

Chlorine Dioxide Disinfection in the Use of Individual Water Purification Devices

Technical Information Paper #31-007-0306

PURPOSE

This information paper provides an in-depth review of chlorine dioxide as a disinfectant in potable water supplies. This paper is intended to assist the reader in evaluating the disinfection capabilities of Individual Water Purification Devices (IWPDs) using chlorine dioxide to kill or inactivate disease-causing bacteria, viruses, and protozoan cysts.

REFERENCES

Appendix A contains a list of references.

INTRODUCTION

Background

Understanding the disinfection capabilities of chlorine dioxide to kill or inactivate diseasecausing microorganisms is important in protecting soldiers, who are considering using this technology, from acute health threats posed by these microorganisms. Soldiers deployed beyond traditional field drinking water supplies must have access to microbiologically safe water. Using IWPDs is one way to provide microbiologically safe water in these situations. These IWPDs must protect the Soldier from acute microbial health threats. The U.S. Environmental Protection Agency (EPA) Guide Standard and Protocol for Testing Microbiological Water Purifiers (reference 1) provides performance standards by which an IWPD using chlorine dioxide can be evaluated. The performance standards are a minimum 6-log reduction/inactivation of bacteria, 4-log reduction/inactivation of viruses, and 3-log reduction/inactivation of protozoan cysts. Chlorine dioxide-using IWPDs meeting these standards are considered effective against disease causing bacteria, viruses, and protozoan cysts. Some IWPD manufacturers test their devices using this protocol. This is the best way to evaluate the IWPDs disinfection capabilities. In the absence of that testing data, this information paper can be used to gain an understanding of chlorine dioxide disinfection capabilities and help determine if an IWPD using chlorine dioxide could successfully meet the EPA Guide's minimum performance standards.

General

Chlorine dioxide (ClO₂) was discovered in 1811 (reference 2). It's widely used in numerous industries including wood pulp processes, wastewater treatment, and food processing. Water treatment plants in the United States first used chlorine dioxide in the 1940s for taste and odor control (reference 3). In addition to taste and odor control, many drinking water systems

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throughout the world today use chlorine dioxide for disinfection, control of organic disinfection byproducts (e.g., trihalomethanes), and oxidation of iron and manganese. Currently, there are only a few Commercial-Off-The-Shelf (COTS) IWPDs using chlorine dioxide for disinfection.

CHLORINE DIOXIDE CHEMISTRY IN WATER

General

Chlorine dioxide exists as an undissociated gas dissolved in water at a near neutral pH range (pH 6-9) (reference 4). Because chlorine dioxide exists as a gas it is vulnerable to volatilization; it can be easily removed from water by turbulent aeration, and is destroyed by ultraviolet light when exposed to sunlight (reference 5). Chlorine dioxide is stable in dilute solution in a closed container in the absence of light (reference 5). One of the advantages of using chlorine dioxide over chlorine for disinfection is the decreased formation of organic disinfection byproducts (DBPs), such as trihalomethanes (reference 3). However, chlorine dioxide is an oxidant and reactions with organic matter form inorganic DBPs including primarily chlorite ion (ClO₂⁻) and to a lesser extent chlorate ion (ClO₃⁻). Chloride (Cl⁻) is also formed to a lesser extent. The reaction of chlorine dioxide in water at pH 6-8 containing organic matter is suggested to be (reference 6):

$$ClO_2 + e^- \rightarrow ClO_2^ ClO_2^- + H^+ \leftrightarrow HClO_2$$
 (chlorous acid)
$$4HClO_2 \rightarrow 2ClO_2 + H^+ + Cl^- + HClO_3 + H_2O$$

Chlorine dioxide reacts rapidly. In drinking water, where typical dosages are 0.07 - 2.0 mg/L, chlorite is the predominant reaction product with approximately 50-70% of chlorine dioxide converted to chlorite, and 30% converted to chlorate and chloride (reference 3). Manufacturer recommended dosages for IWPD use may be similar to those used in water systems or may be much higher. Chlorine dioxide IWPD manufacturers recommend dosages from 0.7 - 4 mg/L for most waters and up to 7.5 mg/L when treating cold and/or cloudy waters (references 7 and 8).

