

Office of Water (4601M) Office of Ground Water and Drinking Water Total Coliform Rule Issue Paper

The Effectiveness of Disinfectant Residuals in the Distribution System

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Background and Disclaimer

The USEPA is revising the Total Coliform Rule (TCR) and is considering new possible distribution system requirements as part of these revisions. As part of this process, the USEPA is publishing a series of issue papers to present available information on topics relevant to possible TCR revisions. This paper was developed as part of that effort.

The objectives of the issue papers are to review the available data, information and research regarding the potential public health risks associated with the distribution system issues, and where relevant identify areas in which additional research may be warranted. The white papers will serve as background material for EPA, expert and stakeholder discussions. The papers only present available information and do not represent Agency policy. Some of the papers were prepared by parties outside of EPA; EPA does not endorse those papers, but is providing them for information and review.

Additional Information

The paper is available at the TCR web site at:

http://www.epa.gov/safewater/disinfection/tcr/regulation_revisions.html

Questions or comments regarding this paper may be directed to TCR@epa.gov

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The Effectiveness of Disinfectant Residuals in Distribution Systems

Executive Summary

Maintenance of a disinfectant residual throughout the distribution system may help to maintain the integrity of the distribution system in the following ways:

- Inactivating microorganisms in the distribution system;
- Indicating distribution system upset; and
- Controlling biofilm growth.

This paper reviews the efficacy of using a disinfectant residual to ensure distribution system integrity. An overview of secondary disinfectants, an overview of existing disinfectant residual guidelines and requirements, a discussion of the three main functions of secondary disinfection, and summaries of research on the efficacy of secondary disinfectants in carrying out these functions are provided. Additionally, this paper provides information on the future research needed to answer more definitively whether provision of a disinfectant residual can meet these expectations.

Disinfectant residual maintenance can be affected by many variables, some associated with distribution system conditions, such as pipe volume, chemical/biological characteristics of treated water entering the distribution system, the type of disinfectant being used, and events introducing contaminants to the distribution system.

There are six pathways by which pathogens can reach the distribution system (USEPA 2002c):

- Treatment breakthrough,
- Leaking pipes, valves, and joint seals,
- Cross-connection and backflow,
- Finished water storage vessels,
- Improper treatment of equipment or materials before and during main repair, and
- Intentional introduction of contaminants into distribution system.

Secondary Disinfectants

The USEPA (1999a) has discussed the efficacy and practicality associated with the use of free chlorine, chloramines, and chlorine dioxide for secondary disinfection. While none of these options is ideal for all systems, each has characteristics that may meet a specific system's needs for secondary disinfection. Selection of the most appropriate secondary disinfectant must be made on a system-by-system basis, with consideration given to the system's concerns regarding inactivation requirements, DBP formation potential, water quality, distribution system condition, and treatment experience and capabilities.

Existing Disinfectant Residual Guidelines and Requirements

In the United States, the Surface Water Treatment Rule requires systems that use surface water (or ground water under the influence of surface water) to monitor and maintain a detectable disinfectant residual throughout the distribution system. This monitoring must be conducted throughout the distribution system at same locations as those used for total coliform monitoring and at entry points. Under the Stage 1 Disinfectant/ Disinfection By-Products Rule, the residual is not to exceed 4.0 mg/L for chlorine and chloramines and 0.8 mg/L for chlorine dioxide in any system based on a running annual average of all measurements in the distribution system calculated each month. States may adopt Federal drinking water regulations or more restrictive drinking water requirements. The TCR lists disinfectant residual as a Best Available Technology for compliance with total coliform Maximum Contaminant Level (MCL).

Disinfection practices vary widely in European countries. The European Union has issued standards for drinking water, and these standards do not require disinfection explicitly. Of the 15 original European Union member states, only Spain and Portugal require secondary disinfection in distribution systems.

Effectiveness in Pathogen Inactivation

A review of the Surface Water Treatment Rule (SWTR) concentration-time (CT) requirements (USEPA, 1991) demonstrates that free chlorine, chloramines, and chlorine dioxide can be used to inactivate viruses and *Giardia lamblia*. This inactivation information helps to inform inactivation capabilities under bulk water conditions.

Distribution systems exhibit varying conditions and multiple forms of microbes that may influence pathogen inactivation. For instance, in the distribution system, bacteria and viruses can be found as part of the bulk water, attached to particles, or as part of biofilms. The literature review found that viruses and bacteria attached to particles or present in biofilms are more protected from inactivation.

Studies were also reviewed to compare the inactivation provided by free chlorine, chloramines, and chlorine dioxide on specific microorganisms. Overall, these studies, which were conducted in laboratory conditions and on bulk water samples, demonstrated that only free chlorine was able to provide 99.99 percent (4-log) inactivation of viruses. To provide 2-log inactivation of most species, free chlorine, chloramine, and chlorine dioxide required a CT of 50, 10,000, and 150 min•mg/L, respectively.

Effectiveness in Indicating Distribution System Upset

Many factors influence the concentration of the disinfectant residual in the distribution system, including the assimilable organic carbon level, the type and concentration of disinfectant, water temperature, and system hydraulics. Entry of foreign material into the distribution system from backflow (or other events) may alter these factors and contribute to a loss of residual. Studies have shown that large episodes of contamination, such as cross-connections, can overwhelm disinfectant residuals, resulting in no residual present in contaminated water.

Since most disinfectants are chemical oxidants that react with many substances, their use as indicators, specifically of microbiological contamination, is not entirely reliable. Inorganic and organic chemicals in the water can present a disinfectant demand that could misleadingly alert operators when no pathogens have been introduced. However, the loss or decrease of the disinfectant residual in this case can serve as an indicator of some contamination events. Furthermore, the presence of disinfectant-resistant pathogens, such as *Cryptosporidium*, and in some instances viruses, may persist in a distribution system despite the presence of a disinfectant residual.

There are several advantages to using disinfectant residual monitoring as a warning mechanism for possible contamination. Residual analysis is inexpensive, results are immediately available, and USEPA-approved methods for analysis already exist. Water system operators are becoming increasingly sophisticated in tracking and measuring disinfectant residuals. Accurate and on-going tracking of disinfectant residuals would assist in detecting sudden changes in residual levels and in using such changes as indicators of contamination.

Effectiveness in Controlling Biofilms

Problems associated with biofilms in distribution systems include enhanced corrosion of pipes and deterioration of water quality. Biofilms can also provide ecological niches that are suited to the potential survival of pathogens. The ability to control (but not eliminate) biofilms using secondary disinfection is impacted by the disinfectant residual concentration used in the system. If concentrations are too low, the disinfectant residual becomes ineffective at controlling excess biofilm growth. The number of variables associated with biofilm control has led researchers to reach differing conclusions regarding the effectiveness of secondary disinfection at controlling biofilm growth.

Several studies have compared the effectiveness of various disinfectants at varying concentrations in controlling bacterial growth. These studies have been performed on different scales, ranging from continuous flow annular reactors to pilot systems to comparisons of full-scale distribution systems. Several studies have concluded that chloramines are more effective secondary disinfectants with respect to biofilm control compared to chlorine. However, in some instances chlorine has been shown to be more effective at physically removing biofilm from pipes.

1 Introduction

Maintenance of a disinfectant residual throughout the distribution system may help to maintain the integrity of the distribution system in the following ways:

- Inactivating microorganisms in the distribution system;
- Indicating distribution system upset; and
- Controlling biofilm growth.

This paper reviews research on the efficacy of using a disinfectant residual to ensure distribution system integrity through pathogen inactivation, indication of distribution system upset (e.g., contamination), and biofilm control. Within the context of this paper, a distribution system is defined as a system of conveyances that distributes potable water. All pipes, storage tanks, pipe laterals, and appurtenances that comprise the delivery system are included in this definition. Appurtenances owned and operated by private customers, such as service lines and plumbing components that are typically not considered the responsibility of the public water system purveyor are also considered in this definition because they are physically attached to the distribution system and could potentially be a source of contamination, through, for example, backflow or leaching of contaminants from service lines. These and similar events may affect the water quality under the purveyor's jurisdiction. This paper provides an overview of secondary disinfectants, an overview of existing disinfectant residual guidelines and requirements, and a discussion of the efficacy of these disinfectants in carrying out the three main functions of secondary disinfection listed above. Additionally, this paper provides information on the future research needed to answer definitively whether provision of a disinfectant residual can meet these expectations.

2 Overview of Available Secondary Disinfectants

Secondary disinfection is the presence of a disinfectant residual in the distribution system (Surface Water Treatment Rule). The USEPA (1999a) has discussed the efficacy and practicality associated with the use of free chlorine, chloramines, and chlorine dioxide for secondary disinfection. This paper focuses on the efficacy of these three secondary disinfectants, describing the application methods and characteristic chemistry of each, dosing mechanisms, reaction chemistry, and distribution system kinetics. There are some alternative secondary disinfectants being investigated by researchers (e.g., potassium permanganate and ozone combined with hydrogen peroxide, copper combined with hydrogen peroxide, silver combined with hydrogen peroxide, and anodic oxidation) but currently there are no indications of their effectiveness within the distribution system.

Selection of the most appropriate secondary disinfectant is made on a system-by-system basis with consideration paid to the system's particular concerns regarding inactivation requirements, DBP formation potential, water quality characteristics, distribution system condition, and treatment experience and capabilities. Exhibit 1 summarizes various aspects of an "ideal" disinfectant residual.

-	Exhibit 1 – Froperties of an Tuear Disinfectant Residuar
The "I	deal" Disinfectant Residual Provides:
•	Protection against distribution system contamination
•	An indication of distribution system upset
•	Biofilm control
The "I	deal" Disinfectant Residual has the Following Chemical Characteristics:
•	Easily measured on-site under field conditions
•	Minimal to no interferences with common constituents in drinking water
•	Generates minimal to no disinfection by-products
•	Long-lasting
•	Selectively reactive (minimal to no corrosion/reaction with dissolved metals, pipe materials, linings, etc.)
•	Provides clear indication of contamination event (is chemically altered rather than consumed)
The "I	deal" Disinfectant Residual has the Following Operational/Physical
Chara	cteristics:
•	Highly soluble in water
•	Safely generated, transported, stored, and fed
•	Cost-effective relative to the application (large- or small-scale)
The "I	deal" Disinfectant Residual has the Following Inactivation Capabilities:
•	Effectively and efficiently inactivates wide range of organisms (bacteria, viruses, protozoa, algae, fungi)
•	Effectively inactivates microorganisms present in the bulk water and those associated with particles/biofilm
•	Achieves desired level of organism inactivation at doses that are safe for human consumption
The "I	deal" Disinfectant Residual has the Following Aesthetic Characteristics:
•	Achieves desired level of organism inactivation without creating tastes and odors
•	Overfeed can be detected by taste, odor, and/or color

Exhibit 1 – Properties of an "Ideal" Disinfectant Residual

2.1 Free Chlorine

Free chlorine is the most commonly used disinfectant in the United States. According to the 2000 *Community Water Systems Survey* (USEPA, 2002a), most surface water and ground water systems that have primary disinfection use chlorine. Of large systems participating in the Information Collection Rule (ICR) study, 83 percent of surface water plant-months and 86 percent of ground water system plant-months used free chlorine for primary disinfection (USEPA, 2003b). One plant-month indicates that a treatment plant used the specified treatment for one month. The ICR data were reported in percentage of plant-months to account for plants that varied disinfectants during the 12-month reporting period. Sixty nine percent of ICR plants used free chlorine as a secondary disinfectant in their distribution systems (McGuire et al, 2002). However, ICR plants were more likely to use primary and secondary disinfectants other than free chlorine if their source water contained high concentration of TOC and bromide. Free chlorine is also currently the most widely used secondary disinfectant in medium systems (USEPA, 2002a;

AWWA Water Quality Division, 2000). This may change, however, due to the implementation of the Stage 1 and 2 Disinfectant/Disinfection By-Product Rules (DBPRs) as systems adjust disinfection treatment to meet THM and HAA requirements.

Reaction Chemistry

Free chlorine reacts with constituents in the water by various mechanisms. It oxidizes soluble iron, manganese, and sulfides typically found in drinking water sources. Once oxidized, these inorganics precipitate and can be removed by clarification and filtration processes. Free chlorine oxidizes ammonia (NH₃) to form chloramines (at Cl₂ to NH₃ ratios less than 8:1) and nitrate and nitrogen gas (at ratios greater than 8:1) (White, 1999). Free chlorine reacts with natural organic matter and bromide to form halogenated organic compounds, such as THMs, HAAs, and chlorophenols, some of which may pose human health risks (USEPA, 1999a; Weisel et al., 1999). Chlorine also oxidizes organic matter to form compounds that do not contain a halogen, such as aldehydes, carboxylic acids, ketones, and alcohols (Richardson, 1998). Of the known halogenated compounds, THMs and HAAs occur in the highest concentrations.

Kinetics in the Distribution System

The chlorine decay rate in water can be described by an initial rate, which is relatively rapid, and a long-term decay rate, which is slower. The initial rate is attributed to substances in water that react rapidly with chlorine and are usually referred to as the chlorine demand. Once this demand has been met, a more persistent residual is established with a slower rate of decay.

Chlorine decay kinetics within the distribution system is governed by both decay occurring in the bulk fluid as well as decay at the pipe walls. A number of factors can affect the kinetics including the water temperature, total organic carbon (TOC) concentration, initial chlorine concentration, biofilms, the rate of pipe corrosion and the presence of corrosion products (Vasconcelos et al., 1996). In general, chlorine decay kinetics increase at higher temperature, chlorine concentration, TOC concentration, biofilm and corrosion product mass, and as pipe corrosion rates increase.

The decay reactions for chlorine in the bulk water and at the pipe wall in some instances can be modeled using a first order rate expression, although the decay constants may vary for each water. The type and concentration of various chemical and biological constituents that exert a chlorine demand (described previously) will impact the decay coefficient. The decay coefficient for a specific bulk water can be determined using bottle decay tests, but the coefficient for pipe wall decay must be determined in the field or with pipe segments taken from the distribution system piping. In general, the relative importance of decay at the pipe wall increases as the pipe diameter decreases because the ratio of volume to pipe surface area decreases (Vasconcelos et al., 1996).

2.2 Chloramines

Initially, chloramines were used to control taste and odor in drinking water; however, they were soon recognized as being more stable than free chlorine in the distribution system and, consequently, were found to be effective in controlling bacterial growth in the distribution system (Kirmeyer et al., 1993). As a result, chloramines were used regularly for secondary disinfection during the 1930s and 1940s. Because of an ammonia shortage during World War II, however, the popularity of chloramination declined.

The recent concern over halogenated organic byproduct (THM and HAA) formation in water treatment and distribution systems has increased interest in chloramines because they react differently with natural organic matter (NOM) compared to chlorine, generally producing lower concentrations of DBPs (Symons et al., 1998). However, chloramines are not as effective as chlorine for primary disinfection, requiring significantly higher concentrations or contact times to achieve comparable levels of inactivation. Therefore, they are used primarily as a secondary disinfectant. Prior to treatment changes to meet the Stage 1 DBPR, chloramines were used by large surface water systems as a secondary disinfectant during 40 percent of the plant-months included in the reporting period, based on data collected under the ICR (USEPA, 2003b). Use of chloramines for secondary disinfection by community and non-transient, non-community ground water plants is less common (about 5 percent of all systems) (USEPA, 2003b). Chloramine use is expected to increase as the Stage 2 DBPR is implemented, with more than half of both large and small surface water plants predicted to be using chloramines for secondary disinfection by the year 2013, in order to comply with the requirements associated with this rule.

Reaction chemistry

Chloramines are formed by the reaction of ammonia with aqueous chlorine. In aqueous solutions, hypochlorous acid from the chlorine reacts with ammonia to form inorganic chloramines in a series of competing reactions. In these reactions, monochloramine (NH₂Cl), dichloramine (NHCl₂), and nitrogen trichloride (NCl₃) are formed. These competing reactions are impacted by bulk water pH and are controlled to a large extent by the chlorine to ammonianitrogen ratio (Cl₂:NH₃-N). Monochloramine is the predominant species formed in the pH range 7.5-9 (Kirmeyer et al., 2004). As the chlorine concentration increases and pH decreases, dichloramines and nitrogen trichloride can form. Temperature and contact time also affect these reactions. Monochloramine is predominately formed when the applied Cl₂:NH₃-N ratio is less than or equal to 5:1 by weight (Kirmeyer et al., 2004). When certain ratios of chlorine and ammonia-nitrogen are present, chloramines may not form, and ammonia and chlorine may be converted to other molecules that do not act as disinfectants and are not detected when chlorine residual is measured. For instance, as the applied Cl₂:NH₃-N ratio increases from 5:1 to 7.6:1, the water approaches breakpoint chlorination, when the residual chloramine and ammonianitrogen concentrations are reduced to a minimum. Breakpoint chlorination results in the formation of nitrogen gas or nitrate and hydrochloric acid. At Cl₂:NH₃-N ratios above 7.6:1, free chlorine and nitrogen trichloride (trichloramines) are present. Trichloramines are quite volatile and will usually dissipate, however, their formation is typically kept to a minimum due to objectionable odor formation (Kirmeyer et al., 1993). To avoid breakpoint chlorination, utilities normally maintain a Cl₂:NH₃-N ratio of between 3:1 and 5:1 by weight.

