to seed processing and dosing solution preparation and the variable treatment performance at low to medium turbidities that is evident with some raw water sources. The extent to which these aspects pose a problem is considered to be dependent primarily on the scale of application. This first section of this chapter will discuss the application of *M.oleifera* seeds to the batch treatment of raw waters at household level. The second section discusses application to continuous flow systems. In addition to operational factors (relating to seed availability, processing and dosing solution preparation), economic aspects and other factors that may be specific to a certain level of application and that may affect the viability of the treatment process as a whole and hence be a potential barrier to implementation will also be discussed.

6.2 THE APPLICATION OF *M.OLEIFERA* TO DOMESTIC HOUSEHOLD WATER TREATMENT PRACTICES

The first step in this discussion element is to define the scale of treatment being considered. For the majority of people in developing countries without access to piped or borehole water supplies, the provision of their daily water requirements involves the collection of water from a local surface water source in containers, the volume of which may vary between 10 and 50 litres. The discussion here centres on the batch treatment of this collected water and restricts itself to the

introduction of such treatment practices to communities that do not already carry out some form of traditional domestic household water treatment.

The tools required to carry out the batch treatment of raw water are basic viz. a container for the raw water, a paddle of some description for providing agitation of the water for rapid mixing and flocculation, a second container for receiving the treated water i.e. settled supernatant, a third, smaller container for preparation of dosing solutions and a source of seed. The water treatment procedures are simply a scale up of the standard jar test procedure outlined in Chapter 4. Seed solution is added to the raw water under rapid agitation then slow mixed to promote flocculation. Following a suitable period of settlement the treated water is carefully decanted to a clean container ready for either direct consumption or some subsequent form of disinfection.

With regards seed requirement, provided the tree is able to grow in the region where implementation is planned this should not pose a problem. For example, to treat 40 litres per day at an average dose of 100mg/l would require 4g of seed kernel powder per day or 1.46 kg per annum. To date, detailed and reliable information on the yield of dry seed per tree is unavailable, however, the following assumptions have been made based on information provided in the literature and on the author's observations of mature trees in Malawi, Kenya and Zimbabwe: Number of pods per tree = 600; Number of seeds per pod = 15; Average weight of seed kernel = 250 mg. Using these values and assuming that the processing methods employed were 100% efficient, each tree would yield 3kg of seed kernel powder per annum. Compare this to the required 1.46kg and it is clear that a single tree should provide more than enough seed for one years worth of treated water. With respect to seed solution preparation, the crushing and resuspending of 4g of kernel material is not a complicated procedure and would be similar to that described in Chapters 4 and 5 viz. water added to seed powder to make a paste, diluted to the required concentration and filtered through cloth prior to addition.

Given the information above it may be considered that the application and implementation of M.oleifera seed to household water treatment practices should be straightforward. In reality, this is perhaps the level at which the most constraints against implementation exist. The constraints do

not relate directly to the effectiveness of the seeds in clarifying water but are practical and socio-cultural in nature.

One of the primary constraints is, perhaps, the determination of optimum dose. Although an advantage demonstrated in this study for *M.oleifera* treatment is the wide dose range over which effective treatment can be achieved, it is clear from the results in Chapter 4 that there is a minimum dose for optimum performance below which effectiveness is greatly reduced. In the developed world, dose determination is a relatively straightforward procedure. With the rapid advance of technology there are some extremely complex batch and on-line techniques that can be utilised, however, still the most widely accepted procedure is the standard jar test. With this in mind, the question must be asked how can the dose required be determined at village level? The application of standard jar test procedures using the equipment detailed in Chapter 4 is neither economic nor practical in the village situation. Even the simplest laboratory jar test apparatus is costly and, more importantly, requires some form of electrical power source for operation. Taking the treatment process a stage further, if one assumes that a minimum dose can be determined, how would this be conveyed to the villagers? It would not be practical to suggest a dose based on a milligram per litre basis since the concepts of a milligram and a litre are unlikely to be understood. One could simplify this to stating the dose as the number of seeds required per container. This would only be effective if every villager used the same type and size of container. In addition, thought would have to be given to who would conduct the 'village jar test' since it would be impractical for each household to be conducting their own tests. A step further, how would one ensure that the mixing parameters required to ensure effective floc formation were achieved? Without the aid of a stopwatch this is extremely complicated and if not carried out correctly can significantly effect treatment performance. As one continues through the treatment train a host of other questions arise. What settlement time should be allowed for full clarification? Should the treated water be decanted following settlement? Should the water be boiled or filtered as a purification stage? What happens to the sludge produced considering the majority of bacteria and viruses present in the raw water will now be concentrated in the sludge?

