Project Summary

Stability and Effectiveness of Chlorine Disinfectants in Water Distribution Systems

Vincent P. Olivieri, Michael C. Sneed, Cornelius W. Kruse', and Kazuyoshi Kawata

A test system for water distribution was used to evaluate the stability and effectiveness of three residual disinfectants—free chlorine, combined chlorine, and chlorine dioxide—when challenged with a sewage contaminant. The test distribution system consisted of the street main and internal plumbing for two barracks at Fort George G. Meade in Fort Meade, Maryland. To the existing pipe network, 152 m (500 ft) of 13-mm (0.5-in.) copper pipe was added for sampling and 60 m (200 ft) of 2.54-cm (1.0-in.) plastic pipe was added for circulation. The levels of residual disinfectants tested were 0.2 mg/L and 1.0 mg/L as available chlorine.

In the absence of a disinfectant residual, microorganisms from the sewage contaminant were consistently recovered at high levels. The presence of any disinfectant residual reduced the microorganism level and frequency of occurrence at the consumer’s tap. Free chlorine was the most effective residual disinfectant and may serve as a marker or flag in the distribution network. Free chlorine and chlorine dioxide were the least stable in the pipe network. The loss of disinfectant in the pipe network followed first-order kinetics. The half-life determined in static tests for free chlorine, chlorine dioxide, and combined chlorine was 140, 93, and 1,680 min, respectively.

This Project Summary was developed by EPA’s Municipal Environmental Research Laboratory, Cincinnati, OH, to announce key findings of the research project that is fully documented in a separate report of the same title (see Project Report ordering information at back).

Introduction

The objectives of this study were to evaluate the stability and effectiveness of residual disinfectants in a test water distribution system when challenged by a sewage contaminant. The level of pathogenic microorganisms that reach the consumer’s tap during cross connection and back-siphonage episodes is a function of dilution of the contaminating material, natural die away, and inactivation by the residual disinfectant. The dilution of the contaminating material depends heavily on the configuration and characteristics of the pipe network and the flow of water in the local area where the integrity of the conduits was breached. Because of the infinite complexity and variety of plumbing and flows found in water distribution systems, the degree of dilution cannot be predicted, even in well-defined systems.

One aspect related to flow as a mechanism for removing contamination from a water distribution system is washout. With continued use and consumption of water, the contaminant slug will be purged from the system in a relatively short time. But dilution and washout cannot be depended on to provide a safe water or a water of good quality at the tap. The enteric microorganisms responsible for transmitting diseases by water tend to die away when introduced into the aquatic environment, and proliferation of these microorganisms has generally not been observed. But the rates of die away are relatively slow, and sufficient levels of
microorganisms can be expected to survive for the time periods found in most water distribution systems.

Experimental Protocol

Test Distribution System

The test distribution system consisted of several hundred feet of 10-cm (4-in.) pipe and the internal plumbing of two army barracks (Figure 1). Each building contained four apartments with the appropriate plumbing for bath and kitchen facilities. The existing pipe network in each building consisted of galvanized pipe ranging in diameter from 5 to 1.3 cm (2 to 0.5 in.) for fixtures. The test system consisted of eight loops derived from the bathroom supplies to each apartment and was plumbed to the sample sink in the laboratory in building T-152 with 1.3-cm (0.5-in.) copper pipe. The total length of new plumbing for the sampling lines added approximately 152 m (500 ft) to the distribution system. The end of the pipe network in each building was connected by 2.5-cm (1-in.) plastic pipe to complete a loop and favor circulation in the test system. The use of cast iron, galvanized copper, and plastic pipe simulated the mixed nature of the materials used in real-world distribution systems. The test system was isolated from the Fort Meade water distribution system by a back-flow preventer and an air gap at the reservoir before the test distribution system and the simulated cross-connection. Pressure was maintained in the test distribution system with a pump and hydropneumatic tank.

Test Protocol

The 1.5 m³ reservoir tank (4000-gal) was filled with water drawn from the Fort Meade water distribution system. Disinfectant residual in the tank was adjusted on a batch basis by the addition of sodium sulfate for dechlorination, chlorine, chlorine plus ammonia, or chlorine dioxide. The pH of the water was not adjusted. Raw sewage was seeded with F2 bacterial virus to a level of 10⁶ plaque forming units (PFU)/ml, and the tracer dye (rhodamine, Tinopal RBS, * or Tinopal CBS) was added. An aliquot was removed to determine the microbiological parameters and actual dye concentration. To contaminate the distribution system, the sewage slug was forced into a tee at the head of the system by air pressure.

The studies of test distribution system were divided into four sections:
1. Single tap, short-term - single home
2. Multi-tap, short-term - neighborhood
3. Multi-tap, long-term - small community, constant flow
4. Long-term, variable-flow - small community, constant flow

In all cases, the reservoir water was adjusted to greater than 30 mg/L free chlorine between each run, and this water was flushed throughout the system for least 24 hr to clean and disinfect the system thoroughly.