Kinetics in the distribution system

Chloramine decay in the distribution system is the result of autodecomposition reactions and reactions with organic and inorganic compounds. Biological nitrification, resulting from chloramine decay or from the presence of excessive ammonia, can cause a large increase in the rate of decay due to consumption of the remaining chloramine residuals. Autodecomposition is highly dependent on pH and temperature, with pH levels above 8 giving the slowest decomposition, and decomposition increasing with increasing temperatures. Higher chloramine residuals also result in an increase in the decay rate. Monochloramine decay has been modeled by Valentine (1998) using a second order rate expression.

2.3 Chlorine dioxide

In a 1998 survey of disinfection practices conducted by the AWWA's Disinfection Systems Committee (AWWA Water Quality Division Disinfection Systems Committee, 2000), approximately 8 percent of 200 large and medium-size respondents reported using chlorine dioxide (ClO₂) as a secondary disinfectant (some surveyed utilities used multiple secondary disinfectants). According to Hoehn et al. (1992), an estimated 700 to 900 U.S. drinking water systems use chlorine dioxide, largely to oxidize iron and manganese, control taste and odor, and reduce THM formation. Nineteen of the more than 500 plants that participated in the ICR reported using chlorine dioxide for at least 9 of the last 12 months of the ICR collection period (USEPA, 2003b).

Although chlorine dioxide is a relatively strong disinfectant, it is not frequently used as a distribution system disinfectant for two reasons: 1) its residual does not last as long as that of other disinfectants, and 2) it breaks down into chlorite (predominantly), a regulated DBP with an MCL. Chlorine dioxide is used more commonly in Europe, even as a secondary disinfectant in France and Germany (Foundation for Water Research, 1993) and the Netherlands (Wondergem and van Dijk-Looijaard, 1991). The USEPA (1999a) recommends that chlorine dioxide use be limited to water suppliers with smaller distribution systems. To ensure a detectable residual at the fringes of the distribution system, a large distribution system may require a larger initial dose of chlorine dioxide than a smaller distribution system. The higher chlorine dioxide dose of the large system might lead to an exceedance of the chlorine MCL as the chlorine dioxide reacts producing chlorate and chlorite ions.

Reaction chemistry

Chlorine dioxide is a neutral compound with chlorine in the +IV oxidation state. Because ClO_2 does not hydrolyze in water, it exists as a dissolved gas as long as the pH of the water ranges from 2 to 10. In strongly alkaline solutions (pH greater than 9 or 10), however, formation rates of DBPs increase with increasing concentrations of ClO_2 . Chlorine dioxide is a volatile free radical that functions as an oxidant by way of a one-electron transfer mechanism in which it is reduced to chlorite $(C1O_2^-)$ (Hoehn and Rosenblatt, 1996; Doerr, 1981). During drinking water treatment, chlorite is the predominant reaction byproduct, with 50–70 percent of the reacted chlorine dioxide converting to chlorite and 30 percent converting to chlorate $(C1O_3^-)$ or chloride $(C1^-)$.

Kinetics in the distribution system

Chlorine dioxide decay in the distribution system is the result of autodecomposition reactions and reactions with organic and inorganic compounds, including biofilms and pipe materials and scales, and also is subject to photolytic decomposition. Several studies using chlorine dioxide as a secondary disinfectant in full-scale distribution systems (Andrews et al., 2001, Volk et al., 2002) have shown that residuals can be maintained throughout these specific systems, without booster stations. Other studies (Gates, 1998) have demonstrated the opposite, that residuals disappear at the ends of the system without booster addition. Residuals decrease faster as the water temperature increases and the size and complexity of the distribution system increase.

3 Overview of Existing Disinfectant Residual Guidelines and Requirements

This section describes the current regulations that address distribution system disinfectant residuals in the United States. Additionally, this section provides a discussion on secondary disinfectants in Europe.

3.1 U.S. Federal Regulations and Guidance

Exhibit 2 provides a summary of Federal regulations that are related to secondary disinfection.

Regulation	Effective	Secondary Disinfection Elements
Surface Water Treatment Rule (SWTR)	1990	 For all systems using surface water or groundwater under the influence of surface water for supply, a detectable disinfectant residual must be maintained within the distribution system in at least 95% of the samples collected (or heterotrophic bacteria counts must be less than or equal to 500 cfu/ml as an equivalent) and at least 0.2 mg/L concentration of residual disinfectant (free or combined) entering the distribution system must be maintained. Monitoring must be conducted throughout the distribution system at same time and locations as those used for total coliform monitoring and continuously at entry point.
Total Coliform Rule (TCR)	1990	 TCR does not require disinfectant residuals or monitoring for disinfectant residuals. TCR lists disinfectant residual as a Best Available Technology for compliance with total coliform Maximum Contaminant Level (MCL).
Stage 1 Disinfectant/Disinfection By-Products Rule (Stage 1 DBPR)	2002	 Establishes Maximum Disinfectant Residual Levels (MRDLs) of 4.0 mg/L as Cl₂ for chlorine, 4.0 mg/L as Cl₂ for chloramine, and 0.8 mg/L for chlorine dioxide. The DBPR also lowers the MCL for total trihalomethanes (TTHMS) from 0.10 mg/L (established in THM Rule) to 0.080 mg/L, and sets new MCLs for haloacetic acids (HAA₅) (0.060 mg/L), chlorite (1.0 mg/L), and bromate (0.010 mg/L). System may use SWTR disinfectant residual monitoring results to determine MRDL compliance. Monitoring must be conducted throughout the distribution system at same time and locations as those used for total coliform monitoring and continuously at entry point

Exhibit 2 - Summary of Regulations for Secondary Disinfectant Residual

3.1.1 Secondary Disinfection in Regulations

Surface Water Treatment Rule (SWTR)

The SWTR was promulgated in June 1989 in 40 CFR Parts 141 and 142. It requires the reduction of Giardia lamblia by 99.9 percent (3-log) and reduction of viruses by 99.99 percent (4-log). The rule applies to all surface water systems, those systems that use ground water under the direct influence (GWUDI) of surface water, and systems that supply surface water to any part of their distribution system or blend surface water with groundwater sources. Systems must filter their water, unless they meet the filtration avoidance criteria, and must disinfect the water sufficiently so that the combination of removal and inactivation achieves the required pathogen reduction levels before the water reaches the first user on the system. This first stage of the disinfection process, before the water enters the distribution system, is referred to as "primary disinfection." The SWTR also requires that systems serving surface water and GWUDI systems maintain a disinfectant residual throughout the distribution system. The free or combined disinfectant residual concentration entering the distribution system must be at least 0.2 mg/L and the system is in violation if it is less than 0.2 mg/L for more than four hours. In addition, the disinfectant residual concentration in the distribution system (known as "secondary disinfection" or "residual disinfection" concentration), measured as free chlorine, combined chlorine, or chlorine dioxide, cannot be undetectable in more than five percent of the samples each month, for any two consecutive months that the system serves water to the public. Water in the distribution system with a heterotrophic plate count (HPC) less than or equal to 500 colony-forming units per milliliter (cfu/ml) is deemed to have a detectable disinfectant residual for purposes of determining compliance with this requirement.

Systems regulated by the SWTR are required to monitor the disinfectant concentration in the water entering the distribution system continuously, and the lowest value must be recorded each day. If there is a failure in the continuous monitoring equipment, grab sampling every four hours can be conducted instead of continuous monitoring, but for no more than five working days following the failure of the equipment. Systems serving fewer than 3,300 people may use grab samples for their disinfectant measurements instead of continuous monitoring. The required number of grab samples taken per day is based on the population served by the system.

Systems complying with the SWTR must measure the disinfectant residual concentration at least at the same points in the distribution system and at the same time as total coliforms are sampled for compliance with the TCR, unless States determine that other sites are more representative of distribution system water quality.

Total Coliform Rule

The TCR was promulgated concurrently with the SWTR in June 1989. Unlike the SWTR, the TCR applies to all public water systems. A public water system is defined as a drinking water supplier that serves at least 25 people or 15 service connections for at least 60 days per year.

The TCR requires systems to monitor for the presence of total coliforms in the distribution system. See the Total Coliform White Paper *Distribution System Indicators of Drinking Water Quality* (USEPA 2006a) for further discussion on the use of total coliforms as indicators. The TCR requires systems to monitor for total coliforms at a frequency proportional to the number of people served. Coliform samples must be collected at sites that are representative of water

throughout the distribution system, according to a written sample siting plan. If any sample tests positive for total coliforms, the system must perform additional tests for either fecal coliforms or *E. coli* and test additional samples for total coliform in response to the positive result.

The TCR does not require the presence of a disinfectant residual but does list maintaining a disinfectant residual throughout the distribution system as a Best Available Technology (BAT) for compliance with the total coliform MCL.

Stage 1 Disinfectants and Disinfection Byproducts Rule

The purpose of the Stage 1 DBP Rule is to improve public health protection by reducing exposure to disinfection byproducts. Some disinfectants and disinfection byproducts (DBPs) have been shown to cause cancer and reproductive effects in lab animals and suggested bladder cancer and reproductive effects in humans. The Stage 1 DBPR, effective in 2002, sets MRDLs and Maximum Residual Disinfection Level Goals (MRDLGs) for chlorine, chloramines, and chlorine dioxide. The MRDLs established by the rule are 4.0 mg/L as Cl₂ for chlorine, 4.0 mg/L as Cl₂ for chloramine, and 0.80 mg/L for chlorine dioxide. The Stage 1 DBPR also lowers the MCL for TTHMs from 0.10 mg/L, established in the 1979 TTHM Rule, to 0.080 mg/L, and sets new MCLs for HAA5 (0.060 mg/L), chlorite (1.0 mg/L), and bromate (0.010 mg/L). Enhanced coagulation or enhanced softening is required to improve removal of DBP precursors for systems using conventional filtration treatment.

Stage 2 Disinfectants and Disinfection Byproducts Rule

For the Stage 2 DBPR, the MCLs will remain at the Stage 1 DBPR levels (0.080 mg/L for TTHM and 0.060 mg/L for HAA5), but compliance will be determined based on locational running annual averages (LRAAs) instead of the RAAs used in the Stage 1 DBPR. Most systems will also be required to conduct Initial Distribution System Evaluations (IDSEs) to identify monitoring locations that represent locations with the highest concentrations of TTHM and HAA5.

Ground Water Rule

The purpose of GWR is to provide for increased protection against microbial pathogens in public water systems that use ground water sources. EPA is particularly concerned about ground water systems that are susceptible to fecal contamination since disease-causing pathogens may be found in fecal contamination. The GWR will apply to public water systems that serve ground water. The rule also applies to any system that mixes surface and ground water if the ground water is added directly to the distribution system and provided to consumers without treatment.

The GWR does not require a disinfectant residual. However, under this rule, ground water systems providing 4-log treatment of viruses using chemical disinfection must monitor for and must meet and maintain a State-determined residual disinfectant concentration (e.g., 4-log inactivation of viruses based on CT tables) or State-approved alternatives every day the GWS serves from the ground water source to the public. Significant deficiencies may include, but are not limited to, inadequate disinfectant residual monitoring, when required.

3.1.2 State Regulations

States may adopt Federal drinking water regulations or adopt more restrictive drinking water requirements, including those for disinfectant residual. At least 34 states have the same regulations as the federal standard (40 CFR 141.72) requiring monitoring, and the same minimum disinfectant concentration at the entrance to the distribution system and within the distribution system. Some states such as Texas, Kentucky, Kansas, and Florida require ground water systems to comply with the disinfectant residual standards as well as surface water and GWUDI systems. Other states have adopted more stringent standards. Several states have increased the minimum disinfectant level to 0.3 or 0.5 mg/L (Delaware and Kentucky, respectively).

3.2 Secondary Disinfection in Europe

The current approaches to secondary disinfection in Europe are influenced by the wide diversity of water resources and supply infrastructures, as well as disinfection philosophy, so European countries vary considerably in their disinfection practices and use of secondary disinfection.

The European Union (EU), which is currently comprised of 25 member states, sets drinking water regulations for its member-states. The European Union Council Directive 98/83/EC was adopted November 3, 1998 to regulate quality of water intended for human consumption. The Directive applies to all water supplies except nationally recognized mineral waters or water used as a medicinal product. Exemptions are allowed where member states are satisfied that the quality of the water has no negative influence on the health of consumers concerned.

The EU Directive does not specifically require water supplies to be disinfected. Residual disinfection is also not required, although the Directive suggests disinfection when necessary. Water must be free of pathogens as measured by *E. coli* and enterococci bacteria (i.e., 0/100 ml mandatory microbiological standard for *E. coli*, and enterococci). The point of compliance with these guidelines is at a customer's tap.

European Union member states may adopt standards and monitoring requirements more stringent than those imposed by the EU Directive. Three countries, Spain, Portugal and the United Kingdom, require primary disinfection for all water supplies. Four countries, Austria, Denmark, France, and the Netherlands, require primary disinfection of surface water, but not groundwater, unless necessary. No other countries in the EU require primary disinfection as a national standard. Out of the 15 original EU member states, only Spain and Portugal require secondary disinfection (or residual disinfection) in distribution systems. Germany and Austria require residual disinfectants as necessary to achieve microbiological standards (no pathogens). Belgium, Finland, France, Ireland, Luxembourg, and Switzerland (not an EU member state) offer guidance on disinfectant residuals.

Some European regulators monitor heterotrophic bacteria while others do not use microorganisms as indicators of water quality (Hydes, 1999).

4 Review of Secondary Disinfection Effectiveness

As discussed earlier, secondary disinfectants have three main functions:

- 1. To inactivate microorganisms in the distribution system,
- 2. To serve as indicators of distribution system upset, and
- 3. To control biofilms.

This section describes each of these functions and what is known about the effectiveness of secondary disinfection using chlorine, chloramine, and chlorine dioxide.

4.1 Inactivation of Microorganisms in Distribution Systems

Studies have shown that disinfectant residuals can be used to inactivate microorganisms in the distribution system. In a study by Snead et al. (1980), researchers showed that a 0.70 mg/L free chlorine residual could effectively inactivate coliform bacteria (3-log inactivation within 30 minutes) when 1% seeded, autoclaved, raw sewage was introduced to tap water. Additionally, more than 1.5-log inactivation of poliovirus 1 was observed after 120 minutes. The initial free chlorine residual lost its effectiveness when challenged with 5% sewage. LeChevallier (1999) states that in cases of massive contamination, the residual may be overwhelmed.

Proponents of maintaining a disinfectant residual point to situations where residuals were not maintained and preventable waterborne disease outbreaks occurred. Haas (1999) argues that both a 1993 *Salmonella* outbreak caused by animal waste introduced to a distribution system reservoir and a 1989 *E. coli* O157:H7 outbreak could have been forestalled if distribution system chlorination had been in effect. Both of these outbreaks were due to bacterial pathogens that are sensitive to chlorine and could have been at least partially inactivated. Whether the extent of inactivation would have been great enough to prevent the outbreak is unknown. Propato and Uber (2004) determined that disinfection practices may provide some public health protection. However, other factors, such as distribution system dynamics and the presence of storage tanks, can affect the vulnerability of consumers to pathogens.

This section focuses on routes by which bacteria enter the distribution system and pathogen inactivation in distribution systems. Estimates of the possible extent of inactivation provided by secondary disinfection and the factors that might influence inactivation are also presented. As with primary disinfection, secondary disinfection effectiveness at pathogen inactivation depends on several factors. For example, turbidity, pH, and chlorine demand of the water containing the pathogens will affect inactivation rates. Pathogen dose and condition will dictate how likely the contamination is to cause waterborne disease. Disinfectant concentration and contact time will impact how strong a treatment barrier the secondary disinfection provides. Exhibit 3 provides a summary of variables that might be considered when evaluating secondary disinfection efficacy.

