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TECHNICAL REPORT 8911

OPTIMUM DISINFECTION PROPERTIES AND
COMMERCIALY AVAILABLE DISINFECTANTS

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JULY 1989

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19. ABSTRACT (Continue on reverse if necessary and identify by block number) Draft criteria were developed for a hypothetical ideal field drinking water disinfectant and submitted for ranking to 18 agencies of the Department of Defense and the U.S. Environmental Protection Agency (USEPA). The consensus placed health considerations first, with efficacy, palatability, and stability next and of approximately equal rank. Candidate replacements for calcium hypochlorite (HTH) were taken from a list of approved drinking water disinfectants provided by the USEPA Office of Pesticide Programs and from five developmental cyclic N-halamines. Based on a preliminary assessment, it is believed that chlorine dioxide and sodium dichloro-s-triazinetrione are strong candidates for field disinfectants which require further evaluation for efficacy, particularly with respect to destruction of water-borne viruses and protozoan cysts. It is also recommended that 3-chloro-4,4-dimethyl-oxazolidinone (Compound I) and related halamines be further investigated for safety and efficacy.					
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INTRODUCTION

Preventive medicine doctrine of the U.S. Army advocates the routine disinfection of military water supplies to prevent transmission of waterborne diseases. The chemicals presently used to accomplish disinfection in the field are calcium hypochlorite (High Test Hypochlorite or HTH, NSN 6810-00-255-0471) and iodine tablets (Globaline, NSN 6850-00-985-7166). These compounds, in use for over 35 years by U.S. military forces, have been found to be effective disinfectants under a wide range of conditions. However, numerous deficiencies of these agents have been identified, namely:

- a. Both exhibit a critical dependence on pH and temperature for optimum disinfection.
- b. Both impart an objectionable taste and odor when used at field concentrations, especially when combined with reactive organic materials (organic demand).
- c. Both provide only low residual activity in water due to volatility and combination with organic demand thereby reducing their ability to protect against post-treatment contamination.
- d. Hypochlorites may form carcinogenic disinfectant by-products in water, principally trihalomethanes.
- e. HTH is corrosive to and reactive with field water equipment and storage containers.
- f. Neither is effective against all waterborne pathogenic protozoans.

Because these deficiencies are perceived to present a significant threat to the health of soldiers in the field, it has been recommended that research be carried out by the U.S. Army Biomedical Research and Development Laboratory (USABRDL) to identify alternative disinfectants, with the principle focus on evaluation of off-the-shelf commercial products (1, 2). The objectives of the research reported herein have been to develop criteria for the hypothetical ideal disinfectant and to identify, through literature and product surveys, promising candidates for testing.

CRITERIA DEVELOPMENT

The following draft criteria for an ideal water disinfection agent were developed by USABRDL:

- a. The basic disinfectant and any disinfectant by-products should cause no adverse health effects to humans at field use conditions (i.e., be non toxic, noncarcinogenic, nonmutagenic, nonteratogenic) yet accomplish quick and complete kill or inactivation of all human pathogens in heavily contaminated water.

b. The disinfectant and its by-products should not impart an objectionable taste, odor, or color to the water when used at field concentrations.

c. The disinfecting agent should be stable under conditions of storage and actual use. When packaged in a suitable container, the agent should meet all military specifications for stability under a wide variety of temperatures and humidity for a specified period. In use, the agent should maintain an adequate disinfecting residual in water, such that the need for repeat disinfection is minimized or eliminated (i.e., have relatively low volatility at high water temperatures and be nonreactive with organic constituents that display a chlorine demand.)

d. Upon addition to water, the agent should dissolve quickly and release its active ingredient(s) rapidly (within 5 minutes) in order to allow as much time as possible for pathogen kill or removal.

e. Ideally, the agent should disinfect waters over a wide range of temperature and pH, preferably instantaneously, but not over 15 minutes.

