Urban Water Research Association of Australia

Assessment of On-line Particle Counters for Routine Control of Microbial Pathogens at Water Treatment Plants

Research Report No. 153

URBAN WATER RESEARCH ASSOCIATION OF AUSTRALIA (UWRAA)

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Foreword

This report is based on UWRAA (now WSAA) Research Project No WS-70: "Assessment of On-line particle Counters for Routine Control of Microbial Pathogens at water Treatment Plants" which was commenced in July 1994. Organisational responsibilities for the project were as follows:

Sponsoring Authority:	Sydney Water
Research Agency:	Centre for Water and Waste Technology The University of New South Wales
Project Officer:	Colin Nicholson, Sydney Water
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EXECUTIVE SUMMARY

Purpose of the Project

It is the purpose of this project to develop the necessary procedures and protocols to enable the routine use of particle counting technology in maintaining optimal treatment plant operation and, hence, water quality within required guidelines. Of major concern is the possible transport of pathogen particles through the deep bed filters under various water treatment plant operating regimes. Attention in this study is thus focussed on i) examining the calibration and optimisation of particle counters for selected particles with particular emphasis given to *Cryptosporidium*, and ii) examining the potential for breakthrough of pathogenic particles as a result of operation of the contact water filtration water treatment process.

To this end, a range of literature, bench and pilot scale studies have been undertaken with the aim of understanding the operation and implementation of particle counters in water treatment and, more specifically, assessing the capacity of particle counters to detect the presence of particles in specific size ranges.

Summary of Key Findings

Instrumentation evaluation

For routine on-line particle counting in a water treatment plant an obscuration sensor would appear to offer the most cost effective sensor type.

The issue of maintenance of floc integrity is critical to the set-up of any counter in a water treatment environment. Special care should be given to sample point placement and construction, sample tubing and joints, choice of a non-pumped flow controller, and proximity to electrical interference from high-load equipment such as backwash pumps.

Although the obscuration counter is the cost-effective option for on-line operation there exists tremendous scope in the laboratory and at the pilot scale for the

application of more sophisticated equipment that provides a greater insight into the mechanisms encountered in water treatment processes.

Pathogen sizing

The sensors of particle counting instruments will soon be routinely calibrated with industry standards for both size and counts. Following such calibration it is essential that the sensor be assessed for pathogen sizing using a water matrix that is representative of that which will routinely pass through the sensor in a water treatment plant.

Jar testing

The importance of coagulant dose to the rate of floc growth and the subsequent relationship to floc size and the ability of the floc to entrap 5μm latex spheres are areas worthy of further investigation. Recent work also suggests that the fractal nature of flocs generated from coagulant addition has a dramatic effect on their ability to capture specifically sized individual particles.

Humic acid was found to have a significant detrimental affect on the coagulation-flocculation process and although this is not a new finding the results served to emphasise the complex nature of the effect of natural organic matter (NOM) on the coagulation process. No doubt this effect will vary from water source to water source.

Filtration studies

Investigations into the filtration process have served to emphasise the knowledge gaps in this area, particularly with regard to the entrapment of flocs generated through the addition of iron salts.

A major finding of the study was the lack of association between particle passage through the filter and coagulant concentration in the finished water. The traditional view of particle breakthrough at the end of a filtration run is that floc material is the major constituent of such particulate material. However, if a filter is allowed to go

through to filter breakthrough before backwashing and if iron is used as the dominant coagulant, it would appear that the particulate material that breaks through is discrete particles with little or no floc attached. If time or headloss triggers for backwashing govern, breakthrough is avoided.

Field trials

Cryptosporidium oocyst seeding runs conducted using the Macarthur WFP pilot filter facility were effective in measuring the *Cryptosporidium* removal performance of the pilot filter under a limited range of operating conditions;

Cryptosporidium oocyst log reductions observed during the three seeding runs, (3.1 to 3.7-logs) were comparable to those reported previously for similar conditions;

The results of pilot filter performance evaluation for the removal of *Cryptosporidium* oocysts by filters of the specific water quality, coagulation, and filter design and operating characteristics of this study were entirely consistent with previously reported information on *Cryptosporidium* and filtration;

The data collected using the Macarthur WFP pilot filter facility during the three brief seeding runs suggest the potential existence of useful correlations between *Cryptosporidium* oocyst removal and removals measured in terms of one or more of the more easily measured conventional performance parameters, turbidity, particle concentration, and aerobic spore concentration.

Recommendations

1. The virtual absence until recently of particle counters in Australian water treatment plants and the absence of guidelines for interpretation of particle counts in Australia has resulted in a lack of knowledge with regard to the day-to-day operation, maintenance and quality assurance procedures for particle counter operation. **It is thus recommended that steps be taken to encourage the wider**

use of particle counters in water treatment and to acquire a significantly larger data base of information on particle counts in treated water (enabling comparison with the more traditional measures of treated water quality such as turbidity).

2. The proposed 3-log reduction rule that has been suggested in the US is clearly invalid for water treatment plants in Sydney where the raw water particle counts are in the vicinity of 3,000 to 5,000 for the majority of the year. A 3-log reduction would require the filtered water to be about 3 to 5 counts/mL, such quality being better than distilled water and comparable to reagent grade water. As such **a guideline value based on a specific particle counts/mL in the finished water would appear to be more achievable and manageable.** Such a recommendation is consistent with views recently reported elsewhere (Hargesheimer et al., J. AWWA, 90, 32-41 (1998)) that log removal is not a good indicator of plant performance because it is influenced more by particle counts in source water than by those in finished water.

3. A variety of studies on the effectiveness of specific particle capture by flocs generated in the coagulation process have been undertaken within this project. The Malvern Mastersizer particle sizing instrument has been used extensively in these studies and was found to produce results of direct application to the water treatment process. As expected from standard Smoluchowski particle-particle interaction kinetics analysis, large flocs provide better entrapment of individual particles with flocs of mean size greater than approximately 100 μm apparently necessary to ensure reasonable capture of individual particles. Further insight into the ability of flocs to capture individual particles will require consideration of the fractal nature of these flocs (which exhibit increasing porosity with increasing size). Recent work suggests that entrapment of individual particles by porous flocs exhibiting fractal characteristics may be significantly less efficient than previously thought. **Considerable additional work in the area of particle entrapment by fractal aggregates is urgently required, particularly extending the work from model systems (latex aggregates) to those typically used in water treatment (iron and aluminium oxides) and further examining**

the effects of surface charge, pH and organic adsorbents on particle capture by fractal aggregates.

4. Preliminary consideration within this project has also been given to particle capture within the deep bed filter and has highlighted the possible importance of floc breakup within the deep bed as a mechanism for release of specific particles (such as those of *Cryptosporidium*) and a reduced likelihood of capture within the filter. Such a mechanism of particle release appears to be particularly problematic for flocs formed from iron salts which are recognised to be generally larger and weaker than their alum-induced counterparts. **Further studies are required to ascertain the significance of floc strength in maintaining contaminant particles within the filter bed.**

5. **Extension of the above issues, related to the coagulation and filtration processes in water treatment, to the development of an accurate description of the principal factors (under given raw water quality and process operating regimes) responsible for lack of capture of specific particles (such as *Giardia* and *Cryptosporidium* oocysts) is also urgently needed.**

6. Recommendations that follow logically from the results of the field seeding trials include the following:

a) The generally interpretable results of the introductory pilot seeding study indicate that further work using the basic elements of approach and methodology as used here would produce similarly effective results;

b) The likely existence of correlations between *Cryptosporidium* removal and other more easily monitored parameters such as turbidity, particle concentration, and aerobic spore concentration suggested by the results of this project indicate the desirability of further developing such correlations for application to treatment process monitoring and control;

c) The effective results of this introductory pilot seeding study strongly suggest the desirability of further describing the *Cryptosporidium* removal

performance of SWC filtration facilities to identify potential problems and the most effective operating strategies.

1. STUDY OVERVIEW AND LITERATURE REVIEW

1.1 Study Overview

It is the purpose of this project to develop the necessary procedures and protocols to enable the routine use of particle counting technology in maintaining optimal treatment plant operation and, hence, water quality within required guidelines. Of major concern is the possible carryover of pathogenic particles under various water treatment plant operating regimes. Attention in this study is thus focussed on i) examining the calibration and optimisation of particle counters for selected particles with particular emphasis given to *Cryptosporidium*, and ii) examining the potential for breakthrough of pathogenic particles as a result of operation of the contact water filtration water treatment process.

To this end, we have undertaken a range of literature, bench and pilot scale studies aimed at understanding the operation and implementation of particle counters in water treatment and, more specifically, assessing the capacity of particle counters to detect the presence of particles in specific size ranges.

1.1.1 Report layout

This report is made up of five stand alone chapters which, while independent, provide complementary information of importance to delivery of the project objectives. A review of literature pertaining to i) the use of particle analysis for water treatment control, ii) treatment strategies for removal of protozoan pathogens, and iii) the use of particle analysis as an indicator for protozoan cyst removal is presented in Chapter 1. In Chapter 2, we delineate the operating principles of the major particle characterisation tools that are available then use these various instruments to charactise particles under conditions typical of those prevailing in water treatment.

The results of more detailed investigations of the impact of system conditions (pH, alkalinity, humic acid content) upon both the size and counts of *Cryptosporidium parvum* oocysts as determined by light obscuration using a Hiac Royco instrument are reported in Chapter 3 while in Chapter 4 we report the extent of capture of Cryptospridium oocyst-sized particles by iron oxide flocs formed under a variety of system conditions (differing doses of ferric chloride, presence/absence of cationic

polymer, increasing concentration of humic acid).

The results of laboratory-based studies into the ability of deep bed filters to remove particles of *Cryptosporidium* oocyst size are given in Chapter 5 and in Chapter 6, we report the results of *Cryptosporidium* oocyst seeding trials undertaken using the Macarthur water filtration plant pilot plant facility where the goals of the trials were i) to define the *Cryptosporidium* removal characteristics of the Macarthur water filtration plant under selected operating conditions, and ii) to examine the relation between particle counts, turbidity and *Cryptosporidium* oocyst concentrations as alternative measures of treatment performance.

A series of conclusions and recommendations are presented in Chapter 7.

1.2 Literature Review

1.2.1 Limits and Context of the Review

This project has the specific aim of assessing the utility of particle analysers in monitoring the removal of the waterborne pathogens *Cryptosporidium* and *Giardia* in water treatment.

By far the greatest concentration of literature on waterborne pathogens is concerned with public health and the microbiology of the organisms. However, this project is concerned only with water treatment processes and as such only the literature that deals directly with particle analysis technology and treatment processes in the context of pathogen removal or monitoring will be discussed in detail. Literature on other issues dealing with waterborne pathogens is only given cursory attention for the sake of completeness.

Although *Cryptosporidium* and *Giardia* are the two protozoan parasites most commonly associated with waterborne disease there are a number of other protozoans, in particular *Cyclospora cayetanensis*, that have been isolated from diarrheal illness patients and which have been shown to have great potential for waterborne

transmission (Sterling, 1994). Irrespective of the array of literature on other waterborne pathogens, only that literature dealing with *Cryptosporidium* and *Giardia* will be reviewed. The work in this report concentrates on *Cryptosporidium* in particular as the removal and inactivation efficiencies reported are far superior for *Giardia* and by inference this organism poses less risk.

The literature reviewed for this report has been drawn from various sources up to and including the WQTC in Denver, Colorado that was held in November 1997. Very little literature that has been published since that date has been included in this report.

1.2.2 Waterborne Protozoan Pathogens

Giardia are flagellated protozoans that have been associated with waterborne disease since 1966 (Moore, 1969). Since this time *Giardia*, in particular *Giardia lamblia,* (also known as *G. intensinalis* and *G. duodenalis*) has been associated with numerous disease outbreaks throughout the world.

The environmental state of *Giardia* is a resilient cyst that may survive for considerable time in the environment though colder climates are more conducive to survival. The cyst has been reported (Lin, 1985) as being ovoid in shape, ranging from 8 to 14μm long and 7 to 10μm wide.

Like *Giardia, Cryptosporidium* has an environmentally resilient stage known as an oocyst. *Cryptosporidium parvum*, one of the two mammalian isolates and the only species associated with disease in humans (Current, 1987, Smith et al., 1995), are somewhat more spherical in shape than *Giardia* with a reported diameter of between 4 and 6μm. The association of *Cryptosporidium* with waterborne disease is more recent than *Giardia* being confirmed only as recently as 1987 (Rose et al., 1988) though the species has been recognised as a cause of diarrhoeal diseases since 1976 (Meisel et al., 1976).

The most widely accepted methodology for the detection of these organisms in water

relies upon a primary concentration technique, followed by a secondary sample preparation technique, followed by a quantification technique based upon immunofluorescence (IF) microscopy (Bee et al., 1991). This methodology requires considerable skill to perform to an acceptable level and currently requires some days before a result is possible. As well as IF microscopy, flow cytometry has become an accepted and more accurate means for both *Giardia* and *Cryptosporidium* (Vesey et al., 1994) though the current cost of the instrumentation is limiting the use of this method to research applications.

Consequently, research has sought to identify an easily measured surrogate for these pathogens. Traditionally turbidity has been the parameter monitored as a pathogen indicator, with the latest US regulations suggesting a limit of 0.1 NTU (SWDA, 1989), more recently however, research in the US is suggesting that particle counting is a more sensitive indicator for the presence of pathogens (Hargesheimer and Lewis, 1992). In contrast the 1996 Australian Drinking Water Guidelines recommend the monitoring of turbidity for aesthetic purposes only, with a recommendation of 5 NTU for unfiltered water and 1 NTU for filtered water.

1.2.3 The Use of Particle Analysis for Water Treatment Control

As technology advances, there should arise products or methodologies that have advantages over existing technology. At present, particle counting technology is seen as the heir apparent to the current turbidity throne, as once turbidity aspired to the ancient throne of gravimetric analysis. At present, particle analysis technology is generally used only on a limited basis, primarily in pilot plant studies of alternative technologies, in the US (Hargesheimer and Lewis, 1992) and in a very limited way in Australia (Murray, 1994).

A recent and extensive evaluation of the value of particle counting technology for water treatment purposes concludes, "...particle counting was not simply a supplementary analysis to turbidity. Particle counting stood alone as a powerful tool for the evaluation of filtered water quality", furthermore, "Particle counting greatly

enhanced the discernment capabilities of the analyst and provided a better margin of control for particulate removal " (Hargesheimer and Lewis, 1992).

Particle counting technology has been applied as a tool to evaluate treatment plant performance sporadically over some twenty years. In the US in 1976, Beard and Tanaka (Beard and Tanaka, 1977) compared the results of turbidity measurements using various Hach models with results obtained by a Hiac PC 320 particle counter which measured particles in twelve discrete sizes, ranging from 1 to 150μm. Their conclusions were that particle counting technology was a useful measurement tool which provided quantitative information on suspended matter. Furthermore, they found particle counting to be a more sensitive measurement technique than turbidity, however they reported particle counting instrumentation to be approximately three times the equivalent cost of turbidity instrumentation and it also required considerably more operator skill and time to produce results. These operational difficulties were largely due to the early particle counters being only batch instruments whereas turbidity could be accessed online.

The year after Beard and Tanaka completed their work the USEPA National Interim Primary Drinking Water Regulations, drafted in 1975, took effect. For the first time authorities realised that there could be an indirect association between turbidity and health as well as aesthetics and a mandatory, rather than recommended limit, was set for turbidity of 1 NTU (Tate and Trussell, 1978). This had the affect of generating research into techniques to produce water with a turbidity less than or equal to 1 NTU at a lower cost.

The following year Tate and Trussel (1978) noted that turbidity measurements were an indirect measure of the particulate matter in solution and did not always provide the information required by operators seeking to optimise pilot plant trials. Their work focused on comparing the information produced by different particle counters, and then choosing an instrument to compare to the traditional turbidity measurements. The researchers were aware of the work of Hopkins and Young (1974) who reported that different particle analysers produced different size results when compared to a microscope, depending on the technology used to size the particles.

The first instrument, a Coulter Counter, counted and sized particles by a change in electrical resistance resulting from a modification in rate of flow of an electrolyte through an orifice (which the particles blocked) and subsequently between two electrodes. The second instrument that was tested was a Hiac Model PC-320, the same instrument used by Beard and Tanaka (1977), which detects and sizes particles due to light obscuration; with the shadow area being proportional to the particle size. A third instrument assessed, which used light from a rotating laser to produce a forward light scatter, was a Spectrex Prototron IL1 100. In this instance the diameter of the conical section striking the photodetector was related to particle size. A fourth instrument was a Royco Liquidborne 345, which used a light obscuration technique similar to the Hiac instrument. Various selection criteria for the choice of particle analyser were described and the instrument that was chosen for the pilot scale testing was the Hiac instrument.

The work of Tate and Trussell (1978) is interesting because they demonstrated the advantages of particle analysis over turbidity. One example of this is that their trials on a typical coagulation, flocculation, sedimentation and filtration process revealed that turbidity measurements were 3.8 NTU in the raw water and 3.9 NTU in the coagulated-flocculated water which reduced to 0.18 NTU in the filtered water. Whilst particle analysis showed that particles less than 25μm passed through the sedimentation process, it also demonstrated that most particles of less than 10μm size passed through the filters as well.

Tate and Trussell (1978) considered that their results demonstrated the greater sensitivity and richer data source of the particle analysis instrumentation as compared to turbidity measurements whereas their results in the modern light of pathogen removal concern would have been quite momentous. Indeed, the work of Tate and Trussell (1978) can be described as the first investigative use of particle analysis for staged process analysis in drinking water treatment.

On a more economical note, the cost savings that may be achieved through the utilisation of particle analysis technology was reported by Monscvitz and Rexing

(1983) and later by Hutchison (1985) in studies undertaken for the Southern Nevada Water System. Hutchison (1985) reported that a reduction in chemical costs of over 30 percent was possible with the use of particle analysis technology to optimise coagulant dosage. The added benefits of less frequent backwashes and lower sludge production were also reported. Hutchison's work was conducted on a direct filtration plant delivering 390 ML/day and using alum as the primary coagulant.

Since this time there have been numerous articles describing the applications of particle analysis technologies to water treatment investigations. However an application of particle analysis technology that has been proposed relatively recently is the use as an indicator for the removal efficacy of the waterborne protozoan pathogens. Such an application has been seen to be viable particularly because the size of *Giardia* and *Cryptosporidium* are comfortably in the range of analysis of most available particle analysers.

1.2.4 Treatment Strategies for the Removal of Protozoan Pathogens

The impact of the waterborne pathogens on the water industry in the US became apparent during the late seventies and eighties with the waterborne transmission of *Giardia* becoming a concern (Craun, 1988; Rose, 1993).

During the early 1980's specific treatment methods for the removal of *Giardia* began to emerge in the literature in the US. Some of the first work that was specifically aimed at *Giardia* removal was reported by Logsdon and coworkers (Logsdon et al., 1981). In this paper, earlier work is discussed which demonstrates that the design of specific treatment strategies for the removal of waterborne pathogens is certainly not novel. In fact work as early as the 1930's and 1940's was aimed at the specific removal of *Endamoeba histolytica* cysts; a pathogen responsible for many dysentery outbreaks including the notorious outbreak in Chicago in 1933 (Spector et al., 1934; Baylis et al., 1936).

This early work in the 1930's demonstrated the effectiveness (99.99% removal) *of E.*

histolytica by direct sand filtration, though it is difficult to know if the prior addition of coagulant assisted this removal. Further work by the US Army during World War II (Department, 1944) demonstrated the necessity of prior coagulation with granular media (sand and gravel) for *E. histolytica* cyst removal. This work also included the development of diatomaceous earth media for drinking water treatment applications.

The investigations of Logsdon and coworkers (Logsdon et al., 1981) built upon these earlier studies with diatomaceous earth and granular media filtration for the removal of pathogen cysts, though *Giardia* cysts were now the subject of interest. Radioactive (Cerium 141) 9μm resin microspheres as cyst models or *Giardia muris* cysts rather than *Giardia lamblia* were used in these investigations. The radioactive microspheres had the advantage of being very simple to detect with one radioactive sphere in a sample detectable by scintillation counting. In contrast, cyst recovery experiments conducted by the researchers for diatomaceous earth filtration produced an average of 10% recovery whereas recoveries for the granular media tests averaged 61% recovery. Such results render the interpretation of removal efficiencies very difficult.

The conclusions drawn from this work were that diatomaceous earth was very effective for the removal of *G. muris* cysts (lowest removal of 99.34%), however the filters required a 1.0 kg/m^2 pre-coat of diatomite plus a continuous diatomaceous earth body feed to prevent filter clogging to obtain this efficiency. Their work with granular media filtration emphasised the need for effective coagulation of the raw water before filtration. Even so, the removal efficiencies reported without coagulation were as high as 94%, however, the removal efficiency could be as low as 59%. When optimum coagulation was employed in conjunction with filtration, greater than 99% removal could be maintained. More importantly their work showed that cyst breakthrough was most likely to occur after backwash, during filter ripening, and that turbidity was only a general indicator for pathogen removal. They emphasised this by reporting that during their studies a turbidity of 0.24 NTU corresponded to 200 cysts/L. On another occasion a turbidity of 0.30 NTU corresponded to 40 cysts/L. The turbidity (maximum contaminant level) guideline was 1.0 NTU at the time, which could represent a solution containing some 4000 cysts/L.

In 1986, Logsdon was again involved in work (Lange et al., 1986) which demonstrated the usefulness of diatomaceous earth for *Giardia lamblia* removal. The use of different grades of diatomaceous earth and the usefulness of alum coating on the media were investigated in this research.

Other work has concentrated on different filtration systems for *Giardia* removal. Al-Ani and Hendricks investigated rapid sand filtration (Al-Ani et al., 1986) while Bellamy and coworkers (Bellamy et al., 1985) demonstrated the effectiveness of slow sand filtration for *Giardia lamblia* cyst removal once a microbial population had been established in the sand bed (achieving almost 100% removal). Their work also reported 96% removal for standard plate count bacteria and 98% removal of particles in the 6.35 to 12.7μm range; particle removal was not emphasised in this work.

Following a winter outbreak of *Giardiasis* in 1983-84 at McKeesport, Pennsylvania, Logsdon produced some further work that investigated the efficiency of sedimentation and coarse anthracite filtration for the removal of *Giardia* cysts (Logsdon et al., 1985). The results showed that this treatment strategy removed between 65 and 93 percent of *Giardia* cysts; *G. canae* and *G. muris* cyst spikes were used. The interesting aspect of their work was the evaluation of different coagulants for cyst removal. Alum alone produced very poor removal while the addition of a high molecular weight, slightly anionic polymer produced markedly better results. Their work again emphasised the need for a filter to waste stage in a filtration run and that turbidity was only a guide to cyst removal. More recent work on the use of particle analysis for comparison of different coagulant performance was conducted in Canada (Jasim et al., 1997). The authors confirm the usefulness of particle analysis technology and demonstrated the improved performance that is achieved by using an aluminium-based coagulant with a higher sulfuric acid content over conventional alum.

A review of different treatment strategies for the removal of *Giardia* cysts was produced by Ongerth in 1990 (Ongerth, 1990). This analysis of the different systems including conventional filtration, in-line filtration and diatomaceous earth filtration again suggested that high removal rates for *Giardia* cysts was possible but

considerable care was required in the treatment plant operation.

In a more recent paper (Roefer et al., 1993), experience with the use of ferric coagulant and direct filtration, optimised through the use of particle counting technology, for the removal of *Giardia* cysts has been reported. Results suggested that removal efficiencies for cysts as high as 99.9994% (5-log) could be obtained, however they reported some interference in cyst enumeration due to the presence of the ferric chloride. Parallel studies with polystyrene beads (though of 5μm diameter) yielded a maximum of only 99.96% (3-log) removal.

In 1995, Timms et al. (1995) demonstrated the effectiveness of slow sand filtration for the removal of oocysts with better than 99.997% removal being achieved. The results are reassuring for consumers of water in London, as slow sand filtration has been the traditional filtration process for the greater London area for at least the past 165 years.

In comparison to this study, Hall *et al.* (1995) investigated the efficiency of oocyst removal using chemically assisted dissolved air flotation. The removal efficiency was found to be better than 99% with no discernable dependence on the media used following DAF clarification. The authors suggest that DAF is a comparable clarification technique to settling and in some instances has the additional advantage of good algae removal. The authors also make reference to the practice of recycling backwash water increasing the risk of protozoa prevalence in the finished water.

1.2.5 The Use of Particle Analysis as an Indicator for Protozoa Cyst Removal

Concerns that *Cryptosporidium* and *Giardia* might induce waterborne disease began to emerge in the mid-1980's. *Cryptosporidium* was also reported as having been transmitted by water that had met or exceeded all water quality guidelines (Rose et al., 1988), including turbidity, although little if any information was available by 1991 for the effective removal of *Cryptosporidium* during water treatment (LeChevallier et al., 1991). The Surface Water Treatment Rule (SWTR) that was enacted by the USEPA was the culmination of many years of concern over the apparent widespread

transmission of waterborne diseases. This rule states that treatment systems are required to ensure overall removal or inactivation of at least 99.9% of *Giardia* cysts and 99.99% of enteric viruses (USEPA, 1989); a recommendation was made for *Cryptosporidium* oocyst removal in the following Enhanced Surface Water Treatment Rule (ESWTR), to be implemented in 1999.

