"Star Glory Technologies Ltd."

04/11/2004 18:25

To: "Environment, Transport and Works Bureau" <hats@etwb.gov.hk>
cc:
Subject: HATS

For the attention of **Dr Sarah Liao (Secretary for the Environment,Transport and Works)**

Dear Dr. Liao,

I understand that you are using sodium hypochloride as the disinfectant for the treated sewage water before discharging into the sea. I wonder if you have ever consider the ecological consequence of the by-products after adding the this kind of chemical.

According to numerous reports of World Health Organisation, chlorine in the disinfectant will combine with ammonia and other organic substances richly consisted in the effluent water to form some toxic and carcinogenic chemicals called Trihalomethanes or THM.

THM will be formed abundantly in those disinfected effluent water and then discharged into the harbour or other waters. In turn, some sea creatures will swallow those chemicals.

If those creatures are eaten by human beings, the chances of intaking THM will then be higher. A harmful food-chain will then be formed.

Moreover, chlorine is quite unstable in non-neutral pH condition and under UV.

Therefore, I plead for your attention on the choice of final disinfectant for the discharge effluent water.

One of the best alternative I would recommend is chlorine dioxide.

Chlorine dioxide

Chlorine dioxide is, together with chlorite and chlorate, one of the best-quality chlorine-oxygen compounds. Especially because of their oxidizing and very little chlorinating effect they are the most effective and, at the same time, environment-friendly disinfectants. They are even suitable to treat organically polluted waste water. Chlorite and chlorate are especially important as starting substances for the production of chlorine dioxide.

Chlorine dioxide (ClO_2) is a very strong oxidant. This yellow-orangeish toxic gas has its melting-point at 59° C and its boiling-point at 9,7° C. Its MAK-value is of 0,1 ppm.

Chlorine dioxide has very high water solubility, at 4° C 20 parts ClO_2 dissolve in 1 part H_2O. Chlorine dioxide is commonly used as a dilute solution in order to ward off great danger of explosion.
General advantages:

- extreme efficacy (up to 250 % higher oxidation as e.g. chlorine)

- effective removal and prophylaxis of biofilms
- constant bactericidal effectiveness at a wide pH range (4-10)
- Highly effective against all microorganisms living in water (bacteria, viruses, protozoa, fungi and yeast)
- no resistance of microorganisms
- low corrosion rates
- highly effective in the removal of iron and magnesium compounds
- very good water solubility and low reaction time

Biocidal effectiveness:

Chlorine dioxide belongs to the oxidising biocides. It is not a metabolic toxic substance, i.e, it destroys the microorganisms by means of interrupting the food transport along the cell walls and not by interrupting the metabolic process (= metabolism).

Danger potential of the chlorine dioxide produced in the traditional way:

Chlorine dioxide is an unstable radical compound which is highly explosive, both in its pure form and in air concentrations down to 10 Vol. %. It disintegrates into chlorine and oxygen under the effect of light and at high temperatures under the release of 102.6 kJ/mol. Therefore, the traditional way of producing chlorine dioxide requires the use of expensive technical equipment and has a high risk potential. These two main burdens have as yet impaired the spread of this substance as a universally applicable disinfectant, especially for small and middle applications.

Chlorine dioxide instead of chlorine

Since the 1950s chlorine has increasingly been replaced by chlorine dioxide for the disinfection of drinking water. Even though free chlorine is also highly effective as a disinfectant, it reacts with water ingredients, like humus acid, bath oils, cleaning additives, algae and nitrogen compounds like urine, ammonia, etc. As a result chlorinated substitution products come about, especially chloramines (bound chlorine) and trihalogenmethanes (THMs). The latter, THMs, are said to have a carcinogenic effect, and the former, chloramines, irritate the mucous membrane, bring about the lost of the skin's natural oil and are responsible for the unpleasant swimming pool smell.

Reduction of bound chlorine:

Due to its high oxidation potential chlorine dioxide can have a negative oxidising effect on bound chlorine according to the following equation:

$$2\ NH_2Cl\ \text{(bound chlorine)} + 2\ ClO_2 \rightarrow N_2 + 4\ H^+ + 2\ Cl^- + 2\ ClO_2^-$$

The resulting chlorite oxidises anew into chlorine dioxide and chlorate by means of the existing chlorine. Bound chlorine is converted into nitrogen and sodium chloride. By continuously adding chlorine dioxide over a longer period of time a chlorine dioxide buffer able to remove the bound chlorine comes about.

Reduction of THMs :

Should trihalogenmethanes once come about, they cannot be removed oxidatively neither with chlorine dioxide nor by point of refraction chlorination.
The use of chlorine dioxide aims at suppressing the formation of haloforms , so that the THMs do not even come about- or at least in very little concentrations. Through the reaction of chlorine dioxide with the water ingredients (see above) these change to such a degree

that their common reaction with free chlorine into THMs is no longer possible.

You are welcome to contact me for obtaining more technical information as our company can supply site mixed chlorine dioxide from Germany without using the chemical plants with expensive capital and running cost.

Should you require any further information, please do not hesitate to contact us.

Regards

PAUL CHAN
Marketing Manager

Water Treatment and Pathogen Control

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WHO Drinking Water Quality Series

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Water Treatment and Pathogen Control

Process Efficiency in Achieving Safe Drinking Water

Mark W LeChevallier and Kwok-Keung Au

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Foreword

Microbial contamination of drinking-water contributes to disease outbreaks and background rates of disease in developed and developing countries worldwide. Control of waterborne disease is an important element of public health policy and an objective of water suppliers.

The World Health Organization (WHO) has developed *Guidelines for Drinking-water Quality*. These guidelines, which are now in their third edition (WHO, 2004), provide an internationally harmonized basis to help countries to develop standards, regulations and norms that are appropriate to national and local circumstances. The latest edition of the WHO *Guidelines for Drinking-water Quality* is structured around an overall "water safety framework", used to develop supply-specific "water safety plans". The framework, which focuses on health protection and preventive management from catchment to consumer, has five key components:

- health-based targets, based on an evaluation of health concerns;
- system assessment to determine whether the drinking-water supply (from source through treatment to the point of consumption) as a whole can deliver water of a quality that meets the health-based targets;

- operational monitoring of the control measures in the drinking-water supply that are of particular importance in securing drinking-water safety;
- management plans that document the system assessment and monitoring plans, and describe actions to be taken in normal operation and incident conditions (including upgrade and improvement, and documentation and communication);
- a system of independent surveillance to verify that the above are operating properly.

Understanding the effectiveness of water treatment is necessary for:

- design of cost-effective interventions
- review of the adequacy of existing structures
- operation of facilities to maximum benefit.

WHO has also developed a series of expert reviews covering various aspects of microbial water quality and health (listed below). This publication forms part of this series of reviews.

- *Managing Water in the Home: Accelerated Health Gains from Improved Water Supply* (M Sobsey, 2002)
- *Pathogenic Mycobacteria in Water: A Guide to Public Health Consequences, Monitoring and Management* (S Pedley et al, eds, 2004)
- *Quantifying Public Health Risk in the WHO Guidelines for Drinking-water Quality: A Burden of Disease Approach* (AH Havelaar and JM Melse, 2003)
- *Safe, Piped Water: Managing Microbial Water Quality in Piped Distribution Systems* (R Ainsworth, 2004)
- *Toxic Cyanobacteria in Water: A Guide to their Public Health Consequences, Monitoring and Management* (I Chorus and J Bartram, eds, 1999)
- *Upgrading Water Treatment Plants* (EG Wagner and RG Pinheiro, 2001)
- *Water Safety Plans* (A Davison et al., 2004)
- *Assessing Microbial Safety of Drinking Water: Improving Apporoaches and Methods* (A Dufour et al., 2003).

Further texts are in preparation or in revision:

- *Arsenic in Drinking-water* (in preparation)
- *Fluoride in Drinking-water* (in preparation)
- *Guide to Hygiene and Sanitation in Aviation* (in revision)
- *Guide to Ship Sanitation* (in revision)
- *Health Aspects of Plumbing* (in preparation)

- *Legionella and the Prevention of Legionellosis* (in preparation)
- *Protecting Groundwaters for Health — Managing the Quality of Drinking-water Sources* (in preparation)
- *Protecting Surface Waters for Health — Managing the Quality of Drinking-water Sources* (in preparation)
- *Rapid Assessment of Drinking-water Quality: A Handbook for Implementation* (in preparation)
- *Safe Drinking-water for Travellers and Emergencies* (in preparation)

Water safety management demands a quantitiative understanding of how processes and actions affect water quality, which in turn requires an understanding of risk assessment. This volume is intended to provide guidance on using risk assessment when selecting appropriate treatment processes, to ensure the production of high quality drinking-water. It is hoped that it will be useful to water utilities, water quality specialists and design engineers.

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This text is one of the supporting documents to the rolling revision of the *WHO Guidelines on Drinking-water Quality*. Its preparation was overseen by the working group on microbial aspects of the guidelines, and thanks are also due to its members:

- Ms T Boonyakarnkul, Department of Health, Thailand (*Surveillance and control*)
- Dr D Cunliffe, SA Department of Human Services, Australia (*Public health*)

- Prof W Grabow, University of Pretoria, South Africa (*Pathogen-specific information*)
- Dr A Havelaar, RIVM, The Netherlands (Working Group Coordinator; *Risk assessment*)
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Acronyms and abbreviations used in the text

AOC	assimilable organic carbon
asu	areal standard unit
AWWA	American Water Works Association
AWWARF	AWWA Research Foundation
BDL	below detection limit
BDOC	biodegradable dissolved organic carbon
CC-PCR	cell culture-polymerase chain reaction
cfu	colony forming unit
DAF	dissolved air flotation
DE	diatomaceous earth
DNA	deoxyribonucleic acid
FAC	free available chlorine
FMEA	failure mode and effects analysis
HACCP	hazard analysis critical control point
HPC	heterotrophic plate count
IDDF	integrated disinfection design framework
IFA	immunofluorescence assay
MF	microfiltration

NA	not applicable
NF	nanofiltration
NR	not reported
NTU	nephelometric turbidity unit
PACl	polyaluminium chloride
pfu	plaque forming unit
PVC	polyvinylchloride
RO	reverse osmosis
RNA	ribonucleic acid
SFBW	spent filter backwash
THM	trihalomethane
UF	ultrafiltration
USEPA	United States Environmental Protection Agency
UV	ultraviolet
WHO	World Health Organization
WTP	water treatment plant

Executive summary

This document is part of a series of expert reviews on different aspects of microbial water quality and health, developed by the World Health Organization (WHO) to inform development of guidelines for drinking-water quality, and to help countries and suppliers to develop and implement effective water safety plans.

Contamination of drinking-water by microbial pathogens can cause disease outbreaks and contribute to background rates of disease. There are many treatment options for eliminating pathogens from drinking-water. Finding the right solution for a particular supply involves choosing from a range of processes. This document is a critical review of some of the literature on removal and inactivation of pathogenic microbes in water. The aim is to provide water quality specialists and design engineers with guidance on selecting appropriate treatment processes, to ensure the production of high quality drinking-water. Specifically, the document provides information on choosing appropriate treatment in relation to raw water quality, estimating pathogen concentrations in drinking-water, assessing the ability of treatment processes to achieve health-based water safety targets and identifying control measures in process operation.

Processes for removal of microbes from water include pretreatment; coagulation, flocculation and sedimentation; and filtration. Pretreatment can broadly be defined as any process to modify microbial water quality before, or at the entry to, the treatment plant. Pretreatment processes include application of roughing filters, microstrainers, off-stream storage and bank infiltration, each with a particular function and water quality benefit. Applications of these pretreatment processes include removal of algal cells, high levels of turbidity, viruses and protozoan cysts.

For conventional treatment processes, chemical coagulation is critical for effective removal of microbial pathogens. Together, coagulation, flocculation and sedimentation can result in 1–2 log removals of bacteria, viruses and protozoa. For waters with high levels of algae, care must be taken to remove these organisms without disrupting the cells, which may release liver or nerve toxins. High-rate clarification using solids contact clarification, ballasted-floc, or contact clarification systems can be as, or more, effective than conventional basins for removal of microbes. Dissolved air flotation can be particularly effective for removal of algal cells and *Cryptosporidium* oocysts. Lime softening can provide good microbial treatment through a combination of inactivation by high pH and removal by sedimentation.

Granular media filtration is widely used in drinking-water treatment. It removes microbes through a combination of physical–hydrodynamic properties and surface and solution chemistry. Under optimal conditions, the combination of coagulation, flocculation, sedimentation and granular media filtration can result in 4-log or better removal of protozoan pathogens. However, without proper chemical pretreatment, this type of rapid rate filtration works as a simple strainer and is not an effective barrier to microbial pathogens. Slow sand filtration works through a combination of biological and physical–chemical interactions. The biological layer of the filter, termed *schmutzdecke*, is important for effective removal of microbial pathogens. Precoat filtration was initially developed as a portable unit to remove *Entamoeba histolytica*, a protozoan parasite. In this process, water is forced under pressure or by vacuum through a uniformly thin layer of filtering material, typically diatomaceous earth. As with granular media filtration, proper chemical conditioning of the water improves the treatment efficiency of precoat filtration. In contrast, membrane filtration removes microbial pathogens primarily by size exclusion (without the need for coagulation), and is effective in removing microbes larger than the membrane pore size.

Oxidants may be added to water for a variety of purposes, such as control of taste and odour compounds, removal of iron and manganese, control of zebra mussel and removal of particles. For microbial pathogens, application of strong oxidizing compounds such as chlorine, chlorine dioxide or ozone will act as

disinfectants, inactivating microbial cells through a variety of chemical pathways. Principal factors that influence inactivation efficiency of these agents are the disinfectant concentration, contact time, temperature and pH. In applying disinfectants, it is important to take into account data on CT (disinfectant concentration multiplied by the contact time) for the specific disinfectant. Ultraviolet light (UV) inactivates microorganisms through reactions with microbial nucleic acids and is particularly effective for control of *Cryptosporidium*.

For control of microbes within the distribution system, disinfectants must interact with bacteria growing in pipeline biofilms or contaminating the system. The mechanism of disinfection within the distribution system differs from that of primary treatment. Factors important in secondary disinfection include disinfectant stability and transport into biofilms, disinfectant type and residual, pipe material, corrosion and other engineering and operational parameters.

Performance models can help in understanding and predicting the effectiveness of granular media filtration processes for removal of particles and microbes. Similarly, equations can be useful in predicting microbial inactivation by disinfectants. It is also useful to consider variability in processes and in measurements to determine the overall effectiveness of treatment to control microbial risk. At present, performance models cannot precisely define microbial treatment effectiveness. This leads the operator back to the monitoring and control of critical points within the treatment process. The combined effect of these control measures ensures that the microbial water quality of the treated water meets or surpasses risk goals for the potable water supply.

A water safety plan combines elements of a "hazard analysis and critical control point" (HACCP) approach, quality managment and the "multiple barriers" principle, to provide a preventive management approach specifically developed for drinking-water supply. It can provide a framework for evaluating microbial control measures by helping to focus attention on process steps such as coagulation, filtration and disinfection, which are important for ensuring the microbial safety of water. Many current practices already employ some elements of a water safety plan, and this type of approach is likely to become more clearly defined in water treatment practices in the future.

1

Introduction

1.1 PURPOSE AND SCOPE

This publication is a critical review of removal and inactivation of microbial pathogens by drinking-water treatment processes. Chapters 2 and 3 focus on removal and inactivation processes respectively, in terms of their operational principles, mechanisms and efficiency. Chapter 4 presents performance models for granular filtration and disinfection, two of the most important barriers for microbes, and illustrates how these models can be used to determine the effects of process variables on treatment efficiency. Chapter 5 looks at measures of process variation, including uncertainty in treatment effects and problems associated with the use of surrogates. Finally, Chapter 6 illustrates how an approach based on a water safety framework can be used to minimize microbial hazards in water.

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The review focuses on bacteria, viruses, protozoan parasites and microbial toxins, and their removal from source water by various treatment processes. The aim is help water utilities to:

- choose appropriate treatment in relation to raw water quality
- estimate pathogen concentrations in drinking-water
- assess the ability of treatment processes to achieve health-based water safety targets
- identify control measures in process operation.

This review does not attempt to cite all the relevant literature; rather, it highlights information that illustrates the performance of each treatment process. Where possible, it provides quantitative information on the removal or inactivation of pathogenic microorganisms and toxins. Also, it considers (and, where possible, quantifies) interactions between the effects of different treatment processes.

The information is given at different levels of detail:

- The first level estimates the order of magnitude of the expected effect under typical process conditions and proper operating conditions. This level of detail allows simple decision trees for the choice of a treatment chain to be constructed.
- The second level identifies the process parameters (both design and monitoring) that are most relevant to the treatment effect, and quantifies the effect of these parameters. Where possible, mathematical models are used to describe these relations. This level of detail allows control measures and their operational limits to be identified. There is an emphasis on physical and chemical parameters; microbiological indicators are discussed in a separate review (Dufour et al., 2003).
- The third level identifies and quantifies any variability and uncertainty in the treatment effect that is not explained by the process parameters. This level of detail allows exposure to pathogens to be assessed within the framework of a formal risk assessment procedure.

1.2 MULTIPLE BARRIERS

For centuries, the process of providing safe drinking-water has relied on the application of the 'multiple barrier concept'. Hippocrates (460–354 B.C.), writes in Air, Water and Places — the first treatise on public hygiene, that 'qualities of the waters differ from one another in taste and weight'. One should 'consider the waters which the inhabitants use, whether they be marshy and soft, or hard and running from elevated and rocky situations, and then if saltish and unfit for cooking .... for water contributes much to health' (Baker, 1948).

The concept of multiple barriers for water treatment is the cornerstone of safe drinking-water production. The barriers are selected so that the removal capabilities of different steps in the treatment process are duplicated. This approach provides sufficient backup to allow continuous operation in the face of normal fluctuations in performance, which will typically include periods of ineffectiveness. Having multiple barriers means that a failure of one barrier can be compensated for by effective operation of the remaining barriers, minimizing the likelihood that contaminants will pass through the treatment system and harm consumers. Traditionally, the barriers have included:

- protection of source water (water used for drinking-water should originate from the highest quality source possible);
- coagulation, flocculation and sedimentation;
- filtration;
- disinfection;
- protection of the distribution system.

If these conventional barriers are thought to be inadequate, it may be advisable to consider adding multiple stages of filtration or disinfection.

The benefit of multiple treatment barriers is illustrated by a recent epidemiological study of a karstic groundwater system where one well was filtered and chlorinated while a second was only chlorinated (Beaudeau et al., 1999). Increases in sales of antidiarrheal drugs correlated strongly with lapses in chlorination of the well that had disinfection as the only treatment. In contrast, no effect could be traced to lapses in chlorination of the filtered well. The combination of filtration and chlorination appeared to provide sufficient duplication in removal of contaminants that temporary lapses in disinfection did not generate a measurable adverse outcome (Beaudeau et al., 1999).

1.3 PROCESS CONTROL MEASURES

There are many different microbes that may be of concern in source waters or within the distribution system. Developing a monitoring scheme for each would be an impossible task; therefore, another approach is needed. The food and beverage industry has used the "hazard analysis critical control point" (HACCP) approach to determine the key points within the manufacturing chain where contamination can be measured and prevented. A similar concept can be used by water utilities, to prioritize the key contamination points within the treatment and distribution system (Bryan, 1993; Sobsey et al., 1993). This approach allows utilities to focus their resources on monitoring these points and correcting any deviations from acceptable limits. The latest edition of the World Health Organization (WHO) *Guidelines for Drinking-Water Quality* (WHO,

2004) incorporates such an approach, providing guidance on the development of a water safety plan. The plan is developed using a water safety framework, which combines HACCP principles with water quality management and the multiple barrier concept.

Most microbiological monitoring programs for drinking-water have not been designed using such a framework. However, many of the relevant concepts are found in the overall process control of water treatment plants and distribution systems. For example, maintaining a disinfectant residual within the distribution system can be considered a control procedure.

The water safety framework is not only applicable to microbial monitoring of drinking-water treatment; it can also be applied to aspects such as turbidity, disinfectant residuals, pressure and particle counts. A strength of the framework is that it allows water utilities to allocate limited laboratory resources to monitoring points within the water supply process where the results will provide the greatest information and benefit.

2

Removal processes

This chapter considers various processes for removal of microbes from water. In particular, it discusses:

- *pretreatment* — broadly defined as any process to modify microbial water quality before, or at the entry to, a treatment plant;
- *coagulation, flocculation and sedimentation* — by which small particles interact to form larger particles and settle out by gravity;
- *ion exchange* — used for removal of calcium, magnesium and some radionuclides;
- *granular filtration* — in which water passes through a bed of granular materials after coagulation pretreatment;
- *slow sand filtration* — in which water is passed slowly through a sand filter by gravity, without the use of coagulation pretreatment.

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2.1 PRETREATMENT

This section describes some of the processes that can be used in pretreatment of water (roughing filters, microstrainers, off-stream storage and bank infiltration), each of which has a particular function and water quality benefit. Applications of pretreatment include removal of algal cells, high levels of turbidity, viruses and protozoan cysts The various options for pretreatment may be compatible with a variety of treatment processes, ranging in complexity from simple disinfection to membrane filtration.

2.1.1 Roughing filters

A roughing filter is a coarse media (typically rock or gravel) filter used to reduce turbidity levels before processes such as slow sand filtration, diatomaceous earth (DE) or membrane filtration. The American Water Works Association Research Foundation (AWWARF) has reviewed design variables for roughing filters (Collins et al., 1994). Such filters typically have a filter box divided into multiple sections containing gravel beds of decreasing particle size, inlet and outlet structures, and flow-control devices. Examples of common configurations are shown in Figure 2.1.

Roughing filters have achieved peak turbidity removals ranging from 60 to 90%; generally, the more turbid the water initially, the greater the reduction that can be achieved (Galvis, Fernandez & Visscher, 1993; Collins et al., 1994; Ahsan, Alaerts & Buiteman, 1996). These filters can achieve similar reductions of coliform bacteria. Pilot studies of various roughing filter configurations (horizontal-flow, up-flow and down-flow) reduced faecal coliform bacteria by 93–99.5% (Galvis, Fernandez & Visscher, 1993). These filters were also combined with a dynamic roughing filter (which contains a thin layer of fine gravel on top of a shallow bed of coarse gravel, with a system of underdrains) to pretreat high turbidity events, and achieved faecal coliform removal of 86.3%. When followed by slow sand filtration, the removal reached 99.8%, with an overall combined treatment efficiency of 4.9–5.5 log units. In a five-month pilot study of a medium gravel (5.5 mm) horizontal roughing filter in Texas City, United States of America (USA), the filter removed on average 47% of total bacteria (as measured by epifluorescence microscopy), 37% of the source water algal cells and 53% of the total chlorophyll (Collins et al., 1994). The researchers found that the roughing filters removed clay particles more effectively when the filter was ripened with algal cells. Addition of alum coagulant before treatment with a horizontal roughing filter improved the filter's performance for turbidity, colour, organic carbon, head loss and filter run length (Ahsan, Alaerts & Buiteman, 1996).