Generation

Chlorine Dioxide Generation for Water Systems

Chlorine dioxide can't be stored commercially or compressed since it is explosive under pressure. Therefore, it must be generated on-site (reference 5). Although there are emerging technologies for chlorine dioxide generation, the two most common methods are (references 2 and 5):

(1) sodium chlorite – acid generation

$$5NaClO_2 + 4HCl \leftrightarrow 4ClO_2 + 5NaCl + 2H_2O$$

(2) sodium chlorite – chlorine generation

$$NaClO_2 + Cl_2 \leftrightarrow 2ClO_2 + 2NaCl$$

Chlorine Dioxide Generation for IWPDs

Chlorine dioxide must also be generated on-site on a much smaller scale or provided in dilute chlorine dioxide solutions for IWPD use. Currently, generating chlorine dioxide on-site for use as an IWPD uses buffered sodium chlorite, generally referred to as "stabilized chlorine dioxide" (references 9 and 10). The sodium chlorite must be "activated" by adding an acid, usually phosphoric or citric acid, resulting in the formation of chlorine dioxide in a reaction similar to the sodium chlorite – acid generation reaction used by water systems (shown earlier). There are health concerns associated with the use of "stabilized chlorine dioxide." "Stabilized chlorine dioxide" can potentially result in little formation of chlorine dioxide, thereby reducing disinfection capability, and can also potentially result in high concentrations of chlorite, which may cause adverse health effects when ingested and also has no disinfection capability (references 3 and 11). Dilute solutions of chlorine dioxide are also used as IWPDs. These solutions lose chlorine dioxide over time, but can be stable for several months and possibly longer. One study showed dilute chlorine dioxide concentrations (approximately 35 mg/L) exhibited variable losses based on the type of container used for storage (reference 12). For example, a 35 mg/L chlorine dioxide solution stored in a high-density Polyethylene Terephthalate (PETE) container for 45 days resulted in a 3% loss of chlorine dioxide (34 mg/L). In contrast, the same study stored chlorine dioxide in a clear glass container for 31 days which resulted in a 12% gain of chlorine dioxide (39 mg/L) possibly due to continuing formation of chlorine dioxide from chlorite. Another study showed a 6.2% overall gain in chlorine dioxide concentration after 252 days of storage in a PETE container (reference 12).

DISINFECTION CAPABILITIES

General

Chlorine dioxide is an effective disinfectant against bacteria, viruses, and many cysts including the capability to disinfect *Cryptosporidium* with realistic (typical to slightly higher water system) dosages (reference 3). A comparison of CTs required for a 2-log inactivation for *E. Coli* bacteria, Poliovirus 1, and *Giardia* cysts showed *Giardia* cysts were 2-5 times more resistant than Poliovirus 1 and 16-22 times more resistant than *E. Coli* bacteria (reference 13). The CT is the product of disinfectant concentration (C in mg/L) and contact time (T in min). The CT

product is a useful way for comparing alternative disinfectants and the resistance of various pathogens (reference 28). Poliovirus was 4-11 times more resistant than *E. Coli* bacteria (reference 13). *Cryptosporidium* oocysts are the most resistant, being 8-16 times more resistant than *Giardia* cysts (reference 5). Chlorine dioxide's general disinfection capability with respect to microorganisms can be illustrated in the following way from most effective to least effective:

bacteria > viruses > Giardia cysts > Cryptosporidium oocysts

Chlorine dioxide is similar to other chemical disinfectants in that its disinfection capability decreases with decreasing temperature, its disinfection capability generally decreases with increasing turbidity, and its disinfection capability is affected by pH (references 3, 4 and 13). Since chlorine dioxide exists as an undissociated gas in water, volatilization and loss of chlorine dioxide and subsequent disinfecting capability is a concern (reference 3). Because chlorine dioxide is an oxidant it will react with organic matter in the water forming primarily chlorite and to a lesser extent chlorate and chloride. Both chlorite and chlorate show no disinfection capabilities and may cause adverse health effects in children, infants, and fetuses (reference 11). Drinking water systems using chlorine dioxide for disinfection are not generally able to provide adequate disinfection per regulations in raw water with high organic carbon (i.e., organic matter) when adding chlorine dioxide in the raw water. This is because the chlorine dioxide is used up by reacting with organic matter, being reduced to primarily chlorite and leaving no chlorine dioxide residual (reference 3). This can be a concern for IWPDs when treating raw, unfiltered water supplies. Higher dosages may be necessary to react with organic matter and provide disinfection.