The difficulty faced by most treatment operations, following the enactment of the SWTR, was that regular monitoring for *Giardia* cysts at environmental concentrations in raw and filtered waters was extremely time consuming, expensive and normally subject to large errors by all save highly trained and experienced staff. At or about the same time the realisation that turbidity may be too coarse a measurement to assess filtered water quality in terms of waterborne pathogens was becoming more widely accepted, many years after the earlier work of Logsdon (Logsdon et al., 1981). The move toward the use of particle removal as a better indicator for pathogen removal was gaining momentum.

Work on particle removal by Lewis and Manz in 1991 (Lewis and Manz, 1991) used the new forward angle light scatter (FALS) technology provided by Hiac-Royco. This technology was reported to measure particles down to 0.5 μm, which not only adequately incorporates the detection of *Cryptosporidium* and *Giardia* particles but was also reported as being superior to the alternative techniques such as Coulter Counting and light obscuration methods.

Lewis and Manz sized *Giardia* in the 1 to 5μm size range using the FALS instrument; however, no sizing was conducted on *Cryptosporidium* in this work. The difference in sizing between the FALS technology and optical microscopy was accounted for by the requirement of the FALS technology that the refractive index of the particle be measured. Their work not only demonstrated the importance of calibrating particle analysis instrumentation with pure solutions of pathogens but also advocated the monitoring of particular size fractions as an indication of pathogen removal efficacy.

The relationships between particle counts, turbidity, *Giardia* and *Cryptosporidium* were investigated by LeChevallier and Norton (1992) in an effort to identify

indicators of treatment efficiency in filtered water supplies. Their work showed that a measure of removal of particles >5μm and turbidity were useful predictors of *Giardia* and *Cryptosporidium* removal. The authors did stress however, that this relationship may not be valid for all treatment plants and that regulating parasite treatment by requiring a certain log removal of turbidity or particle counts was not justified. The work also confirmed the results of previous studies that particle counts were a more sensitive indicator of filter performance than turbidity. LeChevallier and Norton (1993) later amended their recommendation to removal of >3μm as being a better surrogate for protozoa removal than removal of >5 μm particles. The authors also stated that the removal of particles of >3μm was shown to be significantly correlated to treatment of *Giardia* and *Cryptosporidium* by coagulation and filtration and that their results generally supported a linear relationship between removal of particles and parasites.

1.2.6 The Australian Context

Early work on the application of particle counting in water treatment was undertaken by James M. Montgomery Consulting Engineers (now Montgomery Watson) for Wyong Shire Council in 1978 who had, just prior to this involvement, developed an expertise in the area. Further trials were conducted by Montgomery Watson in Newcastle in 1993 (Murray, 1994).

Since then, work has been conducted in 1992-93 by the then Sydney Water Board. This work by Harris (1998) demonstrated the usefulness of a portable particle counting unit for analysing the change in particle size distribution from Warragamba Dam through the Prospect Reservoir to the main city tunnel. In summary the work demonstrated a general decrease in particle size distribution upon chlorination and pumping. The particle size distribution increased again as the water passed through the mains. In 1993 studies utilising the Prospect Water Treatment pilot plant, Entwisle and Murray (1993) reported the usefulness of particle analysis in indicating a likely filter breakthrough before a corresponding increase in turbidity was observed. Some work was also conducted on the removal of *Cryptosporidium* oocysts by coagulation

with alum and cationic polymer followed by direct filtration through deep dual media filters (Murray, 1994, 1995). This process was reported to remove 99.99% (4-log) of formalin-killed oocysts.

The Hunter Water Corporation has also investigated the use of particle analysis technology for treatment process control (Bailey, 1993; Murray, 1994). This work confirmed the results found in the US that particle counting was a more sensitive technique for the investigation of filter performance. Changes in filtration rate were found to be an important factor in particle removal efficiency with sharp changes leading to filter breakthrough whereas gradual change or ramped change allowed for continuous, effective particle removal. The work reported that the removal efficiencies of particles in the 4 to 10μm range was between 96 and 98% for the Grahamstown Treatment Plant and greater than 99% at the Dungog Treatment Plant.

In general, when compared to the UK and the US, the usefulness of particle analysis technology has not been widely accepted by the Australian water industry (Murray, 1995) with no drinking water treatment plant until recently using particle counting as a process monitoring tool.

1.2.7 Current Trends

Within the area of public health related to protozoa, the trend has moved toward developing standards for control of parasites based upon levels of acceptable risk (Teunis et al, 1997, Haas et al, 1996). Some papers have continued to report on levels of the organisms in rivers (States et al., 1997) or in open reservoirs (LeChevallier et al., 1997) or to reevaluate outbreaks (Fox and Lytle, 1996; Roefer et al., 1996). The microbiological research in this area has focused on finding better detection methods (Linquist, 1997) or on reevaluating existing techniques (Faulkner et al., 1997; Sethi et al., 1997).

Over the last three years the trends in particle related research has continued in much the same way involving the evaluation of plant performances and the correlation of

particle removal with protozoan removal (Nieminski and Ongerth, 1995; Li et al., 1997; Hancock et al, 1996; Ongerth and Pecoraro, 1995). Other workers have evaluated different treatment types to the traditional granular media filtration such as DAF (Valade et al., 1996; Plummer et al., 1995) and membrane filtration (Jacangelo et al., 1995).

The emergence of image analysis technology allowed Spicer and Pratsinis (Spicer and Pratsinis, 1996) to develop a better understanding of the relationship of shear force and floc size and structure, as indicated by the fractal dimension during alum flocculation. Their work showed that the average floc structure became less open or irregular as the floc size distribution attained a steady state.

Boller and Kavanaugh (1995) undertook modeling analysis of granular filtration. Their work demonstrated that the rate of headloss build-up is strongly dependent on the size of the particulates in suspension and the size of the granular media. More rapid headloss development was observed in larger sized media due to the lower relative density of deposits that occurs in the larger pore spaces of the larger media. Stevenson (1997) adapted the Carman-Kozeny equations relating flow through porous media in an effort to produce a general model that could be used for filter design. The work was able to simulate such observed phenomena as the formation of wormholes in clogged layers and breakthrough with increased flow on used filters.

As the installation of on-line particle counting has increased, a number of papers have emerged on the investigation of various aspects of the treatment process with a view to improving particle removal. Colton *et al* (1996) studied filter backwash and start-up strategies to identify optimum conditions to minimise the passage of particulates into the filtered water. The work demonstrated that approximately 40% of all particles that pass a filter in a 48-hour run do so in the first hour of operation. The research identified optimum backwash conditions as well as demonstrating the benefits of slow start-up and its dependency on media type.

The usefulness of particle counters to assess the performance of different coagulants on particle removal has been demonstrated (Jasim et al., 1997) whilst other workers

showed the value of having each filter monitored by an independent counter rather than one counter for the combined filtered water flow (Ginn et al., 1997). This latter work emphasised the complementary nature of particle counting and turbidity in that particle counters are more sensitive to filter breakthrough however turbidity was a more sensitive indicator of deviation of optimum coagulation performance. These observations again emphasise the characteristic breakthrough of filters being in the several micron particle size region which particle counters detect but turbidity is insensitive to. It also emphasises that turbidity meters are not redundant when particle counters are installed but rather are a complementary process-monitoring tool. Turbidity meters are sensitive to submicron (such as colloids and viruses) particle breakthrough seen when coagulation is non-optimal whereas current on-line particle counters do not detect particles of this size. This subject will be discussed more fully in section 5. The work by Ginn et al. (1997) also identifies the usefulness of particle counters to detect structural problems within the plant such as filter wall erosion and leaking backwash valves.

Further evaluation of particle counters was carried out by O'Shaughnessy et al. (1997) by comparing the results produced by both scatter and obscuration sensors with that given by electron microscopy. The workers reconfirmed the well-established knowledge of undersizing of particles by particle counters when compared to microscopic sizing. The work also demonstrated good correlation between log removal values determined by the different counting techniques.

At the recent Water Quality Technology Conference in Denver, Colorado (November 1997) a number of authors submitted work that dealt with quality assurance procedures for on-line particle counters. The submissions were divided between those establishing methods for the validation of count correlation (that is, ensuring the ability of counters within a facility to agree with each other when analysing the same water) and those establishing size verification. Traditionally, a particle counter is supplied with factory calibration of the sensor for size, coincidence limits and resolution but not counts (Goldsmith et al., 1997). For counters manufactured in the US, the industry standard for primary calibration by the manufacturer is ASTM F658 where sizing, coincidence and resolution are established with NIST traceable latex

spheres (Goldsmith et al., 1997).

Goldsmith et al. (1997) propose a new field calibration verification method that is not based on the established Master Counter technique in which counters are calibrated against a master counter that has been calibrated to ASTM F658. This master counter calibration is effective for count matching a series of counters but does not allow for individual counters to be tested for resolution and coincidence.

The problem of a count standard was addressed by Vasiliou et al. (1997) who produced both size and count standards for the cross comparison of various counters of different manufacture. The count standards were produced using a gravimetric method that produces a count standard based on the percent solids of monodisperse latex sphere solutions. From the knowledge of percent solids and knowledge of particle density and size, the number of particles per millilitre can be established. The count standards typically had an uncertainty within 20%.

Work of a similar philosophy was conducted by Chowdhury et al. (1997) who identified the need to establish preservation and shipping protocols for particle count samples. Such sample handling may become important as quality assurance programs identify the need for independent verification of particle counts. In summary the researchers found that preserving samples by storing, headspace free in chilled Teflon bottles was currently the best technique although it was no substitute for immediate on-site measurements.

2. Evaluation of Particle Counters

2.1 Introduction

Turbidity is caused by suspended matter such as clay, silt, finely divided organic and inorganic materials together with coloured dissolved materials. It is measured by determining the amount of light that is scattered at 90^{0} to the incident light and the most popular unit of measure is called the nephelometric turbidity unit or NTU.

The meters are normally calibrated against standards that consist of known amounts of suspended, opaque material. As the ability of a particle to scatter light is dependent upon its size, shape and refractive index it is necessary to adopt a standard opaque material with which to produce turbidity standards. The material used in most turbidimeters is a suspension of Formazin polymer, and is prepared by combining known amounts of hydrazine sulfate and hexamethylenetetramine. In practice however, most instruments are supplied with prepared standards that consist of Formazin suspended in a transparent gel.

When the turbidity of a water sample is measured it is necessary to allow any large material to settle or float and a determination of light scattering of just the suspended material performed. The ease of measurement of turbidity is expressed in the popularity of the instruments, whether they be portable models for field measurements, bench scale models for batch sampling throughout a treatment process or online models for determining both treated and raw water quality.

Particle counting is not new technology having been used for many years in other industries such as minerals processing and pharmaceuticals. However, the application as a water quality indicator is relatively new with widespread use only occurring in the 1990's. Few trials have been conducted in Australia and there is no indication that particle counting will become a water quality standard or guideline.

Particle counting is used to both count and size particles in solution, and therefore provides much more information on the nature of the particles than does the simple measure of turbidity. Importantly, particle counting is a two-dimensional measurement, in that both size and counts are recorded whereas turbidity is one-

dimensional with only light scatter measured. As such there is no single conversion factor that can be developed to relate these two parameters. Frequently, the two terms are used interchangeably but this should be avoided, as the relationship between them is empirical at best.

There are three major analysis techniques for particle counting; light obscuration, light scatter and electrical resistance. The essential features of each of these instruments are detailed below.

2.1.1 Light Obscuration Counters

Light obscuration particle counting is also referred to as light blockage or light extinction particle counting. This technique has been in use for assessing drinking water treatment performance since the late 1970's and gained ardent supporters during the 1980's. Its prior use in other research areas such as pharmaceuticals and minerals processing is well documented and an excellent recent edition text in this area is that written by Allen (Allen, 1997).

The basic principle of operation of the particle counting sensor is shown below:

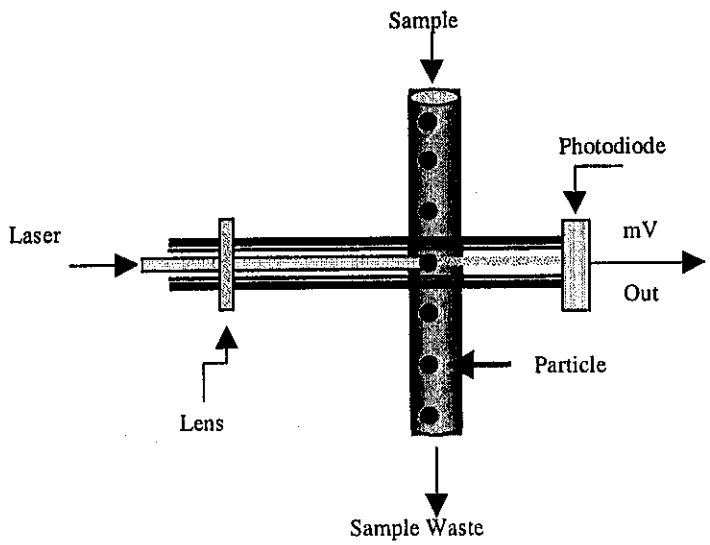


Figure 2.1. Schematic of an obscuration sensor

For light obscuration the sample flows through a sensing zone. In this zone a beam, usually a laser beam, of known intensity shines through the solution to a photodetector. When a particle flows through the sensing zone the beam is intersected and some of the light is scattered or absorbed by the particle resulting in a drop in intensity of the light beam. The change in light intensity is a function of the particle's projected area or shadow and the refractive index of the particle. The change in intensity at the photodetector is transmitted to the data processing unit, which counts and processes pulses to give a particle size distribution. This instrument measures the particle area but the data is usually expressed as particle diameter, which assumes that the particle is spherical.

Light obscuration instruments are capable of measuring particles in the 1 to 500μm size range though a specific sensor configuration will usually have a much smaller range such as 2 to 100μm. The lower limit is determined by the strength of signal that is significantly different to the background noise to be confidently interpreted as a particle. The upper limit of the instrument is determined largely by the size of the sensor region and associated flow channels through which all particles must pass.

Some advantages of this type of sensor are that there is a linear response to particle diameter, good repeatability has been widely reported, a wide size range is able to be determined with this technique and this sensor is amenable to on-line adaptation. Disadvantages of this type of sensor include a lack of submicron sizing capability.

2.1.2 Light Scatter Counters

This technique is somewhat more recent than the light obscuration sensor having been first used in 1977 for medical research. It subsequently found its way to the aquatic environment with oceanographic research in the mid-1980's and now is a viable alternative to the light obscuration technique.

As with the obscuration sensor, light from a laser source passes through a sensing

zone where the sample solution passes in a stream. When a particle intersects the beam some light is scattered and some is absorbed. A photodetector receives the scattered light over a fixed angle. Forward angle scatter between 1 and 19°, side scatter around 80 to 100° and back scatter between 170 and 190° may be measured. Forward angle light scatter (FALS) is the most common of these techniques. After the scattered light is received by the photodetector, the information is processed and a particle size distribution is obtained.

In contrast to the obscuration sensor, submicron particle analysis can be performed with a typical range of analysis of 0.1 to 50μm however most counters have a much more limited range than this. Again, as with the obscuration instrument, the lower size limit is determined by the ability of the sensor to detect a significant signal above the background noise; the upper limit being also set by similar constraints as the obscuration instrument.

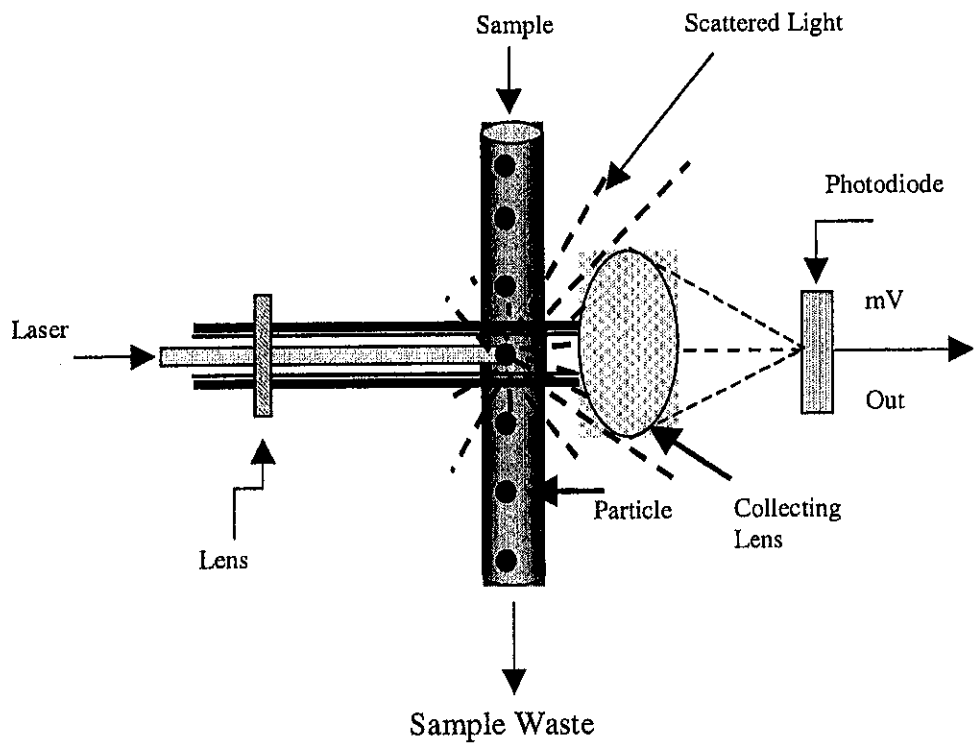


Figure 2.2. Schematic of a scatter sensor

Some of the sources of error for the light obscuration and scatter sensors are described below.

2.1.2.1 Particle Coincidence and Solids Concentration

The success of a particle counting instrument depends on only one particle being in the light path at once. Obviously, if two or more particles are both present, the data supplied to the photodetector will be interpreted as resulting from interference by just one particle.

This potential source of error is minimised by maintaining a balance between the flow rate through the sensor zone and the diameter of the sensor zone such that only one particle will pass through the light at once. Therefore, care should be taken not to introduce particles that may block the sensor zone. Additionally, the solids concentration of the sample solution should not exceed the limit established by the manufacturer. Because of such restrictions on this technique, particle counting is very seldom used in wastewater applications.

2.1.2.2 Sphericity and Refractive Index of Particles

Particle counting and sizing instruments are calibrated for discrete particle sizes using monodisperse (single-sized) particles of a known size and refractive index. Therefore, an assumption is made regarding the refractive index of the particles in the sample being not sufficiently different to result in errors in data interpretation. Additionally not all particles, in fact most, are not truly spherical and the non-spherical shadow cast by such a particle is nevertheless reported as a sphere. Therefore, an average diameter is being reported for roughly spherical particles. For rod and disc shaped particles the error may be quite large. This type of error is discussed in more detail in the sizing of pathogens section below.

2.1.3 Electrical Resistance Counters

In 1956 W.H. Coulter announced the development of a precise counter for discrete microscopic particles. Because of its origins the electrical resistance measurement is commonly referred to as *Coulter counting*. This technique found immediate application in many medical and industrial situations, the most famous of these being the counting of red blood cells. This method, having been used in the 1960's, predates the other two for aquatic applications.

This method requires that the matrix of the sample under analysis be conductive. For seawater this presents no problems but for freshwaters normally an electrolyte such as sodium chloride must be added.

A schematic of the operation of electrical resistance counting is shown below:

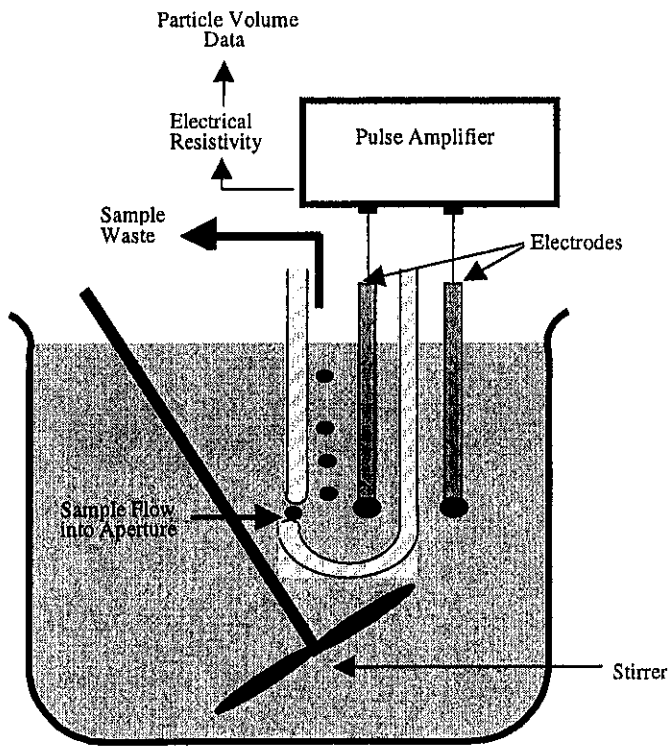


Figure 2.3. Schematic of a Coulter counter

The measurement cell contains a small aperture that is made by placing two electrodes a known distance apart. Between these two electrodes a current is passed. When a particle, sufficiently small, passes through the aperture there is a decrease in current that is proportional to the volume of the particle in the aperture. The current pulses are counted and analysed and the data is processed into a particle size distribution.

The measurement capabilities of this technique depend upon the size of the aperture that is present. A variety of such apertures are usually available and the range of measurement that is possible is in the order of about 0.3 to 1000μm. Of the three techniques discussed here the Coulter technique is the most versatile in terms of range of size determination. In addition the maximum particle concentrations that can be measured by this technique are typically an order of magnitude greater than that able to be measured by light sensors.

As with the light sensors, particle coincidence and deviation from sphericity would be disadvantages with this technique. The need for the addition of an electrolyte prior to measurement for freshwater studies would be considered a major disadvantage for the study of flocculation and coagulation as such an addition would seriously affect the surface charges of both natural particles and flocs. Despite these disadvantages, this technique is still widely regarded as the most accurate for determining the size of a particle. This is confirmed by the work of Hargesheimer and Lewis (1992) who studied a variety of particle counters. A summary of the findings, in terms of size determination comparisons, is provided later in this report. For investigations in wastewater, the technique may come into its own due to the higher ionic strength of wastes.

2.2 Counters Studied in This Report

Three instruments were evaluated in this work with a view to investigating different particle analysis technologies that might be applied to various processes in drinking water treatment. The processes of interest that particularly apply to particulates in

water are coagulation-flocculation, filtration and quality assessment.

To investigate coagulation-flocculation, the instrument must be able to differentiate between individual particulates and floc particles. To investigate filtration the instrument must be sensitive enough to determine the low concentrations of suspended particles that would be expected following an efficient filtration process. To investigate quality aspects, the instrument must again be able to determine low numbers of specifically sized particles in both the raw and filtered water.

A Hiac-Royco particle counting instrument was supplied by the Sydney Water Corporation and is the same instrument that was previously used and described by Murray (Murray, 1994) in his work on the Prospect project. The instrument has two different sensors - a light extinction and a dual extinction-scatter sensor - and is a typical particle counting instrument capable of on-line use. The second instrument was a Malvern Mastersizer E and was supplied by the Water Engineering Department at The University of New South Wales. This instrument is constructed to make use of small angle light scattering and has been used extensively by Amal and Waite (1992) for studies on fractal properties of particle aggregates. The third instrument was a Galai CIS-100 instrument supplied by the Chemical Engineering Department at The University of New South Wales. This instrument uses technology based upon light extinction and has not been extensively used before.

Cryptosporidium parvum oocysts and *Giardia lamblia* cysts were originally supplied by Graham Vesey at Macquarie University. Later studies using *Cryptosporidium parvum* oocysts were done using samples supplied by Jerry Ongerth at The University of New South Wales. These oocysts were isolated from infected calves and supplied weekly and stored at approximately 4° C without further treatment. The oocysts were supplied in lots of approximately 10^{9} oocysts as estimated by optical microscopy.

2.3 The Hiac-Royco

The Hiac-Royco system consisted of a 9600 counter and two sensors. The HRLD-150, which is a light extinction sensor, is designed for counting particles in the 1.2 -

150μm size range in up to 128 channels. The SEL-05, a dual sensor, uses both light extinction and light scatter to count particles over the range of 0.5 - 500μm in up to 128 channels. The crossover between the two sensors is at 2μm and occurs automatically. The system also included an ABS/2 batch sampling and an on-line multi-valve sampler together with a CFC-810 flow controller. The system was controlled and data collected by a Toshiba T1910 laptop operating the Particle Distribution Analysis System (PDAS) software supplied by Hiac-Royco.

The Hiac-Royco was initially used with the HRLD-150 sensor and the ABS/2 batch sampler. The batch sampler uses either compressed air or vacuum to drive the sample through the sensor channel. Initially, batch sampling of polystyrene spheres was used for calibration purposes however, when the batch sampler in the compressed air mode was used for sampling oocysts, there appeared to be a considerable skew in the particle size distribution toward the smallest particle size bin. It was possible that the air pressure of 140 – 420 kPa, which is used to force the sample through the 500μm sensor aperture, was resulting in rupture of the oocysts. The system was then reconfigured to incorporate the CFC 800 flow controller (flow of 25.0 ± 0.2mL/min) with a peristaltic pump or gravity feed. The oocyst size distribution when measured with these two sample introduction methods was more normally distributed.