The questions that have arisen regarding these practical aspects are not unanswerable. A manual produced by the German aid agency, GTZ, concerns itself solely with addressing the problems outlined (Jahn, 1986). From a technical point of view some of the solutions offered appear innovative and eminently feasible, however, there is no evidence to date to suggest that the solutions have been field tested. No detailed discussion of the solutions presented will be entered into here primarily for the lack of evidence that they are applicable in the field. However, the solution to the problem of obtaining a uniform mixing speed for the flocculation stage gives an example of the type of thinking involved. It has been suggested that a chant containing four simple two-syllable words, each syllable representing a half rotation, relating to water treatment and the benefits to be obtained from clean water, be used. An example in English might be "Trea-ted Wa-ter Heal-thy Peo-ple". For this solution to be viable, the tempo of the chant must first be worked out using a stopwatch to ensure that the correct stirring speed is achieved. Again, on paper this would appear to be a relatively effective solution, however, from a practical point of view further questions are raised: What language should the chant be in, bearing in mind cultural conditions? What words should be used in the chant? In what direction should the water be stirred, bearing in mind cultural and religious attitudes? What training is necessary and who should carry out the training, bearing in mind socio-cultural conditions? Although these questions may appear, on the face of it, to be facile these are the type of questions that require consideration to ensure that whatever practices are being encouraged are sustainable.

It is clear, therefore, that the introduction of such household water treatment practices, particularly in those areas where no form of traditional treatment is carried out, is highly problematic. To date there has only been one documented attempt to introduce *M.oleifera* as a water clarifier at household level outside those areas in the Sudan where traditional use has been practiced. In 1986 the Indonesian non-Governmental Organisation Yayasan Dian Desa (YDD) instigated a pilot project in collaboration with GTZ to introduce the techniques of household water treatment to a selected village community. The project had a two year lead in time to enable:

i) The establishment of a *M.oleifera* nursery to provide plant material to enable the target group to cultivate their own trees with the aim of achieving self-sufficiency in later years, and to provide sufficient seed material to allow implementation to be undertaken prior to self-sufficiency;

ii) A survey of the potential target group to be undertaken. The aim of the survey was to gather information on the attitudes and perceptions of the target group to a number of water related aspects, to current knowledge of *M.oleifera* as a tree and to the most effective means by which implementation could be achieved and information communicated. The survey programme also provided an opportunity for the target group to get to know YDD personnel to enable a trusting relationship to be developed;

iii) The development of educational materials.

Implementation itself commenced in 1988 with an extensive training programme that covered not only the treatment itself but also cultivation of the trees and the development and modification of suitable treatment vessels. In order to ensure that all training elements were adhered to and to provide follow up advice, a YDD field worker stayed with the community throughout the first year. After a year all households were continuing the practice, indeed other villagers outside the selected project area had approached YDD with requests to be taught the treatment process, however, problems with the supply of seeds and the requirement of maintaining a constant presence within the target group threatened the success of the project (YDD, 1989). Whether or not the project is still on-going is unknown (the lack of published material on the project would, perhaps, suggest not), however, the YDD experience does highlight the significant amount of time and effort that is required in introducing a 'new' technology to, in particular, communities in rural areas. It is considered unlikely that, given the current state of knowledge of *M.oleifera*, implementation using such an 'isolationist' approach as that taken by YDD will be successful. Although no guarantee of success can be given, this is perhaps a situation where a 'top-down' approach would be more suitable. The cultivation of *M.oleifera* on a large scale, either for large scale water treatment purposes or solely for oil production, would inherently raise awareness amongst governmental and non-governmental organisations to other potential applications of

products from the tree. Large scale cultivation would guarantee that, in the initial stages of any implementation project, seed would be available from alternative sources. Provided implementation programmes were instigated within national water and sanitation frameworks, the application of *M.oleifera* seeds to household water treatment could be considered as a viable short term option, prior to the establishment of a more permanent provision, or a long term option where no feasible alternatives were available. Whatever option may be chosen, assuming that the practical problems outlined can be overcome, any implementation programme must:

i) give due consideration to existing social and cultural practices and traditions that may be associated with the provision of water within the home environment;

ii) be part of a wider programme that includes the provision of adequate sanitation facilities and general hygiene education - significant improvements in the general health of a community will not be brought about by the provision of potable water in isolation;

iii) provide training to core members of the community, and in particular to women's groups with respect to all aspects of treatment practices to be adopted, viz.: tree cultivation, harvesting, crushing and storage of seeds, preparation of dosing solutions, determination of correct dose;

iv) ensure that sufficient seed is available even during extended periods of drought when harvests may be lower than expected - if seed is unavailable for a period people may return to using untreated water and may subsequently reject treatment entirely;

v) ensure that technical assistance may still be readily obtained in the event of any problems that might arise after initial implementation.

Provided that the use of *M.oleifera* is promoted within the context described above there is no reason, technical or otherwise, why such a treatment practice should not be a sustainable, alternative method of providing safe drinking water for rural communities.

6.3 THE APPLICATION OF *M. OLEIFERA* TO CONTINUOUS FLOW SYSTEMS FOR COMMUNITY OR URBAN WATER SUPPLIES

The successful results of the pilot and full scale trials presented in Chapter 5 demonstrate that the ability of the seed material to effect coagulation and hence clarification on continuous flow systems. However, despite the success, the trials have highlighted significant problems that would have to be overcome in order for seed powder solutions to be accepted as an alternative to conventional chemical coagulants for the large scale production of potable water.

6.3.1 Potential constraints to use of powdered seed as a coagulant

i) Reduced clarification efficiency on low turbidity water sources

As shown by the results in Chapter 4 there are clearly problems associated with seed use at low to medium turbidity waters depending on the source. There are two possible ways round this. Firstly, it is conceivable that seed use could be promoted solely during those periods of heavy coagulant demand i.e. the wet season, where seed solutions have been shown to provide similar or superior clarification to that of commercially available coagulants. Secondly, where flocculation aids (typically synthetic polyelectrolytes) have been used with conventional coagulants to assist in floc formation, there exists the potential for a similar treatment regime with *M. oleifera* seed solutions. Under both such conditions significant cost savings, provided in country cultivation was feasible, could still be made.

ii) Seed requirement

The amount of seed kernel material required to ensure that a treatment works can operate effectively year round without interruption can be estimated by assuming an annual average seed dose per litre and multiplying by the volume treated in a year. The land area required to provide the quantity of material calculated may be estimated by assuming the yield from a single tree and the number of trees per hectare. For the purpose of this discussion the following assumptions have been made:

134

a) Average seed kernel dose 100mg/l (assumption based on results presented in Chapters 4 and 5);

b) Yield per tree of 3kg of seed kernel powder (yield obtained from assumptions presented previously in section 6.2);

c) Number of trees per hectare 1089 (using a recommended tree spacing of 3m [KEFRI, 1996]).

Combining b) and c) gives a yield of seed kernel powder per hectare of 3267kg. Table 6.1 details the weight of seed material required and the area under cultivation for two separate water treatment works. The first represents a small community system similar to the main works at Thyolo utilised in the full scale trials and treating on average 900 m^3/day. The second represents a major works similar to that at Lower Ruvu in Tanzania that supplies the city of Dar-es-Salaam and treating 180,000 m^3/day.

Water treatment system m^3/day	Total volume treated (m^3/year)	Total weight of seed kernel required (kg)	Approximate area under cultivation (hectares)
System 1 - 900	328,500	32,850	10.5
System 2 - 180,000	65,700,000	6,570,000	2012

Table 6.1 Seed requirement depending on scale of application

From a practical point of view, cultivation of the areas shown in Table 6.1 is not necessarily a problem (although finding the investment capital to start cultivating from scratch may be) and, as such, providing sufficient quantities of seed is not considered a major constraint to acceptance.

iii) Seed processing

Processing of the seeds to produce powdered seed kernel for use in the pilot plant and full scale studies was relatively straight forward and carried out by hand using equipment that was readily

135

available viz. pestles, mortars and sieves used traditionally for household maize processing. Even for the small quantities required for the pilot and full scale trials (approx. 1200kg was processed) the process was found to be labour intensive and time consuming. Clearly, then, it would be impractical to process by hand the amounts required for both treatment works examples being considered. In order to facilitate such production, mechanical methods would have to be developed for all areas of seed production including harvesting, removal of seed from the pods, de-husking and powder production.