Environmental Effects on Disinfection Capability

Effect of pH on Disinfection Capability

Compared to chlorine, chlorine dioxide is a more effective disinfectant across a broader pH range (roughly between 5 and 10) than free chlorine (reference 3). Several studies have shown the effect of pH on chlorine dioxide disinfection capability, with most results indicating disinfection capability generally increases with increasing pH (reference 14). Numerous studies with viruses (e.g., poliovirus, hepatitis A virus) showed CTs required for a 2-log virus inactivation were 13 – 20 times higher at a pH of approximately 6 compared to a pH of 9 and 10 (references 13 and 15). Another study showed CTs up to 90-100 times higher were required for a 4-log virus inactivation at a pH of 6 compared to a pH of 10 (reference 16). Although these studies showed much higher CTs necessary at lower pHs, CTs were still low at the lower pHs (ranging from approximately 3 – 13 mg-min/L). This indicates chlorine dioxide is a highly effective disinfectant over a broad pH range. In contrast to the previous studies, a study on chlorine dioxide disinfection capability against *Cryptosporidium* oocysts indicated pH does not appear to have a significant effect on *Cryptosporidium* inactivation (reference 17). The degree

of pH effect may be dependent on the targeted organism and in general chlorine dioxide shows an increase in disinfection capability with increasing pH. Chlorine dioxide would likely be effective over the pH range (pH 6-9) for natural, untreated water sources likely to be encountered when using IWPDs.

Effect of Temperature on Disinfection Capability

Like most chemical disinfectants, chlorine dioxide disinfection capability decreases with decreasing temperatures (reference 5). Cold water temperatures slow disinfection and must be compensated for by longer contact times or higher dosages to achieve comparable disinfection at warmer water temperatures (reference 18). A two to three-fold increase in inactivation rates per 10° C water temperature increase seems a generally accepted rule (reference 18). When considering chlorine dioxide, the U.S. Environmental Protection Agency (EPA) developed CT tables for the Surface Water Treatment Rule (SWTR) by assuming a twofold decrease in CT for every 10° increase (reference 19). Research shows a 2-log inactivation of *E. Coli* required four times higher CT at 5° C compared to 20° C (reference 13). A study using *Naegleria* cysts showed at 5° C a CT twice as high than at 20° C was required to provide a 2-log inactivation (reference 5). Using a two-fold CT increase for every 10° decrease in water temperature is a good estimate to use when determining CT requirements for chlorine dioxide disinfection capability.

Effect of Turbidity on Disinfection Capability

Turbidity also has an effect on chlorine dioxide disinfection capability. Turbidity in the form of particulate matter, aggregated or clumped microorganisms, and dissolved organic matter can reduce the effectiveness of chlorine dioxide. One study determined that bentonite clay added to produce turbidity levels up to 2.3 nephelometric turbidity units (NTUs) had no adverse effect on chlorine dioxide disinfection of poliovirus. However, at turbidity levels of 3.2 and 14.1 NTU, poliovirus inactivation was noticeably decreased (references 13 and 20). The study suggested that bentonite appeared to offer protection or shield the viruses from chlorine dioxide disinfection. Another study using bentonite reduced chlorine dioxide disinfection capability against Naegleria cysts by 11% at turbidities less than or equal to 5 NTU and 25% at turbidities between 5 and 17 NTUs (reference 5). Clumped or aggregated microorganisms are also shown to be more resistant to chlorine dioxide disinfection (reference 5). In the presence of organic matter chlorine dioxide rapidly oxidizes the organic matter and is converted to primarily chlorite, and to a lesser extent chlorate and chloride ion (reference 3). This results in loss of chlorine dioxide residual and an increase in chlorite ion leading to reduced disinfection capability. Turbidity does have an effect on chlorine dioxide disinfection capability. Chlorine dioxide disinfection capability decreases in more turbid waters since microorganisms are protected by solid particles in water, protected by aggregation or clumping, and protected by loss of chlorine

dioxide residual from oxidation of organic matter. Higher chlorine dioxide dosages may be necessary when using IWPDs to overcome organic matter oxidation and still provide disinfection when treating raw, unfiltered water supplies.