Cryptosporidium parvum was found to have a modal distribution of between 1.53 and 4.95μm while *Giardia muris* had a modal distribution of between 4.35 and 7.55μm. A comparison with results obtained using other instruments and techniques is given in Table 1.

The Hiac-Royco sizes both pathogens in significantly lower size bins than reported from sizing by flow cytometry. However, the scatter sensor consistently sized the pathogens smaller than the extinction sensor thus, if one considers the flow cytometer as the reference sizing technique, it is apparent that the extinction sensor provides a more accurate result. This may reflect a greater sensitivity of the scatter sensor to semitranslucent particles such as oocysts and cysts.

Table 1. Summary of pathogen sizing results obtained in this study (in bold) compared to size information obtained by Hargesheimer and Lewis (1992).

Instrument	*Giardia Muris* (μm)	*Cryptosporidium parvum* (μm)
Flow Cytometer	8.9	4.3
Hiac-Royco HRLD-150	**4.35 - 7.55**	**1.53 – 4.95**
Hiac-Royco SEL-05	**4.55 – 6.75**	**<1 – 3.25**
Malvern Mastersizer	**7.08**	**4.33**
Galai CIS 100	**7.8 - 8.2**	**4.4 - 4.8**
Climet Instruments	3 -4	<2-3
Coulter Multisizer	8 - 9	4.16 - 4.33
Particle Measuring Systems	8 - 9	2 - 5
Spectrex Corp.	4 - 5	2 - 3 & 4 - 5
Met One	4 -5	2 -3 & 4 - 5
Hiac-Royco HRLD-150	4 - 5	<1 - 2

The results reported by Hargesheimer and Lewis would suggest that the extinction sensor is not suitable as a monitoring tool for *Cryptosporidium* oocysts as the sensor sizes the organisms at times lower than the sizing limit of the sensor, i.e. <1 μm. Our results, with the same sensor, were similar to those of Hargesheimer and Lewis, however the lowest mode obtained was 1.53 μm. Given a calibration on a standard 1μm polystyrene sphere can be obtained where the millivolt signal is clearly distinguishable from the background noise, then sizing oocysts at this level should not present an obstacle to continuous monitoring with this sensor.

The Hiac-Royco was investigated for coincidence error by homogenising a concentrated sample of oocysts. This sample was estimated as having approximately 10^7 oocysts by optical microscopy. This sample was further diluted to obtain oocyst-containing solutions ranging from approximately 100 to 1,000,000 oocysts/mL. A coincidence error of approximately 10% was noted at approximately 12,000 oocysts/mL. Hiac-Royco estimated the coincidence limit (the particle concentration above which coincidence error in unacceptable) of this sensor as 18,000 particles/mL however this was determined with 2μm spheres.

The Hiac-Royco instrument was subsequently used to investigate the flocculation

process. In this work it was found to be impossible to obtain readings that reflected the growth of ferric hydroxide flocs. In other words the instrument registered 0 counts above about 20μm for the 30-minute period that the coagulation and flocculation process was investigated. It is believed that the ferric flocs are transparent with respect to the HRLD-150 (light extinction) sensor. Initial investigations with the SEL-05 (light scatter and extinction) sensor have yielded similar results. Additionally, considerable brown staining of the aperture windows of the sensor cell was noted which would lead to a decrease in the background signal, which in turn would lead to a loss of sensitivity to particle sensing. As such no further investigations of the flocculation process were conducted using the Hiac-Royco instrument.

2.4 The Malvern Mastersizer

The Malvern Mastersizer uses small angle light scattering to determine the size of particles. Scattering is the phenomena of light bending at the boundary of two materials with different refractive indices. A sphere of a certain diameter will scatter light in a known way. The resulting scatter pattern is modeled to fit a theoretical equation that assumes the scattering particles are spherical and the size distribution calculated. The accuracy of size determination of spherical particles above 1μm is very good, as the resulting scatter adheres to the simple Fraunhoffer model very well.

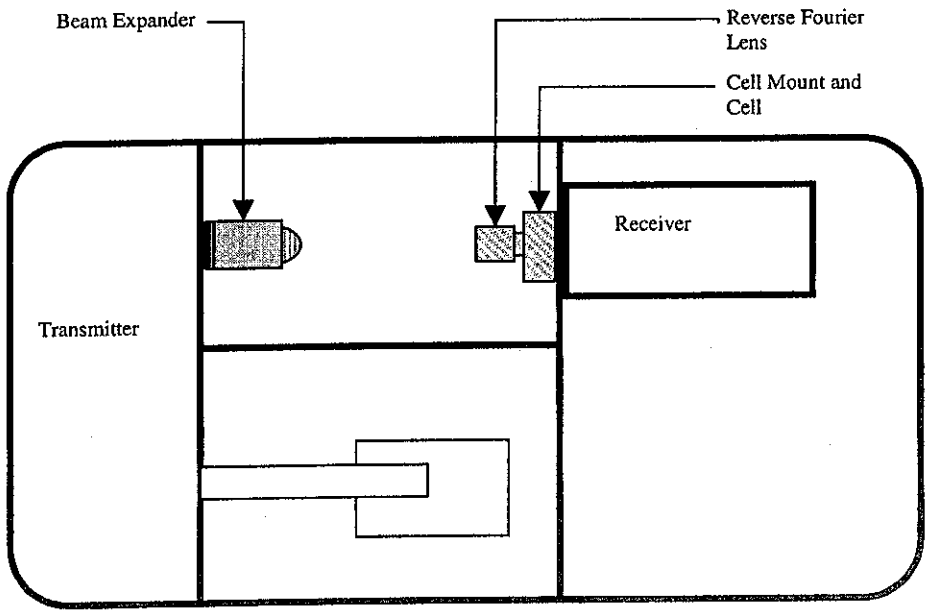


Figure 2.4. Schematic of the Malvern instrument

Non-spherical particles are not catered for by any of the well-known models and work on the interpretation of the scattering patterns of particle aggregates is being conducted by various researchers throughout the world (Amal and Bushell, 1997). An aggregate is a particle composed of smaller sub units. The theoretical model being developed assumes aggregates are composed of uniform spherical primary particles. These aggregates scatter light depending on its primary particle size, total size, shape and compactness of structure.

When any two of the above parameters are classified as small, the Fraunhofer model is inadequate and the refractive index and adsorption parameters must be incorporated into a more appropriate model. If these optical properties can not be determined, then the Fraunhofer model may be used to produce, at best, relative size distributions.

The fundamental instrument measure with the Malvern is one of size based on particle volume. Size information based on particle number can be obtained but an assumption must be made that all particles are spheres. Given the possibility that particles differ substantially from spheres, particular caution must be exercised in drawing conclusions based on number distributions.

As reported earlier (Table 1), the Malvern instrument was used to size both of the protozoan organisms. The results suggest that the Malvern is capable of sizing the organisms yielding dimensions in good agreement to those obtained by the flow cytometry technique. However, the instrument did require a certain number of organisms or particles in the sample cell to produce an interpretable signal. This number was in the order of hundreds of thousands and as such the instrument could not be seen as a monitoring tool for finished water quality. However, the applicability of the instrument to investigation of the coagulation-flocculation process was assessed and, due in part to the greater concentration of solids present in this process, reliable and reproducible results were obtained that will be discussed further in the following section.

2.5 Galai CS-100

Galai markets an 'all in one unit' for particle characterisation. This unit combines a particle sizer, image analyser and zeta potential module.

2.5.1 The Galai particle sizer

The Galai particle sizer technology is based on the extinction principle but uses a time of transition sizing process, somewhat different to that of the Hiac-Royco, that incorporates a laser beam moving rapidly through a circular path in the sample. Because the particles are not moving significantly compared to the expeditious motion of the laser, the technique can be seen as quite different to the obscuration sensor of the Hiac-Royco. Another advantage of the system is that a complex theoretical model is not required to interpret the raw data, unlike the Malvern where an assumption of sphericity in the Fraunhoffer or Mie scattering models is necessary.

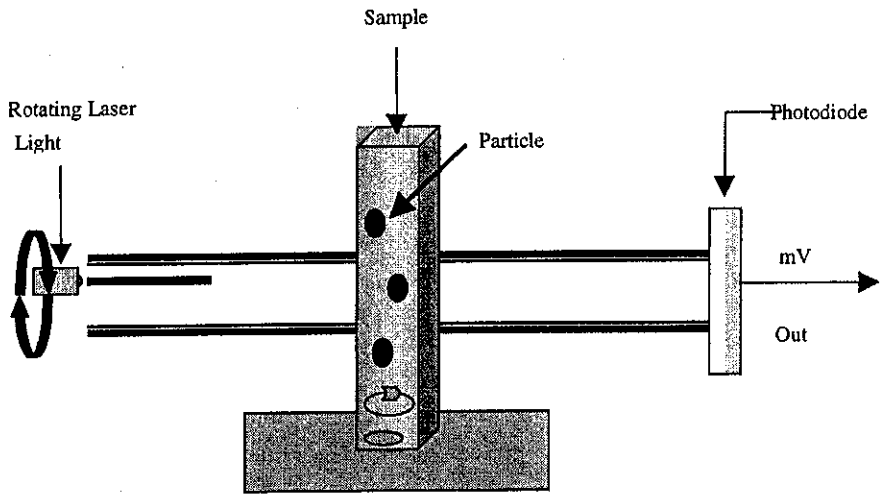


Figure 2.5. Schematic of the Galai particle sizer

When solutions containing the pathogens were introduced into the sample cell the following results were obtained (see Table 1). *Cryptosporidium parvum* had a modal maximum at 4.8μm, *Giardia muris* had a modal maximum at 8.2μm. The comparison with other instruments and techniques showed that the Galai produced results that compared very favourably with those obtained by the flow cytometric technique.

The Galai was then tested as a means of investigating the flocculation process. Due to the translucent nature of flocs, the laser beam is only partially obscured and the

detector was found to be not sensitive enough to define the "true" edge of the floc, thereby under-estimating size. In addition the size analysis using the Galai was slow (up to 2 hours/analysis) and can only be performed at high concentrations of flocs or particles (estimated to require > 10,000 particles/mL). At these concentrations the laser beam is probably being obscured by more than one floc, making results more a product of concentration than size. These problems may be resolved when more sensitive detectors become available.

2.5.2 Galai image analysis

Image analysis involves the recording of real time images and their subsequent computer analyses to determine size and shape parameters. The Galai uses a microscopic camera and numerous different lenses to acquire pictures of particles in a cell on a standard television screen. Because the camera can only focus in a thin plane and the cell is 1 cm^2 there was always a significant number of unfocused images on the screen. The computer analyses the image on the screen after applying graphic alterations to the image such as hole filling, unfocused particle elimination and background light correction. Because of the amorphous nature of ferric flocs they have sections which absorb light and some sections which focus light. As a result it was very difficult to distinguish flocs from unfocused objects and the background with the Galai and so quantitative analysis was not possible.

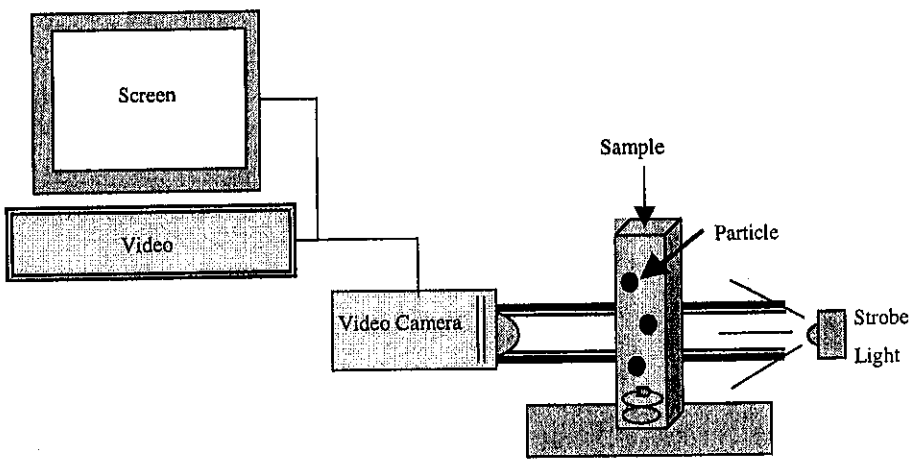


Figure 2.6. Schematic of the Galai image analyser

Despite the difficulties encountered with the Galai, image analysis is an attractive option for floc size measurement since analysis does not involve fitting theoretical (often inappropriate) models to raw data. The image of a floc incorporating 5μm latex spheres, shown in Figures 2.7a & 2.7b demonstrates the quality possible with image capture. The ability to interpret such images to yield quantifiable data was beyond the current capabilities of the Galai. These images are examples of static pictures taken with a phase contrast microscope (courtesy Hans Coster, UNESCO Centre for Membrane Science and Technology, University of NSW). The flocs are placed on a slide, thereby eliminating the possibility of unfocused objects in different planes. The beads are clearly visible and the floc is defined from the background.

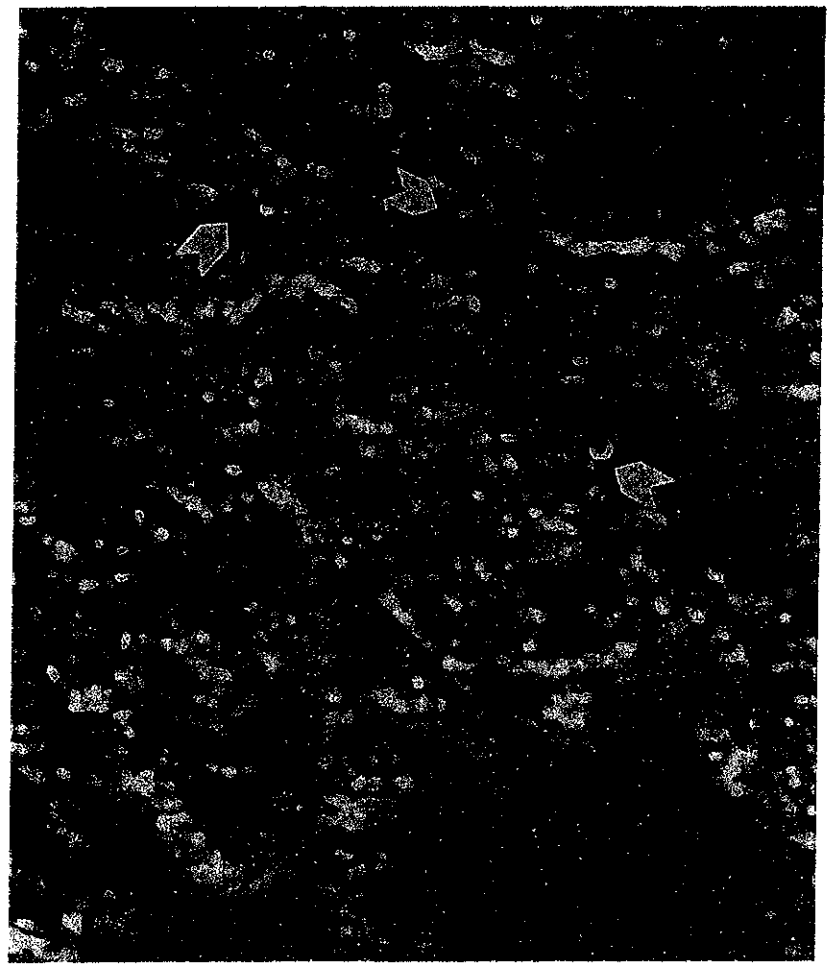


Figure 2.7a. Phase contrast photo of 5 μm latex particles trapped in a hydrous ferric oxide floc.

Figure 2.7b. Phase contrast photo of 5 μm latex particles trapped in a hydrous ferric oxide floc.

2.5.3 The Galai zeta potential module

The zeta potential is the electric potential between the radius of shear of a particle and the bulk solution and reveals information about the surface charge of a particle. It is commonly derived by measuring the electrophoretic mobility of the particle in question in a constant ionic strength solution (often 0.01M or 0.1M sodium chloride)

where the electrophoretic mobility is defined as the velocity of a particle relative to the bulk solution divided by the electric field required to obtain that velocity. Instruments designed to estimate zeta potential via measurement of electrophoretic mobility are available from several companies (including Rank Brothers, Coulter and Malvern). These instruments typically consist of a sample cell that incorporates two electrodes that create an electric field across the cell. A stationary layer of fluid must be found so the true relative velocity can be calculated. Small convection currents that form within the cell, due to localised heating of the sample fluid, will disturb the stationary layer but may by eliminated by good temperature control.

The Galai Zeta Potential Module uses image analysis to derive a value for zeta potential. When the electric field is applied, the particles in the electrolyte solution move, and 'snap shots' of the particles are taken at predetermined time intervals. The direction of the field is reversed occasionally to prevent polarisation of the solution. The total length of the image is analysed and a particle velocity calculated which is subsequently converted into zeta potential. The main problem with the apparatus is the poorly designed optical cell that breaks very easily. The cell also has no temperature control mechanism (only a small static water bath) rendering the stationary layer too unstable to gain meaningful results.

Galai recently supplied a new design for their zeta potential cell, however time did not allow for further investigation.

2.6 Summary

2.6.1 Sizing

The Hiac-Royco instrument, with the extinction sensor engaged, sized oocysts and cysts at much smaller sizes than the flow cytometric technique. The scatter sensor sized the organisms even lower than this, suggesting that the scatter sensor is more sensitive to semi-translucent particles such as natural organisms. As such, calibration of such sensors using size standards that incorporate the size of the organisms of

interest is critical. Our results suggest that the recommendation of a size range to be monitored for water quality assessment, such as 2-6μm for *Cryptosporidium* is flawed. Clearly, the size range for monitoring needs to be established for each individual situation.

In contrast the Malvern Mastersizer and the Galai instrument both produced sizing results for oocysts and cysts that compared more favourably with the flow cytometry method as reported by Hargesheimer and Lewis.

2.6.2 Sensitivity

In terms of sensitivity the Hiac-Royco could provide reproducible results at organism levels of ≥ 10 organisms/mL. In contrast, the Malvern Mastersizer did not produce a signal until the levels of oocysts were in the order of 100,000 oocysts/mL. Similarly, the Galai instrument required excessive accumulation times to produce results on particle numbers of 1000 /mL and higher. As such the Hiac-Royco system, preferably with the extinction sensor engaged, would be the only instrument capable of investigating a filtration process or a water quality monitoring application. It should be noted that neither the Malvern nor the Galai are marketed as particle counting instruments for the drinking water field.

Although particle analysis has very much focused on the application of particle counting for filtration performance and water quality monitoring to date, instruments such as the Malvern are capable of providing valuable, quantitative information on the coagulation and flocculation processes. An example of such an application is given in Chapter 6 on jar testing investigations.

3. Pathogen Sizing by Light Obscuration.

3.1 Introduction

The previous chapter demonstrated the large variation in particle sizes obtained for the same particles (in this case, *Giardia* cysts and *Cryptosporidium* oocysts) between different particle analysis instruments. As well as variation between instruments, the Hiac-Royco particle counting instrument was shown to exhibit a large variation in the measured size of oocysts and cysts.

This difference is most likely attributable to the natural variation in size of the pathogen populations. However, as particle size measurement depends upon the refractive index of the particle surfaces, it is possible that variations in the nature and concentration of surface active agents could also influence the observed particle size and possibly the reproducibility of the particle count.

During this phase of experiments, the solution matrix parameters of pH, alkalinity and humic acid concentration were varied over a range that could be expected in different Australian freshwaters and the influence on apparent particle size of *Cryptosporidium parvum* oocysts investigated. Following this, a set of experiments was designed to investigate the influence of these parameters on particle counts in a set size range.

3.2 Sizing

3.2.1 Methodology

Oocsyts were obtained as previously described. Reagent grade water was obtained by passing Sydney tap water through a Millipore reverse osmosis unit set at 97% flow rejection. This water was subsequently passed through a Millipore Milli-Q system comprised of activated carbon, anion exchange, cation exchange and 0.45μm filtration.

Sufficient oocysts were added to two litres of reagent grade water to produce a solution that exhibited counts of approximately 1000 oocysts/mL. The pH, alkalinity

and humic content of this solution was then adjusted as described below and the modified solution divided into 8 x 250mL samples on which sizing experiments were conducted.

Solutions at varying pH were obtained by addition of 0.1M NaOH or HCl to reagent grade water. Solutions of varying alkalinity were obtained by addition of reagent grade calcium bicarbonate. These solutions were filtered through a 0.2 μm membrane to remove any particulate matter and alkalinity was subsequently confirmed by titration. Solutions of varying humic acid content were obtained by addition of a 0.2 μm filtered solution of approximately 1000mg/L of Aldrich humic acid. True colour was measured using a Hach DR2000 spectrometer. The pH of the alkalinity and humic acid experiments was adjusted with NaOH or HCl to a pH value of 7.0 ± 0.2 before particle counting.

The Hiac-Royco instrument was used with the HRLD-150 sensor for all experiments with the flow controlled by the CFC-800 flow controller as previously described. The extinction sensor was used in these experiments as the scatter sensor not only sized oocysts below the 1.2 μm lower size cutoff but also was expected to be more susceptible to variations in particle refractive index. The sensor was factory calibrated between 1.2 and 12.0 μm with Duke Scientific polystyrene spheres.

3.2.2 **Discussion of Results**

3.2.2.1 Effect of pH variation

Preliminary studies of the effect of pH on measurement of size were undertaken using 10μm polydivinylbenzene spheres from Coulter. The results are shown in Table 3.1 and indicate only a small variation with solution pH, indicating not only a lack of refractive index change but also little or no aggregation of the spheres at the pH values studied.

Also of interest is the difference between the mean and mode of the sizing. Generally the coincidence of these values is quite good (typically within 10%) reflecting a

reasonable Gaussian distribution. These distributions exhibit reasonable width as reflected in relative standard deviations in the vicinity of 20%.

Table 3.1 Variation of size of 10μm PVDB standard spheres with pH (number of replicates in each case = 8)

pH	Mean (μm)	Mode (μm)	Std Dev. (μm)	RSD (%)
5.1	8.29	8.19	1.71	20.7
6.1	8.68	9.31	2.13	24.5
7.1	7.68	8.18	2.02	26.2
8.3	7.80	7.58	1.43	18.4
9.1	7.89	7.58	1.52	19.3

Following this experiment on "model" particles, the procedure was repeated with fresh oocysts. Mean and mode size measurements as a function of pH are given in Table 3.2

Table 3.2 Variation of size of fresh oocysts with pH (number of replicates in each case = 8).

pH	Mean (μm)	Mode (μm)	Std Dev. (μm)	RSD (%)
5.1	2.82	1.85	2.64	93.5
6.2	2.52	1.64	2.27	88.9
7.1	2.87	1.53	2.33	81.4
8.3	3.02	2.00	2.34	77.5

The results show that the oocysts are sized well below their suspected physical size (as determined by optical microscopy or flow cytometry). Also, the mean and mode are well separated with the mode being consistently and significantly of a lower size than the mean. This indicates a non-Gaussian distribution skewed toward higher sizes. These results suggest that, given the significant "skew" in size distribution, the mode value may be a better descriptor of size than the mean value, unless log normalisation is routinely undertaken. No consistent variation in size with pH is evident from these results.

The important outcome of this experiment is that the size range of 2 – 6 μm that has been widely touted as a surrogate measure for oocysts is inappropriate as the organisms may be sized well below this when instruments such as the Hiac-Royco are used. It is therefore critical that verification of the sizing of oocycts by the instrument sensor be part of any quality assurance procedure for a monitoring program that relies on particle counting as a surrogate indicator.

3.2.2.2 Effect of alkalinity variation

Results showing the effects of alkalinity on measured size of fresh oocysts are shown in Table 3.3. A variation in alkalinity could be expected to change the apparent size of the oocysts. The likely mechanism for this would be adsorption of calcium to the surface of the oocyst which could result in either a change in refractive index or the charge destabilisation allowing for oocyst aggregation.

Table 3.3 Variation of size of fresh oocysts with alkalinity (number of replicates in each case = 8)

Alkalinity (mg/L $CaCO_3$)	Mean (μm)	Mode (μm)	Std Dev. (μm)	RSD (%)
0.0	4.94	4.95	3.72	75.4
10.0	3.54	4.61	2.55	71.9
20.0	3.55	4.56	2.65	74.5
30.0	3.54	4.34	2.57	72.5
40.0	3.43	4.27	2.45	71.4
50.0	3.22	3.88	2.30	71.3

In this experiment the results are somewhat different to those reported above (for pH variation) in that the mean and mode for the fresh oocysts are very close together though the peak is still very broad. This result would appear to be the exception rather than the rule with all other results showing a skew of the peak toward higher sizes.

Again any statement about a significant influence of alkalinity on measured size

would be qualitative but it would appear that the influence is to consistently move the mode toward a lower size with increasing alkalinity. This result may well be due to a change in refractive index of the oocycts surface as free calcium ions are adsorbed to the surface as an aggregation of particles would tend to push the mode to a higher value. Again the result emphasises the importance of sensor size verification with oocysts rather than relying on manufacturer's calibration with latex spheres.

3.2.2.3 Effect of humic acid concentration

In contrast to the alkalinity experiments the addition of humic acid would appear to have little effect on the mode of the oocyst size however there appears to be a large shift in the mean toward higher size at the highest humic acid concentration. The result could be explained by some aggregation of the particles in the presence of higher humic acid concentrations with the organic material acting as a flocculating agent.