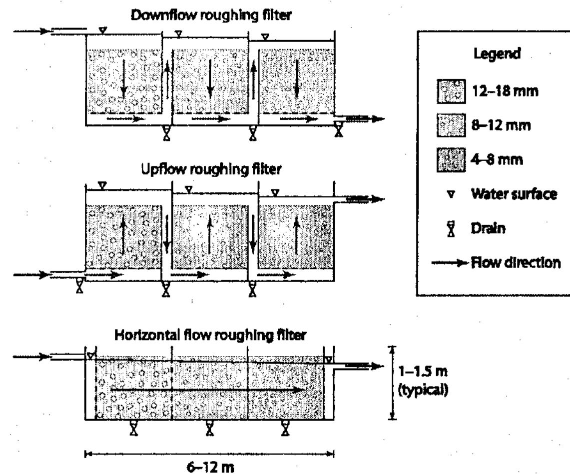


Figure 2.1 Typical roughing filter configurations (Collins et al., 1994)

2.1.2 Microstrainers

Microstrainers are fabric meshes woven of stainless steel or polyester wires, with apertures ranging from 15 to 45 µm (usually 30–35 µm). Such meshes are useful for removing algal cells and large protozoa (e.g. *Balantidium coli*), but have no significant impact on bacteria or viruses. Microstrainers generally remove about 40–70% of algae and, at the same time, about 5–20% of turbidity (Mouchel & Bonnelye, 1998). The performance of microstrainers for specific applications varies, depending on the type of algae present, as summarized in Table 2.1. Although microstrainers can reduce the amount of coagulant needed, they do not remove smaller species or reproductive forms of algae.

Table 2.1 Performance of microstrainers for various algae

Organism	Type	Percentage removal
Diatoms		
Cyclotella	Unicellular	10–70
Stephanodiscus	Unicellular	10–60
Melosira	Filamentous	80–90
Synedra	Unicellular	40–90
Asterionella	Colonial	75–100
Fragilaria	Filamentous	85–100
Chlorophyceae		
Chlorella	Unicellular	10–50
Scenedesmus	Cenobia (4–8 cells)	15–60
Pediastrum	Cenobia (4–64 cells)	80–95
Blue-green algae		
Oscillatoria	Filamentous	40–50
Anabaena	Filamentous	50–70

Adapted from Mouchet & Bonnelye (1998)

2.1.3. Off-stream storage

In this discussion, off-stream storage refers to a storage reservoir that directly or indirectly feeds a potable water intake. The effects of off-stream storage are difficult to generalize because important physical, biological and chemical processes are influenced by hydrological and limnological characteristics of the reservoir. For example, 'round' reservoirs and lowland impoundments influenced by strong winds can be represented as homogeneous biotypes because they are mixed effectively. On the other hand, long reservoirs whose depth increases with length are best represented as a series of interconnected individual basins (Bernhardt, 1995). The characteristics of reservoirs created by construction of a dam will differ from those of a natural or artificial lake.

Oskam (1995) summarized the self-purification processes that improve water quality in off-stream reservoirs (Table 2.2). The major factors that influence these processes are the degree of compartmentalization, the hydraulic residence time, the shape and flow through the reservoir, and the quality of the source water. Certain processes can also degrade water quality; for example, poorer quality of the impounded water can result from failure to:

- manage algal growth;
- control influx of nitrogen, phosphorus or other contaminants;
- limit faecal contamination from run-off of surrounding areas or roosting birds.

Table 2.2 Self-purification processes that improve off-stream reservoir water quality

Type of process	Effects
Physical	Equalization of peak concentrations (e.g. chemicals, microbes) Exchange of oxygen and carbon dioxide with the atmosphere Evaporation of volatile substances (e.g. solvents) Settling of suspended solids and adsorbed substances (e.g. turbidity, heavy metals)
Biological	Biodegradation of organic substances Die-off of faecal bacteria and viruses Nitrification of ammonium to nitrate Denitrification of nitrate to nitrogen Phosphorus elimination by phytoplankton uptake (in pre-reservoirs)
Chemical	Oxidation of divalent iron and manganese Hydrolysis of polyphosphates and organic esters (e.g. phthalates) Photolysis of humic substances and polynuclear aromatic hydrocarbons

Adapted from Oskam (1995)

In a study by Bernhardt (1995), coliform bacteria in dammed reservoirs were reduced by 80–99% when residence times were greater than 40 days, and allochthonous bacteria were reduced by 90–99% when retention times exceeded about 100 days. Kors & Bosch (1995) reported reductions of enteroviruses (1.5 logs), Kjeldahl nitrogen (50%), total phosphorus (60%) and ammonium (70%) for a pumped, off-stream reservoir after about 100 days retention time. Stewart et al. (1997) examined storm events that washed high levels of *Giardia* cysts (up to 17 000 cysts/100 l) and *Cryptosporidium* oocysts (up to 42 000 oocysts/100 l) into receiving reservoirs. Only one of 29 reservoir effluent samples was positive, suggesting that the cysts and oocysts had become trapped in sediments that settled to the bottom of the reservoir, because unattached organisms settle slowly (Medema et al., 1998). Hawkins et al. (2000) reported complete elimination of *Cryptosporidium* spikes (i.e. high concentrations) within three weeks in the 2 million megalitre Lake Burragorang reservoir that provides source water for Sydney (Australia). The authors calculated a settling rate of 5–10 metres/day and postulated that sedimentation was accelerated by oocysts clumping with other suspended particles. In a study of three reservoirs in Biesbosch (Netherlands), storage with long residence times (average 24 weeks) resulted in reductions of 2.3 logs for *Giardia*, 1.4–1.9 logs for *Cryptosporidium*, 2.2 logs for *Escherichia coli* and 1.7 logs for faecal streptococci (Ketelaars et al., 1995; van Breemen & Waals, 1998).

The die-off kinetics for microbes can be modelled as a first-order reaction dependent on the residence time and short-circuiting (i.e. the decrease in hydraulic

residence time in a vessel) (Oskam, 1995). For relatively rapid removal rates (k-values > 0.05/day), the degree of compartmentalization has a positive effect on water quality. Therefore, a series of three or four smaller reservoirs would be better than one large impoundment. With estimated k-values of 0.07/day for removal of *Giardia* and *Cryptosporidium*, and 0.13/day for enteric viruses, compartmentalization in three or four reservoirs would increase the removal effect to 15–230 times that achieved by a single basin (Oskam, 1995).

For reservoirs with short retention times (and therefore limited self-purification), the raw water pumping schedule can be used to improve water quality, by avoiding periods of source water contamination. For example, in a study of the Delaware River (USA), peak levels of microbial contaminants were associated with high levels of turbidity following rainfall events (LeChevallier et al., 1998). By operating the source water pumps to avoid these peak events, levels of *Giardia* and *Cryptosporidium* 12–16 times higher than normal were avoided.

2.1.4 Bank infiltration

Bank infiltration refers to the process of surface water seeping from the bank or bed of a river or lake to the production wells of a water treatment plant. During the water's passage through the ground, its quality changes due to microbial, chemical and physical processes, and due to mixing with groundwater. The process can also be described as 'induced infiltration,' because the well-field pumping lowers the water table, causing surface water to flow into the aquifer under a hydraulic gradient. Bank infiltration can be accomplished through natural seepage into receiving ponds, shallow vertical or horizontal wells placed in alluvial sand and gravel deposits adjacent to surface waters, and infiltration galleries.

Bank infiltration has been widely used in European countries and is of increased interest in many other countries. Variations on the underground passage concept include soil aquifer treatment, injection of surface water for underground passage and aquifer recharge.

The advantages of bank infiltration are summarized in Table 2.3. The efficiency of the process depends on a number of factors: the quality of the surface water (turbidity, dissolved organic matter, oxygen, ammonia and nutrients), the composition and porosity of the soil, the residence time of the water in the soil and the temperature. This efficiency can vary over time, depending on the difference in level between the source water (e.g. river stage) and groundwater. This difference can influence the degree of groundwater mixing and the residence time of the infiltrated surface water.

Table 2.3 Advantages of bank infiltration

A natural pretreatment step requiring little chemical addition
Reduced turbidity and particles
Removal of biodegradable compounds
Reduction of natural organic matter and less formation of disinfection by-products
Reduction of bacteria, viruses and protozoa
Equalization of concentration peaks (e.g. moderation of spills, temperature, etc.)
Dilution with groundwater

Adapted from Kuhn (1999)

Concern about groundwater under the direct influence of surface water has caused some confusion about how to regard bank infiltration. Clearly, this process is under the direct influence of surface water; however, in the USA, the Surface Water Treatment Rule (USEPA, 1989a) does not consider the infiltration process as contributing to water treatment.

In a study of the Grand River in Ontario (Canada), removal of algae and diatoms ranged from 4.8 to 7.2 logs when the quality of the collection well was compared to the raw water (Clancy & Stendahl, 1997). No *Giardia* or *Cryptosporidium* were detected in the collector wells, although these protozoa were frequently detected in the river water. Figure 2.2 shows the relationship between the concentration of algae and the theoretical flow-path distance for wells along the Great Miami River at Cincinnati (USA), with approximately 1 log reduction for every 8.5 m (28 ft) of separation from the source water (Gollnitz, Cossins & DeMarco, 1997). Schijven and Rietveld (1997) measured the removal of male-specific coliphage, enteroviruses and reoviruses at three infiltration sites, and compared the measured values to those predicted by a virus transport model. They found a 3.1-log reduction of bacteriophage within 2 m (6.6 ft) and a 4.0-log reduction within 4 m (13.2 ft) of very fine dune sand. Phage levels were reduced by 6.2 logs through riverbank infiltration over 30 m (98 ft) of sandy soil. In all cases, enteroviruses and reoviruses were eliminated to below detection limits (> 2.6 to > 4.8 log removals). The virus transport model corresponded reasonably well with the measured results, producing calculated removals ranging from 2.5 to 15 logs.

In studies being conducted by the American Water Works Service Company and the Johns Hopkins University, monitoring of three river bank infiltration systems along the Wabash, Ohio and Missouri rivers (USA) have shown complete removal of *Clostridium* and bacteriophage indicators (Table 2.4) and substantial reductions in biodegradable dissolved organic carbon (BDOC) and assimilable organic carbon (AOC), which can stimulate bacterial growth in distribution system pipelines (Ainsworth, 2004). These data indicate that bank infiltration can be highly effective for removal of microbial contaminants.

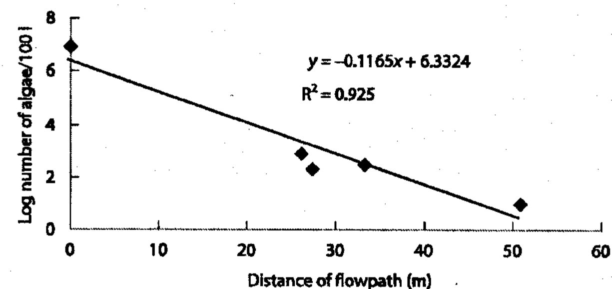


Figure 2.2 Relationship between algae concentration and theoretical flowpath. Adapted from Gollnitz, Cossins & DeMarco (1997)

2.2 COAGULATION, FLOCCULATION AND SEDIMENTATION

Coagulation, flocculation and sedimentation are used in conjunction with subsequent filtration. These processes are summarized below.

- *Coagulation* promotes the interaction of small particles to form larger particles. In practice, the term refers to coagulant addition (i.e. addition of a substance that will form the hydrolysis products that cause coagulation), particle destabilization and interparticle collisions.
- *Flocculation* is the physical process of producing interparticle contacts that lead to the formation of large particles.
- *Sedimentation* is a solid–liquid separation process, in which particles settle under the force of gravity.

Excellent reviews of these processes are available (Gregory, Zabel & Edzwald, 1999; Letterman, Amirtharajah & O'Melia, 1999). With respect to coagulation and flocculation, most bacteria and protozoa can be considered as particles, and most viruses as colloidal organic particles.

Table 2.4 Effects of bank infiltration

Sample	Distance from river (m)	BDOC (mg/l)	Total AOC (µg/l)	Clostridium cfu/100 ml	Bacteriophage Somatic pfu/100 ml	Male-specific pfu/100 ml
Site — Terre Haute						
Wabash River	–	1.35	193	253	129	12
Collector	21–27	0.14	23	0.06	< 0.13	< 0.13
Well #3*	122	0.07	15	< 0.13	< 0.13	< 0.13
Site — Jeffersonville						
Ohio River	–	0.35	58	116	46	10
Well #9	61	0.04	32	< 0.13	< 0.13	0.2
Well #2	177	0.03	19	< 0.13	< 0.13	< 0.13
Site — Parkville						
Missouri River	–	0.33	233	132	42	5.5
Well #4	37	0.28	290	< 0.13	< 0.13	< 0.13
Well #5	37	0.25	201	< 0.13	< 0.13	< 0.13

AOC = assimilable organic carbon; BDOC = biodegradable dissolved organic carbon; cfu = colony forming units.
* Water from this well is not dominated by infiltration.

2.2.1 Conventional clarification

Efficiency of conventional clarification

Conventional clarification typically refers to chemical addition, rapid mixing, flocculation and sedimentation (usually in a rectangular basin). Removal of particles depends mainly on the terminal settling velocity of the particles and the rate of basin surface loading or overflow. The efficiency of the sedimentation process may be improved by using inclined plates or tubes. For conventional treatment processes, chemical coagulation is critical for effective removal of microbial pathogens. In the absence of a chemical coagulant, removal of microbes is low because sedimentation velocities are low (Medema et al., 1998). A chemical coagulant destabilizes microbial particles (e.g. by neutralizing or reducing their surface electrical charge, enmeshing them in a floc particle or creating bridges between them) and allows particles to come into contact with one another. Flocculation of microbial particles creates aggregates with sufficient settling velocities to be removed in the sedimentation basin.

When properly performed, coagulation, flocculation and sedimentation can result in 1–2 log removals of bacteria, viruses and protozoa. However, performance of full-scale, conventional clarification processes may be highly

variable, depending on the degree of optimization. For example, in a report summarizing the performance of treatment plants from various countries, average microbial removals for coagulation and sedimentation ranged from 27 to 74% for viruses, 32 to 87% for bacteria (total coliforms or faecal streptococci) and 0 to 94% for algae (Gimbel & Clasen, 1998). It is difficult to interpret full-scale data for *Cryptosporidium* and *Giardia* because these protozoa are found at very low levels, and methods for their detection have limitations (LeChevallier et al., 1991).

Factors that can result in poor clarification efficiency include variable plant flow rates, improper dose of coagulant, poor process control with little monitoring, shear of formed floc, inappropriate mixing of chemicals, poor mixing and flocculation, and inadequate sludge removal (USEPA, 1991). In addition to metallic coagulants (e.g. alum or ferric), it may be necessary to use polymeric coagulation, filter aids or both to produce low turbidity levels (< 0.1 nephelometric turbidity unit, NTU) especially for high-rate filtration (> 2.71 l/m^2·s). Preoxidation with chlorine or ozone can improve particle removal by sedimentation and filtration (Yates et al., 1997; Becker, O'Melia & Croker, 1998). In some cases, treatment plants are being designed with intermediate ozonation, specifically to aid in particle removal by sedimentation and filtration (Langlais, Reckhow & Brink, 1991).

Using jar tests, Bell et al. (2000) reported removal of bacteria (*E. coli* vegetative cells and *Clostridium perfringens* spores) and protozoa (*Giardia* cysts and *Cryptosporidium* oocysts) as typically of 1–2 logs (Figure 2.3). Overall, iron-based coagulants were slightly more efficient than alum (aluminum hydroxide) or polyaluminium chloride (PACl); however, site-specific water-quality conditions had a greater effect on removal efficiencies than did the type of coagulant. Coagulation conditions (i.e. dose, pH, temperature, alkalinity, turbidity and the level and type of natural organic matter) affected the efficiency of removal, with slightly better overall microbial reductions under pH conditions optimal for removal of total organic carbon (i.e. pH 5–6.5).

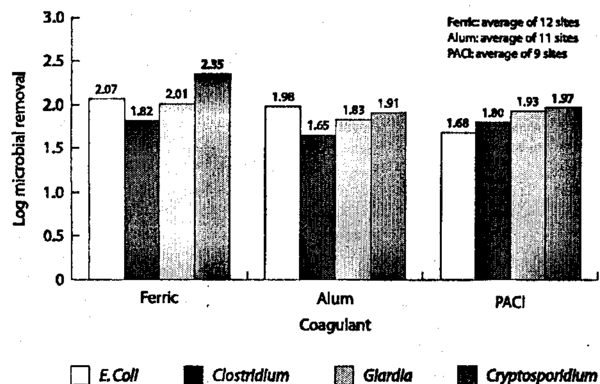


Figure 2.3 Removal of bacteria and protozoa under optimal coagulation conditions. Adapted from Bell et al. (2000)

Viruses

Figure 2.4 shows that different viruses may respond quite differently to coagulation conditions. For example, the bacteriophage MS2 and human enteric poliovirus are removed at a fairly high efficiency (2.6–3.4 logs), whereas the phage PRD-1 and enteric echovirus are removed at a much lower rate (1.1–1.9 logs). The differences in virus removal are most pronounced for alum. Similar differences in virus adsorption have been observed in granulated gels (Mouillot & Netter, 1977). It is evident that the effect of coagulation differs for various viruses, and that it may be unwise to extrapolate the data on viruses to other, untested viruses.

Protozoa

Haas et al. (2000) reviewed data from four bench-scale or pilot-plant studies for coagulation, flocculation and sedimentation of *Cryptosporidium* oocysts. The authors selected data from studies where the coagulant type, coagulant dose, pH, temperature and mixing conditions were described. Using 24 data points, they found that oocyst removal depended on coagulant concentration, polymer concentration and process pH. The model had an excellent fit to the data (R^2 of

0.94); however, the fit decreased when data from other studies were added to the model. The authors concluded that additional data are needed, especially from studies that fully describe coagulation and flocculation conditions.

An optimal coagulation dose is the most important factor for ensuring effective removal of cysts and oocysts by sedimentation and filtration (Logsdon et al., 1985; Al-Ani et al., 1986; Logsdon, 1990; Bellamy et al., 1993). Impaired flocculation was one of the factors in the 1987 outbreak of cryptosporidiosis in Carrollton, Georgia (USA) (Bellamy et al., 1993). In a study of eight water filtration plants, Hendricks et al. (1988) concluded:

> ... without proper chemical pretreatment *Giardia* cysts will pass the filtration process. Lack of chemical coagulation or improper coagulation was the single most important factor in the design or operation of those rapid rate filtration plants where *Giardia* cysts were found in finished water ... with proper chemical coagulation, the finished water should be free of *Giardia* cysts, have few microscopic particles and have turbidity levels less than 0.1 NTU [nephelometric turbidity units].

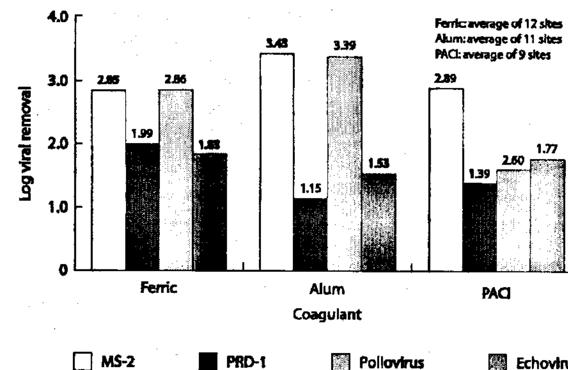


Figure 2.4 Removal of viruses under optimized coagulation conditions. Adapted from Bell et al. (2000).

Algae

Coagulation and sedimentation can be effective for removal of algae, although care must be taken to remove these organisms without disrupting the cells, as this may release liver or nerve toxins. Generally, coagulation appears not to cause the release of algal toxins, provided that oxidants are not added (Yoo et al., 1995b). Coagulation and sedimentation are not very effective at removing algal toxins; studies have shown removal levels ranging from 0 to 49%. However, addition of powdered activated carbon to the clarification process can increase removal levels to 90% or more, depending on the carbon dose, type of carbon, toxin level and organic matrix (Yoo et al., 1995b). A natural coagulant derived from shrimp shells (termed chitosan) was shown to be effective, removing more than 90% of the algae *Chlorella* and *Scenedesmus quadricuda* at neutral to alkaline pH conditions, using chitosan doses of more than 10 mg/l (Chen, Liu & Ju, 1996).

2.2.2 High-rate clarification

High-rate clarification was first used in the 1930s, and it grew in popularity during the 1970s and 1980s. It involves using smaller basins and higher surface loading rates than conventional clarifiers, and is therefore referred to as high-rate clarification. Processes include floc-blanket sedimentation (also known as 'solids-contact clarification'), ballasted-floc sedimentation, and adsorption or contact clarification.

In floc-blanket sedimentation, a fluidized blanket increases the particle concentration, thus increasing the rate of flocculation and sedimentation. Ballasted-floc systems combine coagulation with sand, clay, magnetite or carbon to increase the particle sedimentation rate. Adsorption or contact clarification involves passing coagulated water through a bed where particles attach to previously adsorbed material.