Results

The inactivation of microorganisms contained in contaminating material in the water distribution system depended heavily on the disinfectant residual, contact time, and temperature in the test distribution system. Residual free chlorine, combined chlorine, and chlorine dioxide were compared as to their efficiencies in inactivating natural populations of coliforms from sewage at various contact times. Natural die away over the 240-min test period was insignificant, but the presence of any disinfectant residual dramatically reduced the level and frequency of coliforms at the tap. The residual disinfectant inactivated natural coliforms in the sewage challenge. Free chlorine appeared to be the most effective residual disinfectant for short contact times and consistently yielded the lowest level and frequency of coliform recovery. The level of free chlorine in the pipe network has a relatively short half life in the absence of contamination and was consumed by the added sewage.

The levels of free chlorine appeared to flag the sewage slug. For short contact times, combined chlorine residuals decreased the density of the coliforms at the tap, but the frequency of coliform recovery (80%) was nearly as high as that observed in the absence of residual disinfectant. The levels of combined chlorine residual were high throughout the system. Coliforms were recovered at combined chlorine residuals of 0.7 to 1.0 mg/L. Little suggestion of contamination was indicated by the level of combined chlorine. Chlorine dioxide residuals for short contact times yielded low levels of coliforms at a low frequency of recovery and was more effective than combined chlorine but not as active as free chlorine.

A similar comparison of residual disinfectants was made for seeded F2 virus. Except for combined chlorine, the disinfectant residuals showed activity against viruses. Chlorine dioxide residuals consistently yielded F2-free water at the taps.

The levels of microorganisms observed at the tap are presented as the log of the coliform and F2 survival fraction in Table 1. The N₀ was corrected for dilution by the dye concentration. The mean log survival of coliform and F2 virus observed was < -3.8 and < -3.4, respectively, for free chlorine. Chlorine dioxide yielded a mean log survival of ≤ -2.8 for coliforms and ≤

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* Mention of trade names or commercial products does not constitute endorsement or recommendation for use.

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Figure 1. Schematic of water distribution system in Fort Meade buildings 152 and 162.
Table 1. Log Coliform and F2 Virus Survival Fraction at the Tap in the Test Distribution System During Multi-tap, Short Term Trials*

<table>
<thead>
<tr>
<th>Disinfectant</th>
<th>Residual mg/L</th>
<th>Initial Tap</th>
<th>Log N/N₀</th>
<th>F2 Virus, Log N/N₀</th>
<th>Mean</th>
<th>Range</th>
<th>Mean</th>
<th>Range</th>
</tr>
</thead>
<tbody>
<tr>
<td>Free chlorine</td>
<td>1.00-1.20</td>
<td>0.49-0.95</td>
<td>1.8-2.8</td>
<td>0.5-0.6</td>
<td>1.3</td>
<td>-1.0</td>
<td>0.5</td>
<td>-5.1</td>
</tr>
<tr>
<td>Chlorine dioxide</td>
<td>0.85-0.95</td>
<td>trace-0.94</td>
<td>2.8-4.0</td>
<td>0.8-1.4</td>
<td>4.4</td>
<td>0.3-7.3</td>
<td>0.5-5.3</td>
<td></td>
</tr>
<tr>
<td>Combined chlorine</td>
<td>0.81-1.08</td>
<td>0.49-1.08</td>
<td>3.2-5.3</td>
<td>1.1-1.6</td>
<td>0.9</td>
<td>-0.6-0.04</td>
<td>0.3-2.6</td>
<td></td>
</tr>
</tbody>
</table>

*Temperature, 14°C to 17°C; pH, 7.3 to 7.7; flow, 2 gal/min.

-4.4 for F2 virus. For combined chlorine, the residual log reduction at the tap was ≤ 3.2, but only -0.9 for F2 virus. Free chlorine and chlorine dioxide were effective against coliforms and test virus during short-term trials (240 min). Though combined chlorine residuals were an effective bactericide, they were a relatively poor viricidal. The F2 virus was seeded at high densities (10⁹ PFU/ml) in the added sewage to permit recovery and evaluation of virus inactivation. Densities of natural populations of human enteric viruses in sewage are generally about 1 PFU/ml, and for the sewage in this study, they were 0.01 to 0.1 PFU/ml. The data for F2 exaggerate the virus survival. The level of free chlorine was reduced in each case when the virus were recovered at relatively high densities. The difference between seed virus titer and natural human enteric virus also exaggerates the decreased viricidal activity of combined chlorine since F2 was more resistant than combined chlorine than was human enteric virus.

Clearly, the disinfectant residual represents the primary barrier against post-treatment contamination in a water distribution system for short contact times. The level of sewage used in these trials was about 0.1% of the test distribution system. At greater levels of contamination, the residual disinfectants would afford proportionately less protection. The residual disinfectant will have little effect on the levels of microorganisms contained in a large intrusion of sewage into the water distribution system. But fortunately, high-level contamination would offend the visual and olfactory senses and flag the potable quality of the water.