Table 3.4 Variation of size of fresh oocysts with humic acid (number of replicates in each case = 8).

Humic Acid (mg/L)	True Colour (Pt-Co Units)	Mean (μm)	Mode (μm)	Std Dev. (μm)	RSD (%)
0.0	0	3.33	1.84	2.40	72.0
5.6	72	3.32	1.82	2.43	73.1
11.2	148	3.31	1.84	2.38	71.7
23.8	300	4.21	1.84	7.16	170

The addition of humic acid, as expected, results in an elevation of the matrix colour. The colour and extent of absorbance observed with increasing humic levels is a function of the chomophoric make-up of the organics involved. As a result it is not possible to draw a correlation between work conducted with humic acid and the influence of "colour".

3.3 Counts

3.3.1 Methodology

Following the sizing experiments, the influence of alkalinity and humic acid concentration on particle counts within a set size range was investigated. In this set of experiments a stock solution of 5μm latex spheres from Duke Scientific was homogenised and a small aliquot of this suspension was taken and suspended in 2 litres of tap water for which the particle counts and alkalinity had been previously established. Tap water was used in these experiments to provide a more realistic counting matrix than reagent grade water. The suspension was prepared to have approximately 10,000 counts, toward the higher end of the coincidence level, in the 1 to 5.81μm size bin. This bin or range included 95.5% of the mean size ± 2σ. The mean and mode of the size agreed well and the following statistical analysis assumes a Gaussian distribution.

To these solutions was added various amounts of 0.2μm filtered, reagent grade sodium carbonate and humic acid (Aldrich) to give added alkalinity of 0 to 100 mg/L as $CaCO_3$, (as determined by titration) and 0 to 5 mg/L of humic acid. These suspensions were further divided into 8 x 250mL samples that were analysed for particle counts and the results represented a pooled data set. The sample with no added alkalinity or acid was taken as the reference set. Significance was set at 95% with the null hypothesis being no significant variance; i.e. a rejection of this hypothesis represents a significant variance.

The particle count for the solutions was determined on the Hiac-Royco instrument using the HRLD-150 extinction sensor. The instrument was used in the batch mode and each sample was measured until two replicate analyses were within 1% (usually the second and third runs).

3.3.2 Discussion of results

The results of the alkalinity experiments are presented in Table 3.5 and the results of

the humic acid experiments are presented in Table 3.6.

Table 3.5 Significance of effect of a variation in alkalinity on particle counts.

Added alkalinity (mg/L Ca CO_3)	Mean Counts/mL	Significant Variance (95%)
0	10,971	No
20	10,636	Yes
40	10,335	Yes
60	9,999	Yes
80	9,652	Yes
100	9,489	Yes

Table 3.6 Significance of effect of a variation in humic acid on particle counts.

Added humic acid (mg/L)	Mean Counts/mL	Significant Variance (95%)
0	9,889	No
0.1	9,801	No
0.5	9,619	Yes
2.0	9,606	Yes
5.0	9,531	Yes
10.0	9,388	Yes

The results shown above indicate that a significant variance in the number of counts in a particle size bin occurs when solution parameters such as alkalinity and humic acid concentration are varied. It is difficult to explain the mechanism behind the variance observed in the particle counts as the size range is at the lower end of the sizing capability of the sensor. This makes it impossible to establish whether the matrix change is moving the observed size of the spheres to a slightly lower size range. Another possibility is that the matrix change is broadening the count spread

(increasing the standard deviation of an individual count) and as the size cut-off is at the lower end of the sizing capability the sensor the range can not be broadened to incorporate the broader count.

The result is important not only in terms of establishing a size range to set for water quality monitoring but for verification of particle counts as well as size. Certainly one of the current interests in the US is to establish standards for size and counts. As such it is reasonable to assume that particle counting instruments will be supplied with not only size calibration data but also particle count data. However, even when an operator is supplied with this information it is imperative that verification of oocyst and cyst size and counts are conducted using a representative water matrix rather than purified water.

4. Laboratory Jar Testing Investigations

Much of the previous work has been undertaken utilising the Hiac-Royco instrument which is a typical particle sizing and counting instrument, used almost exclusively in the water industry.

Despite the obvious applicability of such an instrument for water quality monitoring, the information provided is an overall indication of the efficacy of a full treatment process rather than a tool for optimising individual process components.

In this section the more dynamic particle analysis technique offered by the Malvern instrument is investigated for optimisation of the coagulation-flocculation process.

4.1 Introduction

The effectiveness of particle removal from raw water may be considered to be influenced by three interrelated effects:

1. The efficiency, in terms of particle entrapment, of coagulation-flocculation.

2. The efficiency of the filtration media to retain the floc.

3. The integrity of the floc structure, in terms of particle detainment, during the filtration cycle.

The first of these issues has been extensively evaluated using standard jar testing equipment, the purpose of which is to establish the optimal pH and coagulant/flocculent doses to achieve satisfactory water quality in the most cost-effective manner.

The means for investigating the efficiency of the coagulation-flocculation process have traditionally relied on the measurement of colour and turbidity, whereby the impact of the jar testing procedure is a reflection of the process efficiency. These two traditional parameters are probably still the most frequently measured in drinking

water however, newer measurements such as TOC and particle counts provide a means of obtaining more detailed information on the process.

One of the reported benefits of particle counting is that a certain size range of interest can be targeted for removal such as the 2 - 5μm size range which has been correlated with oocysts (Jasim et al., 1997). To use jar-testing equipment for the evaluation of a sedimentation process, the pH and coagulant/flocculent doses are varied and the floc is allowed to settle and the supernatant is extracted and tested for particle counts. However, to investigate a direct or contact filtration process an approximation of the filtration process is required. Clearly the choice of filtration simulation such as filtration through a filter paper or membrane is a very crude approximation. However, in this section the Malvern instrument is investigated for its ability to measure particle entrapment within flocs *in-situ*. The influence of treatment parameters, such as coagulant and flocculent doses and presence of organic matter, on the ability of flocs to entrap 5μm latex spheres has been investigated.

4.2 Methodology

The Malvern Mastersizer E that has been previously described was used in this work for particle size and relative number determination. Particle and floc sizes in the 1.2 - 600μm were measured and latex sphere entrapment was calculated by monitoring the disappearance of the modal count corresponding to the concentration of the spheres and is expressed in a % reduction of the signal. The Hiac-Royco with the HRLD-150 was also used to count the number of added latex particles in the prepared sample solutions allowing a more quantitative evaluation of data.

The raw water used in this work was taken from the weir at Broughton's Pass, south of Campbelltown. This water is the source water for the Macarthur Water Treatment Plant that supplies the Campbelltown area of Sydney. This water was filtered through a 0.45μm membrane filter to remove the natural particulates and then sufficient latex spheres were added to achieve a count of approximately 7,500 particles/mL. This number was chosen as it produced a strong signal from the Malvern detector resulting

in short accumulation times for data collection. This allowed more measurements to be taken during the flocculation process.

All treatment chemicals and dosages were similar to that used at the Macarthur Water Treatment Plant. The ferric chloride used in this work was supplied by ICI and was obtained as liquid from the storage tanks at the Macarthur Water Treatment Plant. This bulk liquid was estimated to contain approximately 42% $FeCl_3$ w/w and was diluted to achieve solutions of 1000 mg/L $FeCl_3$. The polymers LT-35 (polydadmac) and LT-20 (polyacrylamide) were obtained in 25g solid samples from Allied Colloids and 1% w/v solutions were prepared fresh as required.

The coagulation-flocculation process was simulated using a Phipps & Bird 6-paddle jar tester with 1L beakers. The treated sample was withdrawn from the beaker at one minute intervals into the Malvern analysis cell by a peristaltic pump situated after the Malvern cell to minimise floc disturbance. The pH was maintained at 7.0 ± 0.2, reflecting a pH commonly used in water treatment, by the addition of either 0.1M HCl or NaOH and the mixing regime employed was an initial rapid mix (G=300 s^{-1}) for one minute followed by mixing for nine minutes at G=25 s^{-1}.

4.3 Discussion of Results

4.3.1 Effect of Ferric Chloride

Ferric chloride was dosed at 10, 12 and 14 mg/L as 100% $FeCl_3$ and the relationship between dose, floc size and particle entrapment is shown in Figures 4.1 and 4.2.

The results show a strong floc growth dependence on ferric chloride dose with a higher dose resulting in more rapid floc growth and larger eventual flocs. The results also show a strong relationship between floc size, which is dependent on both time and coagulant dose and particle entrapment, with large flocs proving to be better particle scavengers. Such a result is expected from Smoluchowski particle-particle interaction kinetics theory (O'Melia and Tiller, 1993) and fits well with the long-held belief of the benefits of a taper-style flocculation process such as that used in these

experiments. The results also suggest that no significant particle entrapment is achieved until the floc size reaches approximately 100μm. As mentioned the floc size is dependent on both time and coagulant dose. Such results have important implications for a direct filtration process and more particularly a contact filtration process where a limited time for particle entrapment is provided before filtration.

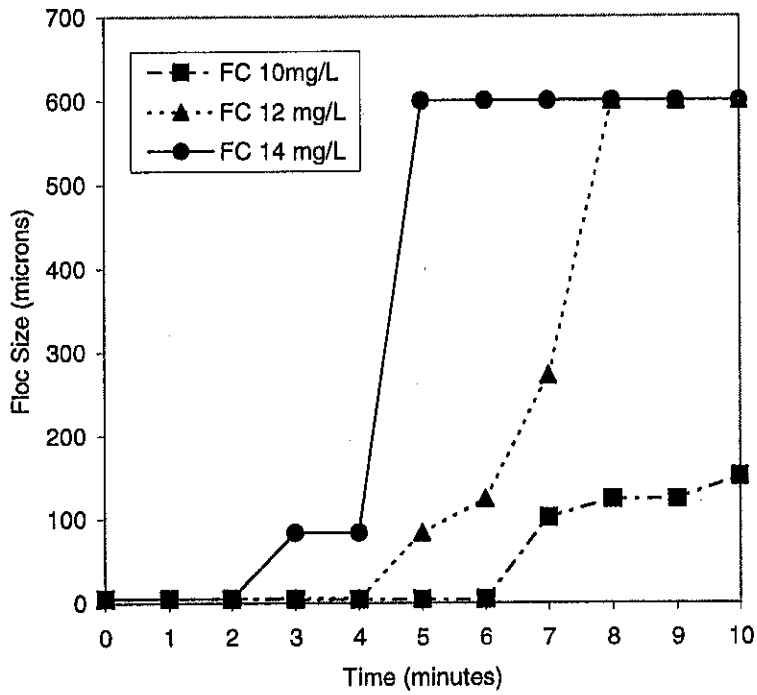


Figure 4.1. Comparison of floc size and growth with time at different ferric chloride (FC) doses.

As mentioned above, that large flocs provide better entrapment of individual particles is not surprising given the increased collision frequency expected. An area that is less understood is the implications of the fractal nature of the coagulating flocs (typically iron or aluminium oxyhydroxides) to particle entrapment. Recent work by Li and Logan (1997a&b) suggests that entrapment of individual particles by porous flocs exhibiting fractal characteristics may be significantly less efficient than previously thought. These workers generated latex aggregates in the 200 – 1000 μm size range of various fractal dimensions and examined entrapment of individual, small (1.48 μm)

particles and showed that particle capture efficiencies decreased significantly as the fractal dimensions of the aggregates decreased. Considerable additional work in the area of particle entrapment by fractal aggregates is urgently required, particularly extending the work from latex aggregates to those typically used in water treatment (iron and aluminium oxides) and further examining the effects of surface charge, pH and organic adsorbents.

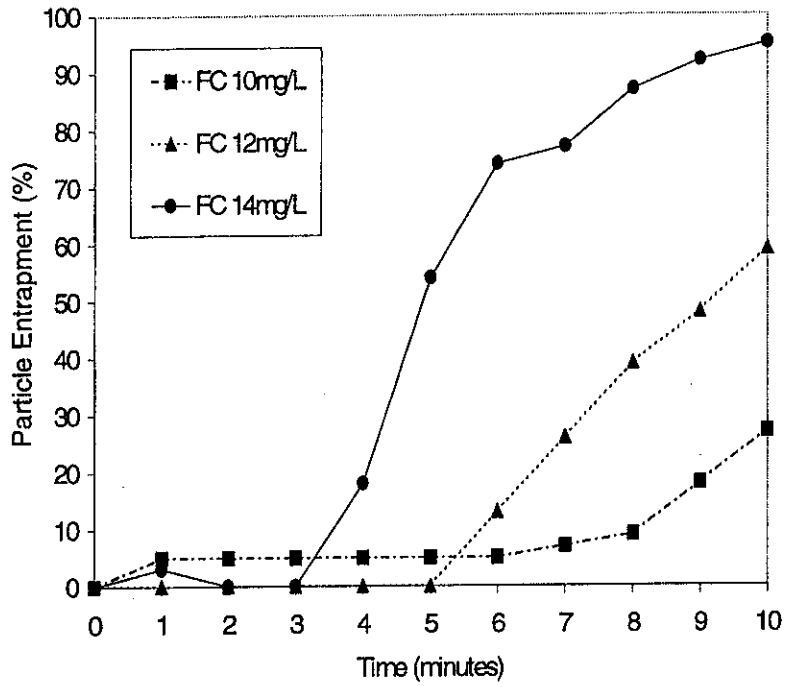


Figure 4.2. Comparison of particle entrapment by ferric chloride (FC) floc as a function of time at different ferric chloride doses.

4.3.2 Effect of Polymer Additives

From the results presented above a ferric chloride dose of 10 mg/L produced slow floc growth and particle entrapment when compared to higher doses. The effect of the polydadmac inorganic coagulant aid LT-35 on the 10 mg/L ferric chloride dose was investigated. In this set of experiments another 1 L sample of water containing approximately 7,500 latex particles/mL was placed on the jar-testing equipment and

ferric chloride was added to the solution at 10mg/L. The same mixing regime of 1 minute rapid mix and nine minutes of gentle mix was repeated and at 40 seconds (toward the end of the rapid mix stage) 0.4 mg/L LT-35 was added. In a further experiment the previous process was repeated with an addition of 0.01 mg/L of the polyacrylamide flocculent LT-20 at three minutes, during the gentle mixing stage.

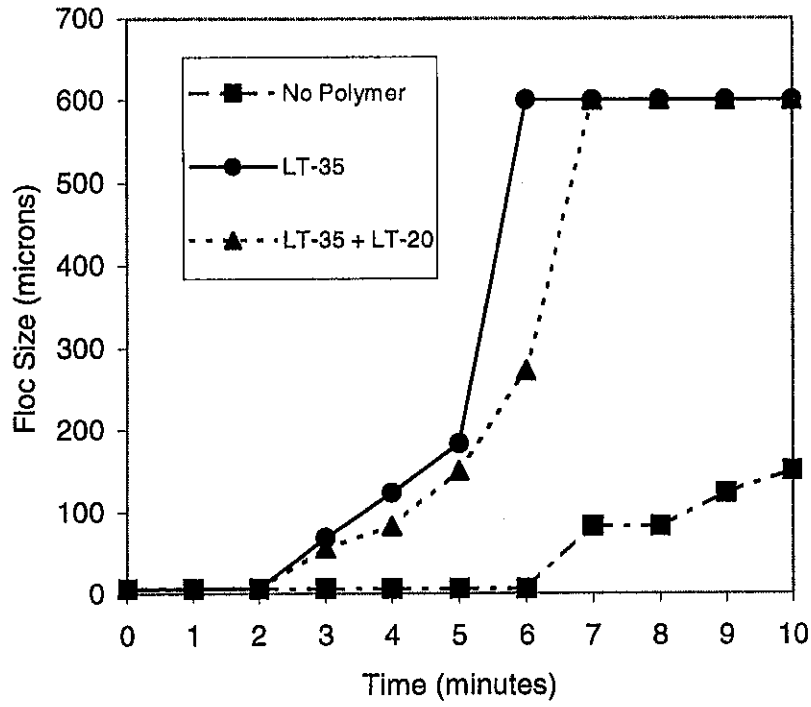


Figure 4.3. Comparison of floc size and growth with time at a ferric chloride (FC) dose of 10mg/L with addition of 0.4mg/L of polydadmac (LT-35) and 0.01mg/L polyacrylamide (LT-20).

The results of these experiments are shown in Figures 4.3 and 4.4 and show the different effects these polymers have on the ferric chloride flocculation. Figure 4.3 shows how the polydadmac dramatically increases both the size and the rate of floc growth. With ferric chloride alone no detectable flocs are produced until after six minutes after which the floc grows slowly to a maximum of about 100μm. With the addition of polydadmac the floc commences to grow rapidly at 2 minutes, 20 seconds after the polymer addition. Such a result is expected and supports the view that a

polymeric coagulant enhances flocculation by forming polymer bridges between destabilised particles. (As mentioned previously, this may be particle destabilisation by charge neutralisation/double layer compression rather than bridging effects)

An interesting result is that the rate and extent of particle capture as shown in Figure 4.4 is somewhat less than for the 14 mg/L ferric chloride dose experiment described in the previous section. For the 14 mg/L ferric chloride dose the floc size reached ≥ 600 μm after 5 minutes. For the 10 mg/L ferric chloride with 0.4 mg/L polydadmac the floc size reached ≥ 600 μm after 6 minutes, whereas the ferric chloride floc achieved 95% particle entrapment after 10 minutes the mixed inorganic and organic floc achieved 84% particle entrapment. It is likely that this result is directly related to the slightly slower floc growth rate noted.

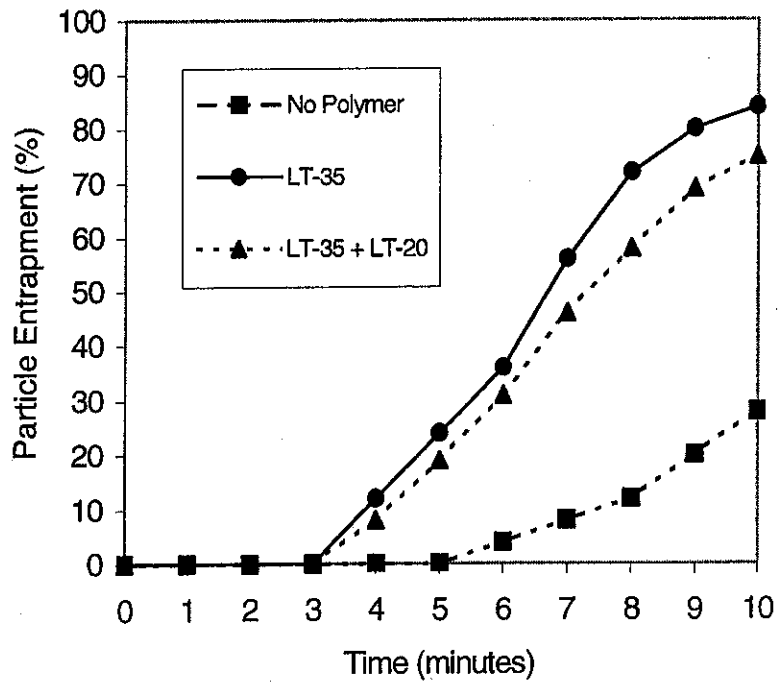


Figure 4.4. Comparison of particle entrapment with time at a ferric chloride (FC) dose of 10mg/L with addition of 0.4mg/L of polydadmac (LT-35) and 0.01mg/L polyacrylamide (LT-20).

The affect of polyacrylamide at a dose of 0.01mg/L appears to be only slight in that there appears to be a small retardation affect of both floc growth and particle entrapment.

4.3.3 Effect of Humic Acid Addition

Humic materials in water often present problems for water treatment processes. For a treatment process involving iron as the major coagulant the humic material may bind to the introduced ferric iron and form soluble iron-organic complexes. Such complexes inhibit the flocculation process as the bound iron can no longer hydrolyse to form the insoluble ferric hydroxide flocs. If the humic material is present in sufficient quantities and insufficient iron is added to exceed the organic demand then it is possible that flocculation will be severely affected.

In this set of experiments a humic acid (Aldrich) solution of 1,000mg/L was prepared by dissolving the appropriate amount of humic acid in reagent grade water. This solution was then filtered through a 0.2μm membrane filter to remove particulate material. Different amounts of this solution were added to the previously prepared raw water solutions containing approximately 7,500 latex spheres/mL. This solution was then treated with 12mg/L $FeCl_3$ and the floc growth and particle entrapment was monitored as described previously. The results of these experiments are presented in Figures 4.5 and 4.6.

The results shown in Figure 4.5 confirm the coagulant demand theory discussed in that a higher humic acid content affected the ability of the iron coagulant to form floc particles. An interesting result was observed for the 0.1 mg/L humic acid experiment where the presence of a small amount of humic material appeared to assist the floc formation process, expressed as a slightly higher growth rate. Such a result has been reported previously (Gray, 1988, Edzwald, 1993) and explained as the humic material, in small quantities, acting in a similar way to inorganic polymers and providing bridges between particles.

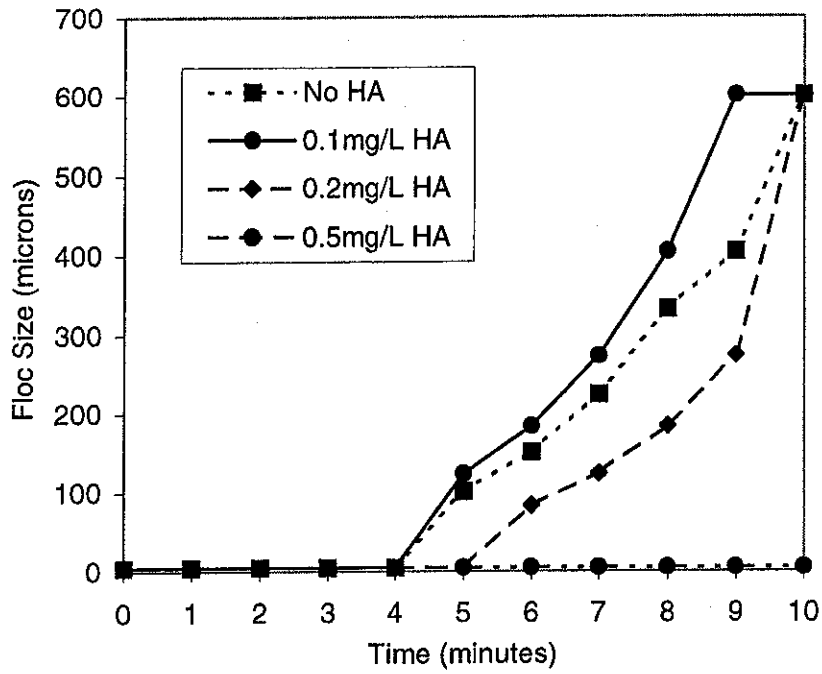


Figure 4.5 Comparison of floc size and growth with time at a ferric chloride (FC) dose of 12 mg/L with addition of up to 0.1 - 0.5mg/L of humic acid.

When the humic acid content is doubled to 0.2 mg/L the opposite affect is observed such that a decrease in floc formation rate is observed. When the humic acid content is raised to 0.5 mg/L a ferric chloride dose of 12 mg/L is not sufficient to exceed the organic coagulant demand and little or no flocculation is observed.

The effect of humic acid concentration on floc growth is reflected by the ability of the flocs to entrap particles. Figure 4.6 shows the result of varying humic acid concentration on the ability of the floc to entrap the latex particles. Once again the floc size appears to be the principal governing factor in efficiency of particle capture. At a humic acid concentration of 0.5 mg/L there is little particle entrapment due to little or no floc formation.

The work by Edzwald (1993) resulted in a hypothesis that natural organic matter and not the turbidity causing particles may in fact control the coagulant dose. This work suggested that a concentration of ≤ 3 mg/L can aid coagulation whereas higher doses

can lead to charge reversal and restabilisation of colloidal particulates (Gray, 1988). For the concentrations of humic material used here the effects of coagulation enhancement by low doses of organic and disruption of coagulation at higher doses were observed for very much lower concentrations of the natural organic material. It is clear that the extent of influence of NOM on the coagulation is very dependent on the nature of the organic and particulates in the water to be treated. If the organic matter has a high complexation potential it could be expected to exert a higher coagulant demand or particle destabilisation. The charge on the particle would also be an important factor, as a more negative particle charge would require more coagulant for destabilisation.