- Harvesting – it is unlikely that this is a process that could be fully mechanised. Firstly, given the length of time required for the pods to dry on the trees and the fact that, depending on climatic conditions, two harvests per year maybe obtained from a crop, it has been frequently observed that new green pods are found growing alongside mature drying pods. Mechanical harvesting methods would be unlikely to discriminate between the two potentially reducing yields from single trees. Results presented in Chapter 5 have demonstrated that harvesting immature green pods and force drying the seeds reduces the effectiveness of the coagulant produced, as such, harvesting all pods from a tree and sorting green from brown pods would not be a viable option. Secondly, any mechanical method of harvesting has the potential for damaging the branches, as they are not considered strong (hence no record of their use as structural members in construction [Morton, 1991]), thereby affecting future tree yields.

- Seed removal from pods – this is an area that could be mechanised. It is envisaged that processes similar to those used for the extraction of peas from their pods could be modified for use with *M.oleifera* pods, however, further work would be necessary to confirm this.

- De-husking (i.e. removal of seed kernel from shell) – this is a commonly used process within the vegetable oil industry and, as such, it is considered that available equipment could be modified for use with *M.oleifera* seeds. Indeed, a small scale mechanical method of dehusking the seeds has been developed in Ghana in association with the American

136

NGOs Appropriate Technology International and the Church World Service. Minimal information is available on the development, however, in its present state it is reported that the device can process approximately 100kg per day with a high efficiency. Developments are continuing to increase the throughput of the system (Fuglie, 1997).

- Production of seed powder – there are a wide variety of methods by which seed powder may be produced on a large scale, however, it is possible that the high oil content may pose problems with some methods (for example, traditional maize mills). This element would require further investigation.

Given the results presented in Chapter 5, the use of presscake material, produced following the oil extraction process, as a coagulant offers perhaps the most effective method of overcoming the problems associated with de-husking seed and the production of seed powder. The seed shell is required within physical oil extraction processes as a source of fibre to promote the release of oil from the kernel. In addition, the significantly reduced oil content within the presscake would be unlikely to hinder the production of a powdered material for water treatment application.

iv) Seed dosing solution preparation

Essentially the preparation of dosing solutions *per se* is not necessarily problematic since most large treatment works, particularly in developing countries, have large mixing tanks used to prepare solutions of aluminium or iron salts. Given this, solution preparation would be similar to that used in both the full scale and pilot plant trials. The major problem associated with the preparation of dosing solutions involves that seed material that is not water soluble and that would have a tendency to settle out in the solution containers. Given the organic nature of the material it would degrade with time potentially producing by-products that could significantly effect treated water quality. As such, solution preparation tanks would have to be cleaned prior to the preparation of each new dosing solution batch (such a situation does not exist for the preparation of conventional coagulants such as the aluminium or iron salts). Further, the waste solids material produced following cleaning would have to be disposed of. In an ideal situation

such waste could be applied to the land as a soil conditioner / fertiliser, however, more often than not in developing countries all wastes from water treatment plants (including sludge produced during the clarification and filtration processes) are returned to local water sources for dispersal. Although the quantities of waste seed material may not be large, the potential toxic nature of seed material to fish (Grabow *et al*, 1985) would bring into question whether such a disposal route was 'good practice', given the promotion of *M.oleifera* use as an environmentally sound alternative to conventional coagulants.

Given the above discussion, provided a water utility was prepared to modify its work practices to take into account the need for regular cleaning of dosing solution tanks and to consider a more 'environmentally friendly' method of waste disposal, then the problems outlined would not be a constraint to *M.oleifera* seed powder use at large scale.

v) The potential effects of the addition of an organic material on treated water quality

Although seed solutions have been shown to be non-toxic, there are major concerns about the utilisation of such an organic material and the potential effect it may have on final water quality. Specific concerns relate to possible increases in the Total Organic Carbon (TOC) content of treated water. Although an increase in the TOC level itself does not necessarily imply that water would be unfit for consumption, such increases could cause related problems:

- As organic matter degrades there exists the possibility that the decay products formed could cause undesirable taste and odours in the treated water;