High-rate clarifiers can be as effective as or even more effective than conventional basins for removal of microbes. The choice of an appropriate blanket polymer is important for optimal operation (Gregory, Zabel & Edzwald, 1999). Bell, Bienlien & LeChevallier (1998) reported turbidity removals of 98% for a solids-contact, sludge blanket clarifier (raw water turbidity 20–50 NTU, settled water 0.6–0.75 NTU), 89% for internal slurry recirculation (raw water turbidity 4–10 NTU, settled water 0.5–0.9 NTU) and 61% for circular floc-blanket purification unit clarification (raw water turbidity 1.2–16 NTU, settled water average 0.97 NTU). Baudin & Laîné (1998) evaluated three full-scale treatment plants and found complete removal (> 2–2.8 logs) of *Giardia* and *Cryptosporidium* by pulsator clarifiers. The units produced a 1.0–2.7 log removal of turbidity. Other investigators (Hall, Pressdee & Carrington, 1994) have reported similar efficiencies for floc-blanket clarifiers. A combination of preozonation and use of a solids-contact sludge blanket reportedly improved clarification of *Giardia* and *Cryptosporidium*-sized particles by about 1.5–2.5 logs (Wilczak et al., 1991). Pilot plant studies of a sand ballasted-floc system showed effective removal of turbidity and particle counts (Jeschke, 1998). In addition, microscopic particulate analysis of raw and settled water showed an average 3.9-log removal of algae, and 4.5-log removal of diatoms (Jeschke, 1998). Floc formed on magnetic particles can be rapidly removed by using magnets within the sedimentation process (Gregory, Maloney & Stockley, 1988; Bolto, 1990; Anderson et al., 1993). The magnetic particles can be collected and regenerated for reuse.

2.2.3 Dissolved air flotation

In dissolved air flotation (DAF), bubbles are produced by reducing pressure in a water stream saturated with air. The rising bubbles attach to floc particles, causing the agglomerate to float to the surface, where the material is skimmed off (Gregory, Zabel & Edzwald, 1999). DAF can be particularly effective for removal of algal cells and *Cryptosporidium* oocysts. It is most applicable to waters with heavy algal blooms or those with low turbidity, low alkalinity and high color, which are difficult to treat by sedimentation because the floc produced has a low settling velocity.

The effectiveness of DAF for treating algal-laden, humic, coloured water is illustrated by the comments of Kiuru (1998), who indicated that the only type of treatment plants built in Finland since the mid-1960s have been DAF plants. A 1.8-log removal of the algae *Aphanizomenon* and *Microcystis* was achieved by pilot-scale DAF. Similar results (1.4–2.0 log removals) have been obtained in full-scale studies (Mouchet and Bonnelye, 1998). DAF is also effective in the removal of cell-associated algal toxins (Mouchet and Bonnelye, 1998).

Plummer, Edzwald & Kelley (1995) reported that, depending on the coagulant dose, DAF achieved 2–2.6 log removal of *Cryptosporidium* oocysts, whereas conventional sedimentation resulted in 0.6–0.8 log removal. The performance of DAF for oocyst removal depended on the pH, coagulant dose, flocculation time and recycle ratio of the saturated water stream. Other researchers have confirmed the effectiveness of DAF for oocyst removal, particularly when polyelectrolyte coagulant aids are added to help stabilize the floc (Hall, Pressdee & Carrington, 1994).

2.2.4 Lime softening

Precipitative lime softening is a process in which the pH of the water is increased (usually through the addition of lime or soda ash) to precipitate high concentrations of calcium and magnesium. Typically, calcium can be reduced at pH 9.5–10.5, although magnesium requires pH 10.5–11.5. This distinction is important because the pH of lime softening can inactivate many microbes at the higher end (e.g. pH 10–11), but may have less impact at more moderate levels (e.g. pH 9.5). In precipitative lime softening, the calcium carbonate and magnesium hydroxide precipitates are removed in a clarifier before the water is filtered. The microbial impact of lime softening can, therefore, be a combination of inactivation by elevated pH and removal by settling.

Logsdon et al. (1994) evaluated the effects of lime softening on the removal and disinfection efficiency of *Giardia*, viruses and coliform bacteria. Coliform bacteria in river water (spiked with raw sewage) were inactivated by 0.1 log at pH 9.5, 1.0 log at pH 10.5 and 0.8–3.0 logs at pH 11.5 for 6 hours at 2–8°C. Bacteriophage MS2 was sensitive to lime softening conditions, demonstrating more than 4-log inactivation in the pH range of 11–11.5 within 2 hours. Hepatitis A virus was reduced by 99.8% when exposed to pH 10.5 for 6 hours. Poliovirus was the most resistant virus tested, requiring exposure to a pH level of 11 for 6 hours to achieve a 2.5-log inactivation. Reductions were less than 1 log when exposed for 6 hours to a pH of less than 11. The viability of *Giardia muris* cysts (measured by excystation) was not significantly affected by exposure to pH 11.5 for 6 hours. *Cryptosporidium* viability (measured using dye exclusion) was not affected by exposure to pH 9 for 5 hours (Robert, Campbell & Smith, 1992).

Jar tests of precipitative lime softening at pH 11.5 resulted in 4-log removal of viruses and bacteria, and 2-log removal of *Giardia* and *Cryptosporidium*, due to combined effects of removal by sedimentation and inactivation through high pH (Bell et al., 2000). Limited full-scale data suggest that 2-log removal can be achieved through sedimentation by precipitative lime softening (Logsdon et al. 1994).

2.2.5 In-line coagulation

In-line coagulation can be used with high-quality source waters (e.g. those where turbidity and other contaminant levels are low). The coagulants are added directly to the raw water pipeline before direct filtration. Typically, the coagulants are added before an in-line static mixer, and it is not necessary to use a basin for sedimentation. In-line coagulation permits the particle destabilization

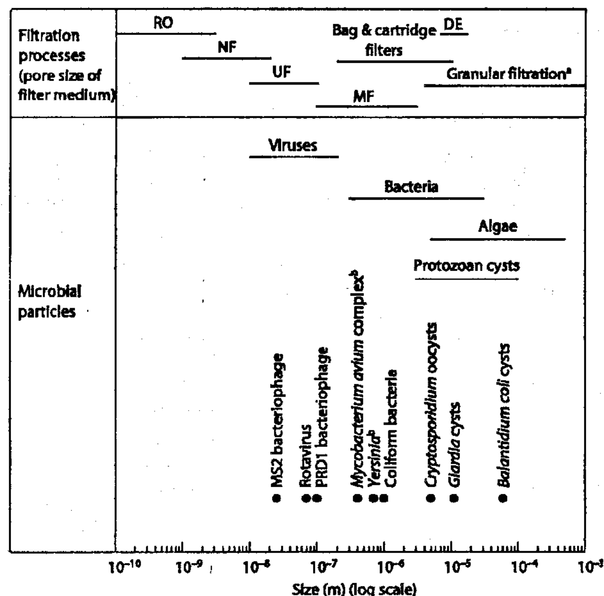
necessary for effective particle removal by filtration, but does not remove microbes by sedimentation.

2.3 ION EXCHANGE

Ion exchange is a treatment process in which a solid phase presaturant ion is exchanged for an unwanted ion in the untreated water. The process is used for water softening (removal of calcium and magnesium), removal of some radionuclides (e.g. radium and barium) and removal of various other contaminants (e.g. nitrate, arsenate, chromate, selenate and dissolved organic carbon). The effectiveness of the process depends on the background water quality, and the levels of other competing ions and total dissolved solids. Although some ion exchange systems can be effective for adsorbing viruses and bacteria (Semmens, 1977), such systems are not generally considered a microbial treatment barrier, because the organisms can be released from the resin by competing ions. Also, ion exchange resins may become colonized by bacteria, which can then contaminate treated effluents (Flemming, 1987; Parsons, 2000). Backflushing and other rinsing procedures, even regeneration, will not remove all of the attached microbes. Impregnation of the resin with silver suppresses bacterial growth initially, but eventually a silver-tolerant population develops. Disinfection of ion exchange resins using 0.01% peracetic acid (1 hour contact time) has been suggested (Flemming, 1987).

2.4 FILTRATION

Various filtration processes are used in drinking-water treatment. Filtration can act as a consistent and effective barrier for microbial pathogens. Figure 2.5 shows the most commonly used filtration processes in potable water treatment, the pore size of the filter media and the sizes of different microbial particles. These size spectra are useful for understanding removal mechanisms and efficiencies, and for developing strategies to remove microbes by different filtration processes.



DE = diatomaceous earth; MF = microfiltration; NF = nanofiltration; RO = reverse osmosis; UF = ultrafiltration.

Figure 2.5 Pore size of filter medium and size of microbial particles

2.5 GRANULAR HIGH-RATE FILTRATION

Granular media filtration is the most widely used filtration process in drinking-water treatment. A comprehensive review of granular media filtration processes is provided by Cleasby and Logsdon (1999). Under optimal conditions, a combination of coagulation, flocculation, sedimentation and granular media filtration can result in 4 logs or better removal of protozoan pathogens with chlorine-resistant cysts. This section discusses granular filtration other than slow

sand filtration (described in Section 2.6). Slow sand filtration is discussed separately because the low filtration rate (< 0.4 m/h) used in this process means that the design and operating criteria, and the mechanisms for removal of microbes are considerably different from those of 'rapid-rate' granular filtration.

2.5.1 Design of granular filtration

In granular filtration, water passes through a filter consisting of a packed bed of granular materials. Microbes or microbe-associated particles are removed as they deposit on the filter medium. The removal occurs within the granular medium (depth filtration) rather than on the top layer only (cake filtration). After a period of operation, the head loss increases (i.e. the pressure increases) or the effluent quality deteriorates to an unacceptable level. The filter then has to be cleaned by backwashing, after which it performs poorly during a 'ripening period' before achieving a stable level of performance. Passage of microbial pathogens during the ripening period can be very high. Various strategies are used to minimize this effect, including:

- *filter to waste* — wasting the initial filtered water;
- *slow start* — limiting the initial filtration rate until the filtrate quality is acceptable;
- *delayed start* — leaving the filter inactive for a time following backwash, before bringing it into operation;
- *filter aid* — adding a filter aid to the wash water supply.

Granular filters can be constructed as monomedium (e.g. silica sand), dual media (e.g. anthracite coal and sand) and trimedia (e.g. coal, sand and garnet). Granular activated carbon is used when both filtration of particles and adsorption of organic compounds are desired. Depending on raw water quality, granular filtration can be operated in three different modes:

- *conventional*, which includes addition of coagulants (rapid mixing), flocculation (slow mixing), sedimentation and filtration;
- *direct filtration*, in which the sedimentation step is omitted;
- *in-line filtration*, in which both flocculation and sedimentation steps are omitted.

Conventional treatment is appropriate for most source waters, whereas direct and in-line filtration are used for raw waters with a consistently good quality (low turbidity and colour).

2.5.2 Mechanism of action of granular filtration

Removal of microbial pathogens by granular filtration does not rely on physical processes alone. Comparing the pore size of granular filters with the size of most types of microbe (as in Figure 2.5), it is evident that effective removal of microbes by granular filtration cannot rely on physical straining alone, at least at the initial stage of a filter run. The removal of particles by granular filtration is considered to involve two steps: transport of particles from suspension to filter medium, followed by attachment of particles to the medium (Yao, Habibian & O'Melia, 1971).

The transport step depends on the physical and hydrodynamic properties of the system. Transport mechanisms include diffusion, interception and sedimentation. Factors such as size and density of microbes, size and depth of filter medium, and filtration rate affect transport efficiency. In the case of motile microorganisms, an additional mechanism is the active movement of the cell (Hozalski & Bouwer, 1998). Attachment is determined by the surface and solution chemistry of the system (Tobiason & O'Melia, 1988). Unfavorable interactions between particles and the filter medium must be avoided so that particles can attach to the medium. Chemical coagulation is used before filtration to destabilize particles; this step is the single most important factor in determining filtration efficiency. Without proper chemical pretreatment, rapid rate filtration works as a simple strainer and is not an effective barrier for microbial pathogens.

2.5.3 Importance of chemical coagulation pretreatment

The importance of chemical coagulation pretreatment for removal of microbes by granular filtration has been emphasized by numerous studies. Al-Ani et al. (1986) conducted a pilot-scale filtration study for low turbidity waters (< 1 NTU). Without chemical pretreatment, the removal by filtration averaged 69% (0.51 logs) for *Giardia* and 28% (0.14 logs) for turbidity. Adding alum and polymer filter aids increased the removal efficiency to more than 95% (1.30 logs) for *Giardia*, 99% (2 logs) for total coliform bacteria and 70% (0.52 logs) for turbidity. Other process variables such as filtration mode (direct and in-line filtrations), filter media (monomedium with sand, and dual-media with sand and anthracite) and temperature (5°C and 18°C) did not significantly affect the filtered water quality. Varying the filtration rate from 5 to about 20 m/h had little effect on removal of *Giardia*, total coliform bacteria and heterotrophic bacteria, but increased turbidity in the filtered water.

Robeck, Clarke & Dostal (1962) reported that, when alum was used as a coagulant, the removal of poliovirus type 1 by a pilot-scale dual-media filter was more than 98% (1.70 logs). Ongerth (1990) conducted pilot studies for conventional and in-line filtration. Without any chemical addition, removal of *Giardia* cysts averaged 75% (0.60 logs) for conventional treatment and 64% (0.44 logs) for in-line filtration. With optimal chemical pretreatment, the removal increased to 98% (1.70 logs) for conventional treatment and 93.6% (1.19 logs) for in-line filtration.

Nieminski & Ongerth (1995) evaluated the removal of *Cryptosporidium* oocysts and *Giardia* cysts over two years at pilot and full-scale filtration plants, operated under direct filtration and conventional treatment modes. Dual-media filters with anthracite and sand were used. *Giardia* and *Cryptosporidium* were effectively removed when coagulation conditions produced filtered water of low turbidity (0.1–0.2 NTU). Under optimal coagulation conditions, the average removal of *Giardia* was 3.3 logs or more, and the average removal of *Cryptosporidium* was 2.3 logs or more in both pilot and full-scale plants, regardless of the treatment modes (direct filtration or conventional treatment). The authors also investigated correlations between the removal of cysts and surrogate parameters. A high correlation was found between removal of cysts and particles of 4–7 µm and 7–11 µm ($R^2 \geq 0.79$). There was a lower correlation between removal of *Giardia* and *Cryptosporidium* and removal of turbidity ($R^2 \leq 0.64$). Particle counting was a better indicator of cyst and oocyst removal than turbidity. Log removal of seeded cysts did not correlate with log removal of heterotrophic bacteria ($R^2 \leq 0.08$), suggesting that heterotrophic plate counts (HPCs) are not a good surrogate to use in evaluating cyst removal. A recent WHO publication (Bartram et al., 2003) discusses the significance of HPCs for water quality and human health.

2.5.4 Effect of filter media design

Swertfeger et al. (1999) evaluated the effect of filter media design on cyst and oocyst removal. Designs included monomedium (sand with a depth of 750 mm), fine dual-media (anthracite and sand with depths of 900 mm and 300 mm, respectively) and deep dual-media (anthracite and sand with depths of 1500 mm and 300 mm, respectively). The feedwater to the pilot systems was taken from the effluent of a sedimentation unit in a full-scale water treatment plant and was in optimal coagulation condition. The authors found no statistical difference in the filtration performance for the different media. Removal of *Giardia* was 4.4 logs or better, with greater removal efficiency

in the summer than in the winter. Removal of *Cryptosporidium* was similar in summer and winter, and averaged 2.7 logs or more.

Payment et al. (2000) reported water-quality monitoring results for a full-scale conventional water treatment plant using dual-media filters, with coagulation provided by alum and activated silica. Prechlorination was applied at 1 mg/l. The results confirmed that a properly operated conventional treatment plant provided a substantial barrier to microbial pathogens. *Giardia* cysts were detected in only 1 of 32 filtered water samples, with a mean removal of 3.6 logs after filtration (including removal by coagulation and sedimentation). Removal of *Cryptosporidium* oocysts was lower than for *Giardia*. Oocysts were detected in 7 of 32 filtered water samples, with a mean removal of 2 logs. *Clostridium perfringens* was detected in 9 of 33 filtered water samples, with average removal of 4.4 logs. No human enteric virus was detected in 32 filtered water samples, with average removal of 3.1 logs (assuming that the concentration of humic enteric virus in filtered water was equal to the detection limit). Somatic coliphage were detected in 24 of 32 filtered water samples, with average removal of 3.5 logs.

2.5.5 Importance of filter backwash

When solids accumulate within a filter bed, they create a resistance to flow. This resistance is measured as loss of head (pressure increase) for the filter bed. The filter is backwashed, usually with finished water, to remove the accumulated particles. The need for backwashing may be determined using various criteria — a terminal head loss, a fixed time interval, or a breakthrough of solids (measured as turbidity or particle counts). Options for disposal of the spent filter backwash water may include discharge to a sewer or a receiving stream. Because backwash water may contain disinfectants and other chemicals that may be harmful to the biological life of a stream, direct discharge to streams may be restricted. Similarly, discharge to sewers may be restricted, based on the constituents and total quantity of the backwash water.

For many water treatment plants, particularly in arid or water-scarce areas with limited raw water resources, it is often necessary to reuse backwash water. When the water is recycled, accumulation of microbial and algal contaminants is a concern. For example, algal toxins may be released from stored treatment sludges when the overlying water is recycled (Drikas et al., 2001). Because of the resistance of oocysts to conventional disinfectants, *Cryptosporidium* has been a major concern for the handling and operation of recycled process streams. Table 2.5 summarizes data on the

occurrence and concentration of *Giardia* cysts and *Cryptosporidium* oocysts in filter backwash water. The level of treatment required for spent filter backwash water before recycle will vary from site to site depending on the treatment process and water-quality objectives. Equalization of the recycle flow and sedimentation of the backwash solids, aided by the addition of a polymer coagulant, is sufficient to reduce cyst concentrations to raw water levels in most cases (Cornwell & Lee, 1993; Arora, Di Giovanni & LeChevallier, 1999; McTigue et al., 2000).

2.6 SLOW SAND FILTRATION

The use of slow sand filtration to protect drinking-water consumers from microbial risk was well established more than 100 years ago. Two of the earliest successful cases were reductions in cholera in Altona (Germany) and typhoid fever in Lawrence, Massachusetts (USA) in the 1890s (Bellamy et al., 1985). Numerous disease outbreaks due to chlorine-resistant protozoan pathogens in the past two decades have increased interest in slow sand filtration because of its ability to remove parasites.

2.6.1 Design and action of slow sand filters

Slow sand filtration involves passing water through a sand filter by gravity at a very low filtration rate, without the use of coagulation pretreatment. The filter typically consists of a layer of sand supported on a layer of graded gravel. Typical design criteria for slow sand filtration are given in Table 2.6. Detailed design guidelines can be found in Hendricks (1991). As water passes through the filter, microbes and other substances are removed. The removal mechanisms are not well understood, although they are believed to be a combination of biological, physical and chemical mechanisms (Weber-Shirk & Dick, 1997ab). Specific mechanisms may include biological action (e.g. ciliate protozoa acting as bacterial predators), attachment of microbes to sand media (e.g. by electrochemical forces and through bridging by microbial extracellular polymers) and physical straining.

Table 2.5 *Giardia* and *Cryptosporidium* occurrence in filter backwash water

Reference	Location (No. of WTPs sampled)	Sample type (No. of samples)	*Cryptosporidium* oocysts/100l		*Giardia* cysts/100l	
Rose et al. (1986)	USA (2)	SFBW (2)	Sample 1: 686,900 Sample 2: 2,430,600		NR	
Colbourne (1989)	Thames Water, UK (1)	Raw (unknown) SFBW (1) Supernatant [a] (1)	0.2–1400 > 1,000,000 >100,000		NR	
Rose et al. (1991)	USA (17 states)	SFBW (subset of 257 samples)	217[b]		NR	
LeChevallier et al. (1991)	USA (66 in 14 states)	Raw Initial SFBW	7–108 57–61 times raw water level		4–32 12–16 times raw water level	
Cornwell & Lee (1993, 1994)	USA (2)	Plant 1:	Round 1	Round 2	Round 1	Round 2
		Raw (1)	6	140	3	BDL
		Mixed influent [c] (1)	40	45	7	NR
		SFBW (1)	902	850	1350	BDL
		Supernatant [a] (1)	141	750	86	BDL

Table 2.5 (continued) *Giardia* and *Cryptosporidium* occurrence in filter backwash water

Reference	Location (No. of WTPs sampled)	Sample type (No. of samples)	*Cryptosporidium* oocysts/100l		*Giardia* cysts/100l	
Cornwell & Lee (1993, 1994)	USA (2)	Plant 2:	Round 1	Round 2	Round 1	Round 2
		Raw (1)	13	20	290	60
		Mixed influent [c] (1)	30	476	160	79
		SFBW (1)	16,613	NR	16,513	NR
		Supernatant [a] (1)	80	420	70	BDL
Karanis, Schoenen & Seitz (1996)	Germany (1)	*Centrifugation method* Raw (8 positive out of 12) SFBW (8 positive out of 11) Cartridge filter SFBW (33 positive out of 39)	 0.8 to 109 1–69 0.8 to 252		NR	
Karanis, Schoenen & Seitz (1998)	Germany (1)	SFBW [d] (1)	150		NR	
States et al. (1995)	Pittsburgh, USA (1)	Raw (11 positive out of 15)	43		42	
		Filtered (2 positive out of 15)	0.4		BDL	
		SFBW (8 positive out of 15)	321			
		SFBW (2 positive out of 15)			59	

Table 2.5 (continued) *Giardia* and *Cryptosporidium* occurrence in filter backwash water

Reference	Location (No. of WTPs sampled)	Sample type (No. of samples)	*Cryptosporidium* oocysts/100l	*Giardia* cysts/100l
Arora, Di Giovanni & LeChevallier (1999)	USA (25)	*IFA method*		
		Raw (17 positive out of 146)	108	
		Raw (44 positive out of 146)		89
		SFBW (7 positive out of 148)	175	
		SFBW (12 positive out of 148)		203
		CC-PCR method [e]	Qualitative method	NA
		Raw (6 positive out of 122)	Qualitative method	NA
		SFBW (9 positive out of 121)		

BDL = below detection level; CC-PCR = cell culture-polymerase chain reaction; IFA = immunofluorescence assay; NA = not applicable; NR = not reported; SFBW = spent filter backwash.