The extended contact time trials emphasize several important factors in the ability of the disinfectant to respond to a challenge in the pipe network. Comparisons of coliform levels at the tap found after long contact times (72 hr) were similar to the short-term trials (240 min) in that high coliform levels were consistently recovered in the absence of a disinfectant residual. The presence of any free, combined, or chlorine dioxide residual was not essential for the development and frequency of coliform recovery at the tap. Combined chlorine performed most effectively against the coliforms, with only 3 samples of 28 positive for coliforms at low levels. Combined chlorine residuals were effective bactericides given an adequate contact time. Under the conditions of this experiment, free chlorine and chlorine dioxide were not as effective as combined chlorine. Coliforms were frequently recovered in the chlorine dioxide trials and consistently recovered with free chlorine. In the latter trial, coliform levels were reduced markedly, and most samples collected for the free chlorine trial had no free chlorine residual. The free chlorine was consumed during extended contact in the distribution system. Free chlorine was not as stable in the pipes as combined chlorine. This fact has been the bane of water utilities and was responsible for the development of the chloramination process in the 1930's and its continued preference by a segment of the water plant operators. A similar situation existed with chlorine dioxide.

The F2 virus levels at the tap were compared for the multi-tap, long-term trials. The bacterial virus F2 was recovered at high densities at the tap in the test system after 72 hr when no disinfectant residual was present. As for coliforms in the longer-term trials, combined chlorine residuals were effective against F2. Free chlorine and chlorine dioxide were less effective. But the free chlorine and chlorine dioxide were consumed in the distribution system, and little or no residual was observed. The coliform level was compared with the F2 level for long-term (72 hr) trials with an initial free chlorine residual at 19°C and 10°C. At 10°C, the free chlorine was considerably more stable. Though the level of free chlorine decreased, only 7 of 28 samples had no free chlorine residual. At 19°C, free chlorine was absent in 21 of 28 samples and did not effectively represent a free chlorine trial. When conditions favored the stability of free chlorine in the distribution system (decreased temperature), the residual functioned when challenged. As in the short-term trials, the disinfectant residual was the primary barrier against the sewage challenge. But during long-term trials, the stability of the disinfectant became an important factor. When present, free chlorine was a superior residual disinfectant. Similar results were obtained for both chlorine at 10°C and for combined chlorine at 19°C. As an alternative, combined chlorine (given a sufficient contact time) was able to provide water at the tap with low coliforms and F2 virus levels.

Under variable flow conditions, levels of bacteria and virus in the sewage challenge were effectively reduced by dilution, washout, and increased mass of disinfectant. Few samples at the taps were positive for coliforms and F2 virus. Data in Figure 2 show standard plate count (SPC) and disinfectant residual after contamination with sewage for the test distribution system operated under variable flow conditions for extended periods of time. Except for chlorine dioxide, residuals were observed throughout the trials and residuals at the tap were always greater than 0.5 mg/L. The residual of the water entering the distribution system was approximately 1 mg/L for the chlorine residual provided water at the tap with an SPC of less than 10 colony-forming units (CFU)/ml.

The fate of naturally occurring viruses was determined, and attempts were made to distinguish between free and solids-associated virus. In the sewage challenge, a volume of contaminated sewage over which the disinfectant residuals were recovered from either the solids or the prefilted fractions of the distribution system samples when the contaminant contained detectable levels of these viruses. Such was the case even when combined chlorine served as the disinfectant residual in the distribution system and large numbers of enteric virus were present in the sewage contaminant. When compared with free chlorine and chlorine dioxide, chloramines are considered inferior viricides, but it is possible that longer contact times allow these relatively stable residuals to be effective against viruses. Since no surviving viruses were recovered from the solids fraction in any of the test distribution system samples, no conclusive evidence on the protective effects of virus association with particulate matter could be obtained.
5. In the presence of 0.2 to 1.0 mg/L residuals of free chlorine, chloramines, and chlorine dioxide, no surviving enteric viruses could be concentrated from test distribution system samples contaminated with raw sewage containing detectable levels of enteric viruses.

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Figure 2. Mean SPC and halogen residuals for samples collected over the course of the long-term trials.

Conclusions
1. The residual disinfectant in the water distribution system represents the primary barrier against transmission of disease by post-treatment contamination. The presence of any disinfectant residual reduces the level and frequency of occurrence of microorganisms at the consumer's tap.
2. Free chlorine was the most effective residual disinfectant and consistently yielded the lowest level and frequency of microorganisms.
3. Free chlorine can serve as a marker for contamination. In a system where free residual chlorine was normally maintained, its absence is evidence that chlorine-demanding substances may have entered the system. Few differences in combined chlorine residuals were found, even after the addition of sizable amounts of contaminant. The detection of a combined chlorine residual offers little assurance of water potability.
4. Free chlorine and chlorine dioxide were the least stable in the water distribution system, and residuals were difficult to maintain under low-flow conditions.
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The complete report, entitled "Stability and Effectiveness of Chlorine Disinfectants in Water Distribution Systems," [Order No. PB 84-140 201, Cost: $16.00, subject to change] will be available only from:
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