	Туре	Chlorine
		Chloramine
		Chlorine Dioxide
	Dose	Residual
		Booster Disinfection
Disinfectant Properties:		Mixing Behavior
		-Plug flow
		-Well mixed
		-Unknown
	Reactivity	Low/long-lasting
		High/short-lived

Exhibit 3 - Variables for Consideration within a Secondary Disinfection Framework

variables for Collsid	eration within a Secondary I	
	Mixing Behavior	• Plug flow
		• Well mixed
		Convection
	Disinfectant Demand	• Sewage
		Groundwater intrusion
	Volume	Concentration
		Duration
	Entry Points	Number
Contamination Event Properties:		Spatial Distribution
		• Type
		-Backflow
		-Intrusion
		-Other
	Contaminant Water	• pH
	Quality Characteristics	• Temperature
		• Disinfectant demand
		• Available nutrients
Microorganism Properties:	Туре	Virus
	- 5 F	Bacteria
		Protozoa
		Other
	Number of Organisms	Growth
	itumber of organisms	Die-off
	Matrix	Particle associated
		Free floating
		Sheared biofilm
		Intact biofilm
		Aggregation
		Encapsulation
		Incubation time
	Form	• Spore
		• Cyst
		• Cell (vegetative)
	Point of Origination	Treatment breakthrough
		• Intrusion
		Cross-connection
		Storage Tanks
		• Sediment
	Microbial Interactions	Sediment Competitive

Exhibit 3 Continued Variables for Consideration within a Secondary Disinfection Framework

	cration within a Secondary Di	
Distribution System Properties:	Disinfectant Demand	• Pipes
		Gaskets
		Coatings
		Sediments
		Corrosion products
	Pressure Gradients and	Negative pressures (frequency
	Hydraulic Characteristics	and duration)
	Cross-Connections	
	Pipe/Reservoir	
	Volume	
	Population (Consumer)	
	Density	
	Available Contact Time	Looping
		Storage facilities
		Dead-ends
	Water Quality	• pH
	Characteristics	• Temperature
		Disinfectant demand
		Available nutrients
	ļ	

Exhibit 3 Continued Variables for Consideration within a Secondary Disinfection Framework

4.1.1 Impact of Route of Entry on Pathogen Inactivation

As mentioned previously, maintenance of a disinfectant residual throughout the distribution system may help to maintain the integrity of the distribution system in the following three ways:

- Inactivating microorganisms in the distribution system;
- Indicating distribution system upset; and
- Controlling biofilm growth.

Contamination and its interactions with disinfectant residuals can differ significantly depending on the route by which the contamination enters the distribution system, and this may affect the effectiveness of the residual. For example, the route can determine the volume of contaminants reaching the distribution system. A main break may introduce a high volume of contaminated water in a short period of time. A disinfect residual may be unable to inactivate such a load. On the other hand, sediments from the interior of a tank are likely to enter the distribution system in smaller amounts over a long period of time and thus may be not cause a noticeable drop in residual concentration. In these cases, the ability of the residual to inactivate or indicate is limited.

Additionally, the route can also be a factor in the type of contamination reaching the distribution system. Cross-connections could be a source of a high variety of contaminants, while treatment breakthrough would allow contaminants present in the source water to reach the distribution system.

One way to determine the route of entry of pathogens is to examine the causes of outbreaks. Craun and Calderon (2001) reviewed reported waterborne outbreaks attributed to distribution system deficiencies from 1971 to 1998. Exhibit 4 provides a summary of the deficiencies that caused the outbreaks in community and noncommunity water systems. As the exhibit shows, cross-connections have caused more than half of waterborne outbreaks. Additionally, crossconnections, main conditions, and storage contamination have historically resulted in more than 85% of outbreaks.

	Community Wat	er Systems	Noncommunity V	Vater Systems
Deficiency	Outbreaks	%	Outbreaks	%
Cross-Connection	45	50.6	15	62.5
Corrosion/leaching of metals	12	13.5	1	4.1
Broken or leaking water mains	10	11.2	0	0.0
Contamination during storage	9	10.1	6	25.0
Contamination of mains during construction or repair	5	5.6	1	4.2
Contamination of household plumbing	7	7.9	1	4.2
Inadequate separation of water main and sewer	1	1.1	0	0.0
Total	89	100	24	100

Exhibit 4- Distribution System Deficiencies Causing Outbreaks from 1971 to 1998¹

¹ Adapted from Craun and Calderon, 2001.

The Distribution System White Paper *Health Risks from Microbial Growth and Biofilms in Drinking Water Distribution Systems* (USEPA 2002c) identifies six routes by which pathogens can be introduced into distribution systems. Further information describing the probability of waterborne pathogens entering through each pathway is described further in the White Paper. The six routes are:

- Treatment breakthrough,
- Leaking pipes, valves, joints, and seals,
- Cross-connections and backflow,
- Finished water storage vessels,
- Improper treatment of equipment, materials, or personnel before entry, and
- Intentional introduction of contamination into distribution system.

Treatment Breakthrough

It has been shown that the majority of organisms that colonize the pipe materials in a distribution system can be found in the system's source water (Camper, 1996). Some organisms will break through treatment barriers (Schaule and Fleming, 1997), particularly following rainfall events (USEPA, 1992). *Klebsiella pneumoniae* (a coliform, a few strains of which are opportunistic pathogens) are protected from disinfectants by several means, including their attachment to carbon fines used to control taste and odor (Morin et al., 1996). Ineffective source water treatment may also allow fungi and bulk water diatoms to enter the distribution system (Doggett, 2000). High turbidity water can shield pathogens and reduce disinfectant effectiveness (Berman

et al., 1988; Ormeci and Linden, 2002). The turbidity change associated with treatment breakthrough, however, can be so small that it may go undetected.

Leaking Pipes, Valves, Joints, and Seals

As distribution systems age, they become increasingly vulnerable to leaks, water main breaks, and system failures that can result in microbiological contamination. For water systems serving more than 50,000 people in the United States, the average age of the oldest section of the system is more than 50 years. For the largest systems in the country, the average age of the system's oldest section approaches 100 years (Haas, 1999). Even new water main installations can be susceptible to leakage, and therefore many utilities follow the recommendations for hydrostatic testing of new mains according to AWWA Standard C-600 - Installation of Ductile-Iron Water Mains and Their Appurtenances (AWWA, 1999). Failure to conduct adequate hydrostatic testing could result in the installation of many miles of leaking pipe that could be susceptible to intrusion during a transient pressure event (Friedman et al., 2004).

Utilities commonly have a significant amount of leakage throughout the distribution system. In a survey conducted by Kirmeyer et al. (2001), 18 of 26 utilities surveyed had sufficient metering data to determine loss through leaks and breaks in terms of a percentage of total water produced. Seventeen utilities reported that less than 10% of total water produced is lost to leaks and breaks. One utility reported that water loss due to leaks and breaks is 18% of total water produced. Leakage points that are submerged may provide opportunities for intrusion of contaminated water during transient pressure events (Kirmeyer, et. al., 2001). Pressure changes in the distribution system can result in hydraulic surges that create low or negative pressure waves, which often go undetected by water system operators. As a low or negative pressure wave passes through a pipe, it can cause untreated, exogenous water to be drawn into the pipe through points of leakage or cross-connections. Sources of these pressure changes can be the effects of routine distribution system operation, such as pump startup and shutdown, opening and closing fire hydrants, and sudden changes in water demand (Kirmeyer et al., 2001). Further detail regarding the introduction of contaminants through intrusion is provided in the Distribution System White Paper The Potential for Health Risks from Intrusion of Contaminants into the Distribution System from Pressure Transients (LeChevallier et al., 2002).

While LeChevallier (1999) contends that disinfectant residuals may be overwhelmed by large backflow episodes, maintaining a disinfectant residual throughout the distribution system may be effective at providing a barrier to illness in instances of smaller contamination episodes. Payment et al. (1991) studied waterborne endemic gastrointestinal illness in a Canadian system that experienced many pipe breaks and low disinfectant residuals throughout the distribution system network, especially at the ends of the system. LeChevallier et al. (2002) report that analysis of Payment's data shows that people who lived in zones far away from the treatment plant had the highest risk of gastroenteritis. Transient pressure modeling (Kirmeyer et al., 2001) found that the distribution system studied by Payment was extremely prone to negative pressures, with more than 90 percent of the nodes within the system drawing negative pressures under certain modeling scenarios (e.g., power outages). LeChevallier et al. (2002) suggested that low disinfectant residuals and a vulnerability of the distribution system to pressure transients (reported in Kirmeyer et al., 2001) could account for the viral-like etiology of the illnesses observed in the Payment study.

In 1992, in Cabool, Missouri, the city of 5,000 exceeded its sewer capacity and raw sewage backflowed into water main break sites causing an outbreak that killed 4 people, hospitalized 32, and caused diarrhea and other problems in 243 people. The responsible agent was a pathogenic strain of *Escherichia coli* (Geldreich et al. 1992). At the time, the city did not disinfect the drinking water supply composed of groundwater sources, and repaired mains were not chlorinated before being made operational.

Cross-Connections and Backflow

Cross-connection and backflow events have the potential to occur anywhere within the distribution system. These events have introduced contaminants at storage reservoirs, pump stations, hydrants, at repair sites, and on customer property (USEPA 2002b). According to Exhibit 4, contamination from cross-connections caused most of the distribution system outbreaks in both community and noncommunity water systems.

The level of threat posed by biological contaminants varies dramatically depending on the vector of the disease, the concentration and degree of infectivity of the pathogen, the level of disinfectant residual maintained by the water system, and the health of the individual exposed (Rusin et al., 1997). Further details on the risks associated with cross-connections are included in the Distribution System White Paper titled *Potential Contamination Due to Cross-Connections and Backflow and Associated Health Risks* (USEPA 2002d).

Many of the documented waterborne disease outbreaks caused by cross-connection problems resulted from contamination of the water supply with sewage. Sartory and Holmes (1997) found *E. coli* isolates from sewage effluents to be less resistant to free chlorine than were *E. coli* isolated from distribution system bulk water, although the large number of bacteria introduced during a sewage contamination episode may make this point less important.

Finished Water Storage Vessels

The long hydraulic retention times of many storage tanks can be either beneficial or detrimental to distribution system water quality. Storage tanks can provide contact time for pathogen inactivation if contamination has occurred. Alternatively, significantly increased water age through certain storage facilities can deplete disinfectant residuals and provide reaction time for DBP formation.

Finished water storage tanks are locations that simultaneously can result in the introduction of contamination and have significant impacts on disinfectant residual. Sediments at the bottom of tanks can introduce potential water quality problems such as increased disinfectant demand, microbial growth, disinfection by-product formation, and increased turbidity within the bulk water. Further details on the potential for contaminants to reach the distribution system through storage tanks can be found in the Distribution System White Paper *Finished Water Storage Facilities* (AWWA and EES, 2002a).

Improper Treatment of Equipment or Materials Before and During Main Repair

Exposure of piping materials to contaminants can begin at the point of manufacture. Subsequent handling and storage of piping also present opportunities for exposure. The Distribution System White Paper *New or Repaired Mains* (AWWA and EES, 2002b) describes the potential for contamination associated with main break and repair.

In North America, disinfection of new mains is typically performed in accordance with AWWA Standard C651-99. This standard recommends a disinfection dose of 25 mg/L for a 24-hour contact time, which should result in a CT of 36,000 min•mg/L. The AwwaRF report *Development of Disinfection Guidelines for the Installation and Replacement of Water Mains* (Haas et al., 1998) documents the results of actual field evaluations to test the adequacy of AWWA Standard C651 for Disinfecting Water Mains. The researchers concluded that the AWWA standard provides adequate disinfection: The AWWA-recommended disinfection dose of 25 mg/L for a 24-hour contact time provides more than a 4-log (99.99%) inactivation of heterotrophic bacteria. It was also found that approximately 10 mg/L free chlorine inactivates heterotrophic bacteria to less than 100 cfu/ml. Based on a survey of 250 utilities, Haas et al. (1998) found that 75% of respondents reference the AWWA Standard C651 in their construction documents.

Intentional Introduction of Contaminants into Distribution System

As the Distribution System White Paper *Health Risks from Microbial Growth and Biofilms in the Drinking Water Distribution System* (USEPA, 2002c) points out, insufficient distribution system security could lead to microbial contamination through accidental or intentional means. For example, biological and chemical contaminants could be intentionally introduced by causing a flow reversal at vulnerable nodes in the distribution system such as fire hydrants, blowoffs, and potentially at any user connection.

In November 2001, a Water Quality Technology Conference (WQTC) Water Security Monitoring Panel (AWWA, 2001) encouraged water supplies to adopt three practices to ready themselves for contamination threats:

- Pay attention to significant changes in water quality at entry points, finished water storage reservoirs, and key monitoring locations throughout the distribution system.
- Establish a reliable water quality baseline against which one can compare current monitoring results. The amount of data needed to establish a reference baseline depends on the normal variability of the water.
- Use indicator tests that can provide real-time results that signal the need for further investigation or action. Frequent monitoring of pH, turbidity, conductivity, and disinfectant residual can serve as watchdogs for changes in distribution system water quality.

4.1.2 Effectiveness of Secondary Disinfectant Residuals at Pathogen Inactivation

One of the goals of this paper is to review the effectiveness of secondary disinfectant residuals for microbial inactivation within the distribution system. Three approaches were used to compare pathogen inactivation in the distribution system.

First published CT values for primary disinfection of *Giardia*, *Cryptosporidium*, and viruses were used as a basis for comparison of disinfection practices within distribution systems. Differences between controlled conditions at a water treatment plant versus the dynamic conditions within a distribution system may result in some variation in disinfection. For example, during primary disinfection, a disinfectant is applied at a known dosage to achieve at least a specific residual over a given contact time. Within the distribution system, the disinfectant residual can vary from pipeline to pipeline and from the pipe centerline to the pipe

wall. Water users are drawing water at different locations and rates in the distribution system, and therefore the contact time between two fixed locations will vary throughout the day. Contact time for an early user on a distribution main will differ significantly from users further downstream, especially if they are located downstream of storage vessels. There may be multiple contaminant entry points within the distribution system such as cross-connections, intrusion during pressure transients, and through storage vessels. Additionally, primary disinfection is not aimed at biofilm control, and in some instances, primary disinfection may even contribute to increased biofilm growth within the distribution system if nutrients for bacteria growth, such as humic substances, are converted to more readily biodegradable AOC.

Second, a literature review was conducted to identify studies that assessed disinfection efficacy for bacteria and viruses that could be associated with distribution systems. Results were sorted by microorganism and study matrix (e.g., whether the microorganisms were in association with bulk water, biofilms, or particles/aggregated). Where data were available, an attempt was made to quantify the impact of distribution system conditions (i.e., the presence of biofilm/clumping) on disinfection efficacy, compared to the relatively simplistic bulk water CT approach used to assess primary disinfection efficacy.

Third, findings from the literature review for bulk water inactivation studies were categorized as a function of the disinfectant used so that disinfection efficacy for various bacteria and viruses could be compared.

<u>Comparison of Secondary Disinfection to Primary Disinfection CT Values for Inactivation of</u> <u>Giardia, Viruses, and Cryptosporidium</u>

For chlorine, chloramines, and chlorine dioxide, primary disinfection CT tables have been developed and promulgated for *Giardia* and virus inactivation under the SWTR. These tables were developed using data from inactivation studies, conducted under laboratory conditions that more closely resembled the conditions that are seen during primary disinfection (USEPA, 1991).

Exhibit 5 provides CT values for virus inactivation using Hepatitis A virus (HAV), and Exhibit 6 provides CT values for *Giardia lamblia* cyst inactivation for chlorine, chloramine, and chlorine dioxide. The values in both Exhibits refer to water at 10°C with pH 6.0-9.0 (pH 7 for chlorine inactivation of *Giardia*).

The log inactivations in the tables incorporate conservative assumptions and safety factors, resulting in the likelihood that they underestimate the actual inactivation achieved for a given contact time and concentration combination under primary disinfection conditions when free chlorine or chlorine dioxide are used as the primary disinfectant. The chloramine CT values in Exhibit 5 were developed for systems using combined chlorine, where chlorine is added prior to ammonia in the treatment sequence (USEPA, 1991). These CT values should not be used for estimating the adequacy of disinfection in systems applying preformed chloramines, as would be the case in a secondary disinfection scenario using chloramines, since CT values based on HAV inactivation with preformed chloramines may not be adequate for destroying rotaviruses. For example, CTs of approximately $4,000 - 6,300 \text{ min} \cdot \text{mg/L}$ were needed for 2-log inactivation of simian rotavirus at a pH of 8 and a temperature of 5°C when preformed chloramines shown in Exhibit 6 are based on disinfection studies using preformed chloramines (USEPA, 1991).

Disinfectant	CT Values (in min•mg/L)							
	2-log inactivation	(99.0%)	3-log inactivation	(99.9%)	4-log inactivation	(99.99%)		
Chlorine	3		4		6			
Chloramine ²	643 ²		1,067 ²		1,491 ²			
Chlorine Dioxide	4.2		12.8		25.1			

Exhibit 5 - CT Values for Inactivation of Viruses in Water at 10° C with pH 6.0-9.0¹

¹ Adapted from Guidance Manual for Compliance with the Filtration and Disinfection Requirements for Public Water Systems Using Surface Water Sources (USEPA, 1991).

² Inactivation achieved using combined chlorine, where chlorine is added prior to ammonia in the treatment sequence (USEPA, 1991). Do not apply to preformed chloramines. CT values for preformed chloramines would be significantly higher.

Exhibit 6 - CT Values for Inactivation of *Giardia lamblia* Cysts in Water at 10° C with pH 6.0-9.0¹

	CT Values (in min•mg/L)								
Disinfectant	0.5-log (68.0%)	1.0-log (90.0%)	1.5-log (96.8%)	2.0-log (99.0%)	2.5-log (99.7%)	3-log (99.9%)			
Chlorine ²	17	35	52	69	87	104			
Chloramine ³	310 ³	615 ³	930 ³	$1,230^{3}$	$1,540^{3}$	1,850 ³			
Chlorine Dioxide	4	7.7	12	15	19	23			

¹Adapted from *Guidance Manual for Compliance with the Filtration and Disinfection Requirements for Public Water Systems*

Using Surface Water Sources (USEPA, 1991)² at all 7.0 and ablaring radiable (0.4 mg/l

² at pH 7.0 and chlorine residual <0.4 mg/L ³ CT values for chloremines are based on preformed chlor

³ CT values for chloramines are based on preformed chloramines (USEPA, 1991).