Notes:
[a] Supernatant from settling basin treating spent filter backwash water
[b] Geometric mean concentration
[c] Sample after addition of recycle stream
[d] Sample taken 10 minutes after start of backwash cycle
[e] Cell culture-polymerase chain reaction method identifies live, infectious *Cryptosporidium*

Table 2.6 Typical design criteria for slow sand filtration

Design criterion	Normal range
Filtration rate	0.04–0.4 m/h
Sand media	
Depth	0.5–1.5 m
Effective size	0.15–0.40 mm
Uniformity coefficient	1.5–3.6
Gravel media	
Depth	0.2–1 m
Graded	Fine to coarse (top to bottom)

Source: Letterman, 1991; Cleasby & Logsdon (1999)

Removal of particles by slow sand filtration occurs predominantly, if not entirely, in a thin layer on the top of the sand bed. This biologically active layer, composed of living and dead microorganisms and macroorganisms, is termed *schmutzdecke*. As operation progresses, deposited materials and biological growth on the sand medium increase the head loss across the filter. When the head loss reaches the operational limit (normally 1–2 m), the filter is removed from service. It is then usually cleaned by scraping about 2 cm of accumulated material and sand from the top layer of the sand bed, before being returned to service. A typical filter run is from one to six months, depending on the raw water quality and filtration rate. After the sand bed is reduced to a lowest acceptable depth by repeated scrapings, it is necessary to replace the sand down to the gravel support level.

2.6.2 Protection provided by slow sand filtration

Slow sand filtration can provide some degree of protection against microbial pathogens. As coagulation pretreatment is not required, slow sand filtration has little maintenance or chemical cost. If the raw water has a high concentration of suspended particles or algae, physical pretreatment processes (e.g. roughing filter or microstrainers) can be used to prevent clogging of the filter and maintain a reasonable filter run period.

Removal of microbes

In a review by Ellis (1985), virus removal ranging from about 1 to 5 logs was reported for bench and full-scale slow sand filters. Various studies have reported the effective removal of bacteria and protozoa by slow sand filtration in pilot and full-scale systems.

In a pilot-scale study, a new filter removed 0.82 logs of total coliform bacteria and more than 1.7 logs of *Giardia* (Bellamy et al., 1985). Once a microbiological population was established within the sand bed (after two weeks), the removal of total coliforms increased to 4 logs and no *Giardia* was detected in the filtered water. The calculated cyst reduction was more than 2.6 logs, depending on influent cyst concentration. Similar results were found in another pilot study, where the removal of total coliform bacteria, heterotrophic bacteria and turbidity increased with the biological activity of the *schmutzdecke* (Bellamy, Hendricks & Logsdon, 1985).

In a full-scale study of a slow sand filter in Empire, Colorado (USA), *Giardia* cysts were detected in almost half of the influent samples, but not in the effluent (Seelaus, Hendricks & Janonis, 1986). In a full-scale study for three slow sand filtration plants in Idaho (USA), no samples positive for *Giardia* were found in the filtered water from two of the three treatment plants (Tanner & Ongerth, 1990). For the one positive sample found in one plant, 1-log removal of *Giardia* was achieved. In the same study, removal of total coliforms and faecal coliforms varied from 84.35 to 99.5% (0.81–2.30 logs) and from 48.1 to 70.0% (0.29–0.52 logs), respectively. Removal of heterotrophic bacteria (as measured by HPC) varied from 65.8 to 91.0% (0.47–1.05 logs). These differences in removal efficiency were influenced by raw water quality, filtration rate, media size and depth. Removal of *Cryptosporidium* by slow sand filtration is often more difficult than removal of *Giardia*. In a full-scale study in British Columbia, Fogel et al. (1993) reported that the average removal of *Giardia* was 93% (1.16 logs) but was only 48% (0.28 logs) for *Cryptosporidium*.

Removal of turbidity

Although the removal of microbes by slow sand filtration can be substantial, reduction of turbidity may be site specific. In one pilot study, turbidity removal was 97.8% (1.66 logs) or more after a filter-ripening period of about two days (Cleasby, Hilmoe & Dimitracopoulos, 1984); similar to the removal of total coliform bacteria (≥ 99.4%) and chlorophyll-a (≥ 95%). Another pilot study found a 27–39% (0.14–0.22 log) removal of turbidity, whereas the reduction of *Giardia* was up to 4 logs (Bellamy et al., 1985). The authors concluded that the low removal of turbidity was due to the fine clay particles present in the raw water, which penetrated the filter. In a full-scale study, turbidity removal was between 0 and 63% (0.43 logs), due to the fine particles present in the raw water and to the large fraction (4% by weight) of fines in the new sand media used in the study (Tanner & Ongerth, 1990). The fact that slow sand filtration can achieve effective removal of microbial pathogens but not necessarily decreased turbidity indicates that turbidity may not be a suitable surrogate for evaluation of the removal of pathogens by slow sand filtration.

2.7 PRECOAT FILTRATION

Precoat filtration was developed by the US Army during World War II as a portable unit for the removal of *Entamoeba histolytica* (a protozoan parasite prevalent in the Pacific war zone) from drinking-water. The process involves forcing water under pressure or by vacuum through a uniformly thin layer of filtering material precoated onto a permeable, rigid, supporting structure (referred to as a septum). Precoat materials include DE and perlite, with DE more commonly used in drinking-water treatment. As water passes through the filter media and septum, the precoat materials (filter cake) capture microbes and other particles, mainly by physical straining. Often, a "bodyfeed" solution containing the filter media slurry is added continuously to the system, to maintain the permeability of the filter cake. As the cake becomes thicker due to the captured particles, head loss increases until further filtration is impractical. The filter cake is removed from the support septum and disposed of. The filter is then cleaned and precoated with a new layer of coating materials, and a new filter cycle starts. A detailed design and operating manual for precoat filtration has been published by the American Water Works Association (AWWA, 1995).

Because the major removal mechanism is physical straining, efficiency of precoat filtration depends to a large extent on the grade (size) of the coating materials and on the size of the microbes. Other factors influencing the removal efficiency are chemical pretreatment of the filter media, filtration rate and bodyfeed rate. Chemical pretreatment of the raw water is usually not necessary; however, the raw water must be of high quality (low turbidity) to maintain a reasonable filter run time.

2.7.1 Removal of microbes

Diatomite grades used for drinking-water treatment have a mean pore diameter of 7–17 µm (Figure 2.5). Precoat filtration can remove protozoan parasites such as *Giardia* very effectively. A pilot study showed complete removal of *Giardia* for both coarse and fine grades of DE over a wide range of operating conditions (Lang et al., 1986). Removal of *Cryptosporidium* can be significant, but because this organism is smaller than *Giardia*, it is more difficult to remove. Removal of *Cryptosporidium* oocysts by a bench-scale DE filter ranged from 3.60 to 6.68 logs, depending on the media grade and the filtration rate (Ongerth & Hutton, 1997). In a pilot-plant study, filtration with DE gave a consistently complete removal of *Giardia* cysts and a 3-log removal of *Cryptosporidium* oocysts (Schuler & Ghosh, 1990).

2.7.2 Importance of chemical pretreatment

Precoat filters remove smaller microbial particles (e.g. bacteria and viruses) less effectively than they do parasites, unless the coating materials are chemically pretreated; for example, with aluminium or iron coagulants, or with cationic polymers. In the pilot study by Schuler & Ghosh (1990) mentioned above, removal of coliforms with untreated DE was about 0.36 logs, increasing to 0.82 logs with a coating of alum at 1 mg/g DE, and to 2 logs at 3 mg/g DE. This increase was probably due to the trapping of bacteria by the alum. A similar beneficial effect was observed using cationic polymers; at 3.5 mg/g DE, removal of coliforms increased to 3.3 logs. The authors concluded that this increase in removal could be due to an increased site density on the polymer-coated DE for adsorption of negatively charged coliforms. A similar improvement in removal of bacteria was reported for the pilot study conducted by Lang et al. (1986). Alum coating of DE increased removal of total coliforms from 0.16 logs to 1.40 logs, and of HPC bacteria from 0.36 logs to 2.30 logs. Removal of viruses also increased with chemical pretreatment of filter cake (Brown, Malina & Moore, 1974). The removal of bacteriophage T2 and poliovirus was about 90% for an uncoated filter, but increased to more than 98% (1.7 logs) when the filter cake was coated with ferric hydrate or polyelectrolytes.

2.8 MEMBRANE FILTRATION

In membrane filtration, a thin semipermeable film (membrane) is used as a selective barrier to remove contaminants from water. There are very few contaminants that cannot be removed by membrane processes. For the past two decades, the use of membrane filtration in drinking-water treatment (including pathogen removal) has been growing, due to increasingly stringent drinking-water regulations and decreasing costs of purchasing and operating membrane filters.

The membrane processes most commonly used to remove microbes from drinking-water are microfiltration (MF), ultrafiltration (UF), nanofiltration (NF) and reverse osmosis (RO). Detailed descriptions of the fundamentals, design and operation of these processes are available (AWWA, 1996; Taylor & Wiesner, 1999). Table 2.7 summarizes these processes, including operating pressure, pore size, primary application and the type of microorganism that can be removed. Not all of these processes are used primarily for removal of pathogens. For example, RO is used mainly for desalination and NF for softening and for removal of precursors of disinfectant by-products. Nevertheless, the ability to remove pathogens broadens the application of these types of filter when used for these other treatment objectives.

Table 2.7 Major membrane filtration processes used in drinking-water treatment

Type	Operating pressure [a] (kPa)	Pore size [b] (μm)	Primary applications	Microbes removed
MF	30–50	≥ 0.1	Removal of particles and turbidity	Algae, protozoa and most bacteria
UF	30–50	≥ 0.01	Removal of dissolved nonionic solutes	Algae, protozoa, most bacteria and viruses
NF	500–1000	≥ 0.001	Removal of divalent ions (softening) and dissolved organic matter	Algae, protozoa, most bacteria and viruses
RO	1000–5000	≥ 0.0001	Removal of monovalent ions (desalination)	Algae, protozoa, most bacteria and viruses

MF = microfiltration; NF = nanofiltration; RO = reverse osmosis; UF = ultrafiltration

[a] All these are pressure-driven processes; the driving force is a pressure difference across the porous membranes.

[b] Pore size is sometimes described as molecular weight cut-off, which is the degree of exclusion of a known solute, determined under a given set of test conditions in the laboratory. Also see Figure 2.5 for pore size.

[c] These membranes are usually made from organic materials such as cellulose acetate and its derivatives, polyamides, polypropylene and other polymers.

[d] Membranes are assembled in different configurations, with hollow fibre and spiral wound the two most common.

Source: Adapted from AWWARF (1996), Taylor & Weisner (1999).

Membrane filtration removes microbial pathogens mainly by size exclusion; that is, microbes larger than the membrane pores are removed. Chemical coagulation is not usually needed before membrane treatment for the removal of microbes. However, some degree of pretreatment is usually employed to reduce membrane fouling (caused by accumulation of chemicals, particles and biological growth on membrane surfaces) and to avoid membrane degradation from chemical attack (caused by hydrolysis and oxidation). Fouling reduces membrane productivity, and membranes must be chemically cleaned to restore productivity.

Examples of pretreatment processes are microstraining, pH adjustment and addition of biocides (chlorine or copper sulfate). If the source water is of poor quality, advanced pretreatment systems (e.g. conventional coagulation–sedimentation–filtration or other membrane processes) may also be necessary.

Based on pore size, the order of effectiveness of microbial removal is RO, NF, UF and MF, with RO being the most effective. However, this is not always the case, because differences in membrane material or configuration, or failure

in the membrane can affect microbial removal. Discussion of the removal efficiency of these different membrane processes follows.

2.8.1 Microfiltration

MF membranes have pores of 0.1 µm or more (Table 2.7). Theoretically, MF can remove protozoa, algae and most bacteria very effectively, and this has been confirmed in a number of studies, some of which are discussed below. However, factors such as bacteria growing in the membrane systems can lead to poor removal of bacteria. Viruses, which are 0.01–0.1 µm in size, can generally pass through MF membranes, but may be removed by the membrane if they are associated with large particles.

Numerous pilot studies have directly evaluated the removal of *Giardia*, *Cryptosporidium* and other specific microbial pathogens by MF. For example, an extensive study using three MF membranes with pore sizes 0.08–0.22 µm found that *Giardia* and *Cryptosporidium* in the filtered water were below detection levels (< 1 cyst or oocyst/l) with two of the membranes (corresponding to log removals of > 4.7 to > 7.0 for *Giardia* and > 4.4 to > 6.9 for *Cryptosporidium*) (Jacangelo, Adham & Laîné, 1995). No cysts or oocysts were detected in the effluent, indicating that the difference in removal efficiency was a function of the feeding concentration. In the case of the membrane where cysts were detected in the filtered water, the membrane seal was defective, but even so it achieved removal of 4.6 logs for *Giardia* and 4.2 logs for *Cryptosporidium*. The authors concluded that MF could act as an absolute barrier to protozoan cysts, provided that the membrane remained intact. As expected, removal of MS2 bacteriophage by these MF membranes was less than 1 log, because the phage is 0.025 µm and the pore size of the membranes is 0.08–0.22 µm.

In another pilot study, MF membranes with an average pore size of 0.2 µm resulted in significant removal of cyst-sized particles (Karimi, Vickers & Harasick, 1999). The removal of *Giardia*-sized (5–15 µm) particles averaged 3.3–4.4 logs. The removal of *Cryptosporidium*-sized (2–5 µm) particles was lower, averaging 2.3–3.5 logs. These removals were a function of the spiking particle concentration and were independent of the membrane flux used (114–170 l/m^2 per hour). Algae were also effectively removed; the feed water contained 275–700 areal standard unit (asu) count of algae and 10–12.7 µg/l of chlorophyll-a, but these were reduced to below detection (< 25 asu and < 0.5 µg/l, respectively) in the filtered water. However, the HPCs from the filtered water exceeded those of the feed water, probably due to the growth of microorganisms in the pilot system.

A pilot study using two MF membranes with nominal sizes of 0.1 and 0.2 µm also confirmed the complete removal of *Giardia* and *Cryptosporidium* by MF, with neither of these organisms detected in the filtered water (States et al., 1999). Hollow fibre membranes with a nominal pore size of 0.2 µm were used in a pilot-scale study using MF membranes for treating filter backwash water (Parker et al., 1999), pretreated in sedimentation tanks. The MF membranes reliably produced water with turbidity below 0.1 NTU, compared to an influent turbidity of 12.4–88 NTU. Average removal efficiency was 99.46% (2.27 logs) for particles in the size range 3–15 µm and 91.3% (1.06 logs) for heterotrophic bacteria. The MF membranes were also challenged with different microbes. Removal efficiencies were 5.3 logs for *Cryptosporidium parvum*, 6.4 logs for algae, more than 4.3 logs for total coliforms, 3.3 logs for heterotrophic bacteria, more than 3.5 logs for aerobic spores, 2.7 logs for total culturable virus and 3.7 logs for male-specific coliphage.

Excellent removal of turbidity, oocyst-sized particles and indicator bacteria was found in a full-scale study of a 19 000 m^3/day MF plant using 0.2 µm MF membranes (Yoo et al., 1995a). Turbidity of up to 100 NTU was observed in the raw water; however, the finished water was always 0.05 NTU or less. Removal of oocyst-sized (4–10 µm) particles was greater than 3 logs. Neither total nor faecal coliforms were detected in any of the finished water samples. During a subsequent seeded challenge study, greater than 6-log removal was observed for both *Giardia* and *Cryptosporidium* at a flux rate of 0.94 gpm/m^2 (AWWARF, 1999).

2.8.2 Ultrafiltration

UF membranes have pores of 0.01 µm or more, small enough to remove some viruses in addition to bacteria and protozoa (Table 2.7). In the bench and pilot-scale studies discussed above, Jacangelo, Adham & Laîné (1995) found that UF, like MF, could act as an absolute barrier to protozoan cysts as long as membranes remained intact. Three UF membranes (with molecular weight cut-offs of 100 000–500 000 daltons, corresponding to pore sizes of 0.01–0.05 µm) were used in the studies. Neither *Giardia* nor *Cryptosporidium* were detected in the filtered water (corresponding to log removals of > 4.7 to > 7.0 for *Giardia* and > 4.4 to > 7.0 for *Cryptosporidium*). Removal of viruses by UF was significantly better than removal by MF, and depended essentially on the pore size of the membranes. The membranes with the lowest molecular weight cut-offs achieved the highest removal efficiency (6 log or higher) for MS2 bacteriophage in both bench and pilot-scale studies. The authors also concluded that, although physical sieving was the main mechanism for the removal of

protozoan pathogens by UF and MF, cake layer formation and changes in the fouling of the membrane also contributed to the removal of viruses.

A pilot study to investigate the removal of particles and indicator bacteria from two surface water supplies used a UF membrane with a molecular weight cut-off of 100 000 daltons (Jacangelo et al., 1989). The membrane effectively removed particles, turbidity, total coliforms and heterotrophic bacteria, and produced filtered water with turbidity less than 0.04 NTU. Particle removal was from 2.6 logs to greater than 4.6 logs, depending on influent particle concentration. No coliforms were detected in the finished water. Influent HPCs of 4–4500 cfu/ml were reduced to < 1–5 cfu/ml in the effluent. The authors concluded that the heterotrophic bacteria in the filtered water were due primarily to the regrowth of bacteria in the membrane system.

A systematic pilot study to evaluate the use of UF to remove microbial pathogens from four different source waters used membranes with a molecular weight cut-off of 100 000 daltons (Jacangelo et al., 1991). Removal efficiencies for *Giardia muris*, coliforms, heterotrophic bacteria and MS2 bacteriophage were determined. *Giardia muris*, total coliform bacteria and MS2 bacteriophage in the filtered water were below detection (corresponding to reduction efficiencies of > 4 logs, > 7 logs and > 6.5 logs respectively). Differences in water quality or changes in operating parameters did not affect the removal capabilities of the process, but maintenance of membrane integrity was critical to assuring process efficiency. Loss of membrane integrity (fibre breakage) was associated with the detection of both *Giardia muris* and MS2 bacteriophage in the permeate water. Heterotrophic bacteria were found in the permeate water, but this was due to colonization of a section of the sample tap piping rather than to penetration of the bacteria through the membrane.

2.8.3 Nanofiltration and reverse osmosis

The pore sizes of NF and RO membranes are smaller than those of UF membranes. However, NF and RO alone are seldom used to remove microbial pathogens because MF or UF are more cost-effective and can achieve a similar degree of microbial removal. Not surprisingly, there is far less literature on the removal of microbial pathogens by NF and RO than by MF and UF. Representative examples are discussed below.

Bench-scale study

A bench-scale study evaluated virus removal by five different RO membranes (Adham et al., 1998). MS2 bacteriophage was used as the model virus, seeded at concentrations of 10^3–10^8 plaque-forming units (pfu)/ml. Virus reduction was from 2.7 logs to more than 6.5 logs. For the membrane with the highest removal

efficiency, no MS2 was found in filtered water (detection limit < 1 pfu/ml). The authors concluded that an RO membrane was not always an absolute barrier to viruses, and that the levels of removal achieved by each membrane varied, depending on the membrane type and manufacturer.

Pilot-scale study

A pilot study to investigate the efficiency of integrated membrane systems used *Bacillus subtilis* endospores as a surrogate for *Cryptosporidium* and *Giardia* to challenge eight different integrated membrane systems (Owen et al., 1999). The systems included two different NF membranes with two different MF membranes as pretreatment, with and without in-line coagulation pretreatment. The systems did not completely remove spores, but gave overall cumulative removals of 8.0–11.0 logs. There was no difference in spore removal with or without in-line coagulation, but membrane configuration and membrane film significantly affected spore removal. The MF membranes, configured as hollow fibres, achieved 5.6–5.9 log removal of spores. The NF membranes, with an average pore size two orders of magnitude less than the MFs and a spiral wound configuration, achieved 2.2–4.5 logs removal.

The authors concluded that a hollow fibre configuration, which simply seals membrane fibres in a straight line, was unlikely to leak. In contrast, the spiral configurations crease membrane envelopes, and include feed stream and permeate stream spacers. The creases and spacers could compromise membrane integrity. Spore removal by the composite thin film NF membrane exceeded that of the cellulose acetate NF membrane by about 2 logs.

Full-scale studies

Full-scale studies to evaluate the removal of microbial pathogens by integrated membrane systems using NF as the major treatment unit have been reported by Lovins et al. (1999) and Gullick et al. (2000). Two composite thin film NF membranes and one cellulose acetate NF membrane with molecular weight cut-offs of 100–300 daltons were used. Protozoa (*Cryptosporidium* oocysts and *Giardia* cysts), bacteria (*Clostridium perfringens* spores) and bacteriophage (MS2 and PRD1) were used to challenge the different NF membranes. Similar to the finding by Owen et al. (1999), the two composite thin film NF membranes were significantly more effective than the cellulose acetate NF membrane at removing microbes. Removals of about 5.5 logs were achieved with the thin film membranes, with complete removal in more than half of the tests. This compared to removals of about 2 logs with the cellulose acetate membrane, which produced complete removal in less than 10% of the tests.

The efficiency of a UF membrane with molecular weight cut-off of 100 000 daltons was also investigated in this study. The observed microbial removal performance for the UF was similar to that of the two composite thin film NF membranes, and significantly higher than that of the cellulose acetate NF membrane. The authors suggested that this was due partly to the configurations of the membrane (hollow fibre for UF and spiral wound for NF). Integrated membrane systems with different configurations were tested in the study. Pretreatment (before NF) included conventional coagulation followed by sedimentation and sand filtration, hollow fibre MF with pore size 0.2 μm and hollow fibre UF with a molecular weight cut-off of 100 00 daltons. As expected, the highest pathogen removals were achieved by integrated membrane systems with composite thin film NF and UF pretreatment, with 6.3–11.0 log removals. However, some membranes did not remove microbes completely, even at relatively low feed concentrations, indicating that integrated membrane systems are not necessarily absolute barriers to pathogens.

2.9 BAG, CARTRIDGE AND FIBROUS FILTERS

A bag filter is one that has a non-rigid fabric medium for the filter. Water flow is usually pressure-driven from the inside of the filter bag to the outside. A cartridge filter is one that has a rigid fabric medium or membrane for the filter. In this type of filter, water flow is usually pressure-driven from the outside of the filter to the inside. Bag and cartridge filters are often developed for small systems and for point-of-use filtration applications. They are also sometimes applied as a pretreatment process for membrane filtration.

Bag filters and cartridge filters remove microorganisms by physical straining. The removal efficiency thus depends primarily on the pore size of the filter medium and on the size of the microbes. A typical pore size range is from 0.2 μm to about 10 μm. The pore size of the filter medium is usually designed to be small enough to remove protozoa such as *Cryptosporidium* and *Giardia*. Submicron particles, including viruses and most bacteria, can pass through the filters. As water passes through a bag or cartridge filter, pressure drop increases to a level impractical for operation. The bag or cartridge is then replaced by a clean one.

