Stability and Effectiveness of Chlorine Disinfectants in Water Distribution Systems

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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, MD. To the existing pipe network, 152 m (500 ft) of 13-mm (0.5 in.) copper pipe were added for sampling, and 60 m (200 ft) of 2.54-cm (1.0 in.) plastic pipe were 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 in 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 1680 min.

Introduction

The concept of a residual disinfectant in water intended for human consumption is not new. Herodotus, the father of history, described the preparation and distribution of the water consumed by the kings of ancient Persia (1):

"The Great King, when he goes to the wars, is always supplied with provisions carefully prepared at home, and with cattle of his own. Water too from the river Choaspes, which flows by Susa, is taken with him for his drink, so that is the only water which the Kings of Persia taste. Wherever he travels, he is attended by a number of four-wheeled cars drawn by mules in which Choaspes water, ready boiled for use, and stored in flagons of silver, is moved with him from place to place."

The fundamental principles for providing a safe water were practiced and noted in the earliest human records. An adequate quantity of water was taken from a known supply, treated and disinfected, and stored in flagons of silver before consumption by the king. Small quantities of silver in the water provided a disinfectant residual to protect against post-treatment contamination.

Several thousand years later, the lessons of history were slowly learned. As water treatment and distribution practices evolved and the intentional addition of biocides to the water for disinfection became the rule,

the disinfectant residual was carried into the piped distribution network. However, the value of the residual disinfectant remained unclear.

In 1958, at the request of the United States Army, the National Academy of Sciences National Research Council (NAS-NRC) prepared a statement concerning the maintenance of chlorine residuals. Portions of the report are noted below (2):

"Residual chlorine in the concentrations routinely employed in water utility practice will not ordinarily disinfect any sizeable amounts of contaminatory material entering the system, though this will depend on the amount of dilution occurring at the point of contamination, on the type and concentration of residual chlorine and on the time-of-flow interval between the point of contamination and the nearest consumer. . . . It is the opinion of the NAS-NRC that the establishment of a universal standard for maintaining residual chlorine in the water in distribution system is not desirable. . . . The NAS-NRC does not consider maintainance of a residual a satisfactory substitute for good design, construction and supervision of a water distribution system, nor does it feel that the presence of a residual in the system constitutes a guarantee of water potability."

The level of pathogenic microorganisms that reach the consumer's tap during cross-connection and back-siphoning episodes is a function of dilution of the contaminating material, natural die-away, and inactivation by the residual disinfectant. 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.

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Methods

Biological

Total Coliforms. The total coliform count was determined by the multiple tube dilution procedure given in *Standard Methods for the Examination of Water and Wastewater (3)* using lactose broth for the presumptive test and brilliant green lactose broth + 2% bile for confirmation.

Enteric Pathogens. *Salmonella typhimurium* was isolated in our laboratory from raw sewage collected at the Backriver Wastewater Treatment Plant in Baltimore, MD. Attempts to isolate a strain of Shigella was unsuccessful, and therefore a laboratory strain of *Shigella sonnei* was used. *S. typhimurium* and *S. sonnei* were grown overnight in brain-heart infusion at 35°C under aerated conditions, washed three times with saline, and resuspended in a volume of saline equal to that of the original culture. An appropriate dilution was prepared for both cultures in sterile sewage to simulate the contaminated material for the cross connection. In the comparative trials, samples were assayed for coliform, Salmonella, and Shigella by spread plates on xylose lysine agar (Difco). This method enabled the simultaneous determination of coliform (yellow colonies), Salmonella (black colonies), and Shigella (red colonies).

Preparation and Enumeration of Viruses. The f2 bacterial virus was obtained from the American Type Culture Collection (ATCC #15766-B) and virus stocks were prepared by the method given by Loeb and Zinder (*4*). The f2 virus was assayed by the agar overlay technique (*5*) using *E. coli* K-13 (ATCC #15766) as the host bacterium.