The data presented in Exhibits 5 and 6 (and from Berman and Hoff, 1984 for preformed chloramine inactivation of simian rotavirus using preformed chloramines) can be applied to distribution system disinfection scenarios to theoretically assess the potential for inactivation of viruses and *Giardia lamblia*, should they enter the distribution system, with the exception of the chloramine data in exhibit 5 which does not apply to preformed chloramines. As discussed in Section 3, the SWTR establishes a minimum concentration of disinfectant entering systems (0.2 mg/L) and requires a detectable residual throughout the system. In general, the minimum detectable residual may be considered the detection limit of the field test analysis employed. This is assumed to be 0.01 mg/L for all three disinfectants (AWWA/APHA/WEF, 1998). Of the three possible secondary disinfectants, chloramines are the weakest, requiring significantly higher concentrations or contact times to achieve levels of inactivation of *Giardia* and viruses comparable to free chlorine and chlorine dioxide. The contact time to achieve even a 10 percent inactivation of *Giardia* at the minimum allowable (i.e., "detectable") chloramine residual concentration of 0.01 mg/l is 3,000 minutes (or just over 2 days), while a 99 percent (2-log) inactivation requires 123,000 minutes (or 85 days).

The CT values presented in Exhibits 5 and 6 (and from Berman and Hoff, 1984 for preformed chloramine inactivation of viruses) can be applied to other disinfection residual scenarios, such as the minimum allowable level at the point of entry to the distribution system (0.2 mg/L) under the SWTR, or the mean disinfectant residual concentrations reported in the 1998 AWWA survey

(AWWA Water Quality Division Disinfection Systems Committee, 2000). Exhibit 7 provides a summary of contact times that would be needed within a distribution system to provide 2-log inactivation of viruses and *Giardia* under various disinfectant residual scenarios.

	Minimum Detectable Residual Level	Needed Inactivati Minimun	t Time for 2-log on Using Detect. (min)	Minimum Residual Level Allowed at POE	Needed Inactivati Minimum Level Al	t Time for 2-log ion Using Residual lowed at (min)	Mean Residual Level ¹	Needed Inactivati Mean Resi	ct Time for 2-log ion Using idual Level ir)
Disinfectant	(mg/L)	Virus	Giardia	(mg/L)	Virus	Giardia	(mg/L)	Virus	Giardia
Chlorine ²	0.01	300	6,900	0.2	15	345	1.1	3	63
Chloramine ³	0.01	630,000	123,000	0.2	31,500	6,150	2.4	2,625	513
Chlorine Dioxide ⁴	0.01	420	1,500	0.2	21	75	0.26	16	58

Exhibit 7 - Contact Time Needed to Achieve 2-Log Inactivation of Viruses and Giardia Using Various Distribution System Residual Scenarios

¹ Source: AWWA Water Quality Division Disinfection Systems Committee, 2000

² 10°C, HAV used for virus at pH 6-9; pH 7 used for *Giardia*

³ Preformed chloramines used for both virus and *Giardia*. 5°C for simian rotavirus SA11 at pH 8; 10°C, pH 6-9 for *Giardia*.

⁴ 10°C, HAV used for virus at pH 6-9; pH 6-9 used for Giardia

Chlorine dioxide requires more time than free chlorine to inactivate viruses, but can inactivate *Giardia* more quickly than free chlorine. Whereas 1 mg/L of free chlorine can provide a rapid virus inactivation (4-log inactivation in 6 minutes), 0.26 mg/L of chlorine dioxide needs 16 minutes to provide just 2-log virus inactivation, and 97 minutes to provide 4-log virus inactivation. Chlorine dioxide can, however, provide some protection against *Cryptosporidium* oocyst contamination. Peeters et al. (1989) found that 0.22 mg/L of chlorine dioxide provide 94.3% *Cryptosporidium* oocyst inactivation in 30 minutes.

An inherent limitation with using the CT approach described above for assessing log-inactivation provided in distribution systems is the uncertainty associated with calculating contact times within a distribution system given the potential for multiple and unknown contamination entry points within any distribution system. Primary disinfection CTs are most applicable to distribution systems if one assumes a single contamination event at any one time, such as from the source of supply, or at a specific storage facility, etc. In this hypothetical case, all regions of the distribution system can be identified for which a specific contact time is met (under various demand conditions) and a minimum disinfectant residual is maintained. As described in the Secondary Disinfection Framework presented previously in Exhibit 3, the number, spatial distribution, and type of contaminant entry locations (and many other variables) will impact the actual secondary disinfection efficacy.

Impact of Study Matrix on Inactivation of Bacteria and Viruses Potentially Associated with Distribution Systems

Several studies have assessed disinfection efficacy for microorganisms in conditions that could be found in distribution systems. For example, Payment et al. (1985) found that the presence of viable viruses in finished water was due to their occlusion in protective matter. Sobsey et al. (1991) noted that there is considerable evidence that most viruses in water are embedded in or otherwise associated with suspended solids and that such associate often interferes with virus inactivation. For example, Sobsey et al. (1991) found that cell-associated Hepatitis A virus was more resistant to both free chlorine and monochloramine disinfection than was a dispersed form of the virus. Cell association had a significantly smaller influence on inactivation by monochloramine than by free chlorine; free chlorine was found to be more effective than monochloramine by a factor of 600-fold for dispersed virus, but only 60-fold for cell-associated virus.

Medema et al. (1998) found that approximately one-third of *Cryptosporidium parvum* oocysts and *Giardia lamblia* cysts introduced into secondary sewage effluent attached quickly to particles from the secondary effluent. The affiliation of the cysts and oocysts to particles may enhance the likelihood of pathogen settling, but also may increase the resistance of the cysts and oocysts to disinfection. Leclerc (2002) points out that treatment breakthrough of flocculated particles could result in the introduction of particle-associated pathogens with a greater resistance to disinfection. Sartory and Holmes (1997) hypothesized that the sensitivity of coliforms to chlorination may be related to their source and metabolic status. Several strains of coliform bacteria were isolated from sewage effluent, source waters, and bulk water and biofilms from distribution systems. For *E. coli*, the isolates from the distribution system bulk water showed greater resistance to free chlorine than those from sewage effluents, and equivalent resistance to those from river water. Coliforms other than *E. coli* (mainly strains of *Klebsiella*, *Enterobacter*, and *Citrobacter*) from distribution system biofilms showed the greatest sensitivity to free and total chlorine, while those from river water had the greatest resistance.

LeChevallier et al. (1988) showed that the attachment of bacteria to surfaces provided the greatest increase in disinfection resistance. Attachment of unencapsulated *Klebsiella pneumoniae* grown in medium with high levels of nutrients to glass microscope slides afforded the microorganisms as much as a 150-fold increase in disinfection resistance. Other mechanisms which increased disinfection resistance included the age of the biofilm, bacterial encapsulation, and previous growth conditions (e.g., growth medium and growth temperature). These factors increased resistance to chlorine from 2- to 10-fold. The choice of disinfectant residual was shown to influence the type of resistance mechanism observed. Disinfection by free chlorine was affected by surfaces, age of the biofilm, encapsulation, and nutrient effects. Disinfection by monochloramine, however, was only affected by surfaces. Importantly, results showed that these resistance mechanisms were multiplicative (i.e., the resistance provided by one mechanism could be multiplied by the resistance provided by a second mechanism).

A literature review of CT requirements for inactivation of various bacteria and viruses in the presence of free chlorine, chloramine, or chlorine dioxide was conducted. Results were sorted by microorganism and matrix, i.e., whether the microorganisms were in association with bulk water, biofilms, or particles/aggregated. Exhibit 8 provides a summary of the microorganisms for which published inactivation results were available in a variety of matrices. Appendix A provides a complete listing of all results identified in the literature review.

Micro- organism	Disinfectant	Disinfectant Dose (mg/L)	Disinfectant Residual (mg/L)	CT (min•mg/L) or time (min)	Temperature °C	Log Inactivation	Difference in percent inactivation from non- clumping	Test System	Matrix
Coliforms					-		Different		
assoc. w/							contact		Particle
particles ¹	Chlorine	5	1.5	50	5	3	times	Laboratory	association
Coliform ¹	Chlorine	5	4	15	5	3.7		Laboratory	Bulk water
HPCs ²	Chlorine Dioxide	No data	0.23	14	20 +- 0.5	0.3	-48	Model DS, annular reactors Model DS,	Particle association
HPCs ²	Chlorine Dioxide	No data	0.23	14	20 +- 0.5	1.61		annular reactors	Bulk water
HPCs ²	Chlorine Dioxide Chlorine	No data	0.45	27	20 +- 0.5	2.17	-0.67	Model DS, annular reactors Model DS,	Particle association
HPCs ²	Dioxide	No data	0.45	27	20 +- 0.5	4.00		annular reactors	Bulk water
HPCs ²	Free chlorine	No data	0.47	28	20 +- 0.5	1.6	-1.9	Model DS, annular reactors Model DS,	Particle association
HPCs ²	Free chlorine	No data	0.47	28	20 +- 0.5	2.20		annular reactors	Bulk water
HPCs ²	Free chlorine	No data	0.95	57	20 +- 0.5	2.44	-0.3	Model DS, annular reactors Model DS, annular	Particle association
HPCs ²	Free chlorine	No data	0.95	57	20 +- 0.5	3.25		reactors	Bulk water

Exhibit 9 Comparison of Disinfection Detwoon Dully Water and Distribution S	water Water Conditiona
Exhibit 8 -Comparison of Disinfection Between Bulk Water and Distribution S	system water Conditions

$\begin{array}{c c c c c c c c c c c c c c c c c c c $		<u>s Continued - C</u>	Disinfectant	Disinfectant	CT (min•mg/L)			Difference in percent inactivation		
$\begin{array}{c c c c c c c c c c c c c c c c c c c $	Micro-		Dose	Residual		Temperature	Log	from non-	Test	
$\begin{array}{c c c c c c c c c c c c c c c c c c c $	organism	Disinfectant	(mg/L)	(mg/L)	(min)	°C	Inactivation	clumping	System	Matrix
$\begin{array}{c c c c c c c c c c c c c c c c c c c $									Model DS,	
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	2								annular	Particle
$\begin{array}{c c c c c c c c c c c c c c c c c c c $	HPCs ²	Monochloramine	No data	1.85	111	20 +- 0.5	2.15	-0.4		association
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$,	
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	2									
$\begin{array}{c c c c c c c c c c c c c c c c c c c $	HPCs ²	Monochloramine	No data	1.85	111	20 +- 0.5	2.53		reactors	Bulk water
$\begin{array}{c c c c c c c c c c c c c c c c c c c $										
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$									Model	
$\begin{array}{c c c c c c c c c c c c c c c c c c c $									plumbing	Particle
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	pneumophila ³	Free chlorine	2	No data	t = 30 min	25 - 30	4	0		association
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$										
Legionella pneumophila³Model plumbingModel plumbingLegionella pneumophila³Free chlorine4No data $t < 30 \min$ $25 - 30$ 40systemassociaLegionella pneumophila³Free chlorine4No data $t < 30 \min$ $25 - 30$ 49ModelLegionella pneumophila³Model t < 30 min	0								plumbing	
Legionella pneumophila3Free chlorine4No data $t < 30 \text{ min}$ $25 \cdot 30$ 40plumbingPartice systemLegionella pneumophila3Free chlorine4No data $t < 30 \text{ min}$ $25 \cdot 30$ 40SystemBulk wLegionella pneumophila3Free chlorine4No data $t < 30 \text{ min}$ $25 \cdot 30$ 4ModelImage: systemBulk wLegionella pneumophila3Monochloramine2No data $t < 30 \text{ min}$ $25 \cdot 30$ 40systemassocialLegionella pneumophila3Monochloramine2No data $t < 30 \text{ min}$ $25 \cdot 30$ 40systemassocialLegionella pneumophila3Monochloramine2No data $t < 30 \text{ min}$ $25 \cdot 30$ 40systemBulk wModelModelModelModelModelModelModelModel	pneumophila'	Free chlorine	2	No data	t < 30 min	25 - 30	4			Bulk water
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$										
$\begin{array}{c c c c c c c c c c c c c c c c c c c $		F 11 '				25 20		0	1 0	Particle
Legionella pneumophila3Free chlorine4No datat < 30 min25 - 304plumbing systemBulk wLegionella pneumophila3Monochloramine2No datat < 30 min	pneumophila	Free chlorine	4	No data	t < 30 min	25 - 30	4	0		association
$ \begin{array}{c ccccccccccccccccccccccccccccccccccc$	r · 11									
Legionella Model pneumophila ³ Monochloramine 2 No data t < 30 min	0	F 1	4	N. I.	(<u>20</u>	25 20	4		1 0	D 11
LegionellaplumbingParticpneumophila3Monochloramine2No data $t < 30 \min$ $25 - 30$ 40systemassociaLegionellaImplembingpneumophila3Monochloramine2No data $t < 30 \min$ $25 - 30$ 49ModelModelModelImplembingModelModel	рпеиторпиа	Free chlorine	4	No data	t < 30 min	25 - 30	4		2	Bulk water
pneumophila³Monochloramine2No datat < 30 min25 - 3040systemassociaLegionellaImage: system associapneumophila³Monochloramine2No datat < 30 min	Lagionalla									Darticle
Legionella Model pneumophila ³ Monochloramine 2 No data t < 30 min		Monochloramina	2	No data	t < 30 min	25 30	4	0	1 0	
Legionellaplumbingpneumophila ³ Monochloramine2No data $t < 30 \min$ $25 - 30$ 4systemBulk wModel	рпеиторнии	wonochiorannine	2	no uata	ι < 50 mm	25 - 50	4	0		association
pneumophila ³ Monochloramine 2 No data t < 30 min 25 - 30 4 system Bulk w Model	Legionella									
Model		Monochloramine	2	No data	t < 30 min	25 - 30	4		1 0	Bulk water
	раситорини	monocinoramine	2	110 unu	t < 50 mm	25 50	т		2	Buik water
Legionella nlumbing Parti	Legionella								plumbing	Particle
		Monochloramine	4	No data	t < 30 min	25 - 30	4	0	1 0	association

					Duik water a		Difference		
				СТ			in percent		
		Disinfectant	Disinfectant	(min•mg/L)			inactivation		
Micro-		Dose	Residual	or time	Temperature	Log	from non-	Test	36.1
organism	Disinfectant	(mg/L)	(mg/L)	(min)	°C	Inactivation	clumping	System	Matrix
Legionella								Model plumbing	
pneumophila ³	Monochloramine	4	No data	t < 30 min	25 - 30	4		system	Bulk water
рпситорнии	Wohoemoralinie	_	110 data	t < 50 mm	25 50		Different	Model	Duik water
Legionella							contact	plumbing	Particle
pneumophila ³	Chlorine dioxide	2	No data	t < 40 min	25 - 30	3	times	system	association
								Model	
Legionella		_						plumbing	
pneumophila ³	Chlorine dioxide	2	No data	t = 30 min	25 - 30	4		system	Bulk water
1								Model	Dentiale
Legionella pneumophila ³	Chlorine dioxide	2	0.5 mg/L	t < 30 min	25 - 30	4	0	plumbing system	Particle association
рпеиторний	Chiornic dioxide	2	0.5 mg/L	t < 50 mm	25 - 50		0	Model	association
Legionella								plumbing	
pneumophila ³	Chlorine dioxide	2	0.5 mg/L	t < 30 min	25 - 30	4		system	Bulk water
							Different		Particle
Poliovirus ⁴	Free chlorine	0.5	0.44	2.4	5	4	СТ	No data	association
Poliovirus ⁴	Free chlorine	0.50	0.46	2.3	5	4		No data	Bulk water
									Particle
Poliovirus ⁴	Free chlorine	0.46	0.41	3.3	5	4	0	No data	association
Poliovirus ⁴	Free chlorine	0.46	0.41	3.3	5	4		No data	Bulk water

Exhibit 8 Continued - Comparison of Disinfection Between Bulk Water and Distribution System Water Conditions

Micro-	<u>s continued - (</u>	Disinfectant Dose	Disinfectant Residual	CT (min•mg/L) or time	Temperature	Log	Difference in percent inactivation from non-	Test	
organism	Disinfectant	(mg/L)	(mg/L)	(min)	°C	Inactivation	clumping	System	Matrix
				· · · · ·			Different contact	•	Particle
Poliovirus ⁴	Free chlorine	2.8	1.8	t < 15 min	5	4	times	No data	association
Poliovirus ⁴	Free chlorine	2.8	2.6	t < 5 min	5	4		No data	Bulk water
Vibrio cholerae⁵	Free Chlorine	0.5	No data	0.5	20	2	Different CT	Laboratory	Rugose
Vibrio cholerae ⁵	Free Chlorine	0.5	No data	<0.5	20	5		Laboratory	Smooth
Vibrio cholerae ⁶	Free Chlorine	0.5	No data	0.5	20	2	Different CT	Laboratory	Rugose
Vibrio cholerae ⁶	Free Chlorine	0.5	No data	<0.5	20	5		Laboratory	Smooth
Vibrio cholerae ⁷	Free Chlorine	0.5	No data	0.5	20	3.5	-0.03	Laboratory	Rugose
Vibrio cholerae ⁷	Free Chlorine	0.5	No data	0.5	20	5		Laboratory	Smooth
Vibrio cholerae ⁷	Free Chlorine	1.0	No data	0.2	20	2	Different CT	Laboratory	Rugose
Vibrio cholerae ⁷	Free Chlorine	1.0	No data	0.33	20	4		Laboratory	Smooth