Since the removal mechanism is physical straining, chemical pretreatment is usually not required for bag filters and cartridge filters. Straining of large compressible particles can blind the filters and reduce filter life. High turbidity and algae can also clog these filters. These processes are therefore only appropriate for high-quality waters. A prefiltration process may be employed to remove large particles.

In principle, microbes larger than the pore size of the medium will be captured by the filters. The nominal size of a filter medium reported by the manufacturers represents an average size — there is often a pore size distribution, meaning that some pores will be larger than the nominal size. Furthermore, some biological particles such as cysts and oocysts do not have hard shells. These microbes may deform slightly, especially under pressure, allowing them to squeeze through small pores. Li et al. (1997) studied the removal of *Cryptosporidium* oocysts using field-scale bag filtration. A bag filter with a single layer of polypropylene fabric, with a nominal pore size of 1 μm, removed an average of 0.42 log of *Cryptosporidium*. This means that 38% of seeded *Cryptosporidium* oocysts passed through the filter, probably for the reasons mentioned above. When the bag filter was changed to multiple layers, the log removal increased to 1.41.

Arora, Di Giovanni & LeChevallier (1999) evaluated the performance of fibrous filters to treat recycled filter backwash water. Two filters, one made of polybutylene terephthalate with a nominal sizing of 5 μm and the other made of nylon with a nominal sizing of 2 μm, were used. The recycled backwash water was pretreated in a sedimentation tank. Pilot runs with the polybutylene terephthalate filter resulted in an average removal of 3 logs for *Cryptosporidium*, 0.5 logs for *Giardia* and 1 log for *Clostridium*. The nylon filter achieved an average removal of 3 logs for *Cryptosporidium*, 1.2 logs for *Giardia* and 1.5 logs for *Clostridium*.

3

Inactivation (disinfection) processes

This chapter covers the various disinfection processes used in drinking-water treatment to inactivate pathogenic microbes. It looks first at factors affecting the efficiency of disinfection process, and then goes on to consider the following disinfection processes:

- *pretreatment oxidation* — in which oxidants are added to water early in the treatment process.
- *primary disinfection* — a common component of primary treatment of drinking-water, and important because granular filter media do not remove all microbial pathogens from water
- *secondary disinfection* — used to maintain the water quality achieved at the treatment plant throughout the distribution system up to the tap.

3.1 FACTORS AFFECTING DISINFECTION

The principal factors that influence disinfection efficiency are disinfectant concentration, contact time, temperature and pH. Disinfectant concentration and

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contact time are integral to disinfection kinetics and the practical application of the CT concept (CT being the disinfectant concentration multiplied by the contact time). Development and derivations of this disinfection model are discussed in the modelling section below. Temperature, over the range appropriate for drinking-water, affects the rate of disinfection reactions according to the Arrhenius equation, although this may not hold for certain disinfectants at low temperatures. The pH of the disinfectant solution affects the reaction kinetics. For example, the disinfection efficiency of free chlorine is increased at lower pH values, whereas that of chlorine dioxide is greater at alkaline pH levels. Monochloramine is formed within seconds in the pH range 7–9, at chlorine to ammonia nitrogen ratios of less than 5:1 and at 25°C; it is also the predominant species when the pH is greater than 5.

Other factors that influence microbial sensitivity to disinfection include attachment to surfaces, encapsulation, aggregation and low-nutrient growth. Increased resistance to disinfection may result from attachment or association of microorganisms to various particulate surfaces, including:

- macroinvertebrates (*Crustacea, Nematoda, Platyhelminthes* and *Insecta*) (Tracy, Camarena & Wing, 1966; Levy, Cheetham & Hart, 1984);
- particles that cause turbidity (LeChevallier, Evans & Seidler, 1981; Ridgway & Olson, 1982);
- algae (Silverman, Nagy & Olson, 1983);
- carbon fines (LeChevallier et al., 1984; Camper et al., 1986);
- glass (Olivieri et al., 1985).

Ridgway & Olson (1982) showed that the majority of viable bacteria in chlorinated water were attached to particles. Stewart & Olson (1986) reported that aggregation of *Acinetobacter* strain EB22 increased its resistance to disinfection, making the bacteria 100-fold more resistant to hypochlorous acid (HOCl) and 2.3-fold more resistant to monochloramine. Several investigators have isolated encapsulated bacteria from chlorinated water (Reilly & Kippin, 1983; Clark, 1984) and concluded that production of the extracellular capsule helped protect bacteria from chlorine. Carson et al. (1972) reported that *Pseudomonas aeruginosa* grown in distilled water was markedly more resistant to acetic acid, glutaraldehyde, chlorine dioxide and a quaternary ammonium compound than cells cultured on tryptic soy agar. Similarly, Berg, Matin & Roberts (1981) and Harakeh et al. (1985) found that bacteria grown in a chemostat at low temperatures and submaximal growth rates caused by nutrient limitation (conditions thought to be similar to the natural aquatic environment) were resistant to several disinfectants.

3.2 PRETREATMENT OXIDATION

Water utilities often add oxidants early in the treatment process to:

- maximize the contact time with the oxidant;
- oxidize compounds for subsequent removal by the treatment process (e.g. iron or manganese);
- provide initial treatment in sufficient time for water to be further treated if necessary (e.g. oxidation of taste and odour compounds);
- control growth of microorganisms and higher organisms (e.g. zebra mussels) on intake structures and in treatment basins;
- improve particle removal in subsequent clarification and filtration processes.

There are a number of potential problems with pretreatment oxidation. Variable source water conditions mean that variable or high levels of oxidant may be needed. This may lead to overdosing of pre-oxidants, which can result in "pink coloured" water when potassium permanganate is misapplied. Also, the process can produce oxidation by-products such as trihalomethanes (THMs), haloacetic acids and bromate. For example, in using chlorine as a pretreatment oxidant, chlorinated by-products can form rapidly. This often limits the application of chlorine to a later stage of the treatment process, when precursor material has been removed. A further problem is that oxidants can lyse algal cells, releasing liver or nerve toxins, or creating objectionable tastes or odours. (Yoo et al., 1995b; Chorus & Bartram, 1999).

One concern with using pre-oxidants for disinfection is that particulate material may interfere with microbial inactivation. Such material protects bacteria and viruses from disinfectants by creating an instantaneous disinfectant demand (preventing the maintenance of a disinfectant residual in subsequent treatment steps) and by shielding the microbe from the oxidant (Hoff, 1978; LeChevallier, Evans & Seidler, 1981; Berman, Rice & Hoff, 1988).

The effect of particulate material on disinfection of cysts or oocysts has not been widely evaluated. Di Giovanni & LeChevallier (2000) studied the effect of turbidity on disinfection of *Cryptosporidium parvum* oocysts by chlorine dioxide or permanganate, and found that particulate material did not interfere with disinfection once the increase in oxidant demand had been satisfied (Figure 3.1). The authors hypothesized that protozoan cysts were too large to be completely shielded from the disinfectant.

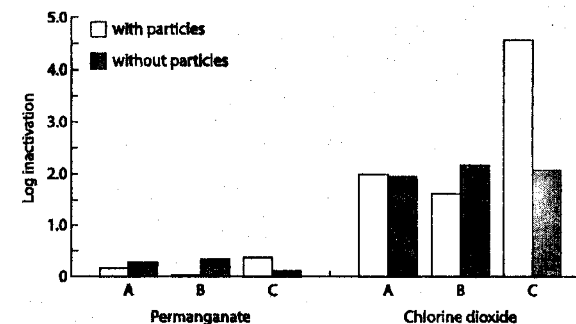


Figure 3.1 Effect of particulate material on disinfection of *Cryptosporidium*. Potassium permanganate applied at 2400 mg/min l^{-1}, chlorine dioxide applied at 120 mg/min l^{-1}. Source: Adapted from Di Giovanni & LeChevallier (2000).

3.3 PRIMARY DISINFECTION

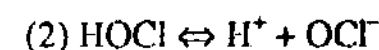
A disinfection barrier is a common component of primary treatment of water. Primary disinfection is typically a chemical oxidation process, although ultraviolet (UV) irradiation and membrane treatment are gaining increased attention. This section looks at different types of disinfectant — chlorine, monochlorine, chlorine dioxide, ozone, UV light and mixed oxidants — in terms of their effectiveness against various pathogenic microorganisms. Further information on selecting a disinfection strategy for a piped distribution system can be found in the WHO publication *Safe piped water: Managing microbial water quality in piped distribution systems* (Ainsworth, 2004).

3.3.1 Chlorine

Mode of action

Chlorine gas and water react to form HOCl and hydrochloric acid (HCl). In turn, the HOCl dissociates into the hypochlorite ion (OCl^-) and the hydrogen ion (H^+), according to the following reactions:

(1) $Cl_2 + H_2O \Leftrightarrow HOCl + HCl$



(2) $HOCl \Leftrightarrow H^+ + OCl^-$

The reactions are reversible and pH dependent:

- between pH 3.5 and 5.5, HOCl is the predominant species
- between about pH 5.5 and 9.5, both HOCl and OCl^- species exist in various proportions
- above pH 8, OCl^- predominates.

The OCl^- and HOCl species are commonly referred to as free chlorine, which is extremely reactive with numerous components of the bacterial cell. HOCl can produce oxidation, hydrolysis and deamination reactions with a variety of chemical substrates, and produces physiological lesions that may affect several cellular processes. Baker (1926) theorized that chlorine destroys microorganisms by combining with proteins to form N-chloro compounds. Chlorine was later found to have powerful effects on sulfhydryl groups of proteins (Green & Stumpf, 1946, Knox et al., 1948; Venkobachar, Iyengar & Rao, 1977) and to convert several α-amino acids by oxidation into a mixture of corresponding nitriles and aldehydes (Patton et al., 1972). The exact product of the reaction depends on chlorine concentration and pH (Dakin 1916, 1917; Wright 1936).

Cytochromes, iron-sulfur proteins and nucleotides are highly vulnerable to oxidative degradation by HOCl, suggesting that chlorine causes physiological damage primarily to the bacterial cell membranes (Venkobachar, Iyengar & Rao, 1977; Camper & McFeters, 1979; Haas & Engelbrecht, 1980; Albrich, McCarthy & Hurst, 1981). Respiration, glucose transport and adenosine triphosphate levels all decrease in chlorine-treated bacteria (Venkobachar, Iyengar & Rao, 1977; Camper & McFeters, 1979; Haas & Engelbrecht, 1980). Electron microscopy of chlorinated bacteria has demonstrated morphological changes in the cell membrane (Zaske, Dockins & McFeters, 1980). In addition, chlorination can kill microbes by disrupting metabolism (Wyss, 1961) and protein synthesis (Pereira et al., 1973), or by modifying purine and pyrimidine bases and thus causing genetic defects (Patton et al., 1972; Hoyano et al., 1973; Haas & Engelbrecht, 1980).

Nearly 100 years of chlorination for disinfection of drinking-water has demonstrated the effectiveness of this process for inactivation of microbial pathogens, with the notable exception of *Cryptosporidium*.

Effectiveness of chlorine against bacteria and viruses

Table 3.1 shows CT values for 99% (2-log) inactivation of bacteria for various chlorine-based disinfectants. In general, the heterotrophic bacteria grown in

drinking-water were more resistant to disinfection than the laboratory-grown *Escherichia coli*.

Table 3.1 Comparative efficiency of disinfectants for the production of 99% bacterial inactivation in oxidant demand-free systems

	Escherichia coli			Heterotrophic bacteria		
Disinfectant	pH	Temp (°C)	CT mg/min l^{-1}	pH	Temp (°C)	CT mg/min l^{-1}
Hypochlorous acid	6.0	5	0.04	7.0	1–2	0.08 ± 0.02
Hypochlorite ion	10.0	5	0.92	8.5	1–2	3.3 ± 1.0
Chlorine dioxide	6.5	20	0.18	7.0	1–2	0.13 ± 0.02
	6.5	15	0.38	8.5	1–2	0.19 ± 0.06
	7.0	25	0.28			
Monochloramine	9.0	15	64	7.0	1–2	94.0 ± 7.0
				8.5	1–2	278 ± 46.0

Source: Adapted from LeChevallier, Cawthon & Lee (1988)

Certain bacteria show a high level of resistance to free chlorine. Spore-forming bacteria such as *Bacillus* or *Clostridium* are highly resistant when disseminated as spores. Acid-fast and partially acid-fast bacteria such as *Mycobacterium* and *Nocardia* can also be highly resistant to chlorine disinfection. One study showed that nearly all of the bacteria surviving chlorine disinfection were Gram positive or acid fast (Norton & LeChevallier, 2000), possibly because Gram-positive bacteria have thicker walls than Gram-negative ones.

Enteric viruses are generally more resistant to free chlorine than enteric bacteria, with CT values for 99% inactivation ranging from about 2 to more than 30 mg/min l^{-1} (Figure 3.2). Viruses associated with cellular debris or organic particles may require high levels of disinfection due to the protective nature of the particle surface (Akin & Hoff, 1986; Hoff, 1992). Chlorination effectively inactivates viruses if the turbidity of the water is less than or equal to 1.0 nephelometric turbidity unit (NTU). It requires a free chlorine residual of 1.0 or greater for 30 minutes, and a pH of less than 8.0. For groundwaters where turbidities are generally low, or for filtered surface water, White (1999) suggests the CT guidelines for the 99% virus inactivation shown in Table 3.2. These data are based on conservative interpretation of inactivation data for Coxsackie A2.

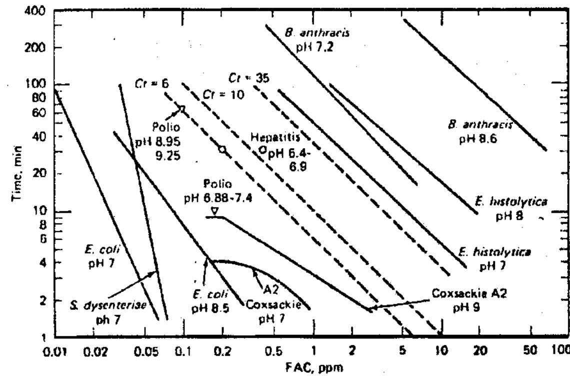


Figure 3.2 Disinfection (2-log) of microorganisms by free available chlorine (White, 1999).

Table 3.2. Disinfection time–chlorine concentration envelopes for 99% virus inactivation at 0–5°C and 10°C

	CT in mg/min l^{-1}	
pH range	0–5°C	10°C
7.0–7.5	12	8
7.5–8.0	20	15
8.0–8.5	30	20
8.5–9.0	35	22

Adapted from White (1999)

Effectiveness of chlorine against protozoa

Protozoan cysts such as *Entamoeba histolytica* and *Giardia lamblia* are highly resistant to chlorine disinfection and may require prolonged contact times at high chlorine residuals (2–3 mg/l) to achieve 99.9% (3-log) inactivation. Clark, Read & Hoff (1989) have described a mathematical model for *Giardia* inactivation that is based on the infectivity data:

$$CT = 0.9847\ C^{0.1758}\ pH^{2.7519}\ temp^{-0.1467}$$

where:

C = the disinfectant residual concentration
temp = the reaction temperature in degrees Celcius

The United States Environmental Protection Agency (USEPA) has published extensive CT tables for *Giardia* inactivation, for different temperature, pH, chlorine residual and other factors (USEPA, 1989b). For example, at a temperature of 25°C and pH 8.0, with a chlorine residual in the range of 1 to 2.6 mg/l, a contact time of 54–65 minutes is needed to achieve a 3-log reduction in *Giardia* (Table 3.3). If the temperature is reduced to 10°C, the contact time increases to 162–194 minutes (Table 3.4), and at 0.5°C it increases further, to 304–368 minutes (Table 3.5).

Table 3.3. Estimated CT values for inactivation of *Giardia* cysts with free chlorine at 25°C

	pH 7 Log inactivation			pH 8 Log inactivation		
Chlorine (mg/l)	1	2	3	1	2	3
1	12	25	37	18	36	54
1.6	13	27	40	19	39	58
2	14	27	41	20	41	61
2.6	15	29	44	22	43	65

Source: Adapted from EPA, 1990.

Table 3.4. Estimated CT values for inactivation of *Giardia* cysts with free chlorine at 10°C

	pH 7 Log inactivation			pH 8 Log inactivation		
Chlorine mg/l	1	2	3	1	2	3
1	37	75	112	54	108	162
1.6	40	79	119	58	116	174
2	41	83	124	61	121	182
2.6	44	87	131	65	129	194

Source: Adapted from EPA, 1990.

Table 3.5. Estimated CT values for inactivation of *Giardia* cysts with free chlorine at 0.5°C

	pH 7			pH 8		
	Log inactivation			Log inactivation		
Chlorine mg/l	1	2	3	1	2	3
1	70	140	210	101	203	304
1.6	75	151	226	110	219	329
2	79	157	236	115	231	346
2.6	84	168	252	123	245	368

Source: Adapted from EPA, 1990.

E. histolytica cysts were inactivated at pH 7.0 in 10 minutes at 25°C with a residual of 3.5 mg/l (Chang, 1982). At pH 4, 30°C and 10 minutes of exposure, 2 mg/l of free chlorine produced a 99.9% reduction of cysts; however, if the pH was increased to 10, a chlorine concentration of 12 mg/l was needed to achieve the same 3-log reduction. Data on other emerging protozoan pathogens are lacking, although a recent report indicated that the microsporidian *Encephalitozoon* syn. *Septata intestinalis* was inactivated by more than 3 logs when exposed to 2 mg/l chlorine for 16 min at pH 7 and 25°C (Wolk et al. 2000).

Chlorine-based disinfectants are generally not effective at inactivation of *Cryptosporidium* (Table 3.6) and early studies found that *Cryptosporidium* oocysts were resistant to a variety of hospital disinfectants, including bleach (Campbell et al., 1982). Chlorine disinfection has not been effective in preventing outbreaks of cryptosporidiosis caused by *Cryptosporidium* in drinking-water and recreational water. Korich et al. (1990) reported that 80 mg/l of free chlorine or monochloramine required 90 minutes to achieve 90% inactivation of oocysts, and suggested that conventional disinfection practices would do little to inactivate waterborne *Cryptosporidium*. However, Rasmussen et al. (1994) examined the disinfection effectiveness of several biocides and found that inactivation of oocysts required an oxidation/reduction potential of about 800 mV, maintained for 30 minutes (Table 3.6). These authors suggest that oxidation/reduction potential is more important than CT for oocyst inactivation.

Table 3.6 Summary of free chlorine and monochloramine disinfection results for *Cryptosporidium*

Chlorine residual (mg/l)	Contact time (min)	CT product (mg/min.l^{-1})	Temp (°C)	pH	Per cent inactivation	Analytical method
Free chlorine						
80[a]	90	7200	25	7	> 99	Mouse infectivity
15[b]	240	3600	22	8	47	Mouse infectivity
968[c]	1440	1,393,920	10	7	85	Excystation
17[d,e]	30	510	NR	NR	99	Excystation
Monochloramine						
80[a]	90	7200	25	7	99	Mouse infectivity
15[b]	240	3600	22	8	99.6	Mouse infectivity
3.75[c]	1440	5400	10	7	80.5	Excystation

NR = not reported
[a] Korich et al. (1990)
[b] Finch, Kathleen & Gyurek (1994)
[c] Ransome, Whitmore & Carrington (1993)
[d] Rasmussen et al. (1994)
[e] Estimated chlorine residual to achieve an oxidation-reduction potential of 800 mV

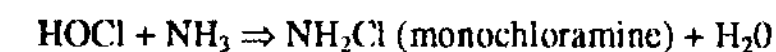
By-products of disinfection with chlorine

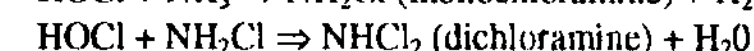
THMs and other halogenated compounds are the main by-products of disinfection with chlorine. Factors affecting the formation of THMs are discussed in *Safe piped water: Managing microbial water quality in piped distribution systems* (Ainsworth, 2004).

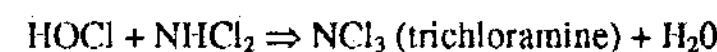
3.3.2 Monochloramine

Mode of action

In dilute aqueous solutions (1–50 mg/l), chlorine reacts with ammonia in a series of bimolecular reactions:


$HOCl + NH_3 \Rightarrow NH_2Cl$ (monochloramine) $+ H_2O$


$HOCl + NH_2Cl \Rightarrow NHCl_2$ (dichloramine) $+ H_2O$



$HOCl + NHCl_2 \Rightarrow NCl_3$ (trichloramine) $+ H_2O$

These competing reactions are dependent upon pH and the relative chlorine to nitrogen concentration (expressed as Cl_2:N). To a lesser degree they are also dependent upon temperature and contact time. The reaction of HOCl and ammonia will convert all the free chlorine to monochloramine at pH 7–8 when the Cl_2:N ratio is equimolar (5:1 by weight) or less.

Ingols (1958) examined the reaction of monochloramine with several amino acids and tripeptides. Exposure of alanine, tyrosine and glycylgylcylgylcine to the disinfectant for several hours at 25°C and pH 8.0 converted these compounds to organic chloramines. The sulfhydryl groups of cystine were oxidized to disulfides (by comparison, exposure of the same compounds to HOCl produced a variety of oxidized, hydrolysed or deaminated reactants). Reaction of monochloramine with hemin (an important component of enzymes such as cytochromes, catalases and peroxidases) resulted in products that could not be reactivated by reducing compounds. The author concluded that monochloramine may kill bacterial cells by reacting primarily with membrane-bound enzymes.

Jacangelo & Olivieri (1985) examined the reaction of monochloramine with amino acids, nucleic acids, nucleotides, nucleosides, purine and pyrimidine bases, and ribose sugars. Monochloramine was most reactive with sulfur-containing amino acids and tryptophan. When the sulfhydryl groups of cysteine were in excess, 1 mol of monochloramine reacted with 2 mol of cysteine to form 1 mol of the cystine disulfide. When monochloramine was in excess, the reaction proceeded beyond the disulfide state.

Watters et al. (1989) extended the observations of Jacangelo & Olivieri (1985) by examining whole cells. They found that *Enterobacter cloacae* could be reactivated after exposure to chloramine by addition of sodium sulfite, and hypothesized that sodium sulfite could reduce oxidized disulfides, or result in other types of oxidative injury. Interestingly, sodium sulfite had no effect on organisms exposed to free chlorine. The results suggest that free chlorine and chloramine react with different functional groups in the cell membrane.