f. In storage and in use, the agent should be nonreactive and noncorrosive to storage/packaging containers and field water equipment (e.g., Reverse Osmosis Water Purification Unit (ROWPU) membranes, Tactical Water Distribution System (TWDS) pipelines, and storage and distribution bladders).

g. The disinfectant should be concentrated such that a small dose (i.e., one or two tablets, packets or ampoules) will ensure adequate disinfection of a small quantity of water (i.e., a canteen) without the need for testing for residual concentration of the disinfectant.

h. The method of application should be simple, substantially foolproof, and not unduly time-consuming.

i. The ingredients required in compounding the disinfectant should be economically and strategically available.

j. Manufacture of the chemical agent should lend itself to economical, large-scale preparation with normally available chemical and pharmaceutical equipment.

k. The disinfectant residual must be readily measurable by analytical procedures that can be used in the field.

These criteria were sent to 18 agencies of the Department of Defense and to the U.S. Environmental Protection Agency (USEPA) Office of Drinking Water for comment and ranking (3); 11 replies, many fragmentary, were received (4-14). Responders recognized that the USABRDL list of criteria contained numerous redundancies, i.e., individual criteria contained several qualifying elements, and the same qualifying elements (such as rapidity of disinfection) were contained in more than one criterion. The ranking matrix is presented in Table 1.

Table 1. Relative Ranking of Draft Criteria

Criterion	4 ^a	5	6 ^b	7	8 ^c	Reference		11	12	13	14 ^b	Relative rank
1			1	1		1	1 ^d	1	1	1	1	1
2		2	2			3	3		2	6	2	2-3
3	1	3	3	2			4		3	2	3	2-3
4			4				9			5	4	8
5	1	1	5				5	3	4	8	5	4
6							10			del ^e	6	11
7	1		7				6		5	7	7	6-7
8			8			2	7		6	3	8	6-7
9			9				8			4	9	9-10
10			10				11			4	10	9-10
11		4	11				2	2		del ^e	11	5

a. Responder did not evaluate criteria directly.

b. Responder accepted USABRDL criteria rank without comment.

c. Responder did not rank criteria

d. Responder proposed modified criterion 1.

e. Responder recommended deletion of criterion.

Among responders there was nearly unanimous agreement that the most important criterion is that the candidate disinfectant do no harm (criterion 1). [One responder considered the risk from trihalomethanes to be acceptable (13).] Next, nearly equal importance was given to the absence of objectionable tastes, odors, or colors (criterion 2) and to stability of the disinfectant under conditions of storage and use (criterion 3). [One responder pointed out that taste and odor are nearly unavoidable, but that objectionable taste can be minimized with proper technique in the case of hypochlorite (probably not suitable for field application) or masked with a flavoring agent in the case of iodine (13).] Criteria 4, 5, and 1 all address elements of efficacy, which may have caused confusion among responders. However, a rapid and complete pathogen kill is ranked among the top criteria, and one responder recommended a minimum log kill of 99.99 percent of all pathogens (4). Some responders placed importance on the ability to measure residual (criterion 11); others considered this to be insignificant (13). The availability of the disinfectant in concentrated form (criterion 7) and simplicity of application (criterion 8) were considered next and given about equal importance. Criteria 9 and 10, which collectively address the question of availability, were ranked low, and corrosivity/reactivity (criterion 6) was not considered important at all. [One responder suggested that the disinfectant should not react with beverage food additives (4).] Finally, one responder suggested an additional criterion: "In storage the disinfectant should exhibit a change in observable characteristics when potency is reduced below an effective level" (8). (This is a characteristic of current iodine tablets.) In consideration of all responses, USABRDL suggests a revised list of criteria and subcriteria, as presented in Table 2. It should be noted that the criteria are not necessarily inflexible; e.g., an otherwise outstanding

disinfectant that has narrow pH limitations can be packaged with a suitable buffer.