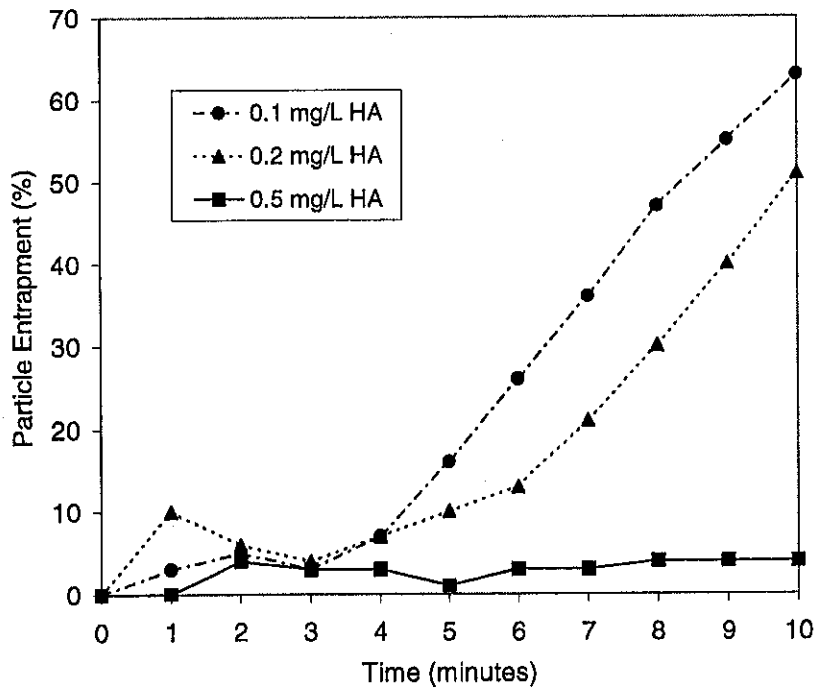


Figure 4.6 Comparison of particle entrapment with time at a ferric chloride (FC) dose of 12 mg/L with addition of 0.1 - 0.5 mg/L of humic acid.

4.3.4 Summary

Although this work has probably served to produce more questions than answers due to the broad nature of the work, the major objective has been achieved in that the

usefulness of the Malvern instrument for assessing the coagulation-flocculation process in drinking water treatment has been demonstrated. The preliminary results from this work have suggested some areas worthy of further investigation. For example the importance of coagulant dose to the rate of floc growth, the subsequent relationship to floc size and the ability of the floc to entrap 5μm latex spheres are certainly areas worthy of further investigation. Such an investigation is of particular importance to a direct filtration process, as there could be a conflict between the desire to produce "pin" flocs to prevent the top layers of the filters from clogging and the desire to enmesh the oocysts in the floc particles before the filtration stage.

The complementary relationship between inorganic coagulant ferric chloride and the organic coagulant LT-35 has been shown to produce a slightly slower rate of floc growth and associated particle entrapment. The presence of the organic flocculent LT-20 at 0.1mg/L was shown to have only a slight affect on floc growth and particle entrapment.

Humic acid was found to have a significant detrimental affect on the coagulation-flocculation process. A concentration of 0.5mg/L humic acid was found to severely inhibit the flocculation process sufficiently to virtually prevent floc growth and subsequent particle entrapment. Such a result has an implication to the observed practical difficulties in obtaining effective coagulation and flocculation of waters with high colour and low turbidity. The significance of the affect of NOM on ferric coagulation is a subject that is currently being pursued in other studies within the Department of Water Engineering at the University of New South Wales (Stokes and Waite, personal communication).

5. Filtration Investigations and Pathogen Association with Particles

In this chapter, we investigate a number of issues of significance in understanding the removal of particles by deep bed filtration: i) the veracity of use of filter paper as an analogue for deep bed filtration is assessed, ii) the importance of depth of deep bed to particle removal is investigated, and iii) the breakthrough of particles that are separated from (or no longer enmeshed in) iron oxide flocs is discussed.

5.1 Laboratory Jar-Testing and Filtration Sizing Trials

5.1.1 A filtration approximation for direct filtration

As mentioned briefly in the previous section, to attempt to optimise a direct filtration process using jar-tests, some form of filtration approximation would be required. To assess a reasonable approximation, studies were conducted with different types of filter papers in the laboratory.

The methodology was as follows: 5μm latex spheres were added to reagent grade water to raise the number of particles to around 7,500 counts/mL; no chemicals were added and the solutions were directly filtered through the filter papers. The results obtained are presented below in Table 5.1 and represent averages of five individual experiments.

Table 5.1 Relationship between various paper filters and removal of 5μm latex spheres (cf 38% removal using 23 m/h direct filtration).

Brand/Type	Rating	% Particle Removal
Whatman 42	slow	90
Whatman 2	medium	88
Whatman 40	medium	91
Whatman 1	**medium fast**	**53**
Asgard 1	medium fast	85
Whatman 41	fast	50
Whatman 54	fast	24
Whatman 541	fast	26

The results show there is significant and variable removal of particles by the different filter papers. The particle removal ability of the different filter papers corresponds well to their manufacturer's rating. However, the difference between the two No. 1 filters suggests that filter paper ratings are not interchangeable between brands.

While a filter paper is never going to simulate very well the behaviour of particles and flocs in a deep bed filter, it would be useful to know the extent of capture of oocysts without and with hydrous ferric oxide coatings or enmeshed in the flocs by the various filter papers.

For direct filtration ideally it would be preferable to use a filter paper that approximated the performance of the treatment plant's filters. During pilot studies at the Macarthur Water Filtration Plant the effectiveness of the filter, without coagulation, to remove particles in the > 2μm size range was investigated.

The filter columns used in the initial Macarthur pilot-scale plant study were perspex cylinders with an internal diameter of 125mm. The columns were packed with 25mm of river pebbles (approximately 8mm in diameter) followed by 150mm of 0.7mm sand and 1850mm of 2.0mm anthracite coal. Such a bed is similar to the Macarthur full-scale plant. The next section deals with the initial pilot scale trial in more detail.

The raw water conditions at the Macarthur site during these initial trials were 1.0 ± 0.1 NTU and true colour 5 ± 1 PtCo units. When this water was allowed to pass through the filter with treatment at a rate of 23 m/h (i.e. at a significantly higher rate than the design filtration rate for the Macarthur water filtration plant), the colour was not significantly affected and the turbidity was reduced to 0.7 ± 0.1 NTU. In terms of particles, it is difficult to correlate particle reduction with turbidity reduction as the variation in the raw water count over a period of three months (October to December, 1996) was from a low of 1,650 counts/mL to a high of 6,300 counts/mL. The variation of turbidity of 0.1 NTU would suggest that the raw water quality was consistent however, in terms of particle counts the quality varied considerably. For the purposes of comparison to paper filters, an average reduction in the particle count in the 2 - 15μm size range for six individual trials was 38 ± 20%. The variation in particle

reduction is quite large but suggests that for laboratory jar testing the traditional use of a medium-fast paper would tend to overestimate the effectiveness of a treatment process and that using a fast paper would be a more conservative approach.

5.1.2 Particle removal with granular media

To investigate the filtration process more carefully a perspex filtration column of 125mm diameter and 1,000mm height was erected in the laboratory and trials were conducted to investigate 5μm latex particle reduction with the different filter media used in the Macarthur full-scale plant. A peristaltic pump was used to introduce water containing approximately 1,000 5μm latex spheres/m L to the top of the column and the Hiac-Royco counter using the HRLD-150 sensor was used to count particles in the 2 – 15 μm size range.

In the first trials, the influence of 100mm of 2.0mm anthracite coal on 5μm particle removal was investigated. It soon became apparent that the filter media itself was a tremendous source of particles which at times yielded more than twice the number of particles in the 2 - 15μm size range than that found in the filtered water. The anthracite used for these experiments was washed three times in reagent grade water to remove the fines, but clearly the anthracite is a continuous source of particles.

This effect increased with increasing depth of anthracite coal. As expected the anthracite coal itself is not effective in removing protozoan sized particles and in fact contributes a significant number of pathogen sized particles into the filtered water. It is theorised that such media may contribute significantly to the large number of particles in the filtered water that are observed after backwash during the filter ripening stage as the backwashing process could well serve to scour the outside surface of the anthracite granules.

The next set of trials was conducted in a similar manner, however the coal was replaced with varying depths of 0.7mm sand. The reduction of latex spheres in the filtered water was then observed. After an initial flushing of the sand to remove inherent micron sized particles, three individual experiments were performed. The

average particle reduction that was observed with varying sand depth is shown in Table 5.2. The filtration was run until a constant particle removal was observed, usually within one hour. At the end of each experiment the sand was replaced, as there was no facility for backwashing the column.

Table 5.2 Reduction in 5μm particles with varying depth of 0.7mm sand.

Sand Depth mm)	Percentage Particle Reduction (%)
50	25 ± 5
100	32 ± 5
150	40 ± 8
200	55 ± 8
250	60 ± 11

Clearly the fine sand layer has the ability to retain a certain proportion of 5μm spheres with a general trend of increasing particle retention with increasing depth. The variability noted in the particle reduction could be a reflection of the sand's ability to shed micron sized particles.

5.2 Particle Association with Flocs

5.2.1 A review of Moran et al's work

In 1993 two papers were produced by workers at the University of Texas, Austin (Moran et al., 1993a&b) that challenged some of the traditional beliefs about the behaviour of particles in deep-bed filtration that have been based on the measurement of the gross parameters of turbidity and suspended solids.

This work was conducted using a heterodisperse effluent from a sedimentation basin that utilised lime precipitation. In this case, the majority of particles were expected to be calcium carbonate and produced a turbidity of between 1.6 and 3.2 NTU. This effluent was filtered without further treatment by a column of 76mm ID that

supported a media bed of 946mm depth. The media for the experiments was spherical glass beads with nominal diameters 0.78mm or 1.85mm. Such media was used to minimise differences in filter performance and to allow comparison with established deep-bed filtration theory that assumes spherical filter media.

During the experiments the filtration velocity for individual experiments varied from a minimum of 6.48 m/h to a maximum of 19.8 m/h. Filtration durations were between 24 and 50 hours. The particles exiting the beds were analysed by a Coulter Multisizer that recorded size range from 1 to 40μm and were monitored at different depths within the filter column. In practice the number of particles greater than 25μm that was observed was negligible and a reduced range from 1 to 25μm was monitored.

The first of the two papers investigated the processes of filter ripening and breakthrough with respect to filter depth, media size and filtration velocity. The basic outcome of the work on filter depth was that particle removal increased with depth, which is in line with accepted filtration theory. The work investigated the performance of the filter at different depths by taking samples and measuring particle size and count distributions. It was found that filtration efficiency decreased in the lower depths of the column, which is in contrast to the accepted uniform clean bed performance. This observed variation in filter efficiency was postulated to be due to two major factors. Firstly, the non-uniform surface chemistry for the influent suspended particles results in preferential capture of lower surface charged particles in the upper levels of the filter. This allows higher surface charged particles to follow through to the lower filter levels. The particles with the higher surface charge would be expected to have a lower collision efficiency and reduced filter capture. The second explanation was that filter ripening could have occurred in the upper filter levels during the sampling time of 15 – 20 minutes and consequently removal efficiency would be greater in the ripened upper levels.

The outcome of the work on media size was that "smaller media resulted in greater removals under clean bed conditions when similar filter depths were compared. Smaller media continued to produce greater removals throughout the experiments except when significant breakthrough was evident."

The outcome of the work for filtration velocity was that "lower filtration velocities resulted in greater removals when similar filter depths were compared, as expected from current theory. Less efficient removal for higher velocity, however, can be offset with greater depth."

Considerable attention is also given in this work to the difference in behaviour of different sized particles. It was noted that the dynamic process of filtration, characterised by ripening and breakthrough, is strongly dependent on particle size. More specifically, smaller particles ripen for the longest duration and exhibit the greatest increase in removal because of ripening, whereas the removal of intermediate sized particles decreased significantly as bed deposition increased. The largest particles gave the most consistent removal but as the bed deposition reached higher values removal efficiency eventually decreased.

Perhaps the most significant result, in terms of implication for water treatment practice was that there appeared to be an early breakthrough of particles in the 3 - 7μm size range which was not detected by turbidity and suspended solids measurement. Such an observation has important implications for water quality monitoring as this size range may include, depending on the sensor used, oocysts.

This observed result could explain the reported greater sensitivity of particle counting, when compared to turbidity for early detection of column breakthrough. Turbidity has a greater sensitivity to smaller particles as the associated surface area of a concentration of smaller particles results in greater perpendicular scatter that turbidity meters are sensitive to.

In the second paper on particle behaviour in deep-bed filtration Moran et al. (1993b) investigated the phenomenon of particle detachment during filtration. Prior to this paper two models existed to explain particle breakthrough after long filtration runs. The first explains breakthrough as occurring after all the attachment sites on the filtration media have been exhausted or saturated. In this model, once media saturation has occurred, breakthrough will occur due to free passage of the influent particles through the filter media. The second model involves the concept of floc

detachment which occurs when the mass of filter deposition becomes large enough to be removed by either the shear of the filtration process or interaction with incoming particles. Such a process has been likened to snow deposition on a mountain with occasional occurrences of small avalanches.

Moran et al. (1993b) used a similar experimental setup and methodology to their previous work and show that detachment of previously retained particles or flocs is a significant factor contributing to filter breakthrough. They further conclude that the predominant breakthrough occurs with intermediate (3 - 7μm) and larger (8 - 25μm) sized particles, with the smaller sizes continuing to exhibit significant removal. Clearly, this result is of significance to water quality monitoring as it suggests that monitoring a specific size range (corresponding for example to the size of individual oocysts) may not provide a valid indication of oocyst removal as the oocyst or oocysts may be attached to floc particles and therefore of a much larger physical size.

The authors noted that the observed detachment could be prevented by increasing the bed depth as the filtration process was found to be one of continual capture and release of particles with detachment occurring at higher levels of the filter and reattachment occurring at lower levels. When clean water was introduced after the passage of significant effluent the extent of detachment was reduced, adding support to the theory that detachment is strongly influenced by incoming particles. A final and interesting note by the authors was that a rise in head loss did not appear to be a major factor in particle detachment.

5.2.2 Investigation of particle association with floc particles

An investigation into the possibility of pathogens or pathogen sized particles detaching with floc particles and entering the filtered water was investigated as part of an initial pilot-scale trial at the Macarthur Water Treatment Plant. The investigation was conducted using a field based pilot-scale plant due to the ready supply of raw water to treat and feed to the filtration columns. A full description of the pilot plant is given in the following section. The columns used have been described previously in the filtration study and consist of dual anthracite sand operated at 23m/h. Such a rate

represents the highest filtration rate possible with this plant and exceeds the upper level of the capacity of the full-scale Macarthur Plant.

During operation of this plant the turbidity, particle counts and total iron residuals were measured on two columns experiencing identical treatment processes in terms of an LT-35 dose of 1.0 mg/L and LT-20 dose of 0.01 mg/L. The ferric chloride dose was 15 mg/L in column 1 and 7 mg/L in column 2. A headloss of 2 metres was attained by column two at 425 minutes and by filter column one at 275 minutes.

The reduction in turbidity and particle counts as a measure of the treatment performance is the basis of the following section dealing more specifically with the pilot-scale trial. The correlation of particles and iron is however, of importance as it is an indication of particle-containing floc breakthrough of the filters. The results of such an investigation are shown in Figure 5.1.

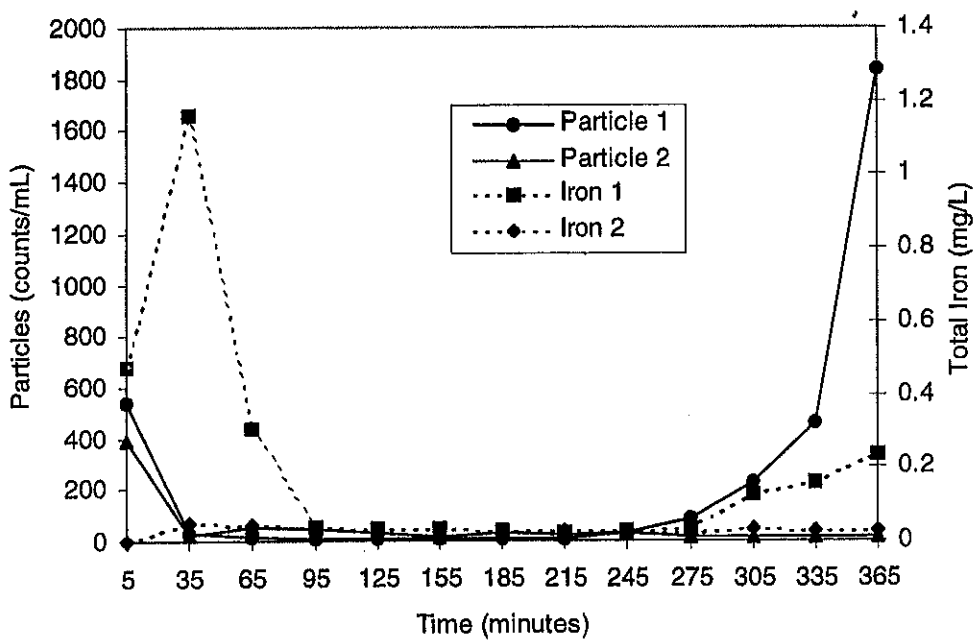


Figure 5.1. Particle counts and total iron versus filter run time in pilot plant studies at Macarthur Water Treatment Plant on 4 December 1996.

The results, in terms of particles and iron, reflect a traditional profile of filter ripening followed by an extended period of good quality water. The raw water conditions at the time of this trial were turbidity of 1.2 NTU and true colour of 5. The particle counts in the raw water for all particles > 2μm was 1,635 counts/mL and the total iron was 0.198 mg/L. The particle counts after ripening were 31 ± 23 counts/mL for column 1 and 33 ± 21 counts/mL for column 2 which reflects a very high quality of water. Of interest is the lack of correlation between iron and particles in the filter ripening stages. In column 1 the level of total iron peaks at 1.16 mg/L however at this stage the particle counts has already reduced to a ripened state at 35 counts/mL. In column 2 both the total iron levels and particle counts had reduced to a ripened state by 10 minutes.

After column 1 had reached the end of the filtration run (headloss of 2m) at 275 minutes a breakthrough was induced by ceasing the lime dose to column 1 resulting in a pH drop from 7.2 to 5.5. This had the result of destabilising the filter floc deposit and a dramatic rise in the passage of particles and iron into the filtered water resulted. At this stage the correlation of particles and iron is more evident, however, the levels of iron are slightly elevated to a level close to that of the raw water. Such a level would not indicate dramatic floc breakthrough but rather a partial solubilisation of the deposited iron oxyhydroxides by the lower pH and subsequent particle release. Such results do not appear to support the theory put forward by Moran et al., which would tend to predict a dramatic rise in iron residuals as floc entered the filtered water. The results are supportive of the earlier suggestion that the particles detected in the filter ripening stage are comprised mainly of anthracite and sand (that is, filter media particles) that have been produced during the backwashing stage through media attrition.

The difference in results can be largely explained by the difference in experimental conditions. The work done by Moran et al. used uniform filter media and the constant build up of floc material on the surface of the media may have created a porous deposition which would have been expected to detain smaller particles in preference to larger particles by well accepted size exclusion principles. Such a situation was not the case for the pilot-scale work as an irregular media shape was employed resulting

in non-uniform shear throughout the column depth. In addition to this the gel-like nature of ferrooxyhydroxide-polymer floc resulted in considerable deposition in the top 30% of the filter column and little deposition below this. This gel-like deposition would appear to establish a porous blanket at the top of the column which makes for a highly efficient particle capture and detainment. The efficiency of capture and detainment meant that all pilot-scale runs that were conducted to 2m headloss, after the pilot-scale plant process was optimised and resulted in no detectable breakthrough in terms of particles or turbidity.

6. Field evaluation of alternative methods for monitoring treatment performance

6.1 Introduction

As part of the WSAA/SWC/NWW-sponsored project on Particle Counting for Application to Water Treatment Control and Optimisation, interest was expressed in the ability of particle counts to serve as a useful indicator of treatment performance for control of *Cryptosporidium* and *Giardia*. These two organisms have been found in local sources of public water supply, and identified as causes of waterborne outbreaks of gastroenteritis elsewhere. Accordingly, public health officials believe water quality monitoring and treatment conditions should be directed to assuring that control of *Cryptosporidium* and *Giardia* is optimised.

6.1.1 Problem statement

Cryptosporidium and *Giardia* are common constituents of raw water fed to water filtration plants similar to the Macarthur Water Filtration Plant (WFP). Accordingly, water quality monitoring and treatment operating conditions are being examined to assure effective control of *Cryptosporidium* and *Giardia*. Previous work has suggested that optimisation of water filtration for turbidity and particle removal provides effective removal of *Cryptosporidium* and *Giardia*. Determining if effective removal is truly optimal would be a desirable ultimate goal, although one far exceeding the capabilities of this preliminary investigation.

Significant work on evaluation of particle counting as a means of assisting in the monitoring and optimisation of Macarthur WFP has been conducted and is described in the preceding chapters. Significant value would result from establishing a direct relationship between particle counting (or other easily monitored parameters) and *Cryptosporidium* oocyst concentrations as means of describing WFP performance.

Removal characteristics can be defined by a suitable testing program on the full-scale works as demonstrated by the 1993 Orchard Hills study. However, basic features of removal characteristics can be established more economically by first conducting seeding studies in the pilot plant on-site. Full-scale works testing can then be more finely focused and conducted to confirm the pilot scale findings.

Elements of works design and operation that should be covered in pilot and full-scale testing to enable comprehensive understanding of treatment performance characteristics and for identification of critical conditions for treatment and operation include the following:

- water quality conditions - Every works will experience a small range of perhaps two to four or five typical water quality conditions. The specific condition will dictate coagulation requirements and will have associated features of filter ripening time, filter run length, and filtered water turbidity. Pilot testing should eventually cover each of the characteristic water quality conditions. Priorities for testing should follow the number of days in the year on which each might occur, and any association with (cycling of) *Cryptosporidium & Giardia* concentrations, if known. A preliminary priority list is suggested as follows:

    1. typical dry weather
        - summer (higher temp. & algae conc.)
        - winter (lower temp. & algae conc.)
    2. typical wet weather
        - summer (short, intense)
        - winter (long, less intense)
    3. reservoir overturn
    4. drought (low reservoir pool depth - higher algae)

- chemical conditioning regime - The nature of chemical conditioning applied at any specific water treatment works is a critical element contributing to filtration performance for any set of water quality conditions. The elements of conditioning that must be taken into account include:

    1. the conditioning chemicals and the concentrations at which they are applied;
    2. pH;
    3. water temperature;

4. physical details of coagulation and flocculation--mixing energies and times.

- filter run cycle - Rapid rate granular media filters have four periods of operation that have been shown to have associated different characteristics for *Cryptosporidium* and *Giardia* removal: a) ripening; b) stable low turbidity production; c) at run termination and d) following periods of intra filter run change in hydraulic or other relevant operating condition. Removal characteristics should be established for each period. Due to the concentration and time proportions of the total cycle, organisms passed during ripening and break through will likely control the overall finished water concentration.

- Characteristics of backwash reclamation should be measured to permit completion of a mass balance (see diagram below).

- Treatment system generalised particle mass balance:

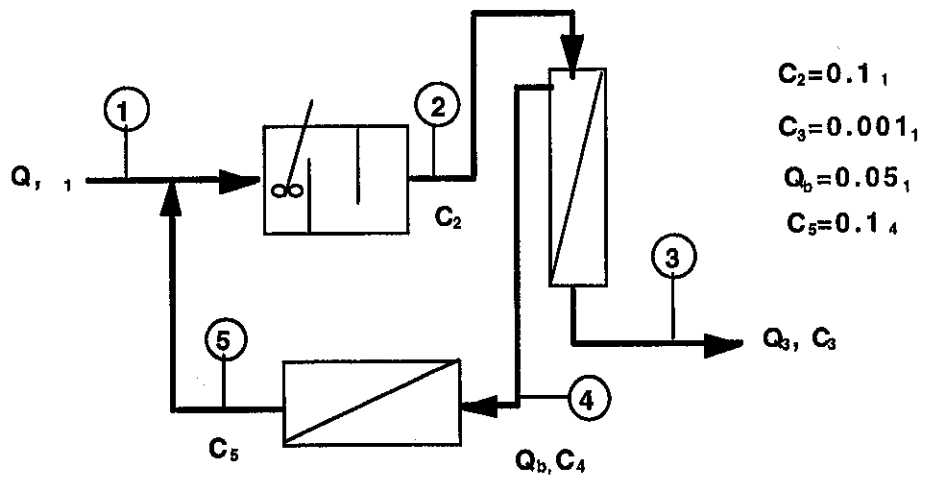


Figure 6.1. Schematic diagram of a filtration treatment works with sedimentation, filtration, and settled backwash return where sampling point 1 is of raw water, sampling point 2 after chemicals addition but immediately prior to filtration, sampling point 3 follows deep bed filtration and sampling points 4 and 5 collect backwash water before and after clarification by settling.

A schematic diagram of the Macarthur works (Figure 6.1) shows the principal components with potential sources and sinks for particulate matter. A backwash reclamation system (settling/thickening) partitions organisms between the sludge and recycled water. Monitoring data on four of the five liquid stream locations shown is required to define a mass balance for the system.

6.1.2 Objectives

Objectives of this project were as follows:

1. Define the *Cryptosporidium* removal characteristics of the Macarthur water filtration system for selected operating conditions and for water quality and treatment conditions occurring at the time of the investigation.

2. Examine the relation between particle counts, turbidity, and *Cryptosporidium* oocyst concentrations as alternate measures of treatment performance.

3. Provide suggestions for further work important to understanding the significance of *Cryptosporidium* concentrations in relation to water treatment objectives and to use of *Cryptosporidium* concentration and/or surrogate measures for monitoring and control of treatment.