Bactericidal Capability

Chlorine dioxide is an effective bactericide. Research on chlorine dioxide bactericidal capability shows bacteria are less resistant than viruses and cysts (reference 13). Studies using *E. Coli* showed 2-log inactivation occurred very quickly in demand-free waters (i.e., no organic matter present) with CT's all less than 1.0 mg-min/L, ranging from 0.25 – 0.48 mg-min/L, at the coldest water temperatures (5° C) and lowest pH levels (6.5 - 7.0) (i.e., worst case conditions, references 13, 21). Another study estimated CTs of 1 or less at 5° C necessary for a 4-log *E. Coli* inactivation (reference 22). Chlorine dioxide should easily achieve a 6-log bacteria inactivation at low temperatures and low pHs if chlorine dioxide is used for disinfection of more resistant viruses and cysts. Highly turbid water may require higher CT (i.e., longer contact time and/or higher dose).

Virucidal Capability

Chlorine dioxide is an effective virucide. Research shows viruses are more resistant than bacteria but less resistant than cysts (reference 13). Similar to bactericidal capability, viruses are rapidly inactivated (reference 13). Experiments conducted under worst case conditions (5° C water temperature in the 6 – 7 pH range) resulted in CT's of 5.5 mg-min/L for a 2-log Poliovirus 1 inactivation and 12.6 mg-min/L for a 4-log Hepatitis A virus inactivation (references 13 and 16). The SWTR provides the following CT values for 4-log virus inactivation at various water temperatures with pH 6-9 (reference 19):

Table 1. EPA Surface Water Treatment Rule (SWTR) Required CT Values for 4-Log Inactivation of Viruses by Chlorine Dioxide for pH 6-9

Temperature (deg C)						
<= 1	5	10	15	20	25	
50.1	33.4	25.1	16.7	12.5	8.4	_

The data used to develop Table 1 were based on experiments conducted in low turbidity waters under otherwise worst case conditions, 5° C water temperature and pH 6. These CT values are based on low turbidity waters since it is assumed water systems provide disinfection after filtration, as the last treatment step prior to distribution. Higher turbidity waters may require

higher CT to achieve the same log inactivation. Separate CT values for different pHs were not developed since chlorine dioxide is generally a more effective disinfectant at higher pHs. Therefore, these CT values are more conservative at the higher pHs (reference 19). A safety factor of 2 was applied to the data to determine CT values in Table 1 (reference 19). The CT values at temperatures other than 5° C in the Table were determined by using a two-fold increase in CT for every 10° C decrease (reference 19). Even at cold water temperatures, low pHs, and low turbidity waters, CTs appear realistic and achievable. Based on a typical chlorine dioxide dosage of 2.0 mg/L for a water system, contact times of 4-25 minutes are necessary to achieve CT values in Table 1. A chlorine dioxide dose of 0.8 mg/L [EPA's Maximum Residual Disinfectant Level (MRDL) for chlorine dioxide] results in contact times of 11-63 minutes which are still reasonable for IWPD use. Highly turbid water may require higher CT (i.e., longer contact time and/or higher dose).

Cysticidal Capability

Giardia Cysts

Chlorine dioxide is effective against *Giardia* cysts. One study showed CTs ranging from 1.7-17.6 mg-min/L necessary for 2-log *Giardia muris* cyst inactivation (reference 23). The SWTR provides the following CT values for 3-log inactivation of *Giardia* cysts at various water temperatures with pH 6-9 (reference 19):

Table 2. EPA SWTR Required CT Values for 3-Log Inactivation of *Giardia* Cysts by Chlorine Dioxide for pH 6-9