Table 2. Criteria for Candidate Disinfectants

Rank	Criterion	Definition
1	Health Effects	Disinfectant and by-products cause no adverse acute or chronic health effects
2.	Efficacy	Disinfectant provides rapid (<15 minutes), complete (>99.99 percent) kill: a. of all waterborne pathogens b. at low temperatures (<40°F) c. at a wide range of pH (5-9)
2.	Palatability	Disinfectant or by-products impart no strongly objectionable taste, odor, or color
2.	Stability	Disinfectant is stable in storage and provides a persistent residual in use.
3.	Analysis	Residual is readily measurable in the field
4.	Ease of Use	Disinfectant is available in concentrated form; method of application is rapid and foolproof
5.	Availability	Manufacture and distribution of disinfectant in bulk is technically feasible and economically reasonable

INITIAL SCREENING CRITERIA

The draft disinfectant criteria submitted for comment were accompanied by suggested draft screening criteria for initial laboratory evaluation of candidates. These were:

- a. A small dose of the agent shall accomplish quick and complete kill or inactivation of representative bacterial, viral, and protozoan pathogens in heavily contaminated water in less than 30 minutes.
- b. The agent should disinfect waters over a wide range of temperature and pH.
- c. The agent shall be stable in water and in storage.

Responders suggested that initial screening criteria also include taste and odor (4, 5, 6, 12), ability to measure residual in the field (5), and ease of use (12).

SURVEY OF CANDIDATE DISINFECTANTS

Approved drinking water disinfectants are registered with the USEPA Office of Pesticides and Toxic Substances (OPTS). Efficacy and toxicological safety must be documented. Because minor ingredients, inert ingredients, concentration of the active agent, and specific use proposed by the manufacturer may be unique to each formulation, each application for a new product must be evaluated independently, even if the active agent is the same and the formulation is substantially identical with existing products. However, OPTS has provided USABRDL a generic list of chemical disinfectants used in registered products (15). These chemicals, presented in Table 3, are categorized according to their approved use: Category A includes chemicals registered for use in municipal drinking water or home well disinfection; Category B includes chemicals registered for emergency treatment of water supplies in the field. The OPTS states that Category B chemicals are primarily limited to military training maneuvers and to backpackers in areas where contamination of natural water sources may be a problem.

Table 3. Listing of Drinking Water Disinfectants

Category	Chemical(s)	Application
Municipal Water and Home Well Disinfection		
A	Calcium hypochlorite	water additive
A	Sodium hypochlorite	" "
A	Chlorine	" "
A	Sodium chlorite	" "
A	Chlorine dioxide	" "
Emergency Water Treatment		
B	Polybromide form of trimethylbenzyl ammonium resin	water filter
B	Halogenated polystyrene divinylbenzene quaternary ammonium anion resin containing iodine	" "
B	Iodine	water additive
B	Tetraglycine hydroperiodide	" "
B	Sodium dichloro-s-triazinetriene	" "
B	Silver	water filter or additive

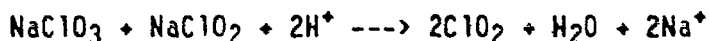
In addition to these agents, Halazone, 4-(N,N-dichlorosulfamoyl)benzoic acid, has been used since World War I, and numerous iodine precursors have been developed (16). Recently, five new cyclic N-haloamines have been synthesized and have undergone preliminary efficacy (17) and toxicity testing (18-22).

PRELIMINARY ASSESSMENT

Category A

Of the Category A disinfectants, sodium hypochlorite and chlorine have no advantages over HTH for field use, and have the disadvantage that they are not practical to use. Sodium hypochlorite is highly unstable unless in aqueous solution, and is thus not available in concentrated form. Chlorine is a gas which requires special equipment for storage and application. Sodium chlorite, not a very effective disinfectant by itself, is used as precursor for chlorine dioxide when combined with chlorine. Chlorine dioxide is the only Category A disinfectant recommended as alternative to HTH or iodine in this study.