- The potential exists for high levels of TOC in treated water supplies to lead to bio-fouling within distribution and storage systems;

- Chlorine, in its many forms, is probably the most widely used disinfectant for water supplies. There is clear evidence that certain organic compounds present in water can react with chlorine to form chlorinated by-products, many of which are known to be harmful to humans. Of the many chlorinated by-product compounds that are now known to exist in water supplies, the trihalomethanes (THMs) are the most common. Those that

occur in drinking water supplies include chloroform, bromoform and bromodichloromethane amongst others. All of these by-products can be harmful to humans. Given this, the concern regarding the trihalomehane formation potential of residual organic material from *M.oleifera* seed solutions is understandable. To date very little work has been done to establish whether or not a problem actually exists. It is possible that the protein fraction of the seeds would not cause problems, firstly because it is likely that the majority of protein material will be attached to the flocculated solids and as such would be removed from the treated water and, secondly, because of the NH or amino-terminal of the protein molecules. There is evidence ammonia will react with free chlorine to form chloramines which cannot undergo further reaction to form THMs and which, in themselves, are still effective disinfectants (Montgomery, 1985). However, it must be noted that seed solutions are a complex mix of organic material that would include proteins, polysaccharides, starches etc., and it is not known to what extent these compounds are present nor the extent to which they may add to the TOC of treated waters. As such, the trihalomethane formation potential related to the use of seed suspensions still requires evaluation.

From the above discussion it is clear that further work is still required to establish the potential impact the use of seed suspensions could have on final water quality.

vi) Cost effectiveness

There are advantages that can be claimed for the large scale use of *M.oleifera* seed suspensions (to be detailed in the next section) one of which is a reduced reliance on imported materials. Provided ecological conditions are such that the tree can be cultivated, coagulant material could be produced in-country providing complete control of the supply. However, in order for *M.oleifera* treatment to be considered viable at large scale it must be able to compete on an economic footing with the commercially available coagulants, even if the coagulants are imported at high cost using valuable foreign exchange. It has been suggested that the availability of an

effective 'green' coagulant in developed countries i.e. the USA, Europe, would be accepted even at a slightly inflated cost over its chemical rivals purely for purposes of public relations, however, it is unlikely that this attitude would be taken in developing countries where chemical costs generally make up a far greater proportion of daily operating costs than in developed countries (Schulz and Okun,1984). Currently there is only limited information available on which to base the potential costs involved in using *M.oleifera* seeds. As such, further work is required in order for cost comparisons to be made with, for example, aluminium sulphate. The main point to note here is that in order for *M.oleifera* seed suspensions to be accepted as a viable alternative, it must be able to compete on a similar economic footing with conventional chemical coagulants.

6.3.2 Potential advantages to the use of powdered seed as a coagulant

Despite the constraints discussed in the previous section, the studies reported in Chapters 4 and 5, have also demonstrated potential advantages that *M.oleifera* seed use has over that of conventional coagulants:

- Treatment performance is not significantly affected by raw water pH nor does treatment affect the pH of the treated water. Given this, use at large scale would not require any additional chemicals to control pH thereby offering potential cost savings;

- Treatment performance on high turbidity raw waters is similar, and in some cases superior, to that of conventional coagulants;

- There is a wide dose range over which effective clarification may be achieved, as such, dose control, provided the dose applied is above the minimum required for optimum flocculation, is not as critical as may be for some of the conventional coagulants;

- A reduced sludge volume following treatment. Both the aluminium and iron salts have a tendency to produce large volumes of sludge. This occurs through the formation of large quantities of insoluble metal hydroxides during treatment and the inclusion of water within the hydroxide floc structure (Montgomery, 1985). The result is a voluminous sludge that can present problems in dewatering and subsequent disposal. As discussed in Chapter 4

M.oleifera, in the absence of high levels of natural organic material, produces floc that are generally smaller and more compact. The proximity of the particles to each other within the floc structure, arising from the proposed mechanism of activity, precludes the inclusion of water and results in a closely packed sludge. It is considered that such a sludge would pose less of a technical problem in terms of dewatering and subsequent disposal. A further benefit is that, unlike the metal hydroxide sludges, the added material contained in the *M.oleifera* sludge is biodegradable. The potential for use of a dewatered slurry or the dry solids as a fertiliser or soil conditioner would be greatly enhanced. It must be noted, however, that no studies have yet been carried out to fully establish either the extent to which *M.oleifera* sludge is easier to handle or the perceived benefits of a biodegradable sludge.