Micro- organism	Disinfectant	Disinfectant Dose (mg/L)	Disinfectant Residual (mg/L)	CT (min•mg/L) or time (min)	Temperature °C	Log Inactivation	Difference in percent inactivation from non- clumping	Test System	Matrix
Hepatitis A ⁸	Free Chlorine	0.5	No data	2.0	5	4	Different CT		Dispersed
Hepatitis A ⁸	Free Chlorine	0.5	No data	27	5	4			Cell
Hepatitis A ⁸	Monochloramine	10	No data	1225	5	4	Different CT		Dispersec
Hepatitis A ⁸	Monochloramine	10	No data	1740	5	4			Cell associatio
Ormeci and Linder Dykstra et al. 2002 Gao et al. 2000 Hoff 1978 Clark et al. 1994 Morris et al. 1996 Rice et al. 1993 Sobsey et al. 1991									

Exhibit 8 Continued - Comparison of Disinfection Between Bulk Water and Distribution System Water Conditions

The protective impact of biofilms or particle association on the inactivation of coliforms and heterotrophic bacteria is clearly shown in Exhibit 8. Ormeci and Linden (2002) found that a free chlorine CT of 15 min•mg/L could provide 3.7-log inactivation of wastewater coliforms that were not associated with particles, but that a CT of 50 min•mg/L was required to provide 3.0-log inactivation of wastewater coliform associated with particles. Thus, those coliforms associated with wastewater particles required more than a three-fold increase in CT to achieve a similar amount of inactivation that occurred for coliforms that were not associated with particles. The authors also suggest that contact time plays an important role in determining the effectiveness of chlorine disinfection in wastewater, and that chlorine dose alone may not be a good indicator of disinfection effectiveness for particle-associated coliforms. The authors concluded that a lower chlorine dose with longer contact time is likely to be more effective on particleassociated coliforms than an identical CT achieved with a higher chlorine dose and shorter contact time.

Dykstra et al. (2002) studied the impact of biofilms on heterotrophic bacteria inactivation using chlorine dioxide, free chlorine, and monochloramine. Greater inactivations were observed for bulk water heterotrophic bacteria compared to those within biofilms, for all three disinfectants, regardless of the CT used.

Gao et al. (2000) compared inactivation of biofilm-associated *Legionella pneumophila* with bulk water *Legionella pneumophila*, using free chlorine, monochloramine, and chlorine dioxide at various disinfectant dosages and a contact time of typically less than 30 minutes. Four-log inactivation was achieved for free chlorine and monochloramine, at all dosages, regardless of biofilm association. For chlorine dioxide at a dosage of 2 mg/L, only 3-log inactivation was observed for the biofilm-associated microorganisms when a disinfectant residual could not be maintained over the full experimental contact time. Comparatively, 4-log inactivation was observed for the bulk water microorganisms when chlorine dioxide was used. A replicate experiment that maintained a disinfectant residual of 0.5 mg/L yielded 4-log removal for both biofilm associated and bulk water microorganisms.

Ormeci and Linden (2002) reported that the decay rate of chlorine was the same for both the particle-associated and non-particle-associated samples, and similar chlorine decay rates were observed at all chlorine concentrations (1 mg/L, 5 mg/L, 10 mg/L, and 15 mg/L). The average total chlorine loss over the duration of the experiment was approximately 3.5 mg/L for the samples that had initial total chlorine concentrations of 5, 10, and 15 mg/L.

Hoff (1978) compared CTs and log inactivations for poliovirus with and without particle association, and for different types of particle matrices. Tests were conducted at a pH of 6 and a temperature of 5°C. To achieve 4-log inactivation of poliovirus, free chlorine CTs in the range of 2.3 to 3.3 min•mg/L were sufficient in the absence of particles, when the virus was associated with bentonite (7.1 NTU), or when the virus was associated with aluminum phosphate (5.0 NTU). However, a free chlorine CT of 23 min•mg/L was required to achieve 4-log inactivation when the virus was associated with cell debris. This and other studies (Hoff and Akin, 1986; Sproul et al., 1979; Hejkal et al., 1979; Stagg et al., 1977; Boyce et al., 1981; Scarpino, 1979), suggest that the effects of

microorganism-particle association on disinfection efficiency are determined by the nature of the association. Viruses associated with cell debris, feces, or wastewater effluent solids are substantially protected where as viruses and bacteria adsorbed on surfaces of particles such as clays or inorganic flocs are only minimally protected.

Berman et al., (1988) compared free chlorine and chloramine disinfection of coliforms associated with particles $< 7\mu$ m and $> 7\mu$ m in size. Sieves and nylon screens were used to separate primary sewage effluent solids into the various particle size fractions. The free chlorine study was conducted at a pH of 7 and a temperature of 5°C, and the chloramine study was conducted over the pH range of 7 to 8.5, at 5°C. To provide 2-log inactivation using free chlorine (0.5 mg/L), a CT of 0.9 min•mg/L was required in association with particles $< 7\mu$ m, compared to a CT of 2.7 in association with particles $> 7\mu$ m. When the larger particles were homogenized, the free chlorine CT required for 2-log inactivation of coliform was reduced to 0.5 min•mg/L. Thus, the authors concluded that larger particles (> 7µm) can have a protective effect against the disinfecting action of chlorine for bacteria and protozoans, due to their larger size as compared to viruses. At pH of 7.0, particle size did not have a significant impact on coliform inactivation using chloramine. However, chloramine inactivated the smaller particles more quickly than those $> 7 \mu$ m at pH of 8.0. Using chloramine as a disinfectant, at either pH, a 99% inactivation necessitated a CT twenty to fifty times greater than that for chlorine at a pH of 7.0.

Hoff and Aiken (1986) reviewed factors affecting the efficacy of chlorine disinfection on microorganisms. The authors concluded that in comparison with growth conditions and aggregation, the association of a microorganism with particulate matter affords the greatest protection from disinfection. The study also found that pathogens are most likely to be introduced to drinking water through an association with particulates, primarily fecal particles. Additionally, the type of particulate matter has an impact on vulnerability to disinfection. For instance, viruses and bacteria adsorbed onto clays are still vulnerable to disinfection. However, viruses associated with cell debris, feces, or wastewater effluent are less vulnerable to disinfection.

Abu-Shkara et al. (1998) tested nutrition (high and low), temperature (6°C and 35°C), and aggregation (0.45-8 μ m-sized aggregates) with selected coliforms to evaluate the impacts of these environmental variables on chlorine resistance. The results of their experiments showed that coliform bacteria grown at lower temperatures are more resistant to chlorination, as are bacteria grown in low nutrient conditions. Predictably, the authors also found that bacterial species that formed aggregates in the water were also more resistant to chlorination.

Vibrio cholerae O1 has both smooth and "rugose" strains that respond differently to disinfection (Rice et al., 1993, Clark et al., 1994, and Morris et al., 1996). The rugose strain appears to produce a mucoid matrix material and has a tendency to aggregate (Rice et al., 1993). Morris et al. (1996) determined that contrary to previous understanding, rugose *V. cholerae* is virulent to humans. Rice et al. (1993) found that 4-log inactivation of smooth strains occurred in 20 seconds with the application of 1.0 mg/L free chlorine. However, with the same application of chlorine, 3-log inactivation of rugose *V. cholerae* was consistently more resistant to free chlorine disinfection than the smooth strain under

differing pH, temperature, and chlorine applications. Rugose strains were composed of larger particles than smooth strains. Broth rugose strains were less chlorine-resistant than those grown on solid media (agar). Clark et al. (1994) and Rice et al. (1993) both indicate that the mucoid matrix and cellular aggregation are the likely cause of the rugose strain's increased resistance. Clark et al. (1994) point out that aggregate rugose strains are less likely to be a problem at the treatment plant due to their size, but could be a potential contamination risk within the distribution system. Clark et al. (1994) indicate that if introduced to the distribution system through a main break or similar incident, it would be very difficult for chlorine to adequately inactivate a rugose strain of V. *cholerae*.

Impact of Disinfectant Type on Inactivation of Bacteria and Viruses Potentially Associated with Distribution Systems

Exhibit 9 summarizes CT values and corresponding inactivation rates for bulk water microbes in the presence of free chlorine. Although the laboratory studies did not typically mimic distribution system conditions, the results present a potential range of CTs that might be required to achieve different levels of inactivation for different microbes that could be associated with distribution systems. It is expected that a wide range of environmental conditions (i.e., pH, temperature, residual level, etc.) would also be encountered in drinking water distribution system.

Of the microbes presented, adenovirus, calicivirus, *Helicobacter pylori*, and *Microsporidia* are included on the USEPA Contaminant Candidate List (CCL), under consideration for additional research and regulatory determination. The data in Exhibit 9 suggest that under the range of conditions tested (shown in Appendix A), a CT of <150 min•mg/L will provide between 2-log and 4-log inactivation of most microbes studied when free chlorine is used as the disinfectant and microbes are not particle-associated or aggregated. Exceptions include *Legionella pneumophila* and *Mycobacterium fortuitum* which required a range of CT values that exceeded 200 min•mg/L to achieve 2-log inactivation using free chlorine.

It should be noted that viruses require 4-log inactivation (under the SWTR). Under the research conditions identified in the literature (and summarized in Appendix A), 4-log inactivation of poliovirus with free chlorine was achieved at CT values of <3.3 min•mg/L. While 4-log inactivation of adenovirus and calicivirus were not observed in the literature reviewed, experimental CT values were typically very low (i.e., 0.01-1.0 min•mg/L). Furthermore, it should be noted that the data summarized in Exhibit 9 represent inactivation under bulk water conditions. As discussed previously, microorganisms that are particle associated are typically less vulnerable to disinfection.

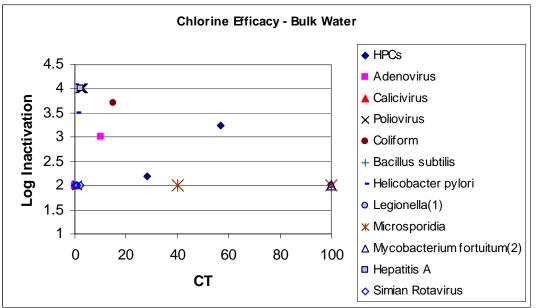


Exhibit 9- Summary of CT and Log Inactivation Data Using Free Chlorine for Various Microbes

Note: Data source, pH, temperature, disinfectant dose, and other information provided in Appendix A.

(1) For *Legionella*, CT range of 100 to 600 min•mg/L required for 2-log inactivation.

(2) For Mycobacterium fortuitum, CT range of 100 to 1000 min•mg/L required for 2-log inactivation.

Exhibits 10a and 10b summarize CT values and corresponding inactivation rates for bulk water microbes in the presence of chloramines. Exhibit 10a shows results over the CT range 0-30,000 min•mg/L, whereas 10b focuses on the CT range of 0-900 min•mg/L. The results presented are from laboratory studies that typically did not mimic distribution system conditions, although the microbes studied could be associated with distribution systems. Of the microbes presented, Aeromonas, adenovirus, and calicivirus are included on the CCL. The data in Exhibit 10a suggest that under the conditions tested (shown in Appendix A), a CT of 10,000 min•mg/L would provide 2-log inactivation of most microbes studied when chloramine is used as the disinfectant, and microbes are not particle-associated or aggregated. Two-log inactivation of *Bacillus subtilis* was achieved over a CT range of 3,200 to 20,000 min•mg/L. Two-log inactivation of Nitrosomonas europaea was achieved over a CT range of 1,900 to 19,000 min•mg/L. As shown in Exhibit 10b, which provides more detail for CT values less than 1,000 min•mg/L, 2-log inactivation of several organisms was achieved by chloramines at CT values less than 150 min•mg/L. It should be noted, however, that 4-log inactivation of viruses was not observed in the literature reviewed when chloramine was used as the disinfectant.

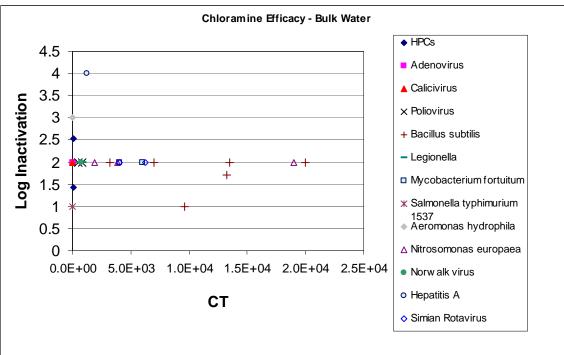
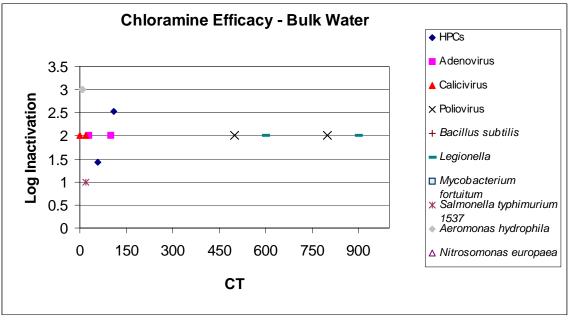


Exhibit 10a - Summary of CT and Log Inactivation Data Using Chloramines for Various Microbes

Note: Data source, pH, temperature, disinfectant dose, and other information provided in Appendix A.

Exhibit 10b- Log Inactivation of Various Microbes at CT Values Less than 1000 min•mg/L



Note: Data source, pH, temperature, disinfectant dose, and other information provided in Appendix A.

Exhibit 11 summarizes CT values and corresponding inactivation rates for bulk water microbes in the presence of chlorine dioxide. The results presented are from laboratory studies that typically did not mimic distribution system conditions, although the microbes studied could be associated with distribution systems. The data in Exhibit 11 suggest that under the conditions tested, a CT of 150 min•mg/L would provide 2-log inactivation of most microbes studied when chlorine dioxide is used as the disinfectant, and microbes are not particle-associated or aggregated. *B. subtilis* required CT values in the range of 40 to 365 min•mg/L to achieve 2-log inactivation. It should be noted, however, that 4-log inactivation of viruses was not observed in the literature reviewed when chlorine dioxide was used as the disinfectant.

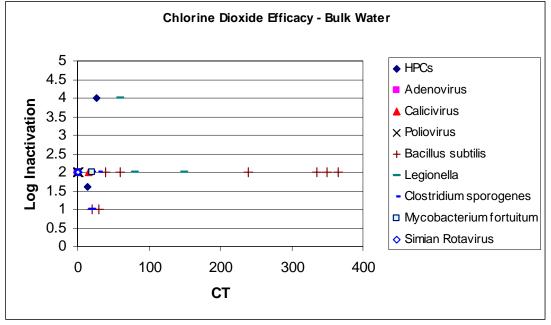


Exhibit 1111 – Summary of CT and Log Inactivation Data Using Chlorine Dioxide for Various Microbes

Note: Data source, pH, temperature, disinfectant dose, and other information provided in Appendix A.

It is important to emphasize that the data presented in Exhibits 9 through 11 were developed under laboratory conditions (as summarized for each data point in Appendix A) and address microbes within the bulk water. While this approach may reasonably represent conditions within storage facilities where the bulk water-to-sidewall surface area ratio is quite large, in light of the secondary disinfection framework variables described previously in Exhibit 3, some variability would be expected in distribution system pipelines. Payment (1999) questioned the effectiveness of free chlorine residuals at providing significant pathogen inactivation in distribution systems. The authors found that sporulated bacteria and viruses added to distribution system water samples containing < 0.9 mg/L free chlorine were nearly unaffected by the residual chlorine. The authors cautioned that, while *E. coli* and thermotolerant coliforms were rapidly inactivated, microorganisms such as *Clostridium perfringens*, somatic coliphages, and poliovirus were almost unaffected by free chlorine for several hours.

4.2 Secondary Disinfectant Residuals as Indicators of Distribution System Upset

Many factors influence the concentration of the disinfectant residual in the distribution system, including the NOM level, the type and concentration of disinfectant, water temperature, and system hydraulics (Trussell, 1999). Entry of foreign material into the distribution system from backflow (or other events) may alter these factors and contribute to a loss of residual. In some cases, reductions in a disinfectant residual can signify the existence of an accidental or intentional contamination problem in the distribution system, including those resulting from cross-connections and backflow (Haas, 1999).