6.1.3 Approach

The approach agreed to for the pilot plant seeding phase of the *Cryptosporidium* removal study included the following elements:

1. Total project scope to be limited to a preliminary screening study only;

2. Project effort would be divided between three seeding runs to be conducted essentially as replicates;

3. Time limitations for completion required that the runs be made at 2-week intervals between the beginning of May and mid June;

4. Organism seeding to be conducted continuously;

5. The seeding concentration to be selected to facilitate measurement of concentration at the most difficult sampling point (filtered water) and to provide for a low relative error in concentration measurement;

6. Sample volumes and analytical procedures to be matched carefully to individual requirements of the sampling points;

7. Analytical procedures for *Cryptosporidium* to be accompanied by quality control (ie. seeded positive controls to demonstrate recovery efficiency which were different for different sampling points). Analysis was to be performed by membrane filtration-IFA-microscopy;

8. Two sampling points to be used to define performance characteristics of the in-line filtration system at the Macarthur Works;

9. Seeding to be conducted with *Cryptosporidium* alone due to time and cost limitations;

10. To provide for safety in relation to use of *Cryptosporidium* oocysts at the water treatment works, the organisms used were to be killed, and all liquid residuals were to be collected and disposed off-site as provided for by the treatment plant operators;

11. Measurements to be made in triplicate to establish performance characteristics with reasonable statistical confidence; and

12. The topics of priority interest in allocating monitoring effort for the seeding trial were as follows: 1) performance during stable operation portion of the filter cycle for water quality conditions occurring during the trial period (May to mid June, 1998); 2) Operation at the design filtration rate of 8 m/hr; 3) Operation using the same chemical regime being used in the full-scale plant at the times of seeding

operation; and 4) Performance during the latter portion of filter ripening and potentially, effects of hydraulic changes during a filter run.

6.2 Methods

6.2.1 Macarthur water treatment works

The Macarthur Water Filtration Plant is of in-line filtration design, providing chemical addition and mixing (coagulation) prior to filtration, but without separate flocculation or settling (see Figure 6.2). The plant was commissioned in 1996, having a current design capacity of 265 ML/day, corresponding to a unit filtration rate of 8 m/hr. During the winter 1998 period of this project, production rates were typically either about 52 ML/day or 96 ML/day. The average plant production from May 14 to June 11 was 76 ML/day.

The filter media design is deep-bed mono-media, using anthracite as the principle media. Filter cycle length is typically based on accumulated headloss or elapsed time, with the time criterion normally reached first. The plant is operated with all filters operating continuously. Exceptions occur when individual filters are taken out of service for maintenance. Control of plant production is achieved by varying of the application rate to the filters.

Chemical conditioning is accomplished using ferric chloride as the primary coagulant, typically in the range of 2 to 6 mg/L (as 40% $FeCl_3$ as received from the supplier). A nonionic polyelectrolyte coagulant aid, polyacrylamide is used at 1 to 1.5 mg/L, and an anionic polyelectrolyte, polydadmac is used as a filter aid. Alkalinity and pH are adjusted to favourable conditions by addition of carbon dioxide and lime.

Raw water quality at the Macarthur filtration plant varies seasonally and with source, but within a relatively narrow range of key parameters including turbidity, pH, alkalinity, TOC and temperature (see Table 6.1). Raw water turbidity is typically near 1 (0.7-1.3) NTU although heavy rainfall (of the order of once per year) may produce periods of turbidities over 20 NTU that may persist for up to several days.

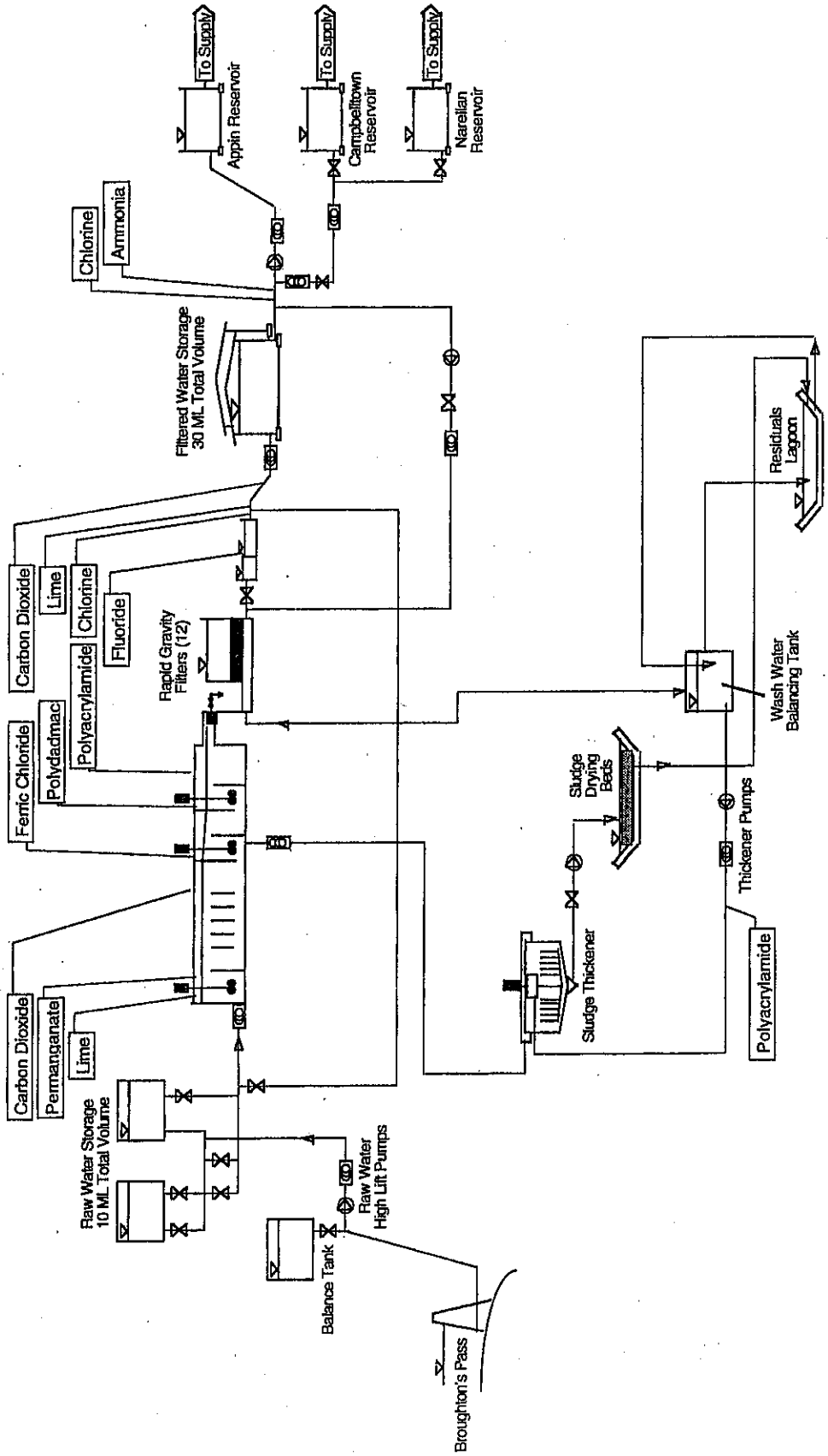
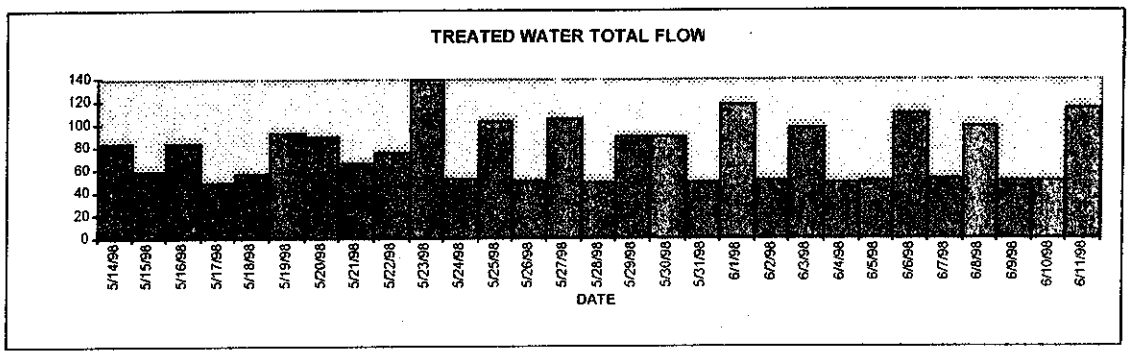


Figure 6.2. Schematic Diagram of the Macarthur Water Treatment Plant Including Major Process Components and Chemical Additions



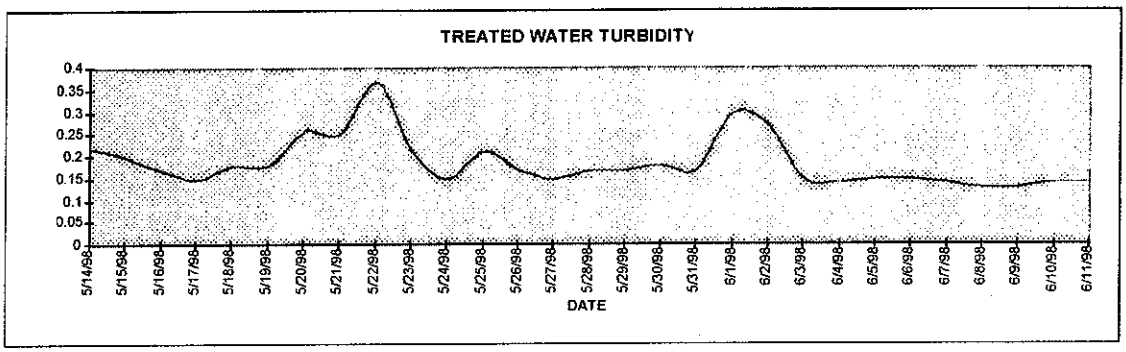


Figure 6.3. Water production and filtered water turbidity at Macarthur Water Filtration Plant, May – June 1998.

6.2.2 Macarthur WTW pilot plant

The Macarthur WTW has a pilot filtration facility for use in research and operational testing. The facility consists of three parallel trains including components analogous to those of the full-scale plant. Raw water from the headworks is delivered to the roof of the lab building (Figure 6.4). Flow can be delivered to any one or combination of the three parallel trains. Flow first passes through float controlled tanks (23 L) in which lime and carbon dioxide are added. Flow is conveyed through 10 m of 40 mm plastic pipe into which ferric chloride is metered followed by a 10 cm static mixing

section. Flow then passes into a second float controlled mixing tank (13 L) immediately after introduction of the coagulant aid. Flow then passes vertically down into the top of the 250 mm dia. (0.1 m^2) perspex filter columns where the pH is monitored and filter aid is added (Figure 6.5). The volume of column above the filter bed is approximately 0.2 m^3.

Table 6.1. Summary of raw and treated water quality at the Macarthur Water Filtration Plant, May – June 1998.

Date	Raw water total flow (ML/day)	Raw water turbidity (NTU)	Post filter turbidity (NTU)	Raw water pH	2nd flash mixer pH	Post filter pH	Raw water temp (°C)	Backwash water consump. (ML/day)
5/23/98	139.9	1.9	0.08	6.91	7.74	8.51	14.6	5.7
5/24/98	51.8	1.2	0.12	6.86	7.73	8.59	14.2	2.2
5/25/98	104.8	1.4	0.18	6.87	7.63	8.57	13.7	3.1
5/26/98	51.4	1.0	0.04	6.41	7.47	7.64	13.6	1.8
5/27/98	107.2	1.1	0.03	6.35	7.49	7.28	13.4	3.9
5/28/98	51.2	1.0	0.04	6.39	7.49	7.41	13.0	1.8
5/29/98	88.7	1.2	0.04	6.37	7.49	7.31	13.0	2.6
5/30/98	91.2	1.2	0.12	6.36	7.41	7.21	12.9	3.1
5/31/98	50.9	1.0	0.09	6.96	7.49	7.42	12.6	0.9
6/01/98	117.9	1.0	0.23	7.16	7.49	7.02	11.7	3.5
6/02/98	51.1	1.0	0.12	7.00	7.59	7.30	12.8	0.9
6/03/98	98.2	1.2	0.05	6.98	7.59	7.40	13.8	4.5
6/04/98	51.1	1.2	0.04	7.02	7.59	7.55	14.2	2.2
6/05/98	51.0	1.0	0.03	6.98	7.59	7.67	14.4	1.3
6/06/98	110.5	1.0	0.03	7.01	7.59	7.47	14.1	4.4
6/07/98	53.3	0.9	0.02	7.00	7.58	7.58	13.8	0.9
6/08/98	98.8	0.9	0.02	7.03	7.59	7.51	13.5	3.9
6/09/98	53.0	0.9	0.02	7.05	7.59	7.57	13.3	3.1
6/10/98	51.0	1.0	0.02	7.03	7.59	7.63	13.2	1.3
6/11/98	114.7	1.0	0.02	6.98	7.59	7.35	13.2	3.9

Figure 6.4. Pilot raw water source, chemical addition and mixing at Macarthur WFP.

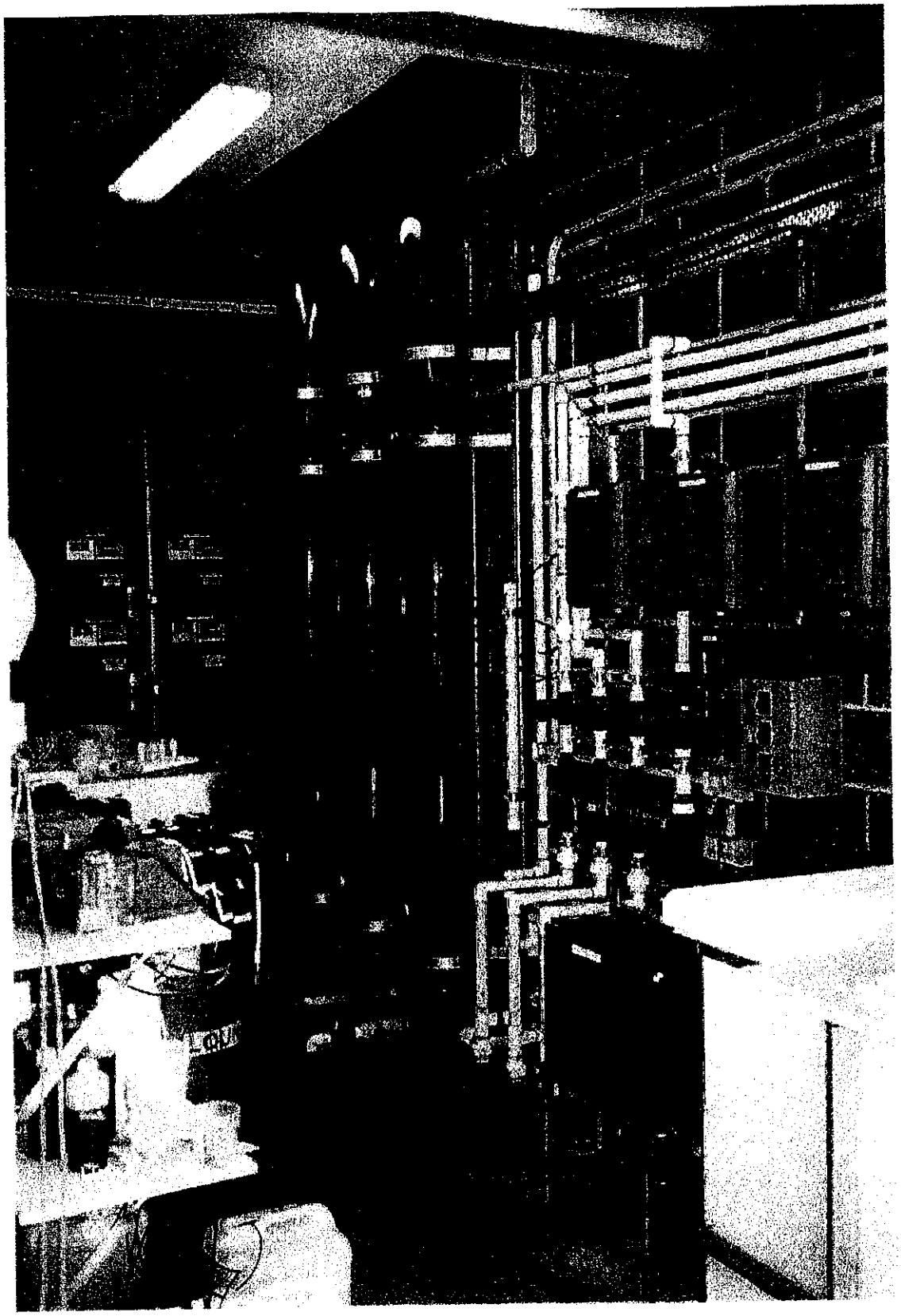


Figure 6.5. Pilot chemical feed, filters and flow controls at the Macarthur WFP.

The pilot filters contain approximately 2m of media identical to that of the full scale filters. Filtration rate is controlled by a variable speed pump on the discharge line from the bottom of each column. Each filter is equipped with separate chemical feed for each of the plant conditioning components. A computer data logging system provides for continuous monitoring of raw and filtered water turbidity, pH, and headloss accumulation.

For this project a single pilot treatment train (Number 3) was used. For each filter run, the pilot filter was operated beginning on the day prior to the run, using the chemical conditioning regimen in use at the full-scale plant. Preliminary operation was continued over-night and terminated with a normal backwash immediately prior to the planned seeding run. The filter was operated throughout each of the three runs at a filtration rate of 8 m/hr corresponding to a flowrate of 400 L/hr.

6.2.3 *Cryptosporidium* seeding

Cryptosporidium oocysts were prepared for the pilot seeding experiments by isolation from fresh dairy calf faeces. Cleaned oocysts were counted by haemocytometer and checked by immunofluorescence assay (IFA) and direct microscopic counting of appropriately sized aliquots. Each experiment required approximately 10^8 oocysts. Oocysts were fixed in 0.1% formalin for at least 48 hours prior to use.

The seed was diluted to a working volume that could be delivered at constant and reliable flow by the available peristaltic pumps normally used to feed chemicals to the pilot filter trains. New tubing was used to avoid any effect of previous chemical use. The stock suspension was located on the lab roof adjacent to the lime addition tank and kept mixed and cool throughout each seeding run. The stock suspension was pumped into the lime addition tank at a constant rate which was verified by the sampling and analysis program described below.

6.2.4 *Cryptosporidium* analysis

Cryptosporidium concentrations were established by sampling and analysis to permit measuring the removal provided by the pilot filter. Feed rates were monitored periodically through each run. Samples of the stock suspension were taken at each sampling period to confirm the feed concentration. Filtration removals were determined by sampling from immediately above and below the filter for measurement of *Cryptosporidium* concentrations. Volumes collected at each sampling location were selected to permit analysis with minimum relative error.

Analysis was performed by IFA on 13 mm dia. 2 μm pore dia., polycarbonate etched-pore membranes (Poretics, Livermore, CA), using direct-labelled (fluorescein isothiocyanate (FITC) monoclonal antibody to *Cryptosporidium* oocysts (Waterborne, Inc. New Orleans, LA). At each sampling time, three independent samples were collected and processed for measurement of *Cryptosporidium* concentration.

6.2.5 Particle counting and particle size distribution analysis

The Hiac/Royco Model 9064 batch type particle counter was used for enumerating particles. This counter is supported by PDAS software and a sensor for particle size detection and data logging. The counter was once calibrated before the start of data collection. The sensor can detect a wide range of particle sizes ie., 1- 250 μm scattered over 50 channels. For this project, particles were counted in 10 channels covering the size range of 1.5 to 12.5 μm. Both differential and cumulative particle counts were recorded. Counts were made on raw and filtered water. Raw water counts were made from four to six representative times over the course of each run. Filtered water counts were taken at times corresponding to each of the *Cryptosporidium* samplings and at up to 50 intermediate times to data corresponding with the continuous turbidity measurements. All samples were processed at the recommended sensor flow rate of 25 mL/min.

The analysis protocol followed for each sample counting time was as follows as

indicated by the Hiac-Royco manual:

1. Connect the counter, the computer and the sensor and turn the computer and the counter ON.
2. Connect the sensor to the fluid supply line through a flow meter. Set the flow meter to 25 mL/min. The flowrate was controlled by means of a manually operated valve and rotameter, making adjustments as necessary to maintain constant flow.
3. Switch the computer to PDAS by typing "cdlpdas" and "pdas" at the C-prompts
4. Check the flow rate and press 'F9'
5. Provide a file name and the sample identification. The counter acquired data and displayed it after the programmed number of repetitions (3) as a table or plot.
6. Save data using "F8".

6.2.6 Aerobic spore analysis

Aerobic spore analysis was conducted by a culturing method described briefly as follows. Samples were collected in sterile glass bottles and maintained on ice until returned to the lab for processing within 8 hrs. Processing consisted of heat shock (emersion in an 75°C water bath) for 10 min, then immediately iced. Appropriate volumes were then filtered using 0.45 μm pore dia. cellulose acetate membranes in a sterile filter holder. Completed membranes were placed on R2A agar (Oxoid). Plates were incubated overnight at 35°C. Resulting colonies were enumerated and the concentration of aerobic bacillus spored deduced by calculation from counts and volumes.

6.2.7 QA/QC

Quality control procedures follow basic NATA requirements for microbiological analysis. Quality procedures to assure interpretability of *Cryptosporidium* analysis consisted of the following elements:

1. Organism stock preparation and enumeration;
2. Organism storage, enumeration, and feeding during a seeding run;
3. Organism quantification in samples collected as part of the experimental design, including appropriate replication; and

4. Data analysis and evaluation.

6.2.8 Data analysis

Two types of calculations were made as an essential part of this project: 1) calculations of log reduction and 2) calculations of *Cryptosporidium* oocyst concentration. Calculations were made as described below.

1. **Log reduction.** Log reductions for this work were calculated as the difference between the $\log_{10}$ of the influent concentration and the $\log_{10}$ of the filtrate concentration.

2. ***Cryptosporidium* oocyst concentration.** For all samples, analytical operations were used involving loss of oocysts. The loss was quantified by measurement of the actual recovery efficiency by means of seeded samples, applying analytical procedure identical to that used in analysis of normal samples. The concentration of oocysts actually present in samples was then calculated as:

   oocyst conc., no./ L = oocysts in sample ÷ (recovery fraction × sample vol., L)

6.3 Results

The project consisted of operating the pilot plant for each of three periods, May 13-14, May 27-28, and June 11-12, 1998. On the initial day of each operating period the pilot plant was started and operation established for chemical conditioning identical to that being used in the full-scale plant. Flow was set at 400 L/hr (8 m/hr) and the pilot filter was allowed to run over night.

In the morning of each second day, oocyst feed was begun, the filter was backwashed, and the seeding run begun, monitoring filtered water turbidity as the key parameter selected as the criterion for picking the initial sampling time.

6.3.1 Run 1, 13-14 May 1998

1. General description of conditions of operation. The raw water turbidity occurring at the Macarthur WFP on May 13-14 was 0.8 NTU, typical of dry late autumn conditions (Figure 6.6). The plant was operating using 2.4 mg/L of ferric chloride, 1.2 mg/L polyacrylamide, and 0.12 mg/L polydadmac. The plant was operating at 50 ML/day and filtered water turbidity was approximately 0.2 NTU.

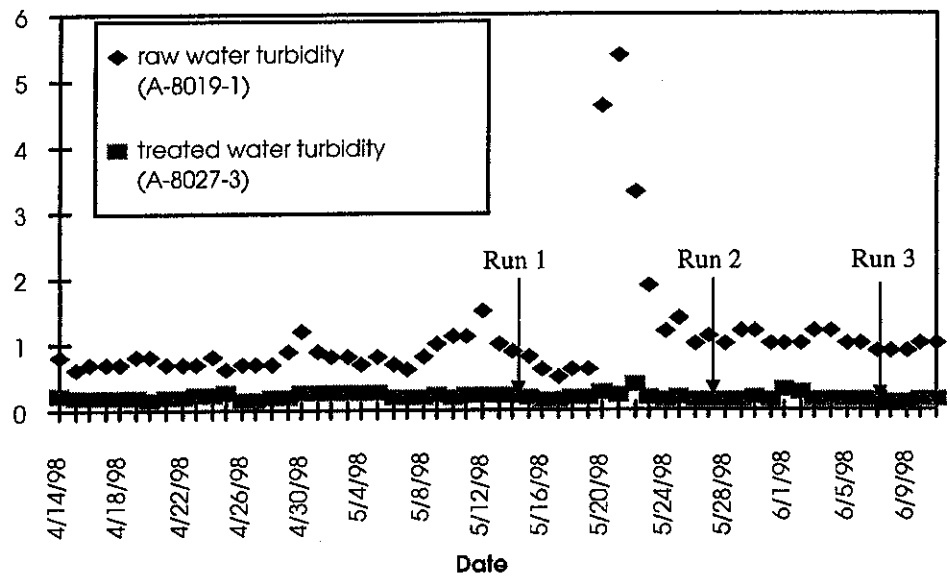


Figure 6.6. Macarthur WFP raw and treated water turbidity record, April-June, 1998

2. Specific run plan

The first seeding run was designed as described above to meet the stated project objective. The pilot plant had not been in use for several months prior to the run. Accordingly, two days before the seeding run the pilot train to be used was put into service beginning by backwashing the filter. Minor modifications to the column fittings were made to permit sampling at the desired points. And, a normal preliminary filter run, without seeding, was initiated on the day preceding the seeding run.