6.3.3 General comments

It is clear from the previous discussion, based on the work carried out over the course of this study, that a significant amount of work is still required in order to overcome the practical constraints to use of *M.oleifera* seed powder use at large scale and to establish whether, financially, its use would in fact be cost effective. It may be that the use of presscake material would offer a more practical and cost effective alternative to seed kernel material, however, the case for use of such a material has yet to be proven.

Perhaps one of the most significant constraints to use is that of regulatory approval, an aspect that the majority of studies that have previously been undertaken seem to ignore when they promote utilisation of *M.oleifera* on a large scale. In all developed countries there are strict guidelines as to what chemicals and other materials (for example plastics used in pipe work) are allowed to come into contact with potable water. In the UK for example, approval for use of chemicals must be obtained from the Drinking Water Inspectorate (DWI) under Regulation 25 of the Water Supply (Water Quality) Regulations 1989 prior to application within water treatment facilities. In order to achieve such approval, the chemical manufacturer must present to the relevant committee within

141

the DWI all information relating to the chemical identity and composition, method of manufacture, possible interactions with other chemicals used in water treatment (e.g. chlorine) and comprehensive toxicological and eco-toxicological data on the components and impurities within the product. Following submission of this information an assessment is made and an approval / refusal notice is issued. It is acknowledged that many developing countries do not have such a strict regulatory structure, however, it is likely that such regulations will come into force in the future. Given such a scenario how would *M.oleifera* seed solutions fare? The toxicological data available to date does not provide sufficient depth of detail and would have to be pursued further. There is minimal information on the interactions between residual seed components in treated water and chlorine (see Section 6.3.1). Questions would undoubtedly be asked regarding quality control of the raw material. For example, are pesticide residues present on the seed and, if so, are they present at levels that may cause concern? How has whole seed or seed powder been stored (fungal growth on whole or powdered seed material has the potential for release of fungal aflatoxins)? Are seeds genetically modified (GM)? Although a question that may be one more likely to be asked with regard potential use in a developed country such GM concerns could well become more widespread. From this discussion it is clear that a significant amount of work would be required to obtain regulatory approval in, for example, the UK. Although it is possible that usage within developing countries may not require such similar regulatory approval (permission for the full scale trials reported in this study was granted based solely on the toxicological information currently available), this situation is likely to change in the future and may prove to be a significant constraint to widespread use.

CHAPTER 7

CONCLUSIONS AND RECOMMENDATIONS

7.1 Determination of treatment effectiveness on a range of raw water sources

The results presented in Chapter 4 obtained from treatment trials undertaken on 4 separate and distinct water sources have demonstrated *M.oleifera* seed solutions to be a highly effective primary coagulant particularly under high raw water turbidity conditions. However, the variability in treatment effectiveness at low turbidity, depending on the water source, does indicate that there are limitations to its use.

From the results presented in Chapter 4 an attempt has been made to further define the mechanism by which *M.oleifera* seed solutions effect coagulation. It has been proposed that the primary mode of action is that of charge neutralisation via the patch mechanism, however, this does not fully explain results obtained in this and other studies that have been reviewed. Given the results from more recent work that has demonstrated a propensity for initially soluble seed material to precipitate at pH levels similar to those of river water it has, therefore, been hypothesised that there exists a mechanism that is a combination of charge neutralisation and precipitation. The extent to which one or other of these mechanisms predominates is unknown.

One of the main findings of the studies in Malawi and Zimbabwe was the observation made regarding the nature of the floc formed at low turbidity. For the Thyolo and Mwanza sources, these were small and compact; whilst for the Walkers Ferry and Morton Jaffray sources they were large and woolly. It has been postulated that the nature of the floc is dependent on the quantity and composition of the natural organic materials (NOM) present in the water being treated. Interactions between the *M.oleifera* proteins and the NOM results in the formation of long chain agglomerates that behave in a similar mode to high molecular weight synthetic polyelectrolytes. Under such circumstances it is considered that the mechanism of activity reverts from that of

solely charge neutralisation to one of charge neutralisation and inter-particle bridging thereby producing the larger floc.