Snead et al., (1980) recognized that a free chlorine residual could be used as an indicator of contamination. If a system that normally has no trouble maintaining a free chlorine residual detects an absence of residual, this may indicate the presence of a contaminant in the system exerting a chlorine demand. Disinfectant residuals can be measured easily, and operators often use residual concentrations as a way to track system operations. While a sudden decrease in the disinfectant residual could be due to other problems such as failure of the feed system, the decrease could reflect the interaction of the disinfectant with material associated with contaminants entering the distribution system, due to a main break, backflow, or sewage leak into the system. Snead et al., (1980) note that combined chlorine residuals may not be effective as indicators of distribution system upset since they are slower to react with constituents in drinking water.

Several studies agree (Craun and Calderon, 2001; Clement et al., 1999) that large episodes of contamination, such as cross-connections through which sewage may enter distribution systems, will overwhelm disinfectant residuals. The chlorine demand of the organic matter carried with sewage may prevent effective inactivation if chlorine or chloramines are being used.

Since most disinfectants are chemical oxidants that react with many substances, their effectiveness as indicators of microbiological contamination may be limited. Inorganic and organic chemicals in the water can present a disinfectant demand that could misleadingly alert operators when no pathogens have been introduced. Furthermore, the presence of disinfectant-resistant pathogens, such as *Cryptosporidium*, may persist in a distribution system despite the presence of the disinfectant. However, the loss or decrease of the disinfectant residual in this case can serve as an indicator of some contamination events. The use of disinfection residual monitoring as an indicator for microbiological contamination, especially in regard to contamination due to treatment breakthrough, is not entirely reliable. The clearest examples of this were the *Cryptosporidium* outbreaks in Georgia (Hayes et al., 1989), Oregon (Leland et al., 1993), and Milwaukee (MacKenzie et al., 1994), during which chlorine residuals were maintained throughout the distribution systems of the supplies delivering contaminated water. Thus, the contamination events did not pose a noticeable disinfectant demand within the distribution system.

Accurate and on-going tracking of disinfectant residuals at critical control points is needed if sudden changes in residual levels are to be identified and used as indicators of contamination. The identification of critical control points within distribution systems is addressed in the Issue Paper *Evaluating HACCP Strategies for Distribution System Monitoring, Hazard Assessment and Control* (USEPA, 2006b). Water system operators

are becoming increasingly sophisticated in tracking and measuring disinfectant residuals. Real-time sensors of chlorine residuals (measuring both free and total chlorine) have been developed, and water suppliers are beginning to couple such monitoring tools with distribution system controls (Haas, 1999). Some advantages to using disinfectant residual monitoring as a warning mechanism for possible contamination are that residual analysis is inexpensive, results are immediately available, and USEPA-approved methods for analysis already exist.

The USEPA Water Protection Task Force (USEPA, 2001) suggested that water supplies increase the frequency and locations of disinfectant residual monitoring in their distribution systems to ensure proper residuals at all points in the system and to establish a baseline and normal fluctuations from the baseline. The Task Force stated that strategically placed residual monitors are an effective way to signal an unexpected increase in disinfectant demand and, possibly, a breach or contamination of the distribution system.

Denver Water, in Denver, Colorado, successfully used on-line chlorine residual monitoring within the distribution system to identify a decrease in total chlorine levels caused by high silt loading after a forest fire within the watershed. The chlorine levels continued to diminish as the water moved further through the distribution system. The increased chlorine demand was linked to high dissolved manganese levels from the silt washed into the reservoir. The online monitoring results enabled staff to take prompt action to increase chlorine residuals leaving the treatment plant (Kirmeyer et al., 2002).

Some disinfectant residual sampling strategies (e.g., grab samples), may not be frequent enough to detect a reduction in disinfectant residual concentrations for transient events, such as many backflow or intrusion incidents. For example, surface water and GWUDI systems are required to monitor disinfectant residuals at the same locations and frequencies as coliform samples under the TCR. Depending on the size of the water supply and population served, disinfectant residual monitoring can be quite infrequent. Since backflow or transient events can occur over a period of seconds, minutes, or hours, it is possible that a grab sampling regime for disinfectant residual monitoring may not detect the potential increases in disinfectant demand that can be associated with certain types of contamination events.

4.3 Biofilm Control

Disinfection of drinking water does not result in the inactivation of all microorganisms. The growth of bacteria and other microbes in distribution systems has been documented for many years. Problems associated with biofilms in distribution systems include enhanced corrosion of pipes and deterioration of water quality. Biofilms can also provide ecological niches that are suited to the potential survival of pathogens (Walker and Morales, 1997). Biofilms are often a consortium of different microorganisms bound to each other and to pipe surfaces by a polysaccharide matrix. Biofilm formation has been shown to be affected by several factors including disinfectant effectiveness, the nature and concentration of biodegradable compounds in the water, pipe materials used for distribution system construction, and water temperature. Proper management of these factors through adequate source water treatment, appropriate materials selection,

maintenance of a clean distribution system, and minimization of water age are all important for biofilm control. Further details on biofilms are included in the Distribution System White Paper *Health Risks from Microbial Growth and Biofilms in the Drinking Water Distribution System* (USEPA 2002c).

Many factors influence the concentration of the disinfectant residual in the distribution system, and therefore the ability of the residual to control microbial growth and biofilm formation. These factors include the AOC level, the type and concentration of disinfectant, water temperature, pipe material, and system hydraulics. The number of variables associated with biofilm control has led researchers to reach differing conclusions regarding the effectiveness of secondary disinfectants at controlling biofilm growth, as illustrated in the discussion below.

Impact of Disinfectant Concentration on Biofilm Growth

The ability to control (but not eliminate) biofilms using secondary disinfection is impacted by the disinfectant residual concentration used in the system. If concentrations are too low, the disinfectant residual becomes ineffective at controlling excess biofilm growth. Several studies have shown that biofilm growth is reduced when sufficient disinfectant residuals are maintained in the bulk water passing through pipes. Zhang and DiGiano (2002) compared bacterial growth in the distribution systems of two North Carolina cities, Durham and Raleigh. The systems delivered water that came from similar surface water sources and received comparable treatment. The systems differed in that Raleigh uses chloramines to maintain its residual and Durham uses chlorine. Although they did not find a difference in heterotrophic bacteria counts between the two systems, the authors did find strong negative correlations between free chlorine residual and heterotrophic bacteria levels (in Durham's system) and between chloramine and heterotrophic bacteria levels (in Raleigh's system).

Momba (1997) also found a large increase in biofilm microorganisms on test coupons in the absence of a disinfectant residual. This study also showed that maintenance of only 0.2 - 0.5 mg/L free chlorine or 0.8 - 1.0 mg/L chloramine could not be relied on to prevent bacterial adhesion onto stainless steel coupons, cement coupons, and glass surfaces. Characklis (1988) found that heterotrophic bacteria levels were controlled in the bulk water but grew in the biofilm when water carried free chlorine residuals of 0.3 - 0.8 mg/L.

LeChevallier et al. (1996) found that distribution systems that maintained dead-end free chlorine residuals less than 0.2 mg/L or chloramine levels less than 0.5 mg/L had substantially more coliform occurrences than systems maintaining higher residuals. LeChevallier et al. (1990) found that systems with high AOC concentrations needed to maintain higher disinfectant residuals to control coliform occurrences, suggesting that maintenance of a disinfectant residual alone will not ensure that treated waters will be free of coliform bacteria. The study suggested that coliform growth in the distribution system could be controlled with a free chlorine residual of 1.0 to 2.0 mg/L at AOC levels less than 5 to 10 μ g/L. Van der Kooij (1987) and Schellart (1986) indicated that no final disinfection is needed in the Netherlands water systems, provided that AOC levels are less than 5 to 10 μ g/L. Gagnon et al. (1998) found that levels of biodegradable organic mater significantly affected distribution system microbial growth if chlorine residual fell

below a critical level, defined as Ccrit. The value of Ccrit was found to be systemspecific, depending on other factors which promote bacterial growth, such as water age or pipe materials.

Impact of Disinfectant Type on Biofilm Growth

Certain disinfectants may have characteristics that make them more effective at controlling biofilms than others. Chloramines, which are less reactive and therefore more persistent than free chlorine, may penetrate biofilms better and thereby control biofilm growth more effectively (Van der Wende and Characklis, 1990). Most research on the effects of chloramines has focused on monochloramine, since it is the preferred form for chloramine disinfection as discussed in Section 2. LeChevallier et al. (1990) found that both free chlorine and monochloramine at fairly low levels (1 mg/L) effectively reduced heterotrophic bacteria associated with biofilms grown on galvanized, copper, or PVC pipe surfaces. Neither free chlorine nor monochloramine, however, were effective at reducing biofilm on iron pipes unless residual concentrations were raised to above 2 mg/L. When residual concentrations were raised, monochloramine out-performed free chlorine in reducing the heterotrophic bacteria levels. It has been suggested that monochloramine does not react with iron pipe material in the same way that free chlorine does, suggesting that monochloramine is more readily available for inactivation of biofilm organisms (LeChevallier, et al., 1990). Momba (1997) also found that monochloramine and hydrogen peroxide were more effective at controlling biofilm growth in laboratory-scale units than were chlorine, ozone, or ultraviolet light (UV).

Some opportunistic pathogens such as *L. pneumophila, M. avium*, and primary pathogens such as *V. cholerae*, and *E. coli* O157:H7 survive and even grow within certain common amoeba (Barker and Brown, 1994; Barker et al., 1999; Wadowsky et al., 1991; Cirillo et al., 1997; Thom et al., 1992) and may be protected from disinfection. Some of the biofilm organisms may even supply an essential nutrient to facilitate the growth of an opportunistic pathogen. In one study, *Legionella* grew only near colonies of the bacterium *Flavobacterium breve* on an L-cysteine-deficient medium (Wadowsky and Yee, 1983).

Several studies have compared the effectiveness of various disinfectants at controlling bacterial growth. These studies have been performed on different scales, ranging from continuous flow annular reactors to pilot systems to comparisons of full-scale distribution systems. Several studies have concluded that chloramines are more effective secondary disinfectants with respect to biofilm control than chlorine in terms of biofilm control (Camper et al., 2000; LeChevallier et al., 1996; LeChevallier et al., 1990). Whereas chlorine is more effective at microbiological inactivation in distribution system bulk water, chloramine may penetrate biofilms and inactivate attached bacteria more effectively. Stewart et al. (2001) state that the penetration of antimicrobial agents into biofilms is controlled by the reactivity of the antimicrobial agent with biofilm components. The high reactivity of chlorine, therefore, blunts its penetration through the biofilm. Disinfectants with lower reactivities, such as chloramine, are more limited in the types of compounds with which they will react, lending them a specificity that may allow them to inactivate microorganisms in complex biofilms. However, there is a lack of agreement among research results on this topic, as illustrated below.

Dykstra et al. (2002) compared log inactivations for heterotrophic bacteria within biofilms in the presence of free chlorine, chlorine dioxide, and chloramine. A "low" and "high" residual concentration was evaluated for each disinfectant. Annular reactors were used for the study, and the pH and temperature were held at 7.5±0.2 and 20±0.5°C, respectively. Exhibit 12 summarizes the required CTs for achieving various log inactivations for each disinfectant type. The results indicate that free chlorine and chlorine dioxide provided equal to or greater log inactivation of heterotrophic bacteria compared to monochloramine for "high" disinfectant residual concentrations tested, and that chlorine was nearly twice as effective as chloramine at the "low" concentrations tested.

Disinfectant	Residual (mg/L)	CT (min•mg/L)	Log Inactivation
Chlorine Dioxide	0.23 Low	14	0.3
	0.45 High	27	2.17
Free Chlorine	0.47 Low	28	1.6
	0.95 High	57	2.44
Monochloramine	0.79 Low	58	0.86
	1.85 High	111	2.15

Exhibit 12 - Comparison of Disinfectant Effectiveness for Biofilm Heterotrophic Bacteria Inactivation

While this study and other studies provide a comparison of disinfection efficacy for the three disinfectant residuals, it is important to note that the CT approach used in the SWTR was developed to assess inactivation of free-floating microorganisms in buffered demand-free water. Thus, the log inactivations cited in biofilm-related studies are not directly comparable to log inactivations presented for various disinfectants and microorganisms in the SWTR.

Gao et al., (2000) compared free chlorine, monochloramine, and chlorine dioxide inactivation of *Legionella pneumophila* within biofilms grown in a model plumbing system. Slug dosages of either 2 or 4 mg/L for monochloramine and chlorine were tested (residual disinfectant levels were not reported), whereas chlorine dioxide was tested at a single dose of 2.0 mg/L and at an initial dose of 2.0 mg/L followed by maintaining 0.5 mg/L residual. A 3-log inactivation of *Legionella* in both biofilm and bulk water phases was observed within 30 minutes of contact for all three disinfectants. Within 30 minutes, more than 4-log inactivation of biofilm-associated *Legionella* was achieved by the 4.0 mg/L monochloramine slug dose, 4.0 mg/L chlorine slug dose, and 2.0 mg/L chlorine dioxide slug dose with 0.5 mg/L residual maintenance. At the lower concentrations (2 mg/L slug doses of monochloramine, chlorine dioxide, and chlorine), only chlorine provided inactivation of all detectable biofilm and bulk water *Legionella* in the 48-hour disinfection period. Monochloramine provided inactivation of all detectable biofilm and bulk water *Legionella* in the disinfection period.

Walker and Morales (1997) studied the impact of biocides on a microbial culture consisting of a mixed microbial consortium obtained from a potable water system. The authors found that 1.0 mg/L of chlorine dioxide was needed to inactivate the bulk water

bacterial population in a continuous culture chemostat model by 99.92% (18-hour contact time), whereas 1.5 mg/L was required to achieve a similar reduction in the biofilm.

LeChevallier et al. (1990) used a model pipe system to compare disinfectant effectiveness at biofilm control. Comparison of equal activities (and equal CT) of hypochlorous acid, hypochlorite, chlorine dioxide, and monochloramine on bacteria grown on various surfaces suggested that monochloramine penetrated and inactivated biofilm bacteria more effectively than the other disinfectants. Moreover, increasing the CT of free chlorine tenfold did not appreciably increase its disinfection efficiency.

Although monochloramine may be more effective at reducing counts of viable bacteria in biofilms, in some instances, chlorine has been shown to be more effective at physically removing biofilm from pipes. LeChevallier et al. (1990) found that TOC and carbohydrate levels increased in the pipe system when free chlorine was used instead of monochloramine, and they attribute this increase to sloughing of material from the pipe surface into the water column. Chen and Stewart (2000), on the other hand, did not find a significant difference in biofilm removal when chlorine was used compared to chloramine. They did, however, find that monochloramine inactivated bacteria in the biofilm better than did free chlorine at neutral pH.

Full-scale comparisons of disinfectants and their effectiveness at limiting bacterial growth have more mixed results than the smaller-scale, controlled studies. Kool et al. (2000) found that hospitals supplied with drinking water containing free chlorine were 10.2 times more likely to have reported an outbreak of Legionnaire's disease associated with potable water than hospitals that used water with monochloramine as a residual disinfectant. Norton and LeChevallier (1997) found substantial decreases in coliform occurrences and heterotrophic bacteria numbers in two distribution systems when they switched from free chlorine to chloramines. They also found improved maintenance of a disinfectant residual and a decrease in disinfection byproducts when chloramines were used. The authors caution, however, that high concentrations of AOC and pitting corrosion appeared to also affect coliform occurrence, reinforcing that disinfection alone may not be enough to control coliform growth in all distribution systems.

Neden et al. (1992) compared bacterial growth in three study areas of the distribution system of the Greater Vancouver (B.C.) Water District, with the following three treatments: 1) chloramine (2.5 - 3 mg/L dose at the plant), 2) free chlorine (0.2 - 0.5 mg/L residuals), and 3) no secondary disinfectant. The investigators looked at what percentage of monthly heterotrophic bacteria samples contained more than 500 cfu/ml and how often the percentage of positive monthly coliform samples exceeded 10%. Findings of the study included:

- The study area with no secondary disinfectant had a higher percentage of heterotrophic bacteria counts that were >500 cfu/ml and a higher percentage of total coliform positive samples than the other two study areas.
- In the area treated with chloramines, the monthly heterotrophic bacteria samples containing more than 500 cfu/ml ranged from 3% to 10% during the study period. In this area, positive coliform samples occurred in more than 10% of all monthly samples during only two months of the 12-month study.

- In the chlorinated area, often more than 20% of monthly heterotrophic bacteria samples contained more than 500 cfu/ml. During the study, the chlorinated area experienced positive coliform levels at a rate of greater than 10% of all monthly samples during six months.
- In the area with no secondary disinfection, a range of 30% 98% of monthly heterotrophic bacteria samples contained more than 500 cfu/ml during the study period. Positive coliform samples exceeded the 10% level for twelve months of the two-year study.
- Chloramine was found to be significantly better at maintaining a residual than chlorine, and chloramine was more effective at controlling coliform and heterotrophic bacteria numbers in pipe biofilms. During the study, chloramine levels of > 2.0 mg/L were maintained, and free chlorine levels ranged from < 0.1 mg/L to 0.5 mg/L.