The seeding run was to be commenced following backwash after termination of the preliminary run. During the backwash period, normal chemical coagulation was maintained with the flow diverted to waste via a bypass valve at the top of the filter. Seeding of the influent flow was commenced during this period to provide for the desired feed concentration to the filter immediately upon beginning the filter run.

Three samplings were planned for the run: 1) the first at the end of ripening, arbitrarily selected at the point when filtrate turbidity reached 0.1 NTU; 2) the second after approximately two hours of operation following the first sampling; and 3) the third after approximately four hours of operation following the first sampling. Turbidity and particle counts were monitored throughout the run.

3. Turbidity, particle counts, *Cryptosporidium* concentrations, and aerobic spore concentrations.

The pilot plant data logging system was not operating during the first run. Raw and filtered water turbidities were recorded manually at times corresponding to filtered water particle counting and sampling for other parameters. The graphical record of filtered water turbidity and particle counts is shown in Figure 6.7 and describes the general pilot filter performance observed during the run. Samples were collected for *Cryptosporidium* and aerobic spore analysis at 25, 140, and 260 minutes after backwashing. The first samples were collected just at the end of the ripening period. For reasons that are not well understood, the filtered water turbidity and particle counts were quite volatile. Although filtered water turbidity after ripening remained below 0.05 NTU compared to ca. 0.2 NTU for the full scale plant, performance was apparently easily affected by minor fluctuations in operation. Two adjustments were made in the pilot plant flowrate, at approximately 140 min., just after the second sampling, and 230 min., just prior to the third sampling. Appreciable effects in the filtered water turbidity

appeared to accompany the adjustments. Effects on particle counts were not clear.

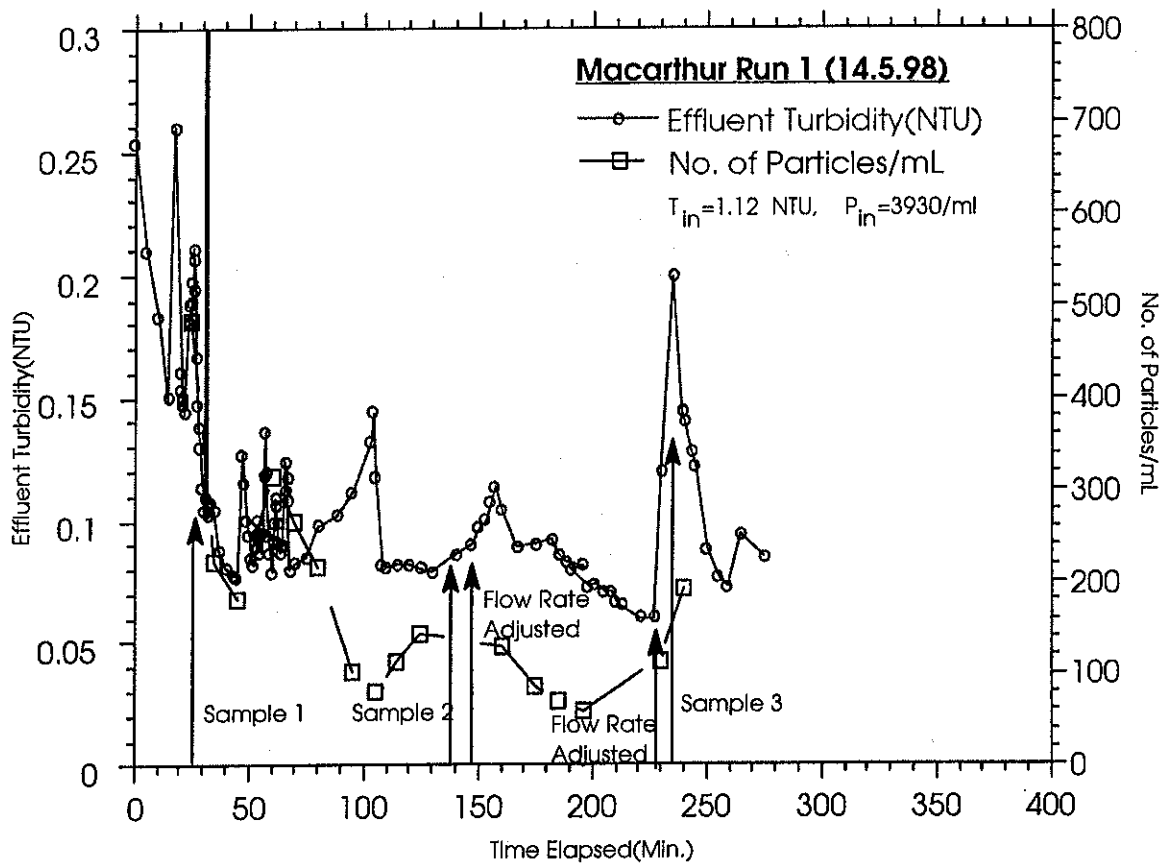


Figure 6.7. Filtered water turbidity and particle counts, Pilot filter 3, 14.4.98.

The reductions observed in each of the four performance parameters measured, turbidity, particles (1.5-12.5 μm), *Cryptosporidium*, and aerobic spores, are summarised in Figure 6.8. The sum of particles in the 1.5 to 12.5μm size range was reduced from ca. 3500/mL in the raw water to ca. 100/mL in the filtered water. *Cryptosporidium* concentrations above the filter ca. 20,000/L were reduced to ca. 100/L. Aerobic spore concentrations ca. 10,000 cfu/L above the filter were reduced to levels slightly below 100 cfu/L.

Log reductions for *Cryptosporidium* averaged 2.35, 2.32, and 2.05 for the three sampling times. Log reductions for aerobic spores averaged 2.16, 2.36, and 2.14 for the same sampling times.

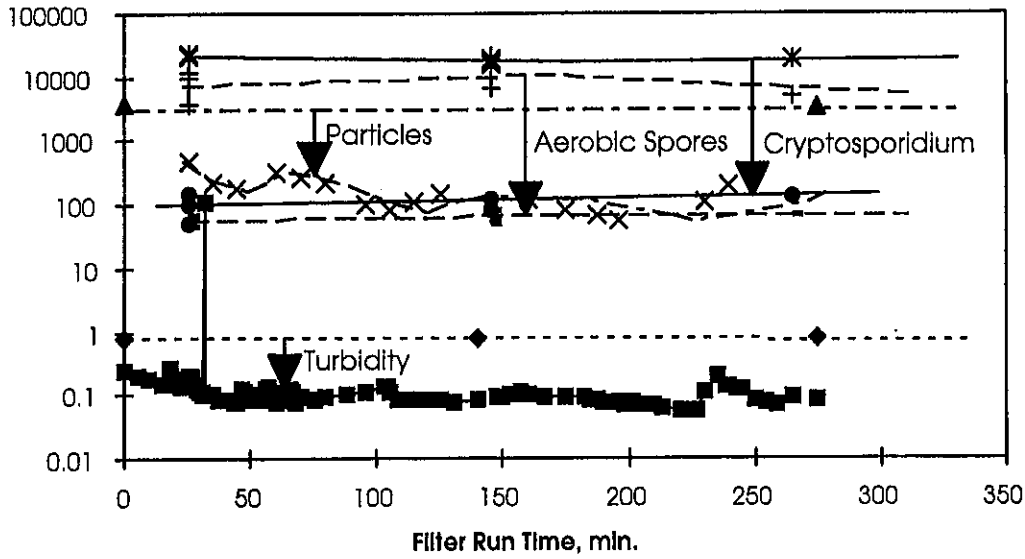


Figure 6.8. Macarthur pilot plant performance (*Cryptosporidium*, particle counts, turbidity, and aerobic spores) during the first seeding run, 14.5.98.

The distribution of particles in the 1.5-12.5 μm size range in raw water during the run was as described in Figure 6.9. The distribution of particle sizes in the same range in the seed suspension is shown on the same figure indicated by the open circles.

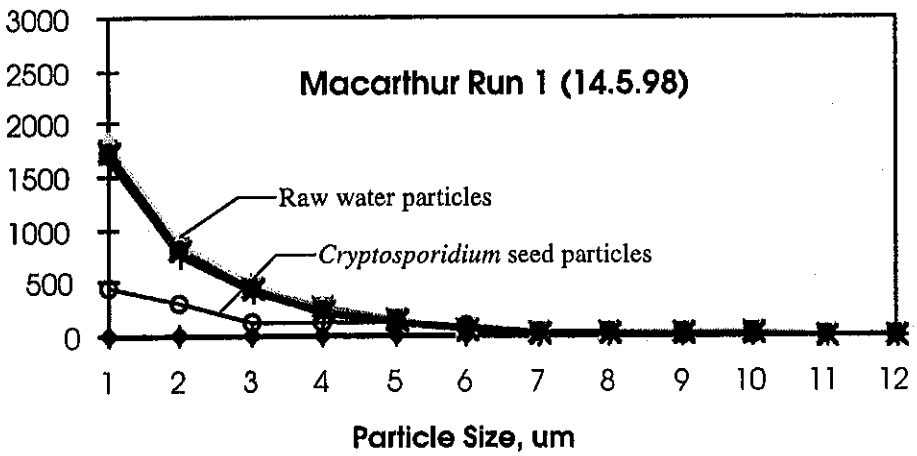


Figure 6.9. Distribution of particles by size (1.5-12.5 μm) in raw water at the Macarthur WFP during the pilot filter seeding run on 14.5.98.

6.3.2 Run 2, 27-28 May 1998

1. General description of conditions of operation.

The raw water turbidity occurring at the Macarthur WFP on May 27-28 was 1.1 NTU. (Table 6.1). This run occurred 8-days after a rainfall of ca. 100 mm on the Monday night of 18-19 May. During that period raw water turbidity at the plant rose to over 30 NTU. During this seeding run the main plant was operating using 6.3 mg/L of ferric chloride, 1.6 mg/L polyacrylamide, and 0.083 mg/L polydadmac. The plant was operating at 50 ML/day with two filters out of service for maintenance. The filtered water turbidity of the main plant during the seeding run was ca. 0.12 NTU.

2. Specific run plan

The second seeding run was designed as described above to meet the stated project objective. The principal difference between the first and second seeding runs concerned the consistency of the flowrate and adjustment of filtered water sampling in relation to flow control. During the second run no adjustments in flowrate were made throughout the run. In fact none was needed. During the second run the filtered water sampling valve was adjusted to provide a continuous flow of 2 L/min. The effluent flow control pump was adjusted accordingly to provide the desired total filter flow, 400 L/hr, corresponding to 8 m/hr filtration rate.

Following the preliminary day's filter operation, the filter was backwashed (10:20 am). The seeding run was commenced at 10:40 am, following backwash. During the backwash period, normal chemical coagulation was maintained with the flow diverted to waste via a bypass valve at the top of the filter. Seeding of the influent flow was commenced during this period to provide for the desired feed concentration to the filter immediately upon beginning the filter run.

Three samplings were planned for the run: 1) the first at the end of ripening, arbitrarily selected at the point when filtrate turbidity reached 0.1 NTU; 2) the second after approximately two hours of operation following the first sampling; and 3) the third after approximately four hours of operation following the first sampling. Turbidity and particle counts were monitored throughout the run.

3. Turbidity, particle counts, *Cryptosporidium* concentrations, and aerobic spore concentrations.

The pilot plant data logging system was operating properly during the second run, providing a continuous record of filtered water turbidity. Raw water turbidity was recorded manually at times corresponding to filtered water particle counting and sampling for other parameters. The graphical record of filtered water turbidity and particle counts shown in Figure 6.10, describes the general pilot filter performance observed during the run. Samples were collected for *Cryptosporidium* and aerobic spore analysis at 35, 185, and 325 minutes after backwashing. The first samples were collected just at the end of the ripening period.

The filtered water turbidity and particle counts were essentially constant throughout the run. The filtered water turbidity fell to about 0.06 NTU, gradually reducing to a low of 0.04 NTU after 200 min, then gradually increasing again to 0.06 at the end of the run. Filtered water particle counts were consistently below 50/ mL throughout the run.

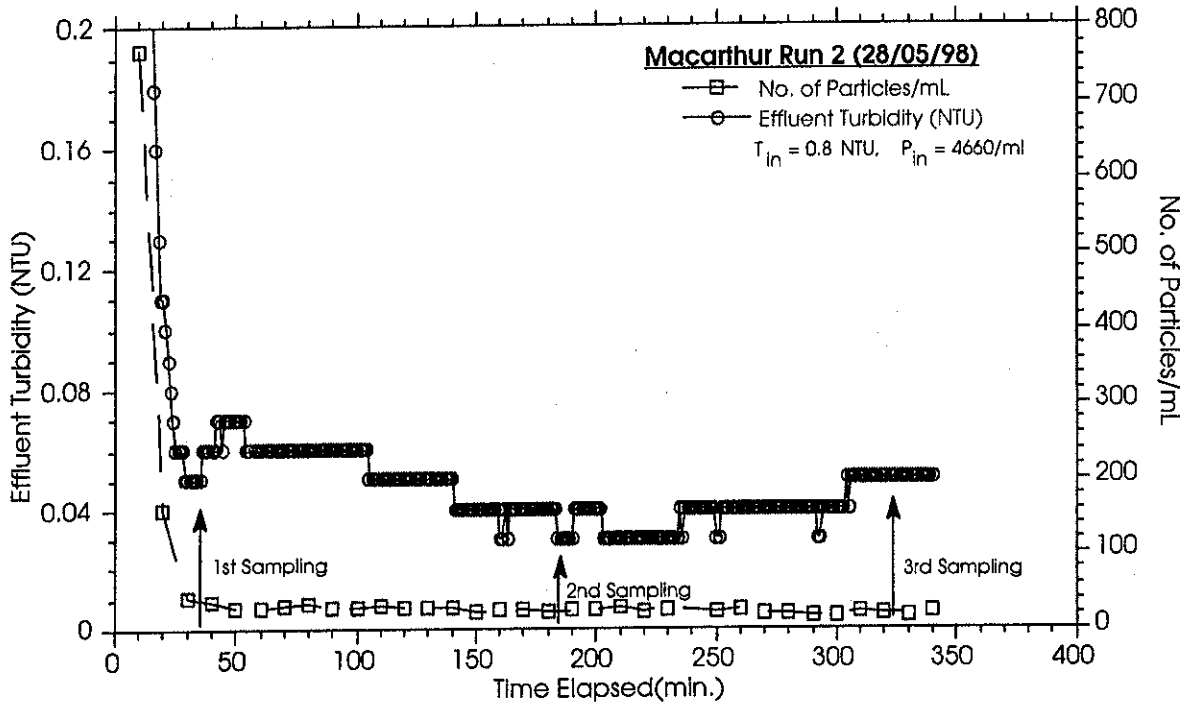


Figure 6.10. Filtered water turbidity and particle counts, Pilot filter 3, 28.5.98.

The reductions observed in each of the four performance parameters measured, turbidity, particles (1.5-12.5 μm), *Cryptosporidium*, and aerobic spores, are summarised in Figure 6.11. The sum of particles in the 1.5 to 12.5μm size range was reduced from ca. 4880/mL in the raw water to ca. 50/mL in the filtered water. *Cryptosporidium* concentrations above the filter ca. 8-9,000/L were reduced to ca. 2-5/L. Aerobic spore concentrations ca. 4-5,000 cfu/L above the filter were reduced to an average of 25 cfu/L.

Log reductions for *Cryptosporidium* averaged 3.33, 3.43, and 3.74 for the three sampling times. Log reductions for aerobic spores averaged 2.19, 2.23, and 2.24 for the same sampling times.

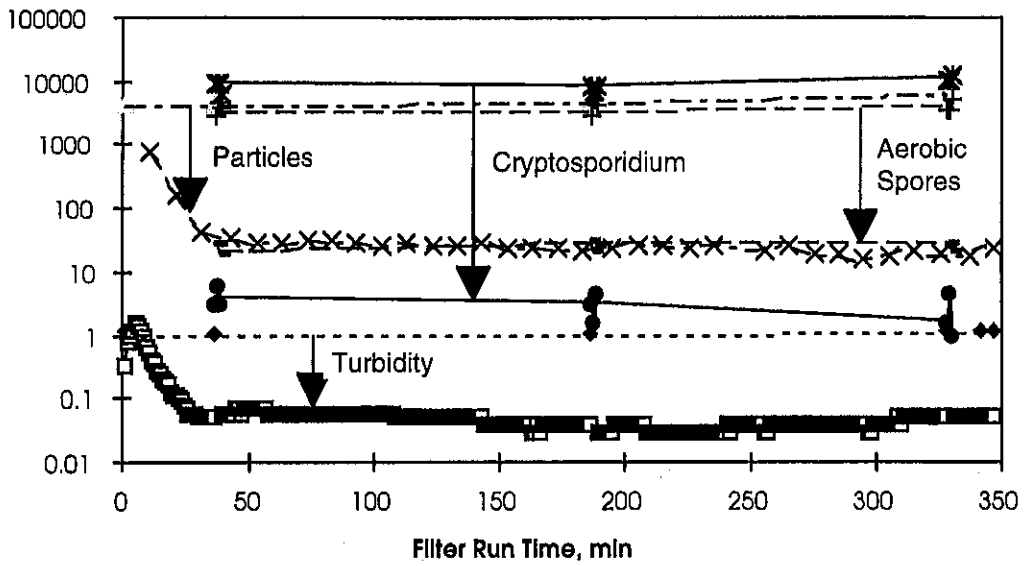


Figure 6.11. Macarthur pilot plant performance (*Cryptosporidium*, particle counts, turbidity, and aerobic spores) during the second seeding run, 28.5.98.

The distribution of particles in the 1.5-12.5 μm size range in raw water during the run was as described in Figure 6.12. The distribution of particle sizes in the same range in the seed suspension is shown on the same figure indicated by the open circles.

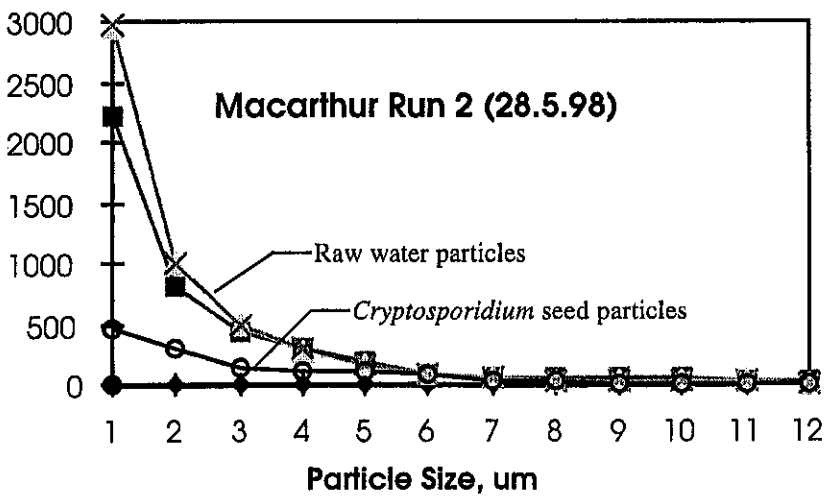


Figure 6.12. Distribution of particles by size (1.5-12.5 μm) in raw water at the Macarthur WFP during the pilot filter seeding run, 28.5.98

6.3.3 Run 3, 10-11 June 1998

1. General description of conditions of operation.

The raw water turbidity at the Macarthur WFP on May 27-28 was 1.1 NTU. (Table 6.1). Weather during this run was cool (15°C) and partly cloudy. Scattered showers had occurred the night before the run but did not appreciably affect the raw water turbidity. During this seeding run the main plant was operating using 5.5 mg/L of ferric chloride, 1.26 mg/L polyacrylamide, and 0.08 mg/L polydadmac. The plant was operating at 50 ML/day. The filtered water turbidity of the main plant during the seeding run was ca. 0.1 NTU.

2. Specific run plan

The third seeding run was designed as described above to meet the stated project objective. The third run was designed to replicate the second run using the same approach to maintaining a consistent flowrate. During the third run no adjustments in flowrate were needed to maintain constant flow through the filter.

Preliminary operation of the filter was begun on Wednesday, 10.6.98, at 1:00 pm. The preliminary operation was concluded with backwashing at 10:45 am. The seeding run was commenced at 11:00 am. As in previous runs normal chemical coagulation was maintained during the backwash period with the flow diverted to waste via a bypass valve at the top of the filter. Seeding of the influent flow was initiated prior to backwash as in the previous seeding runs.

Three samplings were planned for the run and a fourth was added to take advantage of an unplanned opportunity: 1) the first sampling was conducted during ripening and consisted of six individual samples rather than the normal three replicates. The ripening samples were taken at turbidities

ranging from 0.6-0.46 NTU, 0.40-0.318 NTU, 0.20-0.18 NTU, 0.128-0.121 NTU, 0.100 NTU, 0.07-0.068 NTU, and 0.050-0.048 NTU. A sample of equal volume was taken at turbidity 0.150-0.133 NTU for use as the positive control ; 2) the second set of samples was collected at 2:00 pm, after 3 hours of operation following the first sampling; 3) the third set of samples was collected at 4:00 pm, after five hours of operation following the first sampling; 4) After the last (third) normal set of samples, the flowrate through the filter was intentionally increased from 400 to 570 L/hr. Samples were collected to be analysed for *Cryptosporidium* concentration at times corresponding to peak of filtered water turbidity. Particle counts were also made during the turbidity excursion. Turbidity and particle counts were monitored throughout the run.

3. Turbidity, particle counts, *Cryptosporidium* concentrations, and aerobic spore concentrations.

The pilot plant data logging system was operating properly during the third run, providing a continuous record of filtered water turbidity. Raw water turbidity was recorded manually at times corresponding to filtered water particle counting and sampling for other parameters. The graphical record of filtered water turbidity and particle counts shown in Figure 6.13 describes the general pilot filter performance observed during the run. Samples were collected for *Cryptosporidium* and aerobic spore analysis during the first 80 min of operation and again at 180 and 300 min. of operation. The fourth unplanned set of samples was taken 315 min. after the run began.

The filtered water turbidity and particle counts were essentially constant throughout the run. The filtered water turbidity fell to 0.5 NTU at 10 min; 0.1 NTU was reached at 25 min after the run began. The turbidity continued to fall reaching 0.050 NTU at 80 minutes. Throughout the remainder of the run the turbidity continued to fall gradually reaching a low of 0.029 NTU just prior to run termination. Filtered water particle counts paralleled the turbidity, reaching a minimum of 50/mL at the end of the run.

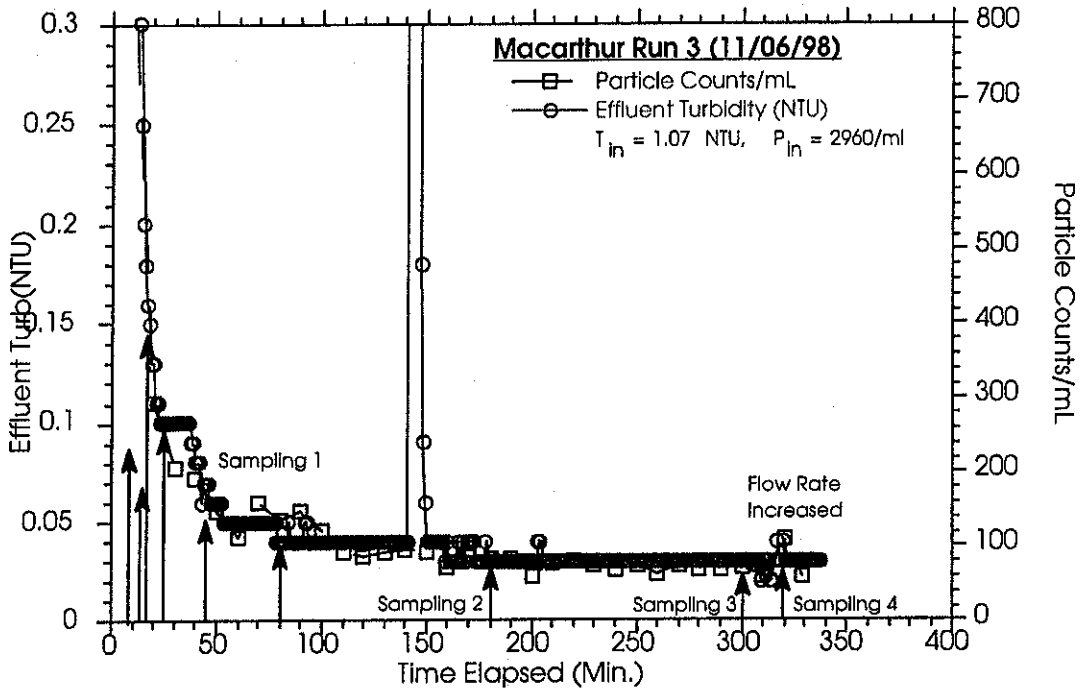


Figure 6.13. Filtered water turbidity and particle counts, Pilot filter 3, 11.6.98.

The spike in filtered water turbidity that occurred at just before 150 min. was caused by the backwash air-scour compressor turning on automatically (by its plenum pressure switch). This caused a small air leakage into the filter effluent line in which the filtered water turbidimeter is located. The excursion in the readings was accordingly not experienced in the filter itself.