Although an attempt has been made to further define the mechanisms of action of seed solutions, it is clear that further work is required. Preliminary work has been undertaken to isolate, identify, sequence and characterise those proteins considered responsible for the coagulant activity of the seeds, however, this is an area that needs to be addressed further. Only by fully characterising the proteins involved and establishing how they behave in isolation and in combination with each other will a better understanding of the mechanisms of activity be achieved.

7.2 Pilot and full scale trials

The work presented in Chapter 5 has examined the potential for using *M.oleifera* within continuous flow water treatment systems. In addition to investigating the physical nature of the floc formed in relation to settlement under non-quiescent conditions, the full scale trial provided an opportunity to establish potential constraints to application resulting from the processing of whole seed to obtain seed powder, and the make up of dosing solutions.

As a coagulant, seed suspensions have been demonstrated to be capable of producing floc under continuous flow conditions suitable for settlement. As for the laboratory studies, results were particularly good under higher turbidity conditions. Under what were classified as medium turbidity conditions, residual turbidity and floc carry over to the sand filters was found to be greater than that observed with alum. This was due to the lighter more compact nature of the formed floc. Under such circumstances, the use of flocculation aids may be considered to increase the size and settleability of seed induced floc. This is an area that has not been considered during the course of this work and needs to be addressed. In particular, attention should be given to the nature of the flocculation aid to be used, as, given the amphoteric characteristics of the proteins, it is possible that cationic polyelectrolytes may be found to be more effective than anionic or non-ionic flocculants.

144

The full scale trials were the first to be carried out using *M.oleifera* powdered seed suspensions and have highlighted a number of potential constraints to utilisation of such material on a large scale. In summary these include:

i)   The requirement for mechanical processing methods to remove seeds from the pod, remove the seed shell and crush the seed to a powder;

ii)   The requirement for regular cleaning of dosing tanks to remove settled seed material. Such material would ideally have to be disposed of in a controlled way in order to minimalise potential environmental problems;

iii)   The potential to increase Total Organic Carbon levels within treated water.

iv)   Cost effectiveness. If *M.oleifera* seed were to be seen, on performance, as a viable alternative to conventional chemical coagulants it would still have to be shown to be as cost effective. This has still to be shown.

v)   Regulatory issues. It is likely that in the future stricter regulations will apply in developing countries regarding materials that may be used within water treatment systems. There are issues, particularly relating to quality control of seed and seed processing, which may hinder the process of obtaining regulatory approval.

Although there are advantages to the use of *M.oleifera* seed solutions over conventional coagulants viz.: pH independence of treatment, wide dose range for effective treatment and the potential for use sludge produced during treatment as a soil conditioner / fertiliser (further work is needed to establish the latter), it has to be concluded that the constraints to use of *M.oleifera* seed suspensions for large scale potable water treatment outweigh the possible benefits. However, despite this conclusion this does not necessarily mean that large scale utilisation of *M.oleifera per se* cannot be considered. Two alternatives exist:

• Utilisation of seed powder or oil free presscake for the treatment of domestic and industrial wastewaters. Increasingly conventional coagulants are being used to treat wastewaters and, as such, the potential for *M.oleifera* seed use as an alternative must also exist. The potential advantages offered by *M.oleifera* use

145

include the production of an essentially bio-degradable sludge for re-use and, given the iron and manganese removal results in Chapter 4, the possibility of the specific recovery of metals and removal of heavy metals. This is an area that requires further investigation.

- The commercial production of a protein extract using seeds as the raw material.

Given the enormous potential of *M.oleifera* as a new cash crop for both large scale and subsistence farmers within developing countries, the opening up of alternative avenues whereby the seeds can still be utilised within water/wastewater applications is considered a priority area for research and development. In addition, it is considered that only by ensuring *M.oleifera* development on a large scale can its use at household level become more acceptable and widespread.

APPENDIX – Published work from this study

The following publications arose directly from work detailed in this study.

Folkard, G.K., Sutherland, J.P. & Jahn, S.A.A. Water clarification with natural coagulants and dissolved air flotation. *Waterlines* 1986, 5, (2), pp.23-26.

Folkard, G.K., Grant, W.D. & Sutherland, J.P. Natural coagulants in water clarification. In: Ince, M. ed. **Rural Development in Africa: Proceedings of the 13th WEDC Conference**, Lilongwe, Malawi, April 6-10, 1987. Loughborough University of Technology Press, 1988, pp.42-49.

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