Jacangelo & Olivieri (1985) found that monochloramine reacted more slowly with nucleic acids and free purine and pyrimidine bases than with amino acids. These results support the observation that many viruses are inactivated more slowly than bacterial cells. Berman & Hoff (1984) showed that simian rotavirus SA11 required more than 6 hours contact with 10 mg/l preformed monochloramine at pH 8.0 to achieve 99% inactivation. Shih & Lederberg (1976) found that exposure of *Bacillus subtilis* deoxyribonucleic acid (DNA) to monochloramine induced single and double stranded breaks, reduced the transforming activity of DNA and enhanced the sensitivity of DNA to endonuclease cleavage.

Effectiveness of monochloramine

Monochloramine is not recommended as a primary disinfectant because of its weak disinfecting power (Table 3.1). This disinfectant is not effective for inactivation of *Cryptosporidium* (Table 3.6). In systems using monochloramine, free chlorine is usually applied for a short time before addition of ammonia, or an alternative primary disinfectant is used (e.g. ozone, chlorine dioxide).

By-products of disinfection with monochloramine

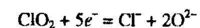
Treatment to produce a monochloramine residual poses the risk of nitrite formation in the distribution system, especially in low-flow stagnant areas, because bacteria on surfaces and in deposits may nitrify any slight excess of ammonia.

3.3.3 Chlorine dioxide

Chlorine dioxide is a strong oxidant that can be used to control iron, manganese and taste and odour causing compounds. It has also been used as a secondary disinfectant in many European countries.

Mode of action

Chlorine dioxide is highly soluble in water (particularly at low temperatures), and is effective over a range of pH values (pH 5–10). Theoretically, chlorine dioxide undergoes five valence changes in oxidation to chloride ion:



$$ClO_2 + 5e^- = Cl^- + 2O^{2-}$$

However, in practice, chlorine dioxide is rarely reduced completely to chloride ion (White 1999). Chlorine dioxide is thought to inactivate microorganisms through direct oxidation of tyrosine, methionyl, or cysteine-containing proteins, which interferes with important structural regions of metabolic enzymes or membrane proteins (Gates 1998). In water treatment, chlorine dioxide has the advantage of being a strong disinfectant, but not forming THMs or oxidizing bromide to bromate.

Effectiveness of chlorine dioxide against bacteria and viruses

Chlorine dioxide is roughly comparable to free chlorine for inactivation of bacteria and viruses at neutral pH (White, 1999), but is more effective than free chlorine at pH 8.5 (Hoff & Geldreich, 1981).

Effectiveness of chlorine dioxide against protozoa

Chlorine dioxide is an effective disinfectant for control of *Giardia lamblia*; the required CT values for 1-log inactivation (pH 6–9) range from 5 mg min/l at 20°C to 21 mg/min l^{-1} at 0.5°C (USEPA, 1989b; White, 1999). The 3-log inactivation CT values (pH 6–9) range from 19 mg/min l^{-1} at 15°C to 63 mg/min l^{-1} at 0.5°C. These values are 3–14 times less than those required for free chlorine, but approximately 20 times more than those required for ozone.

Figure 3.3 summarizes results from various studies of *Cryptosporidium* inactivation by chlorine dioxide. Peeters et al. (1989) reported 1.5 and 1.2-log inactivation of *Cryptosporidium*, using an animal infectivity method, for CT values of 3 and 9.8 mg/min l^{-1}, respectively (average of initial and final concentrations). Korich et al. (1990) reported a CT value of 78 mg/min l^{-1}, with an initial concentration of 1.3 mg/l and a contact time of 60 minutes, for a 90% (1-log) inactivation of *Cryptosporidium*, based on mouse infectivity. The CT for 1-log inactivation was calculated to be 51 mg/min l^{-1} (average of initial and final concentrations). Finch, Liyanage & Belosevic (1995) recalculated the Korich data using a dose–response model developed for CD-1 mice, and estimated a 99% (or 2-log) inactivation. Ransome, Whitmore & Carrington (1993), employing the excystation viability method, reported *Cryptosporidium* inactivation ranging from 0.14 to 1.4-log for average CT values ranging from 6.5 to 67.5 mg/min l^{-1}, respectively. Based on results from 12 animal infectivity experiments, Finch et al. (1997) reported *Cryptosporidium* inactivation ranging from 0 to greater than 3.2-log for average CT values ranging from 12.5 to 212 mg/min l^{-1}. Chlorine dioxide concentration decreased markedly at contact times of more than 30 minutes, a factor that could result in low CT values. LeChevallier et al. (1996) found that oocysts were more rapidly inactivated by chlorine dioxide at pH 8.0 than at pH 6.0, and that effectiveness was reduced by 40% when temperature was reduced from 20°C to 10°C. This finding is supported by other studies (Bernard et al., 1965; Owens et al., 1999; Ruffle, Rennecker & Marinas, 1998). Chlorine dioxide inactivation rates using a cell culture technique to determine infective oocysts were similar to rates generated using animal infectivity tests.

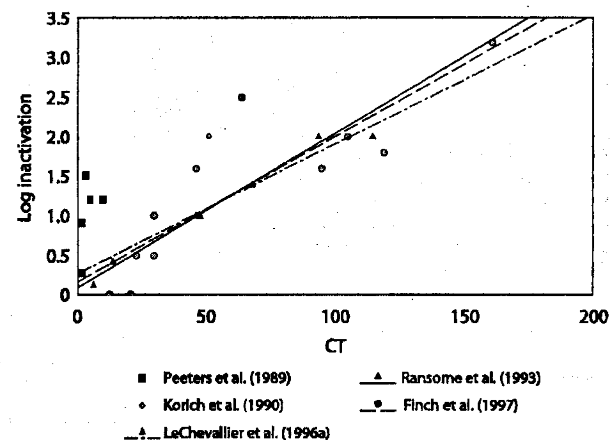


Figure 3.3 Summary of *Cryptosporidium* inactivation by chlorine dioxide

By-products of disinfection with chlorine dioxide

The chlorine in chlorine dioxide exists in a +4 oxidation state, compared to an oxidation state of +1 for chlorine in free chlorine (in hypochlorous and hypochlorite ions). This means that chlorine and chlorine dioxide have different pathways for disinfection and formation of by-products when used in drinking-water treatment. For example, chlorine dioxide does not produce significant levels of halogenated organic by-products.

Chlorine dioxide forms undesirable inorganic by-products (chlorite and chlorate ions) upon its reaction with constituents of water such as dissolved organic carbon, microbes and inorganic ions. Therefore, a water utility may need to provide additional treatment depending on the level of these inorganic by-products and their specific regulatory requirements (Gordon & Bubnis, 1995; WHO, 2000).

3.3.4 Ozone

Ozone has been used for more than a century for water treatment, mostly in Europe, although its use is now spreading to other countries.

Mode of action

The mechanism by which ozone inactivates microbes is not well understood. Ozone in aqueous solution may react with microbes either by direct reaction with molecular ozone or by indirect reaction with the radical species formed when ozone decomposes. Ozone is known to attack unsaturated bonds, forming aldehydes, ketones or carbonyl compounds (Langlais, Reckhow & Brink, 1991). Additionally, ozone can participate in electrophilic reactions, particularly with aromatic compounds, and in nucleophilic reactions with many of the components of the microbial cell. Carbohydrates and fatty acids react only slightly with ozone, but amino acids, proteins, protein functional groups (e.g. disulfide bonds) and nucleic acids all react very quickly with it (Langlais, Reckhow & Brink, 1991). It is likely, therefore, that microbes become inactivated through ozone acting on the cytoplasmic membrane (due to the large number of functional proteins), the protein structure of a virus capsid, or nucleic acids of microorganisms.

Free radicals formed by the decomposition of ozone are generally less effective for microbial inactivation than molecular ozone, because microbial cells contain a high concentration of bicarbonate ions that quench the free radical reaction, and many microbial cells also contain catalase, peroxidase, or superoxide dismutase to control the free radicals produced by aerobic respiration. In addition, some bacteria contain carotenoid and flavonoid pigments that protect them from ozone. These factors can account for reports that heterotrophic bacteria may be less susceptible to ozone inactivation than *Giardia* (Wolfe et al., 1989). Studies of peroxone (a mixture of ozone and hydrogen peroxide that promotes the generation of hydroxyl free radicals) showed that peroxone was comparable to ozone, or slightly more potent, when CTs were based on ozone residuals (Wolfe et al., 1989). These results suggest that free radicals provide little benefit in terms of microbial destruction.

Effectiveness of ozone against bacteria and viruses

Of the vegetative bacteria, *Escherichia coli* is one of the most sensitive (Table 3.7), while Gram-positive cocci (*Staphylococcus* and *Streptococcus*), Gram-positive bacilli (*Bacillus*) and mycobacteria are the most resistant (Langlais, Reckhow & Brink, 1991). *Mycobacterium avium* can be effectively controlled by low doses of ozone ($CT_{99.9}$ of 0.1–0.2 mg/min l^{-1}), whereas the

organism is highly resistant to free chlorine ($CT_{99.9}$ of 551–1552 mg/min l^{-1} for water-grown isolates) (Taylor et al., 2000).

Table 3.7 CT values (mg/min l^{-1}) for 99% inactivation at 5°C

Microorganism	Free chlorine (pH 6–7)	Preformed chloramines (pH 8–9)	Chlorine dioxide (pH 6–7)	Ozone (pH 6–7)
E. coli	0.034–0.05	95–180	0.4–0.75	0.02
Poliovirus 1	1.1–2.5	770–3740	0.2–6.7	0.1–0.2
Rotavirus	0.01–0.05	3810–6480	0.2–2.1	0.006–0.06
Phage f2	0.08–0.18	–	–	–
G. lamblia cysts	47–>150	–	–	0.5–0.6
G. muris cysts	30–630	1400	7.2–18.5	1.8–2.0

Adapted from Hoff (1986)

Viruses are generally more resistant to ozone than vegetative bacteria, although phage appear to be more sensitive than human viruses (Langlais, Reckhow & Brink, 1991).

Effectiveness of ozone against protozoa

For the protozoa *Giardia lamblia* and *Naegleria gruberi*, ozone inactivation (Table 3.7) did not follow linear kinetics, due to an initial latent phase. However, CT products could be reasonably estimated with a CT_{99} (a CT for 99% inactivation) of 0.53 and 4.23 mg/min l^{-1}, respectively, at 5°C (Wickramamayake, Rubin & Sproul, 1984).

Ozone is effective for removal of *Cryptosporidium* (Table 3.8). Noticeable for *Cryptosporidium* is the impact of the analytical method on the CT values. Generally, excystation and vital staining are more conservative measures of oocyst inactivation than animal infectivity. Reliance on excystation and vital staining alone could greatly overestimate disinfection requirements for *Cryptosporidium*. On average, 4.5 mg/min l^{-1} CT was required for 99% oocyst inactivation (measured by mouse infectivity) by ozone at 20–25°C (Table 3.8). However, Finch et al. (1993) indicated that the conventional method of determining CT by using the final concentration of reactants at the end of the contact time overestimates the CT needed for disinfection and unduly increases treatment costs. The authors recommended the Holm disinfection model, which integrates the disinfectant concentration and time throughout the reactor. Using this alternative calculation, CT for *Cryptosporidium* inactivation were 6.9 mg/min l^{-1} at 7°C and 2.4 mg/min l^{-1} at 22°C.

Table 3.8 Summary of ozone disinfection results for *Cryptosporidium*

Ozone residual (mg/l)	Contact time (min)	CT product (mg/min l^{-1})	Temp °C	Per cent inactivation	Analytical method
1[a]	5	5	25	90–99	Mouse
1[a]	10	10	25	>99	infectivity
0.77[b]	6	4.6	'Room'	>99	Mouse
0.51[b]	8	4.1	'Room'	>99	infectivity
0.16–1.3[c]	5–15	7	7	99	Mouse
0.17–1.9[c]	5–15	3.5	22	99	infectivity
2.4 (avg)[d]	2.3	5.5	22–25	99	Mouse infectivity
1.25[e]	15	18.75	10	98.6	Excystation
4 (approx)[f]	2	8	'Room'	>90	Excystation
1–5[g]	10	10–50	5	18–39	Stain
1–5[g]	10	10–50	20	70–>99	
0.7–1.5[h]	14–25	9.8–27	8–10	42–84	Stain

[a] Korich et al. (1990)
[b] Peeters et al. (1989)
[c] Finch et al. (1993)
[d] Owens et al. (1994)
[e] Ransome, Whitmore & Carrington (1993)
[f] Armstrong et al. (1994)
[g] Parker, Greaves & Smith (1993)
[h] Hall, Pressdee & Carrington (1994)

Chlorine dioxide, chlorite, and chlorate

History | Summary

Information extracted from:

Guidelines for drinking-water quality, 2nd ed.
Vol. 2. ***Health criteria and other supporting information.***
Geneva, World Health Organization, 1996. pp. 803-816.

- General description
- Analytical methods
- Environmental levels and human exposure
- Kinetics and metabolism in laboratory animals and humans
- Effects on experimental animals and in vitro test systems
- Effects on humans
- Guideline value

1. General description

Identity

Compound	*CAS no.*	*Molecular formula*
Chlorine dioxide	10049-04-4	ClO_2
Chlorite (sodium salt)	7758-19-2	$NaClO_2$
Chlorate (sodium salt)	7775-09-0	$NaClO_3$

Physicochemical properties (1–3)

Property	*Chlorine dioxide*	*Sodium chlorite*	*Sodium chlorate*
Boiling point (°C)	11	–	>300 (decomposes)
Melting point (°C)	-59	180–200 (decomposes)	248

Density at 0 °C (g/cm³)	1.64 (liquid)	–	2.5
Vapour pressure at 25 °C	–	Negligible	–
Water solubility (g/litre)	3.01 (25 °C)	390 (17 °C)	–

Note: Conversion factor in air for chlorine dioxide: 1 ppm = 2.8 mg/m^3.

Organoleptic properties

The taste and odour threshold for chlorine dioxide in water is 0.4 mg/litre (*3*).

Major uses

Chlorine dioxide is used for disinfection and odour/taste control of water; as a bleaching agent for cellulose, paper pulp, flour, and oils; and for cleaning and detanning leather. Sodium chlorite is used in on-site production of chlorine dioxide; as a bleaching agent in production of paper, textiles, and straw products; and in the manufacture of waxes, shellacs, and varnishes. Sodium chlorate is used in the preparation of chlorine dioxide; in the manufacture of dyes, matches, and explosives; for tanning and finishing leather; and in herbicides and defoliants (*1–3*).

Environmental fate

Chlorine dioxide rapidly decomposes into chlorite, chlorate, and chloride ions in treated water, chlorite being the predominant species. This reaction is favoured by alkaline conditions.

2. Analytical methods

Methods are available for the determination of chlorine dioxide, chlorite, and total available chlorine (*4,5*). The limits of detection for these methods are 8 μg/litre for chlorine dioxide, 4 μg/litre for total chlorine, and 10 μg/litre for chlorite and chlorate.

3. Environmental levels and human exposure

Water

Chlorite occurs in drinking-water when chlorine dioxide is used for purification purposes. The levels of chlorite in water reported in one study ranged from 3.2 to 7.0 mg/litre (*6*).

Food

Chlorine dioxide, chlorite, and chlorate may occur in foodstuffs as a result of their use in flour processing, as a decolorizing agent for carotenoids and other natural pigments (chlorine dioxide), as a bleaching agent in the preparation of modified food starch (sodium chlorite), as an indirect additive in paper and paperboard products used for food packaging (sodium chlorite), and as a defoliant, desiccant, and fungicide in agriculture (sodium chlorate) (*7–9*).

Estimated total exposure and relative contribution of drinking-water

The major route of environmental exposure to chlorine dioxide, sodium chlorite, and sodium chlorate is through drinking-water.

4. Kinetics and metabolism in laboratory animals and humans

Chlorine dioxide is rapidly absorbed from the gastrointestinal tract. No particular organ appears to selectively concentrate the dose following exposure (*10*). Following oral ingestion by monkeys, chlorine dioxide was rapidly converted into chloride ion and, to a lesser extent, chlorite and chlorate (*11*). Excretion is mainly via the urine, smaller amounts being excreted in faeces (*12*).

Chlorite was readily absorbed when administered to rats, then randomly distributed throughout the tissues (*12*). It was transformed mainly into chloride in rats, smaller amounts appearing as unchanged chlorite. Excretion was mainly via the urine, followed by faeces (*13*).

Chlorate was readily absorbed and randomly distributed throughout the tissues of rats (*12*). It was excreted mainly in the form of chloride in the urine, smaller amounts appearing as chlorite and chlorate (*13*).

5. Effects on laboratory animals and *in vitro* test systems

Chlorine dioxide

Chlorine dioxide

Short-term exposure

Drinking-water containing 0, 10, or 100 mg of chlorine dioxide per litre (equivalent to approximately 0, 1.5, or 15 mg/kg of body weight per day) was administered to mice (10 per dose) for 30 days with no apparent effects on blood parameters. The NOAEL for this study was 15 mg/kg of body weight per day (14).

A total of 12 African green monkeys were exposed to water containing chlorine dioxide at concentrations of 0, 30, 100, or 200 mg/litre (corresponding to measured doses of 0, 3.5, 9.5, or 11 mg/kg of body weight per day) using a rising dose protocol. Each dose was maintained for 30–60 days. A slight suppression of thyroid function (decreased thyroxine) was observed in monkeys receiving the two highest doses. No other effects were noted. The NOAEL was 3.5 mg/kg of body weight per day (11).

Six monkeys were treated for 8 weeks with drinking-water containing chlorine dioxide at 100 mg/litre, corresponding to an average measured dose of about 4.6 mg/kg of body weight per day. Thyroxine level was reduced after 4 weeks of treatment but rebounded after a further 4 weeks. In the same study, drinking-water containing chlorine dioxide at 0, 100, or 200 mg/litre was administered to male rats (12 per dose) (equivalent to 0, 10, or 20 mg/kg of body weight per day). A dose-dependent decrease in thyroxine levels was observed after 8 weeks of treatment; there was no rebound. The exposure level of 100 mg/litre, equivalent to a dose of approximately 10 mg/kg of body weight per day, was the LOAEL in this study (15).

Sprague-Dawley rats (10 per sex per dose) were exposed to 0, 25, 50, 100, or 200 mg of chlorine dioxide per litre in drinking-water for 90 days (approximate dose levels of 0, 2, 4, 6, or 12 mg/kg of body weight per day for males and 0, 2, 5, 8, or 15 mg/kg of body weight per day for females). Water consumption was decreased in both sexes at the three highest dose levels, probably due because of its reduced

palatability. Food consumption was decreased in males receiving the highest dose. Goblet-cell hyperplasia was significantly increased in the nasal turbinates of females given 100 or 200 mg/litre and males at all doses. Inflammation of the nasal cavity was observed in males at 25 mg/litre and in both sexes at higher doses. The authors concluded that the lowest dose (2 mg/kg of body weight per day) was a LOAEL (16).

Long-term exposure

In a drinking-water study, chlorine dioxide was administered to rats (7 per sex per dose) at concentrations of 0, 0.5, 1, 5, 10, or 100 mg/litre (highest dose equivalent to about 13 mg/kg of body weight per day) for 2 years. At the highest dose level, survival rate was substantially decreased in both sexes, and mean life span was reduced compared with that for control animals. No correlation was observed between treatment and histopathological findings. In this study, a NOAEL of 10 mg/litre (1.3 mg/kg of body weight per day) was identified (17).

Reproductive toxicity, embryotoxicity, and teratogenicity

Female rats were exposed to 0, 1, 10, or 100 mg of chlorine dioxide per litre in drinking-water (equivalent to 0, 0.1, 1, or 10 mg/kg of body weight per day) for 2.5 months before mating and throughout gestation. At the highest dose, there was a slight reduction in the number of implants and live births per pregnancy. No effects were observed at 1 mg/kg of body weight per day, which was identified as the NOAEL (18).

Female Sprague-Dawley rats (13–16 per dose) were supplied with drinking-water containing 0, 2, 20, or 100 mg of chlorine dioxide per litre from 2 weeks before mating to gestation and lactation until pups were weaned on postnatal day 21. No significant effect on the body weight of either the dams or the pups was observed at any dose tested. At 100 mg/litre (14 mg/kg of body weight per day for the pregnant dam), a significant depression of serum thyroxine and an increase in serum triiodothyronine were observed in the pups at weaning, but not in the dams. Neurobehavioural exploratory and locomotor activities were decreased in pups born to dams exposed to 100 mg/litre but not to those exposed to 20 mg/litre (3 mg/kg of body weight per day), which was considered a NOAEL (19).

In a second experiment, rat pups were exposed directly (by gavage) to 14 mg of chlorine dioxide per kg of body weight per day (equivalent to the dose received by a pregnant dam drinking water containing 100 mg of chlorine dioxide per litre) on postnatal days 5–20. In this study, serum thyroxine levels were depressed, a somewhat greater and more consistent delay in the development of exploratory and locomotor activity was seen, and pup body weight gain was reduced. The decrease in serum triiodothyronine levels was not statistically significant. Based on decreased pup development and decreased thyroid hormone levels, a LOAEL of 14 mg/kg of body weight per day (the only dose tested) was identified (19).

Cell number was significantly depressed in the cerebellum of 21-day-old rat pups born to dams supplied during gestation and lactation with water containing 100 mg of chlorine dioxide per litre (about 14 mg/kg of body weight per day to the dam). A group of 12 rat pups dosed directly by gavage with 14 mg/kg of body weight per day had depressed cell numbers in both the cerebellum and forebrain at postnatal day 11 and displayed decreased voluntary running-wheel activity at postnatal days 50–60, despite the fact that chlorine dioxide treatments were terminated at 20 days of age. These data suggest that chlorine dioxide is capable of influencing brain development in neonatal rats. In this study, a LOAEL of 14 mg/kg of body weight per day, the only dose tested, was identified (20).

The developmental neurotoxic potential of chlorine dioxide was evaluated in a study

in which it was administered to rat pups by oral intubation at 14 mg/kg of body weight per day on postnatal days 1–20. Forebrain cell proliferation was decreased on postnatal day 35, and there were decreases in forebrain weight and protein content on postnatal days 21 and 35. Cell proliferation in the cerebellum and olfactory bulbs was comparable to that in untreated controls, as were migration and aggregation of neuronal cells in the cerebral cortex. Histopathological examination of the forebrain, cerebellum, and brain stem did not reveal any lesions or changes in these tissues. In this study, a LOAEL of 14 mg/kg of body weight per day (the only dose tested) was identified (18).