The types of chloramines present may also influence their effectiveness. USEPA (1999a) indicates that studies have not been able to definitively determine which chloramine exhibits greater disinfection efficacy. Dichloramine has exhibited better inactivation efficiency in some tests (Esposito, 1974) and monochloramine has in others (Dorn, 1974; Esposito, 1974; and Olivieri, 1980). Additionally, investigators have demonstrated that solutions containing equal amounts of monochloramine and dichloramine provide better disinfection than those with only one of the chloramines (Weber and Levine, 1944). Monochloramine is the preferred chloramine. Additionally, dichloramines are more corrosive and decrease in predominance at pH values of 7 to 8. One study found that neither chlorine residuals nor chloramine residuals alone were able to control biofilm development, however when used in combination (i.e., free chlorine followed by monochloramine), biofilms were controlled (Momba and Binda, 2002).

Little information is available about the effectiveness of chlorine dioxide at controlling biofilms. Since biofilm biocides appear to favor more specific reactants that can diffuse more readily into the biofilm, chlorine dioxide's high level of specificity suggests that it could be very effective at inactivating biofilm bacteria. Walker and Morales (1997) found that chlorine dioxide was effective at inactivating biofilm bacteria, but only when the chlorine dioxide concentration was held at 1.5 mg/L. This concentration exceeds the MRDL for chlorine dioxide of 0.8 mg/L. Chen and Stewart (2000) and Simpson et al. (2002) found that chlorine dioxide was effective at inducing biofilm sloughing as well as bacterial inactivation.

Role of Pipe Material in Disinfectant Effectiveness for Biofilm

Pipe material plays an important role in biofilm growth and disinfectant effectiveness. In some instances, pipe material may be more influential than the level of organic matter in the system (Volk and LeChevallier, 1999). Some materials provide the microbes a protective niche where growth can occur, while some provide nutrients to support microbial growth. Chlorine's ability to control biofilm depends on the pipe material, because different pipe materials demonstrate different levels of chlorine demand. LeChevallier et al. (1990) found that free chlorine residuals achieved greater biofilm

inactivation compared to chloramine for PVC and copper pipes. For galvanized pipes, monochloramine provided greater biofilm inactivation than free chlorine. Iron pipes seem to exert the greatest disinfectant demand. In the same study, the disinfectant demand of biofilm on iron pipes was as much as ten times greater than for biofilms grown on other pipe materials. Concentrations of 1 mg/L of either free chlorine or chloramine could reduce viable counts of heterotrophic bacteria and coliforms by more than 2-log in biofilms grown on galvanized, copper, or PVC pipe surfaces. For iron pipes, however, free chlorine residuals from 3-4 mg/L were ineffective for biofilm control, and only monochloramine residuals greater than 2 mg/L succeeded at reducing viable counts of heterotrophic bacteria and coliform. Monochloramine residuals ranging from 0.33 mg/L to 1.11 mg/L did not significantly reduce biofilm counts, even when applied for seven days. LeChevallier et al. (1990) found that corrosion control improved the efficiency of free chlorine disinfectant efficiency.

The bacterial levels on disinfected iron pipes generally exceed those on disinfected PVC pipes (Norton and LeChevallier, 2000). Biofilms also develop more rapidly on iron pipes, even with corrosion control (Haas et al., 1983; Camper, 1996). In addition, iron pipes support a more diverse microflora compared to PVC pipes (LeChevallier, 1999a). Iron pipes facilitate the development of tubercles, which are primarily iron oxides (Tuovinen et al., 1980), and these tubercles can adsorb organic material (Geldreich, 1996; Geldreich and LeChevallier, 1999). In this manner, the level of corrosion and tuberculation (i.e., buildup of corrosion pitting products) affect biofilm development. Sloughing of biofilms into the water column can also occur as a result of elevated biofilm levels on iron pipes (Norton and LeChevallier, 2000).

5 **Opportunities for Additional Research**

There are several areas where opportunities for additional research exist regarding disinfectant residuals and their multi-purpose role of inactivating microorganisms in the distribution system, serving as indicators of distribution system upset, and controlling biofilms. A few of these areas include:

- To what extent does microbiological contamination chronically or sporadically enter distribution systems through leaking pipes and valves or as a result of pressure transients?
- Do inactivation rates of pathogens differ based on their route of entry? Are pathogens entering via treatment breakthrough either hardier or more vulnerable to disinfection?
- More full-scale studies are needed that evaluate the effectiveness of disinfection on biowarfare agents in water.
- What level of chlorine demand is associated with different types of contamination events? How do chlorine and chloramine differ in this capacity?
- If a public water system intends to use reduction in disinfectant residual as an early warning of distribution system upset, where should residual monitoring take place in the system and how frequently? How can a system determine how large

a reduction in residual needs to take place in order for it to be considered a significant indication of contamination?

- How accurately does bulk water sampling of heterotrophic bacteria, coliforms, or other microbes reflect biofilm composition and the potential threat posed by pathogens in biofilms?
- More full-scale distribution system studies could be carried out that consider the effectiveness of different disinfectants and different residual concentrations on biofilm composition and growth.
- If pathogens are present in biofilms, to what extent does a disinfectant residual inactivate or injure impair them? How is infectivity affected by pathogen exposure to residual disinfection?
- How do distribution system disinfection regimens that switch disinfectants at certain times of the year affect pathogens, coliforms, and heterotrophic bacteria in biofilms and the bulk water of distribution systems?

References

Abu-Shkara, F., I. Neeman, R. Sheinman, and R. Armon. 1998. The effect of fatty acid alteration in coliform bacteria on disinfection resistance and/or adaptation. *Water Science & Technology* 38(12):133-139.

Anderson, E., M.A. Larson, B. Corona-Vasquez, and B.J. Mariñas. 2001. A comparative study of the inactivation kinetics of *Cryptosporidium parvum* oocysts and *Bacillus subtilis* spores with chemical disinfectants. In *Proceedings of 2001 AWWA Water Quality Technology Conference*. AWWA: Denver, CO.

Andrews, R. et.al. 2001. Chlorine dioxide trial as a post disinfectant in Wiarton, Ontario. In *Proceedings of the 4th International Symposium on chlorine dioxide*. February 15-16. Las Vegas, NV.

AWWA and EES. 2002a. *Finished Water Storage Facilities*. Distribution System White Paper. <u>http://www.epa.gov/safewater/tcr/pdf/storage.pdf</u>. Accessed November 16, 2004.

AWWA and EES. 2002b. *New or Repaired Mains*. Distribution System White Paper. http://www.epa.gov/safewater/tcr/pdf/maincontam.pdf. Accessed November 16, 2004.

AWWA 2001. WQTC Water Security Monitoring Panel. November, 2001.

AWWA Water Quality Division Disinfection Systems Committee. 2000. Committee report: disinfection at large and medium-size systems. *Journal AWWA*, Vol 92(5):32-43

AWWA. 1999. AWWA Standard C-600 - Installation of Ductile-Iron Water Mains and Their Appurtenances. AWWA: Denver, CO.

AWWA/APHA/WEF. 1998. *Standard Methods for the Examination of Water and Wastewater*. 20th ed. American Public Health Association, American Water Works Association, Water Environment Federation: Washington DC.

Barbeau, B., I. Myre, N. Facile, R. Desjardinsand, and M. Prévost. 1998. Evaluating disinfection processes: aerobic spore formers as a surrogate for *Giardia* and *Cryptosporidium*. In *Proceedings of 1998 Water Quality Technology Conference*. AWWA: Denver, CO.

Barker, J, TJ Humphrey, and MWR Brown. 1999. Survival of Escherichia coli O157 in a soil protozoan: implications for disease. *FEMS Microbiol. Letters*. 173:291-295.

Barker, J, and MRW Brown. 1994. Trojan horses of the microbial world: protozoa and the survival of bacterial pathogens in the environment. *Microbiology*. 140(6):1253-1259.

Baumann, E. Robert and Daniel D. Ludwig. 1962. Free available chlorine residuals for small nonpublic water supplies. *Journal AWWA*. 54 (11): 1379-1388.

Berman, D., Hoff, J. 1984. Inactivation of simian rotavirus SA11 by chlorine, chlorine dioxide, and monochloramine. *Applied and Environmental Microbiology*, 48 (2): 317-323

Berman, D., E.W. Rice, J.C. Hoff. 1988. Inactivation of particle-associated coliforms by chlorine and monochloramine. *Applied and Environmental Microbiology*, 54 (2): 507-512.

Boyce, D. S., O.J. Sproul, and C.E. Buck, 1981. The effect of bentonite clay on ozone disinfection of bacteria and viruses in water. *Water Research*, 15: 759-767.

Brown, N.P., J.G. Jacangelo, C.N. Haas, and C.P. Gerba. 2002. Assessment of existing disinfection practices for inactivation of emerging pathogens. In *Proceedings of 2002 AWWA Annual Conference*. AWWA: Denver, CO.

Burrows, W.D. and S.E. Renner. 1999. Biological warfare agents as threats to potable water. *Environ. Health Perspect*. 107(12):975-984.

Camper, A.K., P. Butterfield, B. Ellis, W.L. Jones, W.B. Anderson, P.M. Huck, R. Slawson, C. Volk, N. Welch, and M. LeChevallier. 2000. *Investigation of the Biological Stability of Water in Treatment Plants and Distribution Systems*. AwwaRF and AWWA: Denver, CO.

Camper, AK. 1996. Factors limiting growth in distribution systems: laboratory and pilot-scale experiments. AWWARF: Denver, CO.

Chauret, C., C. Radziminski, R. Creason, and R. Andrews. 2000. Chlorine dioxide inactivation of *Cryptosporidium parvum* oocysts and bacterial spores in natural waters. In *Proceedings of 2000 AWWA Annual Conference*. AWWA: Denver, CO.

Chen, X. and P.S. Stewart. 2000. Biofilm Removal Caused by Chemical Treatments. *Water Research*, Vol. 34(17):4229.

Cirillo, JD, S Falkow, LS Tompkins, and LE Bermudez. 1997. Interaction of *Mycobacterium avium* with environmental amoebae enhances virulence. *Infec. and Immunity*. 65:3759-3767.

Clark, Robert and Walter Grayman. 1998. *Modeling Water Quality in Drinking Water Distribution Systems*. AWWA: Denver, Co.

Clark, Robert M., E. Rice, B. Pierce, C. Johnson, and K. Fox. 1994. Effect of aggregation on *Vibrio cholerae* Inactivation. *Journal of Environmental Engineering*, 120 (4):875-887.

Clement, J., C. Haas, N. Kuhn, M. LeChevallier, R. Trussell, D. Vander Kooij. 1999. Roundtable: the disinfectant residual dilemma. *Journal AWWA*, 91(1): 24-30. Craun, G.F. and R.L. Calderon. 2001. Waterborne disease outbreaks caused by distribution system deficiencies. *Journal AWWA*, 93 (9):64-75.

Doggett, MS. 2000. Characterization of fungal biofilms within a municipal water distribution system. *Applied and Environmental Microbiology*, 66 (3): 1249-1251.

Doerr, R. L. 1981. Reactions of chlorine, chlorine dioxide and mixtures thereof with humic acid. *In Water chlorination, environmental input and health*, vol. 2. Edited by R. C. Jolley. Ann Arbor Science Publishing, Inc.: Ann Arbor, MI

Dorn, J. M. 1974. A Comparative Study of Disinfection on Viruses and Bacteria by Monochloramine. Master's thesis, Univ. Cincinnati, Ohio

Dykstra, Trevor, K. O'Leary, C. Chauret, R. Andrews, and G. Gagnon. 2002. Impact of UV disinfection on biological stability in distribution systems. In *Proceedings of the Water Quality Technology Conference*. American Water Works Association: Denver, CO.

Esposito, M.P. 1974. *The Inactivation of Viruses in Water by Dichloramine*. Master's thesis, Univ. Cincinnati, Ohio.

Finch, G. R., L. L. Gyürék, L. R. J. Liyanage, and M. Belosevic. 1997. *Effect of various disinfection methods on the inactivation of Cryptosporidium*. AwwaRF and AWWA: Denver, CO.

Foundation for Water Research. 1993. *Policy and practice of drinking water disinfection in France, Germany, and the Netherlands*. Report FR0370. Summary at www.fwr.org/environs/fr0370.htm. Accessed August 27, 2002.

Friedman, M., L. Radder, S. Harrison, D. Howie, M. Britton, G. Boyd, H. Wang, R. Gullick, M. LeChevallier, D. Wood, and J. Funk. 2004. *Verification and Control of Pressure Transients and Intrusion in Distribution Systems*. AwwaRF and AWWA: Denver CO

Gagnon, G.A., P.M. Huck, G.S. Irwin and PlJ. Ollos. 1998. Defining the relationships between BOM, chlorine residual, and bacterial growth. In *Proceedings of Protecting Water Quality in the Distribution System: What is the Role of Disinfection Residuals?* AWWA: Denver, CO

Gao, Yang, R. Vidie, R. McCall, J. Stoul, and V. Yu. 2000. Monochloramine and chlorine dioxide as alternative disinfection method for Legionella control: results of pilot-studies in model plumbing. In *Proceeding of the AWWA 2000 Annual Conference*. AWWA: Denver, CO

Gates, D.J. 1998. *The chlorine dioxide handbook*. Ed: B. Cobban. American Water Works Association. Denver, CO.

Geldreich, E.E., and M. LeChevallier. 1999. Microbiological quality control in distribution systems, Chapter 18. pp. 18.1-18.49. In *Water Quality and Treatment* (5th ed.). Edited by R.D. Letterman. McGraw-Hill, Inc: New York, NY.

Geldreich, E.E. 1996. *Microbial quality of water supply in distribution systems*. Lewis Publishers: Washington, DC.

Geldreich, E. E., K. R. Fox, J. A. Goodrich, E. W. Rice, R. M. Clark, and D. L. Swerdlow. 1992. Searching for a water supply connection in the Cabool, Missouri disease outbreak of *Escherichia coli* O157:H7. *Water Research*, 26(8): 1127-1137.

Haas, Charles N. 1999. Benefits of using a disinfectant residual. *Journal AWWA*, 91 (1):65-69.

Haas, C. N., R. B. Chitluru, M. Gupta, W. O. Pipes, and G. A Burlingame. 1998. *Development of Disinfection Guidelines for the Installation and Replacement of Water Mains*. AwwaRF and AWWA: Denver, CO.

Haas, C.N., M.A. Meyer, and M.S. Paller. 1983. The ecology of acid-fast organisms in water supply, treatment and distribution systems. *Journal AWWA*, 75:139-144.

Hambsch, B. and P. Werner. 1996. The removal of regrowth enhancing organic matter by slow sand filtration. Chapter in *Advances in slow Sand and Alternative Biological Filtration*. Edited by N. Graham and R. Collins. John Wiley & Sons: Chichester, UK.

Hayes, E.B. et al. 1989. Large community outbreak of cryptosporidiosis due to contamination of a filtered public water supply. *New England Journal of Medicine*, 320(21):1372-1376.

Hejkal, T.W., F.M. Wellings, P.A. LaRock, and A.L. Lewis, 1979. Survival of poliovirus within organic solids during chlorination. *Applied and Environmental Microbiology*, 38(1) 114-118.

Hoehn, R.C., and A.A. Rosenblatt, 1996. Full-scale, high-performance chlorine dioxide gas generator. Poster presented at *AWWA Engineering and Construction Conference*. AWWA: Denver, CO.

Hoehn, R.C., A.A. Rosenblatt, and D.J. Gates. 1992. Considerations for chlorine dioxide treatment of drinking water. In *Proceedings of 1992 AWWA Water Technology Conference*. AWWA: Denver, CO.

Hoff, J.C. and E.W. Akin. 1986. Microbial resistance to disinfectants: mechanisms and significance. *Environmental Health Perspectives*, 60: 7-13

Hoff, J.C. 1978. The relationship of turbidity to disinfection of potable water. In *Evaluation of the Microbiology Standards for Drinking Water*. Edited by C.H. Hendricks. USEPA 570/9-78-00C. U.S. Environmental Protection Agency, Washington DC.

Hydes, Owen. 1999. European regulations on residual disinfection. *Journal AWWA*, 91 (1): 70-74.

Johnson, C.H., E.W. Rice, and D.J. Reasoner. 1997. Inactivation of *Helicobacter pylori* by chlorination. *Applied and Environmental Microbiology*, 63(12): 4969-4970.

Kirmeyer, G., K. Martel, G. Thompson, L. Radder, W. Klement, M. LeChevallier, H. Baribeau, and A. Flores. 2004. *Optimizing Chloramine Treatment: Second Edition*.

Kirmeyer, G., M. Friedman, K. Martel, G. Thompson, A. Sandvig, J. Clemente, and M. Frey. 2002. *Guidance Manual for Monitoring Distribution System Water Quality*. AwwaRF: Denver, CO.

Kirmeyer, G.K., M. Friedman, K. Martel, D. Howie, M. LeChevallier, M. Abbaszadegan, M. Karim, J. Funk, and J. Harbour. 2001. *Pathogen Intrusion into the Distribution System*. AwwaRF and AWWA, Denver, CO.