Temperature (deg C)						
<= 1	5	10	15	20	25	
63	26	23	19	15	11	

Data used to develop Table 2 were based on experiments conducted in low turbidity waters at pH 7 and water temperatures ranging from 1 - 25° C for 2-log *Giardia* cyst inactivation (reference 19). Determining 3-log inactivation at all temperatures listed in Table 2 required extrapolation using first order kinetics and applying a safety factor of 1.5 (reference 19). Based on Table 2 it appears chlorine dioxide is effective against *Giardia* cysts at realistic and achievable CT values. Based on a typical chlorine dioxide dosage of 2.0 mg/L for a water system, contact times of 6 - 32 minutes, depending on temperature, are necessary to achieve the CT values in Table 2. These contact times are also reasonable for IWPDs. A chlorine dioxide

dose of 0.8 mg/L (EPA's MRDL for chlorine dioxide) results in contact times of 14 - 79 minutes which are still reasonable for IWPD use. Highly turbid water may require higher CT (i.e., longer contact time and/or higher dose).

Cryptosporidium Oocysts

Chlorine dioxide appears effective against *Cryptosporidium* oocysts at CT values achievable by water systems. Studies show 3-log *Cryptosporidium* inactivation varied from a CT of 70 mg-min/L to 400 mg-min/L under various water quality conditions (reference 5). *Cryptosporidium* is more resistant than *Giardia* cysts; up to 8-16 times more resistant (reference 5). Similar to bacteria, viruses, and other cysts, chlorine dioxide, in general, is more effective against *Cryptosporidium* oocysts at higher pHs and higher temperatures (reference 5). However, there is data suggesting pH has a negligible effect on inactivation of *Cryptosporidium* (reference 17). Pursuant to the Long Term 2 Enhanced Surface Water Treatment Rule (LT2ESWTR), the EPA proposed chlorine dioxide CT tables for various log inactivations of *Cryptosporidium* (reference 24) based on studies conducted using low turbidity waters. The proposed CT values for 3-log *Cryptosporidium* inactivation are shown in Table 3. These doses are conservative and were developed using a safety margin to account for variability and uncertainty in the experimental data (reference 24).

Table 3. EPA Proposed CT Values for 3-Log Inactivation of *Cryptosporidium* Oocysts by Chlorine Dioxide for pH 6-9

_	1	5	10	15	20	25	_
	1830	1286	830	536	347	226	

Based on a typical chlorine dioxide dosage of 2.0 mg/L for a water system, contact times of 115 - 915 minutes (2 - 15 hours), depending on temperature, are necessary to achieve the CT values in Table 3. For water systems, these CT values are realistic and achievable at warmer water temperatures. Higher than typical chlorine dioxide dosages would be necessary for a water system to achieve the proposed CTs in colder waters (i.e., less than 10° C). Based on this Table, use of an IWPD would be practical in only warmer waters (i.e., above 10° C). Highly turbid water may require even higher CT values (i.e., longer contact time and/or higher dose). Chlorine dioxide is effective against Cryptosporidium oocysts in warmer, low turbidity waters.

CHLORINE DIOXIDE TOXICITY

Health Effects of Chlorine Dioxide and Chlorite

Chlorine dioxide and its byproducts, chlorite and chlorate ion can result in adverse health effects when consumed at large enough quantities. The EPA regulates chlorine dioxide and chlorite ion in drinking water for systems using chlorine dioxide for disinfection. The EPA established a MRDL of 0.8 mg/L for chlorine dioxide and a maximum contaminant level (MCL) of 1.0 mg/L for chlorite (reference 25). The most common adverse health effects of chlorine dioxide and chlorite ion are oxidizing effects seen in the blood, either as methemoglobinemia or hemolytic anemia (reference 3). Children, infants, and fetuses, a more susceptible subpopulation may experience adverse neurotoxic effects (reference 26). When a regulated water system using chlorine dioxide is out of compliance with the chlorine dioxide MRDL or chlorite MCL, the EPA considers this to have a significant potential to have serious adverse health effects as a result of short-term exposure (reference 27). However, the short-term adverse health effects are limited to children, infants, and fetuses. It is these groups that may be susceptible to adverse nervous system effects from short-term exposure (reference 27). Health effect data for healthy adults appear to indicate that short-term exposure does not result in adverse health effects. Several clinical studies assessing the acute and subchronic effects of chlorine dioxide, chlorite, and chlorate have been conducted (reference 3). Healthy adults consuming 2.5 mg daily of either chlorine dioxide, chlorite, or chlorate for 12 weeks showed no clinically significant adverse health effects (reference 3). Another study had healthy adults consuming 0.1 to 24 mg/L concentrations of either chlorine dioxide, chlorite, or chlorate daily for 3 weeks, again resulting in no clinically significant adverse health effects. Based on this information, it is not likely that healthy adults consuming water containing chlorine dioxide concentrations recommended by IWPD manufacturers (0.7 - 7.5 mg/L) for a short duration (e.g., < 3 weeks) would experience any adverse health effects from ingestion of chlorine dioxide, chlorite, or chlorate. However, adverse health effects could occur if higher chlorine dioxide dosages are used for treating highly turbid and/or colder water to kill Cryptosporidium. To avoid potential adverse health effects, longer contact times should be used in place of higher chlorine dioxide dosages, provided sufficient chlorine dioxide remains after oxidizing organic matter.