The reductions observed in each of the four performance parameters measured, turbidity, particles (1.5-12.5 μm), *Cryptosporidium*, and aerobic spores, are summarised in Figure 6.14. The sum of particles in the 1.5 to 12.5μm size range was reduced from ca. 2960-3500/mL in the raw water to ca. 50-100/mL in the filtered water. Corresponding log reductions ranged from 1.06 to 1.68. *Cryptosporidium* concentrations above the filter ca 5-6,000/L were reduced to ca. 1-7/L. Aerobic spore concentrations ca. 2-4,000 cfu/L above the filter were reduced to 5-50 cfu/L.

Log reductions for *Cryptosporidium* averaged 3.24, 3.64, and 3.58 for the three sampling times. During the concluding period of flow increase, the log reduction was 3.17. Log reductions for aerobic spores averaged 1.47, 2.25, and 2.41 for the three sampling times. During the period of flow increase the aerobic spore log reduction was 2.33.

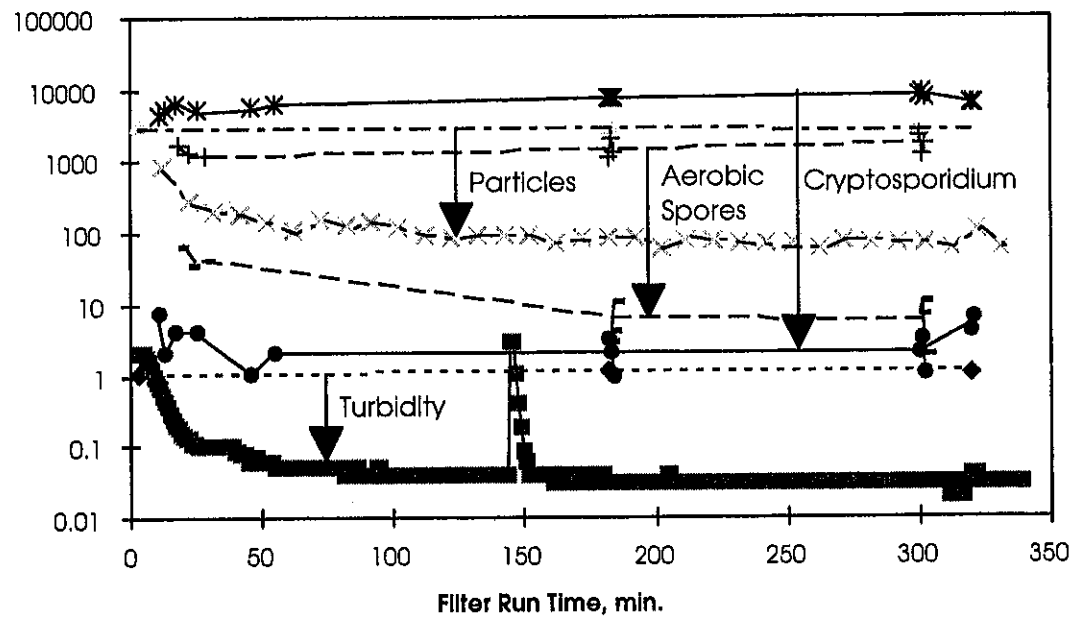


Figure 6.14. Macarthur pilot plant performance (*Cryptosporidium*, particle counts, turbidity, and aerobic spores) during the second seeding run, 11.6.98.

The distribution of particles in the 1.5-12.5 μm size range in raw water during the run was as described in Figure 6.15. The distribution of the same particle sizes in the seed suspension is shown on the same figure indicated by the open circles.

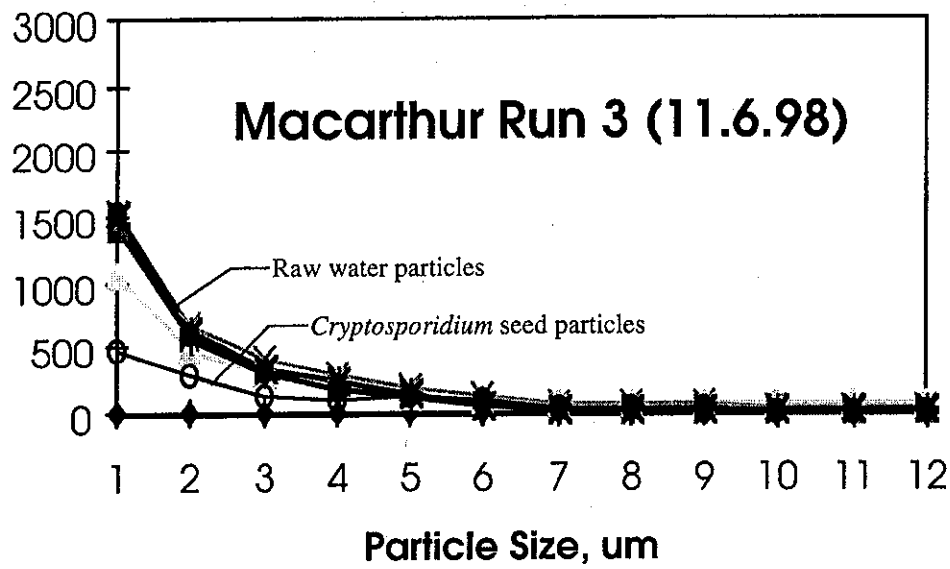


Figure 6.15. Distribution of particles by size (1.5-12.5 μm) in raw water at the Macarthur WFP during the pilot filter seeding run, 11.6.98

6.3.4 Summary of pilot plant performance (Runs 1, 2, and 3)

The performance of the pilot filter measured in terms of log reductions in turbidity, particle concentration, *Cryptosporidium* oocyst concentration, and aerobic spore concentrations summarises the observations during the three seeded runs and is shown in Table 6.2.

Log reductions were all calculated on the same basis, ie. the difference between the log of the prefiltration parameter value minus the log of the filter effluent parameter value. Although this is somewhat unconventional it permits comparing performance for the different parameters on a comparable basis.

It is interesting to note that the log reductions in turbidity generally had the smallest numerical values. This is due obviously to the relative insensitivity of turbidity as a measure of water quality. Of equal interest were the relatively small numerical values of log reductions calculated for particles and for aerobic spores. It appears that these values are quite sensitive to the raw water values. Or looking at it from the other end of the process, reductions appear to be limited to minimum values in filtered water

that were reached more or less independently of treatment conditions. If raw water concentrations were higher, it may be that the log removals would have been correspondingly greater.

Table 6.2. Summary of log reductions in turbidity and in concentrations of *Cryptosporidium* oocysts, particles and aerobic spores for seeded runs made at the Macarthur WFP in May-June, 1998

Run No.	*Cryptosporidium* Log Red.	Turbidity Log Red.	Particle Log Red.	Aerobic Spore Log Red.
1	2.35	1.12	1.09	2.16
	2.32	1.30	1.22	2.36
	2.12	1.09	0.94	2.14
2	3.33	1.51	2.04	2.19
	3.43	1.31	2.35	2.23
	3.74	1.28	2.43	2.24
3	3.09	0.34	1.06	
	3.14	0.87	1.24	1.47
	3.50	1.26	1.54	
	3.64	1.44	1.68	2.24
	3.58	1.57	1.68	2.41
	3.17	1.42	1.50	

6.4 Discussion

6.3.1 Macarthur WFP operation and performance

Five major factors of design and operation were identified as relevant to *Cryptosporidium* removal. One major unknown factor remains to be defined. The five relevant factors include:

1. The raw water turbidity is consistently low, ie. 1.0 ± 0.2 NTU, with peak periods during heavy runoff as high as 20 NTU lasting for at least several hours. The raw water particle concentration (1.5-12.5 μm) observed during the six week period of the study was 3-5000/mL. Particle concentrations in peak periods have not been defined.

2. Raw water *Cryptosporidium* oocyst concentrations as defined by SWC (1993, Hutton, P.H) and by a confirmatory measurement during this study is ca. 1.0 oocyst/L. The variation in oocyst concentration and its potential relation to turbidity, particle counts, and aerobic spore concentrations has not been defined.

3. Coagulation of natural particles is affected by the presence of significant dissolved organic carbon (DOC), monitored in terms of colour, typically 3-5 TCU with peaks to 50 TCU.

4. Coagulation is accomplished by addition of ferric chloride and polydadmac cationic polymer. A polyacrylamide nonionic polymer is added above the filters as a filter aid. Coagulation is accomplished with addition of lime at ca. 35 mg/L with pH adjusted to 6.7-7.2 by manipulation of lime and carbon dioxide addition.

5. The filters are of a deep bed monomedia anthracite of 1.2-1.4mm median size by 1.6 m deep over a second supporting layer of sand.

Performance of the full-scale plant, as measured by the filtered water turbidity, is controllable by manipulation of coagulant doses. Turbidity as low as the monitoring instrument's limit of detection (ca. 0.01-0.02 NTU) is achievable. Filtered water turbidity is typically maintained ca. 0.1-0.2 NTU to meet water quality goals at minimum cost.

6.4.2. *Cryptosporidium* oocyst removal

Cryptosporidium oocyst removals observed ranged from 2.12 to 3.74 logs among nine

sets of triplicate measurements during all three runs. This was comparable to the range reported in the literature for similar studies. We believe that measurements in the range of 2.12 to 2.35 logs during the first run were adversely affected by variations in the pilot filter flowrate that is not experienced in full-scale filters. Removals of that magnitude are thus not likely to be characteristic of the full-scale plant.

It may be noted that, in comparison to other reports in the literature, the log reductions reported here are based throughout on true concentrations accounting for recovery efficiency. The actual recovery efficiency efficiencies for C_{in} samples was 100% (the samples were applied to filters directly incurring no losses. The average recovery efficiency for C_{out} samples was 40.5%, s.d.=8.6%, n=6.

Cryptosporidium oocyst removals observed during the second and third runs ranged from 3.2 to 3.7 logs. Removals in this range are well within the range of previously reported measurements for comparable plants. Performance of the full-scale plant would be expected to be in this range.

Rough correlations were observed between *Cryptosporidium* oocyst removals and the removals of turbidity, particles (1.5-12.5 μm), and aerobic cyst concentrations (Table 6.2). The correlations between reduction in particles and *Cryptosporidium* and between reduction in turbidity and *Cryptosporidium* were relatively good based on the very limited data of this study. This result suggests that more detailed data would likely produce correlations that would be of significant predictive value. However, with respect to the data of this study, unavoidable differences between the pilot and full-scale system would require verification at full-scale to provide reliable predictive tools.

6.4.3 Turbidity reduction

The filtered water turbidity differed significantly between the first and the next two seeded runs. During the first run unfamiliarity of the operators with the pilot system led to uncontrolled variations in flowrate during the run. This resulted in poor and

variable filtered water turbidity. The resulting data may be useful in that they provide an indication of *Cryptosporidium* removal performance under poor turbidity removal conditions. Otherwise, we believe that inferences regarding the likely capabilities of the full-scale plant should not be made based on performance during the first run.

In general the performance of the pilot filter as measured by turbidity and turbidity reduction appeared to provide a reasonably sensitive measure of treatment performance. When treatment conditions were impaired, as in the first run, during the ripening period of each run, and following the flow increase at the end of the third run, the turbidity measured in filtered water was higher. The potential relation of turbidity as a measure of performance to *Cryptosporidium* reduction will be discussed below.

The ability of the pilot filter to achieve low filtered water turbidity apparently exceeded that of the full-scale plant. To the extent that the chemical coagulation, mixing and flocculation conditions of the pilot treatment train matched those of the full-scale plant, the lower filtered water turbidity observed in the second and third runs in the pilot filter effluent compared to the full-scale plant suggests that pilot plant results may be expected to exceed performance of the full-scale plant. An alternative to the current pilot treatment scheme that might assist in making pilot and full-scale trains more nearly equivalent would be to use coagulated filter influent piped directly from the full-scale plant to the top of the pilot filters.

6.4.4 Particle removal

Overall, particle removals provided by the pilot filter appeared to have excellent characteristics. However, significant differences were observed between particle removals occurring during the three seeded runs. During the first run minimum particle concentrations reached during periods prior to a flowrate change were comparable to those reached during the other more consistent runs, eg. ca. 50/mL.

During the second run the raw water particle concentration was ca. 1000/mL higher than in the other runs, yet minimum particle concentrations reached in filtered water

were consistently lowest, ca. 25/mL. The filtered water particle concentrations during the third run were stable but at a somewhat higher level, ca. 50/ mL.

The reason for better particle removal during the second run can not be deduced from the available data. For example, coagulation conditions used in the two runs were nearly the same and performance measured by filtered water turbidity was nearly the same. Differences may likely have been due to differences in the particle assemblages during the two periods and to levels of other coagulation-related water quality parameters such as the concentration of dissolved organics (colour and humic substances).

Based solely on observations made during the three *Cryptosporidium* seeding runs, choosing between particle counts and turbidity as measures of treatment performance would be difficult. Both appeared to be sensitive measures of performance. Relations between particle concentrations, turbidity, and *Cryptosporidium* removal are discussed below.

6.4.5 Aerobic spore removal

The removal of aerobic spores differed little between the three seeded runs, with average log removals of 2.2, 2.2, and 2.0 respectively. The average for the third run is biased to the low side because the first measurement, 1.47, was taken during ripening when the filtered water turbidity was ca. 0.4 NTU, significantly earlier during ripening than the first measurement made during the other two runs.

While the log removal of aerobic spores did not differ between the three runs, within each run the log removals observed changed generally corresponding to intuitive understanding of filter performance. That is, log removals were lowest during the ripening period of each run. They were also highest at the end of each run, consistent with a gradual improvement in performance during the essentially stable operating period. During the first run in which an unplanned flowrate change was made near the end of the run resulting in increases in both turbidity and particle counts, the removal of aerobic spores decreased accordingly.

The results of this work demonstrate general presence of aerobic spores in raw water at levels sufficient to provide two to three logs of measurement sensitivity across filtration. The simplicity and low cost of the analytical procedure, and the small amount of data collected in this project on which judgements must be based all argue for additional effort to determine the potential utility of this parameter for characterising filtration performance.

6.4.6 Relationships between performance measures

The four parameters, turbidity, particle concentration, aerobic spore concentration and *Cryptosporidium* oocyst concentration were compared as alternate means of characterising filtration performance (Figure 6.16). Using simple linear correlations to describe the observed interrelations, reasonably strong relationships were apparent between turbidity and *Cryptosporidium* and between particle concentration and turbidity (Table 6.3). The lower set of correlation descriptors was calculated without the two points earliest in the ripening period of the third run. The effect on the correlation coefficient is relatively large.

The observations of this work suggest that useful relationships could be developed between *Cryptosporidium* oocyst removal and other more easily monitored parameters. Based on existing information in the literature this would appear logical and worth pursuing.

Surface charge characteristics (such as electrophoretic mobility) of *Cryptosporidium* oocysts have been reported as similar to most particles. Nothing in the literature suggests that *Cryptosporidium* oocysts should behave in unique ways that would affect their removal in filtration.

The likely existence of reasonably strong correlations between the parameters used in this work to characterise filtration performance suggests that further work planned specifically to determine the quality of such relationships and their potential utility as operational tools would be worth while.

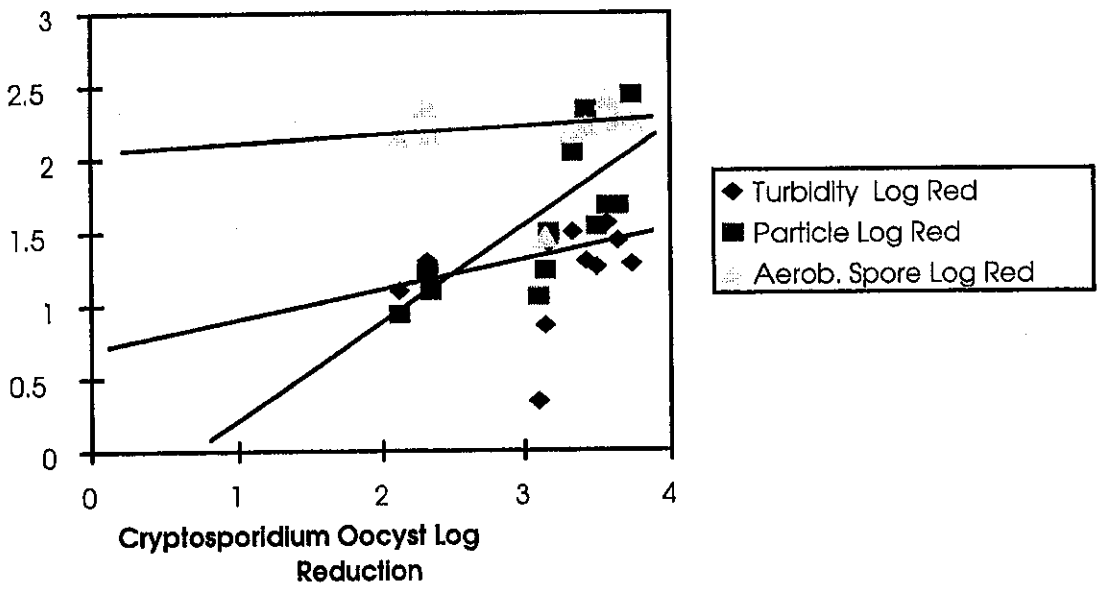


Figure 6.16. Relationships observed between log removals of turbidity and concentrations of particles and aerobic spores and the concentration of *Cryptosporidium* oocysts in three seeded runs at Macarthur pilot plant, May-June, 1998.

Table 6.3. Statistical summary of simple linear correlations between *Cryptosporidium* oocyst concentration and turbidity, particle concentration, and aerobic spore concentration. (The lower set of correlation descriptors was calculated without the two points earliest in the ripening period of the third run).

Parameter	Turbidity	Particle Concentration	Aerobic Spore Concentration
Slope	0.1738	0.6656	0.0250
Intercept	0.6698	-0.5108	2.0833
Correl Coef	0.2459	0.2876	0.0580
Slope	0.1688	0.6655	0.0249
Intercept	0.8035	-1.4787	2.083
Correl. Coef.	0.6615	0.7431	0.0579

6.4 Conclusions

A limited range of conclusions can be drawn based on the observations resulting from the preliminary pilot *Cryptosporidium* seeding runs made in this project:

1. *Cryptosporidium* oocyst seeding runs conducted using the Macarthur WFP pilot filter facility were effective in measuring the *Cryptosporidium* removal performance of the pilot filter under a limited range of operating conditions;

2. *Cryptosporidium* oocyst log reductions observed during the three seeding runs, (3.1 to 3.7-logs) were comparable to those reported previously for similar conditions;

3. The results of pilot filter performance evaluation for the removal of *Cryptosporidium* oocysts by filters of the specific water quality, coagulation, and filter design and operating characteristics of this study were entirely consistent with previously reported information on *Cryptosporidium* and filtration;

4. The data collected using the Macarthur WFP pilot filter facility during the three brief seeding runs suggest the potential existence of useful correlations between *Cryptosporidium* oocyst removal and removals measured in terms of one or more of the more easily measured conventional performance parameters, turbidity, particle concentration, and aerobic spore concentration.

7. Conclusions and Recommendations

7.1 Summary of Findings

7.1.1 Instrumentation evaluation

For routine on-line particle counting in a water treatment plant an obscuration sensor would appear to offer the most cost effective sensor type.

The issue of maintenance of floc integrity is critical to the set-up of any counter in a water treatment environment. Special care should be given to sample point placement and construction, sample tubing and joints, choice of a non-pumped flow controller, and proximity to electrical interference from high-load equipment such as backwash pumps.

Although the obscuration counter is the cost-effective option for on-line operation there exists tremendous scope in the laboratory and at the pilot scale for the application of more sophisticated equipment that provides a greater insight into the mechanisms encountered in water treatment processes.

7.1.2 Pathogen sizing

The sensors of particle counting instruments will soon be routinely calibrated with industry standards for both size and counts. Following such calibration it is essential that the sensor be assessed for pathogen sizing using a water matrix that is representative of that which will routinely pass through the sensor in a water treatment plant.

7.1.3 Jar testing

The usefulness of the Malvern instrument for investigating the coagulation-flocculation process in drinking water treatment has been demonstrated.

The importance of coagulant dose to the rate of floc growth and the subsequent relationship to floc size and the ability of the floc to entrap 5μm latex spheres are certainly areas worthy of further investigation. Recent work also suggests that the fractal nature of flocs generated from coagulant addition has a dramatic effect on their ability to capture specifically sized individual particles. Further work in this area is urgently needed.

Humic acid was found to have a significant detrimental affect on the coagulation-flocculation process and although this is not a new finding the results served to emphasise the complex nature of the affect of NOM on the coagulation process. No doubt this effect will vary from water source to water source.

7.1.4 Filtration studies

Investigations into the filtration process have served to emphasise the knowledge gaps in this area, particularly with regard to the entrapment of flocs generated through the addition of iron salts.

A major finding of the study was the lack of association between particle passage through the filter and coagulant concentration in the finished water. The traditional view of particle breakthrough at the end of a filtration run is that floc material is the major constituent of such particulate material. However, if a filter is allowed to go through to filter breakthrough before backwashing and if iron is used as the dominant coagulant, it would appear that the particulate material that breaks through is discrete particles with little or no floc attached. If time or headloss triggers for backwashing govern, breakthrough is avoided.

7.1.5 Field trials

The key findings of the field seeding trials are summarised below.

- *Cryptosporidium* oocyst seeding runs conducted using the Macarthur WFP pilot filter facility were effective in measuring the *Cryptosporidium* removal performance of the pilot filter under a limited range of operating conditions;

- *Cryptosporidium* oocyst log reductions observed during the three seeding runs, (3.1 to 3.7-logs) were comparable to those reported previously for similar conditions;

- The results of pilot filter performance evaluation for the removal of *Cryptosporidium* oocysts by filters of the specific water quality, coagulation, and filter design and operating characteristics of this study were entirely consistent with previously reported information on *Cryptosporidium* and filtration;

- The data collected using the Macarthur WFP pilot filter facility during the three brief seeding runs suggest the potential existence of useful correlations between *Cryptosporidium* oocyst removal and removals measured in terms of one or more of the more easily measured conventional performance parameters, turbidity, particle concentration, and aerobic spore concentration.

7.2 Recommendations

The virtual absence until recently of particle counters in Australian water treatment plants and the absence of guidelines for interpretation of particle counts in Australia has resulted in a lack of knowledge with regard to the day-to-day operation, maintenance and quality assurance procedures for particle counter operation. It is thus recommended that steps be taken to encourage the wider use of particle counters in water treatment and to acquire a significantly larger data base of information on particle counts in treated water (enabling comparison with the more traditional measures of treated water quality such as turbidity).

The proposed 3-log reduction rule that has been suggested in the US is clearly invalid for water treatment plants in Sydney where the raw water particle counts are in the vicinity of 3,000 to 5,000 for the majority of the year. A 3-log reduction would require the filtered water to be about 3 to 5 counts/mL, such quality being better than distilled water and comparable to reagent grade water. As such a guideline value based on a specific particle counts/mL in the finished water would appear to be more achievable and manageable. Such a recommendation is consistent with views recently reported elsewhere (Hargesheimer et al., 1998) that log removal of particles is not a good indicator of plant performance because it is influenced more by particle counts in source water than by those in finished water.

A variety of studies on the effectiveness of specific particle capture by flocs generated in the coagulation process have been undertaken within this project. The Malvern Mastersizer particle sizing instrument has been used extensively in these studies and was found to produce results of direct application to the water treatment process. As expected from standard Smoluchowski particle-particle interaction kinetics analysis, large flocs provide better entrapment of individual particles with flocs of mean size greater than approximately 100 μm apparently necessary to ensure reasonable capture of individual particles. Further insight into the ability of flocs to capture individual particles will require consideration of the fractal nature of these flocs. Recent work suggests that entrapment of individual particles by porous flocs exhibiting fractal characteristics may be significantly less efficient than previously thought. Considerable additional work in the area of particle entrapment by fractal aggregates is urgently required, particularly extending the work from model systems (latex aggregates) to those typically used in water treatment (iron and aluminium oxides) and further examining the effects of surface charge, pH and organic adsorbents on particle capture by fractal aggregates.

Preliminary consideration within this project has also been given to particle capture within the deep bed filter and has highlighted the possible importance of floc breakup within the deep bed as a mechanism for release of specific particles (such as those of *Cryptosporidium*) and a reduced likelihood of capture within the filter. Such a mechanism of particle release appears to be particularly problematic for flocs formed

from iron salts which are recognised to be generally larger and weaker than their alum-induced counterparts. Further studies are required to ascertain the significance of floc strength in maintaining contaminant particles within the filter bed.

Extension of the above issues, related to the coagulation and filtration processes in water treatment, to the development of an accurate description of the principal factors (under given raw water quality and process operating regimes) responsible for lack of capture of specific particles (such as *Giardia* and *Cryptosporidium* oocysts) is also urgently needed.

Recommendations that follow logically from the results of the field seeding trials include the following:

- The generally interpretable results of the introductory pilot seeding study indicate that further work using the basic elements of approach and methodology as used here would produce similarly effective results;

- The likely existence of correlations between *Cryptosporidium* removal and other more easily monitored parameters such as turbidity, particle concentration, and aerobic spore concentration suggested by the results of this project indicate the desirability to further develop such correlations for application to treatment process monitoring and control;

- The effective results of this introductory pilot seeding study strongly suggest the desirability of further describing the *Cryptosporidium* removal performance of SWC filtration facilities to identify potential problems and most effective operating strategies.

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