Kirmeyer, G.J., G.W. Foust, G. L. Pierson, J.J. Simmler, M.W. LeChevallier. 1993. *Optimizing Chloramine Treatment*. AwwaRF and AWWA: Denver, CO

Kool, J.L., J.C. Carpenter, and B.S. Fields. 2000. Monochloramine and Legionnaires' disease. *Journal AWWA*, 92(9): 88-96

Larson, M.A. and B.J. Mariñas. 2000. Comparing the inactivation kinetics of *Bacillus* subtilis spores and *Cryptosporidium parvum* oocysts with ozone and monochloramine. In *Proceeding of 2000 AWWA Annual Conference*. AWWA: Denver, CO.

LeChevallier, M., Gullick, R., Karim, M. 2002. The Potential for Health Risks from Intrusion of Contaminants into the Distribution System from Pressure Transients. Distribution System White Paper. <u>http://www.epa.gov/safewater/tcr/pdf/intrusion.pdf</u>. Accessed on November 16, 2004.

LeChevallier, M.W. 1999. The case for maintaining a disinfectant residual. *Journal AWWA*. 91(1): 86-94.

LeChevallier, M.W., N.J. Shaw, and D.B. Smith. 1996. *Factors limiting microbial growth in distribution systems: full-scale experiments*. AWWARF: Denver, CO

LeChevallier, M.W., C.D. Lowry, and R. G. Lee. 1990. Disinfecting biofilms in a model distribution system. *Journal AWWA*, 82: 87.

LeChevallier, M.W., C.D. Cawthon, and R. G. Lee. 1988. Factors promoting survival of bacteria in chlorinated water supplies. *Applied and Environmental Microbiology*, 54 (3): 649-654

Leclerc, H 2002. Relationships between water bacteria and pathogens in drinking water. In *Proceedings of the NSF International/WHO Symposium on HPC Bacteria in Drinking Water: Public Health Implications?* April 22-24, 2002. Geneva Switzerland. Leland, D., J. McAnulty, W. Keen, and G. Stevens. 1993. A Cryptosporidiosis outbreak in a filtered water supply. *Journal AWWA*, 85(6): 34-42.

MacKenzie, W.R. et al. 1994. A massive outbreak in Milwaukee of Cryptosporidium infection transmitted through the public water supply. *New England Journal of Medicine*, 331(3):161-167.

McGuire, M.J., McLain, J.L., Obolensky, A. 2002. Information Collection Rule Data Analysis. AwwaRF and AWWA: Denver, CO.

Medema, G.J. et al. 1998. Sedimentation of Free and Attached *Cryptosporidium* Oocysts and *Giardia* Cysts in Water. *Appl. Envir. Microbiol.* 64(11):4460-4466.

Momba, M.N.B., and M.A. Binda. 2002. Combining chlorination and chloramination processes for the inhibition of biofilm formation in drinking surface water system models. *J. Appl. Microbiol.* 92:641-648.

Momba, M.N.B. 1997. *The Impact of Disinfection Processes on Biofilm Formation in Potable Water Distribution Systems*. Ph.D. Thesis, Univ. of Pretoria, South Africa.

Morin, P., A. Camper, W. Jones, D. Gatel, and J.C. Goldman. 1996. Colonization and disinfection of biofilms hosting coliform-colonized carbon fines. *Appl. Environ. Microbiol.* 62:4428-4432.

Morris, J.G., M. Sztein, E.W. Rice, J.P. Nataro, G.A. Losonsky, P. Panigrahi, C.O. Tacket, and J.A. Johnson. 1996. *Vibrio cholerae* 01 can assume a chlorine-resistant rugose survival form that Is virulent for humans. *Journal of Infectious Diseases*, 174: 1364-1368.

Neden, D.G., R. Jones, J. Smith, G. Kirmeyer, and G. Foust 1992. Comparing chlorination and chloramination for controlling bacterial regrowth. *Journal AWWA*, 84: (7):80

Norton, C. and M. LeChevallier. 1997. Impact of Biological Filtration. In *Proceedings* of 1996 Water Quality Technology Conference. AWWA: Denver, CO.

Norton and LeChevallier. 2000. A pilot study of bacterial population changes through potable water treatment and distribution. *Appl. Environ. Microbiol.* 66:268-276.

Oldenburg, P.S., J. M. Regan, G.N. Harrington, and D.R. Noguera. 2002. Kinetics of *Nitrosomonas europaea* inactivation. *Journal AWWA*, 94(10): 100-110.

Olivieri, V.P., 1980. Reaction of Chlorine and Chloramines with Nucleic Acids Under Disinfection Condition. In *Water Chlorination: Environmental Impact and Health Effects*, Vol. 3. Edited by R.J. Jolley. Ann Arbor Science Publishers, Inc.: Ann Arbor, MI.

Ormeci, B. and K.G. Linden. 2002. Comparison of UV and chlorine inactivation of particle and non-particle associated coliform. *Water Science and Technology: Water Supply*, 2 (5-6): 403-410.

Parker, D.L., B.R. Schram, J.L. Plude, and R.E. Moore. 1996. Effect of metal cations on the viscosity of a pectin-like capsular polysaccharide from the cyanobacterium *Microcystis flos-aquae* C3-40. *Appl Environ Microbiol*. 62(4):1208-1213.

Payment, P. 1999. Poor efficacy of residual chlorine disinfectant in drinking water to inactivate waterborne pathogens in distribution systems. *Canadian Journal of Microbiology*, 45(8):709-715.

Payment, P. et al. 1991. A Randomized trial to Evaluate the Risk of Gastrointestinal Disease due to Consumption of Drinking Water Meeting Current Microbiological Standards. *Am. Journal of Public Health* Vol. 81, No. 6: 703-708.

Payment, P. et al. 1985. Elimination of Viruses and Indicator Bacteria at Each Step of Treatment during Preparation of Drinking Water at Seven Water Treatment Plants. *Appl. Environ. Microbiol.* 49(6):1418-1428.

Peeters, J.E., E. Ares Mazas, W.J. Masschelein, I. Villacorta, and E. Debacker. 1989. Effect of disinfection of drinking water with ozone or chlorine dioxide on survival of *Cryptosporidium parvum* oocysts. *Applied and Environmental Microbiology*, 55(6):1519-1522

Propato, M. and J. G. Uber. 2004. Vulnerability of water distribution systems to pathogen intrusion: How effective is a disinfectant residual? *Environ. Sci. & Technol.* In Press.

Richardson, S.D. 1998. Drinking water disinfection by-products. In *The Encyclopedia of Environmental Analysis and Remediation*. Edited by R. Meyers. John Wiley and Sons: Hoboken, NJ.

Rusin, P.A., J.B. Rose, C.N. Haas, and C.P. Gerba. 1997. Risk assessment of opportunistic bacterial pathogens in drinking water. *Reviews in Environmental Contamination Toxicology*. 152:57-83.

Safe Drinking Water Committee. Board on Toxicology and Environmental Health Hazards, Assembly of Life Sciences, National Research Council. 1980. *Drinking water and health. Vol. 3.* Washington, DC: National Academy Press.

SAIC (Science Applications International Corp.). 1996. *Ultraviolet Light Disinfection Technology in Drinking Water Application*. An Overview. EPA/811/R-96/002. Washington, DC: U.S. Environmental Protection Agency.

Sartory, D.P., and P Holmes. 1997. Chlorine sensitivity of environmental, distribution system, and biofilm coliforms. *Water. Science and Technology*. 35(11-12): 289-292.

Scarpino, P.V. 1979. *Effect of Particulates on Disinfection of Enteroviruses in Water by Chlorine Dioxide*. USEPA-600/2-79-054. US Environmental Protection Agency: Cincinnati, OH.

Schaule, G, and H-C Fleming. 1997. Pathogenic microorganisms in water system biofilm need biofilm sampling. Ultrapure Water. Corrosioneering - microorganisms in water system biofilm. <u>http://www.clihouston.com/microrg.htm</u>. April, 1997.

Schellart, J.A. 1986. Disinfection and Bacterial Regrowth: Some Experiences of the Amsterdam Water Works Before and After Stopping the Safety Chlorination. *Water Supply*, 4: 217-225.

Shin, G.A., G. Ishida, K.G. Linden, and M.D. Sobsey. 2002. Sequential disinfection with UV irradiation and chlorine species on several important waterborne pathogens. In *Proceedings of 2002 AWWA Water Quality Technology Conference*. AWWA: Denver, CO.

Simpson, G.D., R. Miller, G. Laxton, and W. Clements. 2002. *A Focus on Chlorine Dioxide: The "Ideal" Biocide*. <u>http://www.clo2.com/reading/waste/corrosion.html</u>. Accessed May 2004.

Sirikanchana, K., L. Raskin, and B.J. Mariñas. 2002. Inactivation kinetics of *Aeromonas hydrophila* with monochloramine. In *Proceedings of 2002 AWWA Water Quality Technology Conference*. AWWA: Denver, CO.

Snead, M.C., V. Olivieri, C. Kruse, and K. Kawata. 1980. *Benefits of Maintaining a Chlorine Residual in Water Supply Systems*. USEPA 600/2-80-010. USEPA: Washington, DC.

Sobsey, M.D., J.A. Kase, M.E. Anderson, M.J. Casteel, C. Likindopulos, and E. Sickbert-Bennett. 2000. Inactivation of *Cryptosporidium parvum* oocysts and other waterborne microbes by oxidants generated electrochemically from sodium chloride from portable pen and bench scale systems. In *Proceedings of 2000 AWWA Water Quality and Technology Conference*. AWWA: Denver, CO.

Sobsey, M.D., T. Fuji and R.M. Hall. 1991. Inactivation of Cell-Associated and Dispersed Hepatitis A Virus in Water. *Journal AWWA*, 83 (11):64-67.

Sproul, O.J., C.E. Buck, M.A. Emerson, D. Boyce, D. Walsh, and D. Houser, 1979. *Effect of Particulates on Ozone Disinfection of Bacteria and Viruses in Water*. USEPA-600/2-79-089. U.S. Environmental Protection Agency: Cincinnati, OH.

Stagg, O.H., C. Wallis, and C.J. Ward, 1977. Inactivation of clay-associated bacteriophage MS-2 by chlorine. *Applied and Environmental Microbiology*. 33 (2): 385-391.

Stevens, M., N. Ashbolt, and D. Cunliffe. 2001. Microbial Indicators of Water Quality: An NHMRC Discussion Paper. National Health and Medical Research Council. Canberra, Australia.

Stewart, P.S. et al. 2001. Biofilm penetration and disinfection efficacy of alkaline hypochlorite and chlorosulfamates. *J. Appl. Microbiol.* 91(3):525-532.

Symons, J.M., G.E. Speitel Jr., C. Hwang, S.W. Krasner, and S.E. Barrett. 1998. *Factors affecting disinfection byproduct formation during chloramination*. Project #803. Report 90728. AwwaRF: Denver CO.

Thom, S., D. Warhurst, and B. S. Drasar. 1992. Association of *Vibrio cholerae* with fresh water amoebae. *J. Med. Microbiol.* 36:303-306.

Trussell, RR. 1999. Safeguarding distribution system integrity. *Journal AWWA* 91(1):46-54.

Tuovinen, O.H., K.S. Button, A. Vuorinen, L. Carlson, D. Mair, and L.A. Yut. 1980. Bacterial, chemical, and mineralogical characteristics of tubercles in distribution pipelines. *Journal AWWA*, 72 (11):626-635.

Tyrrell, S.A., S.R. Rippey, W.D., Watkins, and A.L. Marcotte Chief. 1995. Inactivation of bacterial and viral indicators in secondary sewage effluents, using chlorine and ozone. *Water Research*, 29:2483-2490.

USEPA. 2006a. *Distribution System Indicators of Drinking Water Quality*.. Total Coliform Rule White Paper. (Not yet posted)

USEPA. 2006b. Evaluating HACCP Strategies for Distribution System Monitoring, Hazard Assessment and Control. Total Coliform Rule White Paper. (Not yet posted)

USEPA. 2003a. Introduction to SDWA: Part 2. History Part 1 Before 1974. Website: www.epa.gov/safewater/dwa/electronic/presentations/sdwa/pt2/sdwa34.html.

USEPA, 2003b. Occurrence Assessment for the Stage 2 Proposal. USEPA: Washington DC. Accessed on <u>http://www.awwa.org/countterr/docs/epa11_08_01.pdf</u>.

USEPA. 2002a. *Community Water System Survey*. Vol 2 Office of Water. USEPA 815-R-02-005B.

USEPA. 2002b. Information Collection Request for the Stage 2 Disinfectants and Disinfection Byproducts Rule. Prepared by The Cadmus Group.

USEPA. 2002c. *Health Risks From Microbial Growth and Biofilms in Drinking Water Distribution Systems*. Distribution System White Paper. http://www.epa.gov/safewater/tcr/pdf/biofilms.pdf. Accessed on November 16, 2004. USEPA 2002d. Potential Contamination Due to Cross-Connections and Backflow and the Associated Health Risks: An Issue Paper. Distribution System White Paper. http://www.epa.gov/safewater/tcr/pdf/ccrwhite.pdf. Accessed on November 16, 2004.

USEPA. 2001. *EPA Water Protection Task Force Notice #V: Water Supply Systems*. Accessed on <u>http://www.awwa.org/countterr/docs/epa11_08_01.pdf</u>.

USEPA. 1999a. Guidance Manual for Alternative Disinfectants and Oxidants. Office of Water. EPA 815-R-99-014.

USEPA. 1992. Seminar Publication: Control of Biofilm Growth in Drinking Water Distribution Systems. EPA/625/R-92/001. Washington, DC.

USEPA. 1991. Guidance Manual for Compliance with the Filtration and Disinfection Requirements for Public Water Systems Using Surface Water Sources. Science and Technology Branch Criteria and Standards Division Office of Drinking Water.

USEPA 1985. Turbidity Criteria Document. Draft.

Valentine, R.L., et al. 1998. *Chloramine Decomposition in Distribution System and Model Waters*. Denver, Colo.: AwwaRF and AWWA (Order #90721).

van der Kooij, D. 1987. The Effect of Treatment on Assimilable Organic Carbon in Drinking Water. In *Proceedings Of the Second National Conference on Drinking Water*, Pergamon Press. London, England.

van der Wende, E. and W.G. Characklis. 1990. Biofilms in potable water distribution systems. pp. 249-268. In *Drinking Water Microbiology*. Edited by G.A. McFeters. Springer-Verlag: New York, NY.

Vasconcelos, J.J., P. F. Boulos, W. M. Grayman, L. Kiene, O. Wable, P. Biswas, A. Bhari, L. A. Rossman, R. M. Clark, and J. A. Goodrich. 1996. *Characterization and Modeling of Chlorine Decay in Distribution Systems*. AWWA and AwwaRF: Denver CO.

Volk, C.J., R. Hoffmann, C. Chauret, G.A. Gagnon, G. Ranger, and R.C. Andrews. 2002. Implementation of chlorine dioxide disinfection: Effects of the treatment change on drinking water quality in a full-scale distribution system. *Journal of Environmental Engineering and Science*, 1(5): 323-330.

Volk, C.J. and M.W. LeChevallier. 1999. Impacts of the reduction of nutrient levels on bacterial water quality in distribution systems. *Applied and Environmental Microbiology*. 65(11):4957-4966.

Wadowsky, R.M., A.J. West, J.M. Kuchta, S.J. States, J.N. Dowling, and R.B. Yee. 1991. Multiplication of Legionella spp. in tap water containing *Hartmannella vermiformis*. *Appl. Environ. Microbiol.* 57:1950-1955.

Wadowsky, R.M. and R.B. Yee. 1983. Satellite growth of *Legionella pneumophila* with an environmental isolate of Flavobacterium breve. *Appl. Environ. Microbiol.* 46:1447-1449.

Walker, J.T. and M. Morales. 1997. Evaluation of chlorine dioxide (ClO2) for the control of biofilms. *Water Science and Technology* 35(11-12):319-323.

Wannemacher, R.W. Jr., R.E. Dinterman, W.L. Thompson, M.O. Schmidt, and W. J. Burrows. 1993. *Treatment for Removal of Biotoxins from Drinking Water*. US Army Biomedical Research and Development Laboratory: Ft. Detrich, MD

Weber, G.R. and M. Levine. 1944. Factors affecting the germicidal efficiency of chlorine and chloramine. Amer. J. Public Health: 32:719.

Weisel, C.P., H. Kim, P. Haltmeier, and J. Klotz. 1999. Exposure estimates to disinfection by-products of chlorinated drinking water. *Environmental. Health Perspectives*, 107(2):103-110.

White, G.C. 1999. *Handbook of chlorination and alternative disinfectants*. Fourth Edition. John Wiley & Sons, Inc., New York, New York.

Wondergem, E., and A.M. van Dijk-Looijaard. 1991. Chlorine dioxide as a postdisinfectant for Dutch drinking water. *The Science of the Total Environments*, 102:101-12.

Zhang, W. and F.A. DiGiano. 2002. Comparison of bacterial regrowth in distribution systems using free chlorine and chloramine: a statistical study of causative factors. *Water Research* 36 (6): 1469-1482.