Chlorine dioxide, a disinfectant of about the same potency as chlorine, is superior to HTH in that it oxidizes common water contaminants such as phenol without producing intermediates with offensive tastes and odors, and destroys humic substances without producing significant levels of trihalomethanes. It must be generated in situ. One method is to combine sodium chlorite and sodium chlorate:



As indicated, chlorine dioxide is stable in acid solution; in basic solution it reverts to chlorite and chlorate ions, the combination of which is a much weaker disinfectant than chlorine dioxide. In most cases it will be necessary to add a buffer to the water along with the precursors. Aqua-Chem has developed individual water disinfectant kits which consist of packages with two bubble capsules, one containing buffer solution, the other containing precursor solution. Breaking a seal between the two capsules results in generation of chlorine dioxide, which can be added to a canteen. To most palates, the finished water quality is far superior to water containing iodine. One reference notes that few subjects found ClO_2 , ClO_2^- , or ClO_3^- to have an objectionable taste at levels up to 24 mg/L (23). Principal drawbacks to ClO_2 are its volatility and the absence of a field method for residual measurement. However, it may be possible to develop a simple colorimetric field test based on bleaching of chlorophenol red or a similar dye (24).

The toxicities of chlorine dioxide and its byproducts have received considerable attention (25). In one series of studies, "administration of ClO_2 , ClO_2^- and ClO_3^- in drinking water for up to 84 days to human volunteers at increasing doses (up to 24 mg/L, 600 mL/day) did not result in detectable alterations on blood parameters, serum or urine chemistry values or adverse physical symptoms. For individuals identified as being deficient in glucose-6-phosphate dehydrogenase (which makes such individuals more sensitive to the oxidative stress of disinfectants) significant changes in several blood and serum chemistry parameters were observed after an 84-day exposure to ClO_2^- at 5 ppm" (25). However, in no case was any detrimental physiological effect observed (23). Because of concern about hemolytic effects in experimental animals (at ClO_2^- levels >50 mg/L) and thyroid effects in nonhuman primates (at ClO_2 levels >100 mg/L), it has been recommended that levels of ClO_2 and ClO_2^- not exceed 1 mg/L in finished drinking water (24). This applies to the

population at large, some of whom may be unusually sensitive to hemolytic agents. Maximum contaminant levels for ClO_2 , ClO_2^- , and ClO_3^- in drinking water are pending (26). Carcinogenesis assays with chlorine dioxide and its byproducts have been negative (25).

Category B

Sodium dichloro-s-triazinetriene, also known as sodium dichloroisocyanurate, is the only promising alternative disinfectant from Category B. The bromine- and iodine-containing resins may be effective disinfectants, but they are not, in themselves, alternatives to HTH or Globaline because they involve filtering action and require special equipment. Silver has been used as bactericide in home and portable water filters. It can also be used as a water additive; however, the required dose for efficient disinfection far exceeds the tentative USEPA maximum contaminant level of 0.05 mg/L (since withdrawn). Tetraglycine hydroperiodide is the active constituent of Globaline, one of the substances for which an alternative is sought. Elemental iodine has all the disadvantages of Globaline and lacks the ease of use of Globaline.

At this time sodium dichloro-s-triazinetriene is used as the active constituent in individual water supply disinfection tablets, viz. Chlor-Floc and Aquatabs. It also has many nonpotable applications. We are not aware of its use for disinfection of bulk drinking water supplies; however, it has been used for disinfecting swimming pools and cooling water. In water, half of the chlorine is released as free available chlorine; the remainder is combined available chlorine (27). Thus, its disinfection efficacy is similar to HTH. Advantages over HTH and/or Globaline are the relative absence of taste and odor, stability in use, and nonformation of trihalomethanes. It is a moderately toxic material, with LD50 of 1400 mg/kg (rat).