Health Concerns of Stabilized Chlorine Dioxide

The use of "stabilized chlorine dioxide" products for IWPD use may expose the user to significant chlorite concentrations. The "activation" of stabilized chlorine dioxide (i.e., sodium chlorite) with an acid can result in high levels of chlorite remaining after activation and relatively low chlorine dioxide concentrations compared to typical chlorine dioxide generating systems (reference 3). Use of these products may result in the direct application of several hundred mg/L of chlorite to the water, much higher than typical drinking water chlorite levels (reference 3).

CONCLUSIONS

Chlorine dioxide as an IWPD can be effective against bacteria, viruses, *Giardia* cysts, and to a limited extent, *Cryptosporidium* oocysts. Very high CT values are estimated for a 3-log *Cryptosporidium* inactivation in colder waters, requiring very high chlorine dioxide dosages and/or very long contact times. Colder temperatures, lower pHs, and higher turbidity all tend to have an adverse effect on disinfection capability. Health concerns of ingesting chlorine dioxide and chlorite ion are likely minimal for healthy adults over a short-term duration (e.g., < 3 weeks) for IWPD manufacturer-recommended chlorine dioxide dosages of 0.7 – 7.5 mg/L. However, adverse health effects could occur if higher chlorine dioxide dosages are used for treating highly turbid and/or colder water to kill *Cryptosporidium*. To avoid potential adverse health effects, longer contact times should be used in place of higher chlorine dioxide dosages, provided sufficient chlorine dioxide remains after oxidizing organic matter. IWPDs using "stabilized chlorine dioxide" may result in exposure to high levels of chlorite. Table 4 provides a summary of chlorine dioxide's disinfection capabilities.

Table 4. Chlorine Dioxide Disinfection Capabilities

Parameter	Chlorine Dioxide Disinfection
General Disinfection Capability	Cysts most resistant. Achieving cyst inactivation will ensure adequate bacteria and virus inactivation. Disinfection capability generally follows: Bacteria > viruses > Giardia > Cryptosporidium
Bacteria	Effective at reasonable CT values for IWPD use
Viruses	Effective at reasonable CT values for IWPD use. Use EPA SWTR CT table for recommended CT values (Table 1).
Giardia Cysts	Effective at reasonable CT values for IWPD use. Use EPA SWTR CT table for recommended CT values (Table 2).
Cryptosporidium Oocysts	Effective at high CT values. Use Table 3 as guide for CT values. If possible, use longer contact times instead of higher dosages to achieve adequate CT values.
Effect of Temperature	Colder water temperatures require higher CT values. Use a two-fold increase in CT for every 10° C decrease. Use longer contact time instead of higher dosages to achieve higher CT values.

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Effect of pH	Effective over typical pH levels for raw, untreated natural waters. Disinfection capability generally increases with increasing pH.
Effect of Turbidity	Higher turbidity generally reduces disinfection capability. Use longer contact time instead of higher dosages in more turbid waters to achieve CT values. Higher dosages may be necessary to ensure chlorine dioxide remains after oxidation of organic matter.
Health Effects	Chlorine dioxide and chlorite are potential health concerns. IWPD manufacturer-recommended dosages are not likely to cause adverse health effects for healthy adults. Exposure to much higher chlorite concentrations may occur when using stabilized chlorine dioxide products.

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