Mutagenicity and related end-points

Chlorine dioxide was mutagenic in Salmonella typhimurium strain TA100 in the absence of a metabolic activation system (21). No sperm-head abnormalities were observed in male mice following chlorine dioxide gavage (22). No chromosomal abnormalities were seen in either the micronucleus test or a cytogenetic assay in mouse bone marrow cells following gavage dosing with chlorine dioxide (22).

Carcinogenicity

Tumours were not observed in rats following 2-year exposures to chlorine dioxide in drinking-water (17).

Chlorite

Acute exposure

An oral LD_{50} of 105 mg/kg of body weight has been reported in rats (23). Quail were more resistant than rats; the LD_{50} was 493 mg/kg of body weight (24).

Short-term exposure

Single doses of sodium chlorite administered orally to cats produced methaemoglobinaemia (25). A dose of 20 mg of chlorite per litre (equivalent to approximately 1.5 mg of chlorite per kg of body weight) caused up to 32% of the haemoglobin to be in the methaemoglobin state and was considered to be the LOAEL. A dose-dependent increase in methaemoglobinaemia and anaemia was observed in 12 African green monkeys treated with sodium chlorite at 0, 25, 50, 100, or 400 mg/litre in drinking-water using a rising dose protocol. Doses of chlorite were approximately 0, 3, 6, 13, and 50 mg/kg of body weight per day, and each dose level was maintained for 30–60 days (11).

Rats were exposed to chlorite ion at 0, 10, 50, 100, 250, or 500 mg/litre in drinking-water (equivalent to 0, 1, 5, 10, 25, or 50 mg/kg of body weight per day) for 30–90 days. Haematological parameters were monitored, and the three highest concentrations produced transient anaemia. At 90 days, red blood cell glutathione levels in the 100 mg/litre group were 40% below those of controls; there was at least a 20% reduction in the rats receiving 50 mg/litre. In this study, a NOAEL of 1 mg/kg of body weight per day was identified (25).

Long-term exposure

The effect of sodium chlorite in drinking-water at 0, 1, 2, 4, 8, 100, or 1000 mg/litre on the survival and postmortem pathology of albino rats (7 per sex per dose) was examined in a 2-year study. The life span of the animals was not significantly affected at any dose. No effects were observed in animals exposed to 8 mg/litre (0.7 mg/kg of body weight per day) or less. Animals exposed to 100 or 1000 mg/litre (9.3

or 81 mg/kg of body weight per day) exhibited treatment-related renal pathology; the author concluded that this was the result of a nonspecific salt effect (17).

Reproductive toxicity, embryotoxicity, and teratogenicity

Female mice (10 per dose) were treated with sodium chlorite at 0 or 100 mg/litre in drinking-water (equivalent to 0 and 72 mg/kg of body weight per day) from day 1 of gestation and throughout lactation. Conception rates were 56% for controls and 39% for treated mice. The body weights of pups at weaning were reduced in treated mice relative to controls, so that 72 mg/kg of body weight per day is the LOAEL for this study (14).

In a series of experiments, sodium chlorite was administered to male rats (12 rats per dose) in drinking-water for 66–76 days at concentrations of 0, 1, 10, 100, or 500 mg/litre (equivalent to 0, 0.1, 1, 10, or 50 mg/kg of body weight per day). No compound-related abnormalities were observed on histopathological examination of the reproductive tract. Abnormal sperm morphology and decreased sperm motility were seen at the two highest dose levels, but no sperm effects were observed at 1 mg/kg of body weight per day, which can be identified as the NOAEL. In another part of the same study, male rats were bred with female rats treated at the same dose levels for 2 weeks before and throughout a 10-day breeding period. Females were exposed to sodium chlorite throughout gestation and lactation until the pups were weaned on day 21. There was no evidence of any adverse effects on conception rates, litter size, day of eye opening, or day of vaginal opening. Based on reproductive effects, a NOAEL of 10 mg/kg of body weight per day, the highest dose tested, was identified (26).

Treatment of maternal mice with 100 mg of sodium chlorite per litre in drinking-water (equivalent to 14 mg of chlorite per kg of body weight per day) throughout gestation and lactation resulted in pups with decreased body weights (14% below those of controls) at weaning. In this study, a LOAEL for developmental effects of 14 mg/kg of body weight per day was identified (14).

Fetuses from maternal rats exposed to chlorite ion via drinking-water at levels of up to 10 mg/litre (about 1 mg/kg of body weight per day) were examined. No compound-related skeletal or soft-tissue anomalies were observed. A NOAEL of 1 mg/kg of body weight per day was identified (27).

Mutagenicity and related end-points

No chromosomal abnormalities were seen in either the micronucleus test or a cytogenetic assay in mouse bone marrow cells following gavage dosing with chlorite (22).

Carcinogenicity

In a long-term study in which mice received sodium chlorite in drinking-water for 85 weeks, there was no significant increase in tumours as compared with controls at a dose of 250 mg/litre (about 36 mg of chlorite ion per kg of body weight per day). Although treated male mice exhibited an increased incidence of lung and liver tumours, tumour rates were within historical ranges for control mice, increases in the liver tumours did not display a typical dose–response pattern, and significant increases were seen only for benign tumours (28). Tumours were not observed in rats following 2-year exposures to sodium chlorite in drinking-water (17).

Chlorate

Acute exposure

An acute oral dosing study in dogs demonstrated lethality at levels of sodium chlorate as low as 600 mg of chlorate ion per kg (29).

Short-term exposure

Beagle dogs (4 per sex per dose) were exposed by gavage to sodium chlorate at doses of 0, 10, 60, or 360 mg/kg of body weight per day for 3 months. There was no significant effect at any dose level on body weight, food consumption, clinical chemistry, organ weights, ophthalmic effects, gross necropsy, or tissue histopathology. Haematological changes were limited to a slight elevation in methaemoglobin level in highest-dose animals, but this appeared to be within normal limits and was not judged to be treatment-related. In this study, a NOAEL of 360 mg/kg of body weight per day in dogs was identified (30).

Sprague-Dawley rats (14 per sex per dose) were exposed by gavage to sodium chlorate at doses of 0, 10, 100, or 1000 mg/kg of body weight per day for up to 3 months. No treatment-related effects were observed on mortality, physical appearance or behaviour, body weight, food consumption, clinical chemistry, gross necropsy, or organ histopathology. At the highest dose, haematological changes indicative of anaemia included decreases in erythrocyte count, haemoglobin concentration, and erythrocyte volume fraction (haematocrit). In this study, a NOAEL of 100 mg/kg of body weight per day was identified (31).

Reproductive toxicity, embryotoxicity, and teratogenicity

Sodium chlorate was administered to pregnant CD rats by gavage at doses of 0, 10, 100, or 1000 mg/kg of body weight per day on days 6–15 of gestation. There were no maternal deaths in treated animals or treatment-related effects on maternal body weight gain, food consumption, clinical observations, number of implantations, or gross necropsy. Examination of fetuses on day 20 revealed no effects on fetal weight or sex ratio, and no external, visceral, or skeletal abnormalities were detected. In this study, a developmental NOAEL of 1000 mg/kg of body weight per day in rats was identified (32).

Mutagenicity and related end-points

No chromosomal abnormalities were seen in either the micronucleus test or a cytogenetic assay in mouse bone marrow cells following gavage dosing with chlorate (22).

6. Effects on humans

Chlorine dioxide

Six different doses of chlorine dioxide (0.1, 1, 5, 10, 18, or 24 mg/litre) in drinking-water were administered to each of 10 male volunteers using a rising dose protocol. Serum chemistry, blood count, and urinalysis parameters were monitored. A treatment-related change in group mean values for serum uric acid was observed, which the authors concluded was not physiologically detrimental. The highest dose tested, 24 mg/litre (about 0.34 mg/kg of body weight per day), can be identified as a single-dose NOAEL (33).

The same male volunteers drank 0.5 litres of water containing 5 mg of chlorine dioxide per litre each day for approximately 12 weeks, and were then kept under observation for 8 weeks. Serum chemistry, blood counts, and urinalysis revealed no abnormalities, except for a slight change in blood urea nitrogen, which the authors concluded was of doubtful physiological or toxicological significance. This exposure, equivalent to 36 µg/kg of body weight per day, can be considered a

NOAEL (33).

In a prospective study of 197 persons, a portion of the population of a rural village exposed for 12 weeks to a chlorine dioxide-treated water supply (containing 0.25–1.1 mg of chlorine dioxide per litre and 0.45–0.91 mg of free chlorine per litre) experienced no significant changes in haematological parameters, serum creatinine, or total bilirubin (6).

Chlorite

The effects of sodium chlorite on humans were evaluated in 10 male volunteers on a rising dose protocol. Single doses of 0.01, 0.1, 0.5, 1.0, 1.8, and 2.4 mg of chlorite ion per litre in 1 litre of drinking-water were ingested by each subject. Changes in group mean values for serum urea nitrogen, creatinine, and urea nitrogen/creatinine ratio were observed, which the authors concluded were not adverse physiological effects. The highest dose tested, 2.4 mg/litre (0.034 mg/kg of body weight per day), can be identified as a single-dose NOAEL (33).

The same volunteers ingested 0.5 litres of water per day containing 5 mg of sodium chlorite per litre for approximately 12 weeks, and were then kept under observation for 8 weeks. Treatment was associated with a change in group mean corpuscular haemoglobin; however, as there was no trend over time for this change and values were within the normal ranges, the authors were reluctant to attach physiological significance to the observation. The dose tested, equivalent to 36 µg/kg of body weight per day, was identified as the NOAEL (33).

Chlorate

Because of its use as a weed killer, a large number of cases of chlorate poisoning have been reported (3). Symptoms include methaemoglobinaemia, anuria, abdominal pain, and renal failure. For an adult human, the oral lethal dose is estimated to be as low as 20 g of sodium chlorate (230 mg of chlorate per kg of body weight) (34).

Ten male volunteers were given six separate doses of sodium chlorate following a rising dose protocol, single doses of 0.01, 0.1, 0.5, 1.0, 1.8, and 2.4 mg of chlorate ion per litre in 1 litre of drinking-water being ingested by each volunteer. Very slight changes in group mean serum bilirubin, iron, and methaemoglobin were observed, but the authors concluded that they were not adverse physiological effects. The highest dose tested, 2.4 mg/litre (34 µg/kg of body weight per day), can be identified as a single-dose NOAEL (33).

The volunteers also ingested 0.5 litres of water per day containing 5 mg of sodium chlorate per litre (36 µg/kg of body weight per day) for approximately 12 weeks, and were then kept under observation for 8 weeks. Treatment was associated with slight changes in group mean serum urea nitrogen and mean corpuscular haemoglobin, but the authors concluded that these were not physiologically significant as values remained within the normal range for each parameter. The NOAEL was 36 µg/kg of body weight per day (33).

7. Guideline values

Chlorine dioxide

Chlorine dioxide has been shown to impair neurobehavioural and neurological development in rats exposed perinatally. Significant depression of thyroid hormones has also been observed in rats and monkeys exposed to it in drinking-water studies.

A guideline value has not been established for chlorine dioxide because of its rapid breakdown and because the chlorite provisional guideline value (see below) is adequately protective for potential toxicity from chlorine dioxide. The taste and odour threshold for this compound is 0.4 mg/litre.

Chlorite

Chlorite affects the red blood cells, resulting in methaemoglobin formation in cats and monkeys. IARC has concluded that chlorite is not classifiable as to its carcinogenicity to humans (Group 3) (35).

The TDI for chlorite is 10 µg/kg of body weight, based on the NOAEL of 1 mg/kg of body weight per day for decreased red blood cell glutathione levels in a 90-day study in rats exposed to chlorite in their drinking-water (25) and applying an uncertainty factor of 100 (to account for inter- and intraspecies variation). Owing to the acute nature of the response and the existence of a 2-year rat study, an additional uncertainty factor of 10 was not incorporated to account for the short duration of the key study. The TDI derived in this manner is consistent with the NOAEL (36 µg/kg of body weight per day) in a 12-week clinical study in a small number of human volunteers (33).

On the assumption that drinking-water contributes 80% of the total exposure, the provisional guideline value is 0.2 mg/litre (rounded figure). This guideline value is designated as provisional because use of chlorine dioxide as a disinfectant may result in the chlorite guideline value being exceeded, and difficulties in meeting the guideline value must never be a reason for compromising adequate disinfection.

Chlorate

Available data on the effects of chlorate in humans and experimental animals are considered insufficient to permit the development of a guideline value. Data on accidental poisonings indicate that the lethal dose to humans is about 230 mg/kg of body weight per day. This is of the same order of magnitude as the NOAELs identified from studies in rats and dogs. Although no effects were observed in a 12-week clinical study in a small number of human volunteers ingesting 36 µg/kg of body weight per day, a guideline value was not derived from these results because no adverse effect level was determined.

Further research is needed to characterize the nonlethal effects of chlorate. Until data become available, it may be prudent to try to minimize chlorate levels. However, adequate disinfection should not be compromised.

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Chapter 5

TREATMENT EFFICIENCY

G. Stanfield, M. Lechevallier and M. Snozzi

5.1 Introduction

The primary purpose of water treatment is to provide drinking water to consumers that is free of waterborne pathogens. Because no single treatment process can be expected to remove all of the different types of pathogens that can be found in water (under all conditions), multiple barriers are desirable. Multiple barriers will also ensure additional safety in the case that a single treatment step is not working optimally. The number of treatment processes (technical barriers) required is influenced by the quality of the source water (see Chapter 4). Groundwaters that are protected from surface influence are usually of relatively good quality and so traditionally few, if any, treatment processes are required. Lowland surface water sources are usually of much poorer quality and more treatment processes are needed to provide an acceptable level of safety.

A number of treatment processes are also designed to modify the chemical and physical properties of the water (rather than pathogen elimination). State of the art treatment includes techniques to reduce AOC and reducing matter, so that on the one hand the regrowth of the pathogens in the distribution system is low and on the other hand the disinfection is more effective. This chapter, however, does not detail such processes but concentrates solely on the reduction of faecal-oral infection risk.

A wide spectrum of pathogenic agents can be found in water and monitoring for their presence on a routine basis is impracticable. Traditionally (as outlined in Chapter 1) microbial safety of drinking water has been confirmed

by monitoring for the absence of microorganisms of faecal origin. Bacteria such as *E. coli*, faecal streptococci and *Clostridia* have been used for this purpose, because they are consistently present in high numbers in the faeces of warm-blooded animals and are relatively easy to detect in water. These bacteria and groups of bacteria are microbial indices of faecal pollution and form the basis of guidelines and national standards.

It has been recognised that the microbial indicator parameters do not necessarily behave in the same way as certain pathogens in water treatment processes. The ability of treatment processes to remove specific pathogens has been directly measured, with such studies typically conducted at bench or pilot scale some of them using water spiked with pathogens (Sommer and Cabaj, 1993; Jacangelo *et al.*, 1995; Bellamy *et al.*, 1985; Hunt and Mariñas, 1999). The potential removal determined in such pilot studies will, however, not necessarily be achieved in full-scale treatment. Therefore, there is a need for alternative parameters that correlate more closely with the behaviour of specific pathogens both to assess the disinfection potential of full-scale treatment and to measure process performance during treatment.

Safe drinking water is the result of careful evaluation of source water quality and variation (as outlined in Chapter 4) and adequate, reliable treatment processes combined with performance monitoring to assure that treatment is within operating parameters. The focus for the control of process operation should be put on simple measurements, which can be done on-line. If the input to the system and its normal performance is known, the on-line measurement will be a perfect indication of disturbances and changes in the water quality. This shifts the emphasis of quality control of drinking water from end product testing (*i.e.* testing for failure) to the testing and control of treatment processes (*i.e.* preventing failure). Current treatment processes and appropriate indicators of performance are discussed below.

A verification of the quality at the end of the treatment chain is necessary. For this purpose non microbial parameters like flow, colour and disinfectant residual (where appropriate) are suitable (see also Chapter 2, Table 2.4). Microbial parameters for the validation of the treatment process include *E. coli*, total and thermotolerant coliforms, heterotrophic bacteria and aerobic spore-forming bacteria. However, it should be stressed that this verification should not be mistaken as a determination of the safety of the drinking water.

5.2 Microbial treatment efficiency

A review of the available data on treatment efficiencies has been published by LeChevallier and Au (2002). Disinfection can be achieved in two ways:

- The physical removal of the pathogens.
- The inactivation (death) of the pathogen.

Apart from careful characterisation of the disinfection potential of a given treatment process (which in many cases involves experimental determinations) it is also important to identify simple measurements that give information rapidly on whether the treatment process is working properly. For the latter, physical and chemical measurements (preferably on-line measurements) are often better than microbial determinations.

A review of potential inactivation rates for different disinfection treatments has been published by Sobsey (1989). More recently, the United States Environmental Protection Agency (USEPA, 1999) has compiled data on achievable disinfection efficiencies for various processes and combinations of treatment steps. Although the actual inactivation will be influenced by many factors (including the ability of many of the microbial parameters to remain viable while becoming non-culturable), the following subsections list typical ranges reported for each treatment process. Although retention of water in reservoirs and impoundments can bring about significant improvements in quality as a result of inactivation, sedimentation and predation this process is not discussed here. For more details the reader is referred to the review by LeChevallier and Au (2002). For more precise evaluation of the reduction of individual pathogens by a treatment process, specific experimental determination is necessary.

5.2.1 Coagulation and sedimentation

The most common coagulants in use throughout the world are aluminium sulphate, ferric sulphate, ferric chloride and poly-aluminium chloride. These coagulants are mixed into the water where they produce hydroxide precipitates that are 'fluffy' and enmesh particles and microbes along with some of the dissolved organic carbon. In some circumstances, flocs generated by aluminium and ferric salts can be strengthened by the addition of coagulant aids such as long chain organic polymers. The flocs formed by this process must be removed. This can be achieved by sedimentation or, if the flocs are very light,

fine air bubbles may be used to carry them to the surface (air flotation) where they are skimmed off. They can also be removed by direct filtration.

Various forms of coagulation and sedimentation are used in water treatment and there are differences in general practices between countries, which makes the comparison of data difficult. However, published data indicate that this process may remove between 40% and 99% of bacteria, which translates into 0.2 and 2 logs of removal. Removal of viruses is rather poor, below 1 log, whereas for parasites such as *Cryptosporidium* removal of up to 2 logs has been reported.

The retention of the formed flocs is very important because of the accumulation of pathogens, since even single flocs may contain sufficient numbers of pathogens to be of hygienic importance (Gale *et al.*, 1997). Continuous measurements of turbidity or particle counts are useful for monitoring the efficiency of this process.

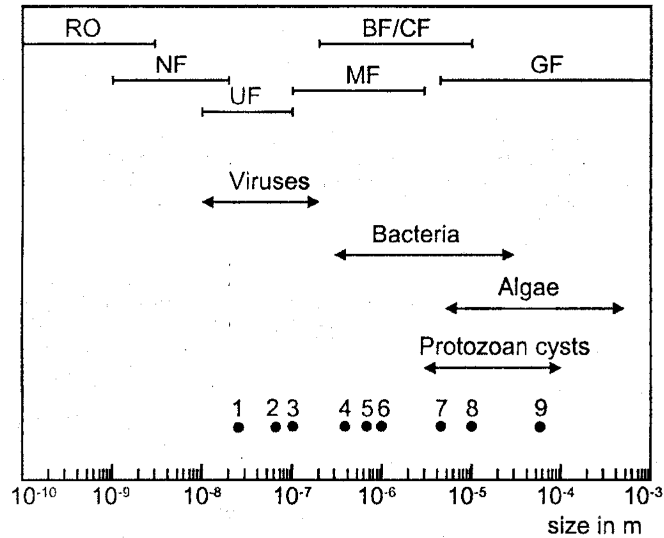
5.2.2 Filtration

Various filtration processes are used in drinking water treatment. Used with proper design and operation, filtration can act as a consistent and effective barrier against microbial pathogens. Filtration processes that are used in potable water treatment and the pore size of filter medium are shown in Figure 5.1, along with the sizes of selected microbial particles. This provides an insight into the removal mechanisms and likely efficiencies of the different filtration processes.

Filtration is a physical removal of organisms together with other particulate matter. On-line measurements of turbidity or particle counts, as well as determination of particle size distribution are excellent control parameters for this process. If parallel filtration units are operated, it is essential that each unit is measured separately in order to ensure the recognition of poor performance in an individual filter unit.

Figure 5.1. Filter medium pore sizes and the size of microbial particles (with selected microorganisms marked with numbers)

(Adapted from LeChevallier and Au, 2002)



Key:
RO: reverse osmosis. NF: nanofiltration. UF: ultrafiltration. MF: microfiltration. BF/CF: bag and cartridge filters. GF: granular filtration including slow sand filtration (slow sand filters have lower pore sizes than rapid-rate filters)
1. MS2 bacteriophage. 2. Rotavirus. 3. PRDI bacteriophage. 4. *Mycobacterium avium* complex (represents smallest size). 5. *Yersinia* spp. 6. Coliform bacteria. 7. *Cryptosporidium* oocysts. 8. *Giardia* cysts. 9. *Balanthidium coli* cysts.

5.2.2.1 *Rapid filtration*

Rapid filters are deep beds (0.6-1.0 metres) of sand, anthracite and sand or granular activated carbon. The particle size of the medium is usually about 1 mm. They are operated at flow velocities of about 5-15 metres per hour. Rapid filters retain most of the flocs and other particles that escape chemical coagulation and sedimentation. The size of particles that can be removed in deep-bed filtration can be much smaller than the pore size of the filter (Hall,

1998). This is due to electrostatic adhesion causing adsorption of particles that are in close proximity to the filter medium. Rapid filters are stopped and backwashed according to a time cycle (usually 24 hours), when flow becomes excessively restricted due to clogging or when the turbidity or particle counts of the filtrate become unacceptably high.

The microbial removal efficiency of rapid filters can be influenced by a number of factors. Correct operation and maintenance of rapid filters is essential otherwise performance may be lost. In poorly maintained filters, cracks have been observed particularly near the walls, which allow unfiltered material to pass through, decreasing the bacteriological quality of the filtrate. Changes in the flow rate can dislodge deposits containing microorganisms causing them to pass into the filtrate. When a filter is put back into service after backwashing, the initial filtrate is of poor quality in terms of turbidity and bacterial numbers. This is due to displacement of residual backwash water, and the lower efficiency of the clean filter media, compared with a partly used (ripened) filter (Amirtharajah and Wetstein, 1980). For this reason the initial filtrate may be run to waste or returned to the start of the treatment processes for a period of up to 30 minutes. Alternatively a 'slow start' procedure may be used in which the flow rate through the filter is restricted until the filtrate becomes of acceptable quality. Additionally, backwash water should not be recycled within the treatment plant.