Developmental and Others

A number of new N-haloamines have been examined as potential alternatives to HTH (17). These include 3-chloro-4,4-dimethyloxazolidinone (Compound I) and four closely related nitrogen heterocycles. Compound I, which has received most attention, is about as effective as HTH as bactericide (*Staphylococcus aureus* and *Shigella boydii*), much less effective as virucide (poliovirus type 1, rotavirus), but more effective as cysticide (*Giardia lamblia*, *Entamoeba invadens*). It has long-term stability in water at temperatures as high as 37° and is largely taste and odor-free at concentrations equivalent to 10 mg/L available chlorine. Based on an oral LD50 of about 300 mg/kg for rats and mice, it is classified as "very toxic" (20,21). (This does not necessarily disqualify Compound I; more toxicity testing is needed.) Compound I is commercially available and could readily be manufactured in bulk.

An individual water disinfectant used by the Australian Army uses a mixture of potassium iodide and potassium iodate to generate elemental iodine. Its advantages are ease of production and storage stability; its disadvantages are those of all iodine-based disinfectants.

SUMMARY AND RECOMMENDATIONS

Summary

There are two strong off-the-shelf candidates and at least one developmental candidate to be considered as alternatives to HTH and Globaline. Qualities of the off-the-shelf disinfectants relative to criteria are summarized in Table 4. The advantages of chlorine dioxide are that it has an extensive data base on health effects and efficacy and that it produces no objectionable tastes or odors. Its disadvantages are that it is volatile and, like HTH, can rapidly lose its residual. There is no field test for residual, and storage stability and ease of application are uncertain. Sodium dichloro-s-triazinetriene appears to have no strong disadvantages; however, its data base is relatively limited. The most advanced of the developmental candidates is Compound I (Table 5). Its principal disadvantage may be its toxicity, which is as yet inadequately explored.

Table 4. Criteria Compliance for Disinfectant Candidates

Criterion	Chlorine dioxide	Sodium dichloro-s-triazinetriene
Health effects: toxicity of chemical and byproducts	less than HTH	no more than HTH
Efficacy		
Against all pathogens	ca. same as HTH	similar to HTH
pH dependence	acid pH required	same as HTH
Temperature dependence	unknown	use $\geq 22^{\circ}$
Palatability		
Taste and odor	absent	slight
Color	absent	absent
Stability		
In storage	unknown	96.8 percent at 40° , 3 mo.
In use	volatile	stable
Analysis of residual	no field method	standard method
Ease of use		
In bulk	unknown	more conc. than HTH
Individual	Globaline is easier	same as Globaline
Availability	unrestricted	unrestricted

Table 5. Criteria Compliance for Compound I

Criterion	Compliance
Health effects: toxicity of chemical and byproducts	5x more toxic than dichloro-s-triazine-trione; no haloforms produced
Efficacy	
Against all pathogens	variable; similar to dichloro-s-triazine-trione
pH dependence	alkaline pH favored
Temperature dependence	high at low residual
Palatability	
Taste and odor	claimed to be largely absent
Color	absent
Stability	
In storage	unknown; probably same as dichloro-s-triazinetriene
In use	37°C: half-life more than 1200 hours at pH 7, ca. 200 hours at pH 9.5
Analysis of residual	standard method
Ease of use	
In bulk	same as dichloro-s-triazinetriene
Individual	same as Globaline
Availability	commercial process available

Recommendations: Research Needs

1. For both chlorine dioxide and sodium dichloro-s-triazinetriene bactericidal properties are well known; however, there is a requirement to define the efficacy of inactivation of water-borne viruses and destruction of protozoan cysts (*Giardia* and *Cryptosporidium*) at various temperature and pH levels.
2. A field procedure for measuring disinfectant residual is needed for chlorine dioxide.
3. Stability in storage and application methods need clearer definition for chlorine dioxide.
4. Compound I and the related cyclic haloamines deserve continued study. More toxicology is needed.

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GLOSSARY OF TERMS

EPA	U.S. Environmental Protection Agency
HTH	high test hypochlorite
LD50	lethal dose, 50 percent
OPTS	Office of Pesticides and Toxic Substances (EPA)
ppm	parts per million (mg/L)
ROWPU	reverse osmosis water purification system
TWDS	tactical water distribution system
USABRDL	U.S. Army Biomedical Research and Development Laboratory

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