Published data indicate that coagulation combined with rapid filtration may remove between 2 and 3 logs of bacteria, while reported removal of viruses range from 1 to 3 logs and for parasites such as *Cryptosporidium* 2 to 3 logs. Continuous measurements of turbidity and/or particle counts are important for monitoring.

5.2.2.2 *Slow sand filtration*

Slow sand filtration is a biological treatment process, which has to be used without coagulation pre-treatment. Other pre-treatment, particularly rapid filtration, may be used to remove high particle loads. Typically, a slow sand filter has a depth of about 0.7 metres and is operated at flow rates of 0.1 to 0.3 metres/hour compared to 5-15 metres/hour in rapid filters. The sand is mixed in size ranging from 0.15 to 0.35 mm. The pores are still quite large at about 60 µm. Although there is some filtration in depth, as in rapid sand filtration, the vital process is the formation of a biologically active layer (the Schmutzdecke) in the top 20 mm. Optimum treatment performance is dependent on a well-established Schmutzdecke. This provides an effective surface filtration of very small particles, including bacteria, parasites and viruses.

Any particles that pass through the Schmutzdecke may be retained in the remaining depth of the sand by the same mechanisms as exist in rapid filtration. The growth of the Schmutzdecke and its retention of particles cause a loss of permeability in the top layer of sand so that after some weeks of operation, flow rates decline. When this occurs, the filter is taken out of service and the top 20-30 mm removed by skimming. Slow sand filters are known for their high efficiency in removing bacteria and parasites, but small channels can occur in the filter if not properly operated and maintained which influence performance. In well-maintained systems with slow sand filtration it is possible to achieve a performance similar to a combination of coagulation and filtration. Continuous measurements of turbidity and/or particle counts are important for monitoring.

5.2.2.3 *Activated carbon filtration*

Activated carbon filters are predominantly used to remove organic compounds. However, they may also affect counts of microbial organisms including reduction of viruses and parasites. Due to growth in the filters, increased heterotrophic plate counts and total coliform counts can sometimes be observed.

5.2.2.4 *Membrane filtration*

In membrane filtration water is passed through a thin film, which retains contaminants according to their size. Membrane filtration has been playing an increasing role in drinking water treatment, including pathogen removal. The most commonly used membrane processes in drinking water treatment for microbial removal are microfiltration (MF) and ultrafiltration (UF) (see Figure 5.1). Detailed description of the fundamentals, design and operation of these processes are available in the literature (AWWARF, 1996; Taylor and Wiesner, 1999). Other membrane processes such as reverse osmosis (RO) and nanofiltration (NF), which are used primarily for other purposes, also remove pathogens.

Membrane filtration removes microbial pathogens primarily by size exclusion; microbes with sizes greater than the membrane pore size are removed. Chemical coagulation prior to the membrane is not a requirement for microbe removal. However, some degree of pre-treatment must be employed to reduce membrane fouling. Fouling arises from accumulation of chemicals, particles and the growth of organisms on membrane surfaces, resulting in reduced membrane productivity. Once fouling accumulates to such a level that the productivity of the system is unacceptable, the membranes must be

chemically cleaned to restore productivity. Advanced pre-treatment systems such as conventional coagulation-sedimentation-filtration or other membrane processes may also be considered, depending on the quality of the source water.

Published data indicate that membrane filtration may remove up to 6 logs of bacteria, viruses or parasites. Process performance is generally monitored by measurement of physical parameters such as pressure drops across the membrane.

5.2.3 *Chemical inactivation*

Chemical disinfection to inactivate pathogens is an important treatment barrier. Chemicals used include chlorine, chloramine, chlorine dioxide and ozone. Treatment effectiveness is a function of dose, contact time, temperature and sometimes pH. Chemical disinfection can be placed at different positions in the treatment train and more than one disinfectant can be used, however it is important to note that organisms entrapped in particles may be shielded from the action of the chemicals. Primary disinfection is the process by which microorganisms are inactivated during the treatment process, while a secondary disinfectant can be added prior to distribution to maintain the water quality within the distribution system. Secondary disinfection provides a final barrier against bacterial contamination and regrowth within the distribution system. The practice of residual disinfection is, however, controversial (IWSA, 1998). It has been suggested that if biological stability is achieved and the system is well maintained then the disinfectant is unnecessary and may mask ingress into the distribution system by killing the bacterial indicators (but not the more robust pathogen microorganisms).

The concept of disinfectant concentration and contact time is integral to the understanding of disinfection kinetics and the practical application of the CT concept (which is defined as the product of the residual disinfectant concentration [C in mg/l] and the contact time [T in minutes], that residual disinfectant is in contact with water – USEPA, 1999) is important. Allowance must be made for the decline in concentration over time and in measuring time it is important to take account of the hydraulic behaviour of the treatment plant (in particular any short-circuiting). Temperature, over the range appropriate for drinking water, affects the rate of disinfection reactions according to the Arrhenius Law, although some deviations have been noted for certain disinfectants at low temperatures. The pH of the disinfectant solution also affects reaction kinetics. Table 5.1 outlines CT values for inactivation of viruses.

Table 5.1. CT values for virus inactivation

(USEPA, 1999)

Disinfectant	Units	Inactivation 2-log	3-log	4-log
Chlorine[1]	mg min/l	3	4	6
Chloramine[2]	mg min/l	643	1 067	1 491
Chlorine dioxide[3]	mg min/l	4.2	12.8	25.1
Ozone	mg min/l	0.5	0.8	1.0
UV	mW s/cm^2	21	36	not available

1. Values based on a temperature of 10 °C, pH range 6 to 9, and a free chlorine residual of 0.2 to 0.5 mg/l.
2. Values based on a temperature of 10 °C and a pH of 8.
3. Values based on a temperature of 10 °C and a pH range of 6 to 9.

5.2.3.1 Chlorination

Chlorination can take a number of forms including the use of chlorine, chloramines and chlorine dioxide. Each chemical has different disinfecting properties. Monochloramine (formed by the combination of chlorine with nitrogenous compounds) has a lower disinfection activity than chlorine but is more stable. Chlorine dioxide may be chosen because of its greater effectiveness against parasites.

Nearly 100 years of drinking water chlorination has demonstrated its effectiveness in the inactivation of microbial pathogens and the benefits of chlorination out-weigh any disadvantages, such as production of trihalomethanes. Enteric viruses are generally more resistant to chlorine than enteric bacteria, and viruses associated with cellular debris or organic particles may require high levels of disinfection due to the protective nature of the particle surface. Chlorination is considered to be highly effective for virus inactivation if the water has a turbidity of ≤ 1.0 nephelometric turbidity units (NTU), a free chlorine residual of 1.0 or greater for at least 30 minutes, and a pH of < 8.0. Protozoan cysts such as those of *Cryptosporidium* and *Giardia lamblia*, however, are highly resistant to chlorine disinfection (USEPA, 1989). Other factors that influence microbial sensitivity to chlorine include surface attachment, encapsulation, aggregation and low-nutrient growth.

Chlorine is a strong disinfectant that is effective at inactivating bacteria and viruses and, under certain circumstances, Giardia. CT values for 2 log inactivation of vegetative bacteria may vary between 0.02 and 200 mg min/l

(Grohmann, A; 2002) This wide range depends on a number of factors particularly the presence of reducing matter. One purpose of water treatment, therefore, is to eliminate such matter from water prior to chlorination. Residual levels of reducing matter can be determined by electrochemical methods such as oxidation-reduction-potential (ORP) measurements. The use of high dosage of chlorine is, therefore, by itself not a guarantee of safe drinking water as the presence of reducing matter may result in high concentrations of disinfection by-products (DBP), such as trihalomethanes (THM), which are toxic.

No significant reduction of Cryptosporidium is achieved with conventional CT values. Since pH, temperature and chemical composition will influence the disinfection potential they need to be monitored together with the CT measurements.

Because of the weak disinfecting power of monochloramine, it is not recommended as a primary disinfectant and it is ineffective in the inactivation of *Cryptosporidium*. Most systems using monochloramine apply a short period of free chlorine prior to ammonia addition or use an alternative (*e.g.* ozone, chlorine dioxide) primary disinfectant. Chloramines have CT values of more than 80 mg min/l for a 2 log inactivation of bacteria; values for the same inactivation of viruses are above 600 mg min/l and, therefore, they are only suitable for the inactivation of bacteria.

Chlorine dioxide is a strong oxidant as well as a powerful disinfectant and, therefore, can be used for the control of iron, manganese and taste and odour causing compounds as well as a primary disinfectant. It has also been used as a secondary disinfectant in many European countries. However, chlorine dioxide forms inorganic by-products (chlorite and chlorate ions) upon reaction with water constituents, and a water supplier may need to provide additional treatment depending on the level of these inorganic by-products and specific regulatory requirements. Chlorine dioxide is roughly comparable to free chlorine for inactivation of bacteria and viruses at neutral pH (), but it is more effective than free chlorine at an alkaline pH of 8.5 (Hoff and Geldreich, 1981). CT values for chlorine dioxide resulting in a 2 log inactivation of vegetative bacteria are less than 1 mg min/l. While values around 4 mg min/l have been reported for viruses and those for *Giardia* inactivation are around 15 mg min/l. Temperature and chemical composition need to be monitored together with the CT measurements (or calculations) and chlorine residual.

Chlorination usually takes place at a central treatment point but, particularly in developing countries, there is growing interest in applying it at household level. Sachets or tablets of a chlorine compound (sometimes together with a coagulant to remove turbidity) are sometimes used. Decentralised

production of sodium hypochlorite is now possible from the electrolysis of a solution of common salt and this may provide a cost-effective source of chlorine solution. Combined coagulant-disinfectant tablets or powders or use of a solution of sodium hypochlorite are available for household water treatment (Sobsey, 2002).

5.2.3.2 Ozonation

Ozone has been used for more than a century for water treatment, mostly in Europe, but this usage is spreading to other areas. Despite this long use, the exact mechanism of how ozone inactivates microbes is not well understood, although it is known that ozone in aqueous solutions may react with microbes by direct reaction with the molecular ozone, or via reaction with the radical species formed on ozone decomposition.

Of the vegetative bacteria, *E. coli* is one of the most sensitive to ozone disinfection, while Gram-positive cocci (*Staphylococcus* and *Streptococcus*), the Gram-positive bacilli (*Bacillus*) and the mycobacteria are the most resistant. *Mycobacterium avium* can be effectively controlled by low doses of ozone, whereas the organism is highly resistant to free chlorine. It has been reported that heterotrophic plate count bacteria may be less susceptible to ozone inactivation than other indicator organisms. Viruses are generally more resistant to ozone than vegetative bacteria, although phages appear to be more sensitive than human viruses. Ozone is effective against *Giardia* and to a lesser extent *Cryptosporidium*. Because ozone does not produce a stable residual it is frequently followed by chlorination to produce a residual disinfectant for distribution. Due to the relatively fast decay of ozone even in pure water, hydraulics of the ozonation reactor are very important (see below).

Ozone will oxidise organic components present in the water, such as natural organic matter to produce smaller organic substances. Since these are usually more biodegradable, ozonation will increase bacterial growth after treatment. To prevent this, post ozonation removal of the oxidation products is necessary.

Ozone is a very powerful disinfectant for inactivation of vegetative bacteria. CT values below 0.5 mg min/l are reported for 2 log reduction of bacteria. CT values between 0.5 and 1 mg min/l are required for a 2 log inactivation of viruses. Inactivation of protozoa like *Giardia* is possible at temperatures above 15°C with CT values of 0.7 mg min/l for 2 log inactivation, while at 5°C the CT value increases to 1.3 mg min/l. For the same inactivation of *Cryptosporidium* the CT values required are about ten times higher. Content

of organic carbon will also influence the disinfection efficiency. Therefore the measurement of CT values needs to include control of temperature and quality of water entering the ozonation reactor.

Case study: Hydraulics of an ozonation reactor

In Switzerland, food-related laws and regulations require health risks assessment and the evaluation of critical treatment steps in drinking water production. In the city of Zurich, a considerable fraction of drinking water is produced from lake water following a multistage procedure. During a health risk assessment the hydraulics of the ozonation reactor were evaluated by addition of a concentrated sodium chloride solution to the water inlet of the reactor for a period of two hours. Five sampling points along the water flow allowed the spread of the addition through the reactor to be followed (Kaiser *et al.*, 2000). Modelling of the experimental data showed that the reactor was best described by a series of four mixed reactors followed by a plug-flow reactor with considerable back flow. The model was confirmed by the comparison of modelled and measured ozone profiles and atrazine concentrations.

Modelled inactivation of microorganisms showed a remarkable difference between a single plug-flow model and the model derived from the experimental measurements. According to the model, the ozonation should reduce vegetative bacteria and viruses by more than 6 logs, spores of *Bacillus subtilis* will be inactivated by 1.5 logs, whereas the inactivation of *Cryptosporidium* is less than 1 log.

5.2.3.3 UV disinfection

UV action results from absorption by nucleic acids (DNA and RNA), leading to the dimerisation of pyrimidine bases, and all organisms are susceptible to UV light. Exposure to UV results in reduced viability of the treated cells. However, most bacteria have evolved different repair systems to cope efficiently with UV damage to their genetic material, for example, thymine dimers can be repaired both in the presence ('photoreactivation') or absence of light ('dark repair') (Jagger, 1967). Thus, UV doses in a certain range will only transiently reduce the ability of bacteria to form colonies without having a long-term effect on their survival (Mechsner *et al.*, 1991). Therefore, for the UV disinfection of drinking water it is essential to treat each volume part with a sufficient light dose to kill the bacteria. Usually a dose of 400 J/m^2 (40 mW s/cm^2) is accepted as being sufficient for efficient treatment.

Three types of light source are used for UV disinfection, namely:

- Low-pressure mercury lamp.
- Medium-pressure mercury lamp.
- Pulsed lasers.

The most popular so far is the low-pressure mercury lamp, which emits light at the wavelength of 254.7 nm, almost exclusively. Due to the rather low light intensity of such lamps, radiation times required for efficient disinfection are substantially higher than those for the second type, the medium pressure mercury lamp, which emits light of higher intensity and also of longer wavelength. It is sometimes claimed that the medium-pressure lamps have a better performance, because they may act in a dual way, damaging both DNA and proteins, some of which might be involved in the DNA-repair process. On the other hand, due to the much higher light intensity of medium-pressure lamps, the required contact time is much shorter with a concomitant risk of volume parts not being treated sufficiently. Recently, the use of pulsed UV lasers has been suggested. It is claimed that the same extent of cell inactivation can be achieved with this light source at less than one tenth of the dose of low-pressure mercury lamps. Rubin *et al.* (1982) showed a dependence of photoinactivation of yeast cells on the UV light intensity at the same dose. A similar dependence was observed for the photoprotection. Therefore, at high light intensity more dead cells were found at lower doses.

Another factor interfering with this type of disinfection is the UV transmission of the water. For treatment process evaluation the minimal UV dose for water with different UV transmission characteristics must be known. Biodosimetric determination of the UV dose under production conditions has been proposed as the best method for determining efficiency (Sommer and Cabaj, 1993). This procedure includes the addition of spores of *Bacillus subtilis* to the water before treatment; from the difference between the colony counts before and after treatment the UV dose in the reactor can be inferred from a dose-response curve determined in the lab. Similar dose-response curves can be determined for other organisms of interest (*e.g.* pathogens) and the reduction potential of the treatment system can be evaluated.

The transmission of the water should be monitored on-line with the help of an UV detector. The determination of colony forming units of coliform bacteria is not a satisfactory measure of UV inactivation because of the possibility of repair mechanisms coming into play (Mechsner *et al.*, 1991). If a microbial indicator parameter is required, the reduction of spores should be measured since they are easy to measure and at the same time quite resistant to UV light.

UV disinfection has been proven to be adequate for inactivating bacteria and viruses. UV doses of 400 J/m^2 will reduce vegetative bacteria by 4 to 8 logs. Virus inactivation is by 3 to 6 logs. Protozoa are more resistant to UV disinfection, but newer studies showed that in neonatal mouse infection studies with UV treated *Cryptosporidium oocysts* at a UV dose of 410 J/m^2 a 4 log reduction in infection occurred. Similar UV doses are required for a 4 log reduction in spores of *Bacillus subtilis*.

UV disinfection case study

In Austria, Germany and Switzerland certification requirements have been established for the UV disinfection of drinking water, which typically require biodosimetric determination of the disinfection efficiency under production conditions (Snozzi *et al.*, 1999). Spores of *Bacillus subtilis* are used for this process since repair mechanisms are not important and can be neglected. The water entering the UV plant is inoculated with the spores and their concentration is determined before and after UV treatment. The UV dose can be calculated from the reduction of viable spores and a dose-response curve measured previously in the lab. Variation of the light intensity and the flow rate allows the definition of the range of flow rate with turbulent mixing within the reactor.

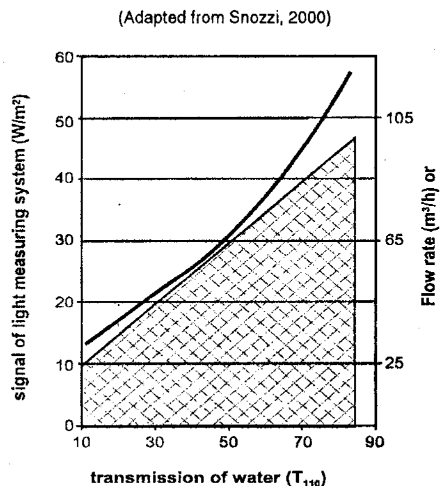
The result of the experimental determination of the disinfection efficiency can be represented in a graph (Figure 5.2) showing the maximal flow rate as a function of the UV transmissions of the water, which will ensure a minimal radiation dose of 400 J/m^2. If operation remains within these limits, the predetermined reduction of the number of viable pathogens can be ensured.

This experimental determination of the UV disinfection potential of a given reactor is very reproducible. Deviations between different determinations several months apart were found to be less than 2% (Snozzi, 2000).

Measurements of UV light intensity in the reactor serves as a control for process performance (it is important that the measuring point should be positioned such that changes in the UV transmission of the water will influence the reading of the light meter).

Figure 5.2. Measured UV light intensity as a function of UV transmission of the water

(Adapted from Snozzi, 2000)



The dashed area represents combinations of UV transmissions and flow rates, which result in reliable disinfection. The solid curve represents the signal of the light measuring device as a function of the UV transmission of the water. UV transmission (T_{100}) is given as percent transmission using a 100 mm light pass.

5.2.3.4 *Solar water disinfection*

Solar panels can be used to generate electricity to power the UV lamps mentioned in the previous section but in low-income countries the sunlight alone can be used to kill or inactivate many, if not all, of the pathogens found in water. Solar water disinfection is a method of treating relatively small amounts of water at the point of use. There are three ways in which solar radiation can be used to eliminate pathogens. The first is through heating, the second through the effect of the natural UV radiation and the third through a mixture of both thermal and UV effects. None of these methods is yet widely used but laboratory experiments and field programmes show that some systems have good potential to produce potable water. Solar disinfection is included in the

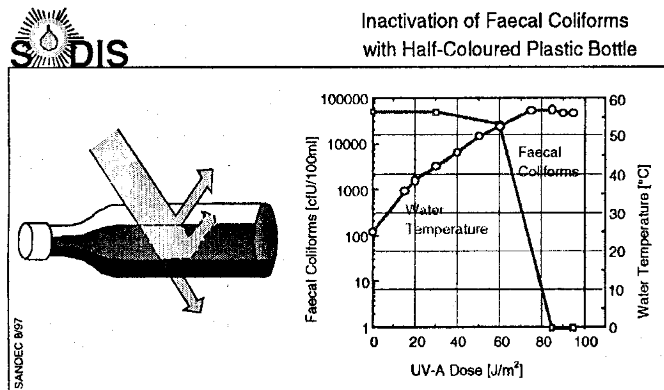
technologies reviewed by WHO for household water treatment and storage (Sobsey, 2002).

Thermal heating from the sun can be via solar cookers (which concentrate the rays of the sun with reflectors) or from simply exposing black-painted containers to the sun. In many systems temperatures can reliably reach over 55°C killing many pathogens. With the cookers and some of the other systems the temperature of the water can easily exceed 65°C, a pasteurisation temperature capable of inactivating nearly all enteric pathogens. Achievement of specific temperatures can be monitored using simple low-cost re-usable water pasteurisation indicators, based on the visible melting of wax in a clear plastic tube.

The use of heating and UV radiation to simultaneously disinfect water is used by a number of different solar treatment systems. The widest known is the SODIS system (Figure 5.3), which is suitable for low-income countries. The only equipment required is locally available bottles to contain the water (which needs to have a turbidity <30 NTU). This technique is now being field tested in various parts of the world and increasing amounts of data are becoming available on its effectiveness. Obviously for the UV to be effective the bottle material needs to be transparent to the useful wavelengths of the UV rays. The promoters of SODIS suggest the use of thin PET plastic bottles rather than PVC ones because the former material is more chemically stable. The half of the bottle furthest from the sun should be painted with black paint to improve the heat gain from the absorption of thermal radiation, and the bottle can be laid on a dark roof to further increase the potential temperature rise in the water. Shaking a partly filled bottle to aerate the water before filling it completely has been found to give a faster pathogen kill rate (Reed, 1997). The water requires several hours of exposure to strong sunlight to obtain the advantageous synergy between UV dosage and temperature rise (Wegelin *et al.*, 1994, Sommer *et al.*, 1997). In cloudy weather a much longer period (such as two days or more) is required because of the lower level of UV radiation and the reduced likelihood of the temperature of the water ever exceeding 50°C.

Figure 5.3. Schematic representation of solar water disinfection and the influence of the water temperature on the UV-inactivation of bacterial cells

(Printed with permission of M. Wegelin)



5.3 Summary

This chapter reviews the different treatment barriers available to ensure the production of safe drinking water. The choice of which barriers to implement depends on a number of considerations including the source water quality. Non-microbial indicator parameters that can be measured on-line are most useful for assessing process performance and such monitoring is important within the total system approach to risk management. Treatment steps with relevant pathogen removal or inactivation are described together with possible indicators for the measurement of process performance.

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