Poliovirus 1 (vaccine strain) was prepared in Buffalo green monkey (BGM) cells (*6*) grown in roller bottles in Eagle's minimal essential medium containing 5% fetal calf serum. The poliovirus was grown without antibiotics, since the presence of antibiotics would preclude mixing the poliovirus with the bacterial strains in the inactivation experiments. The virus was harvested using three freeze-thaw cycles, followed by centrifuging to remove cell debris. Poliovirus plaque assays were done using BGM cells. All experiments were performed using aliquots from a single virus preparation.

Standard Plate Counts. Standard plate counts were performed using the pour plate procedure (*3*). The medium used was plate count agar, with incubation at 35°C for 48 hr.

Chemical

Free and Total Chlorine and Chlorine Dioxide. Free and total chlorine and chlorine dioxide residuals were determined by amperometric titration using a Fisher-Porter amperometric titrator. Phenylarsine oxide (PAO) titrant was standardized using potassium iodate as a primary standard. All chlorine concentrations were reported as milligrams per liter available chlorine (*3*).

Turbidity. Turbidity was measured by nephelo-

Table 1.

Dye	Excitation filter	Emission filter
Rhodamine WT	546 nm	23A[b] and 3-66[a]
Fluorescein	45B[c] and 3[c]	4-97[a], 2A[a], and 12[c]
Tinopal RBS	7-37[a]	47B[b]
Tinopal CBS	7-37[a]	47B[b]

[a] Corning color specification number.
[b] Kodak color specification number.
[c] Wratten color specification number.

metric method using a Hach model 2100A turbidimeter (*3*).

Tracer Dye. Tracer dye concentrations were determined with a Turner Model III fluorometer using the filters listed in Table 1.

Experimental Protocol

Holding Tank Studies. A 30-L volume of Baltimore City tap water was drawn and brought to the desired temperature. The chlorine (Cl) residual was measured by amperometric titration and adjusted to the level required for the experiment by adding sodium sulfite or chlorine as required. When a combined chlorine residual was desired, ammonium chloride was added to a threefold molar excess of ammonia. The water ws buffered by adding 0.001 M phosphate; the pH was also adjusted. The schematic of the test protocol used during the experimental runs is shown in Figure 1. Four-liter aliquots were dispensed into polypropylene containers and held in a constant temperature water bath. The pH
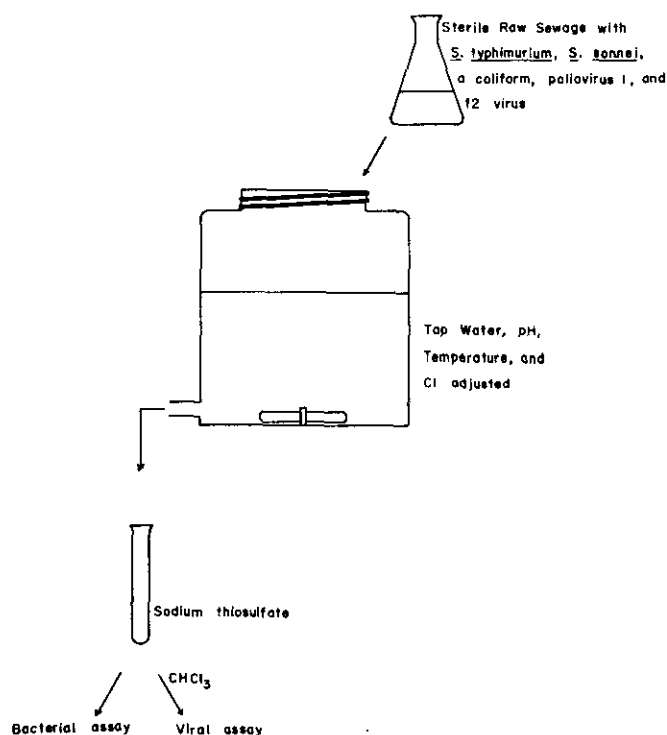

FIGURE 1.   Schematic of holding tank experimental protocol.

and temperature of the autoclaved raw sewage were adjusted, and the sewage was seeded with test organisms (coliform, *S. typhimurium*, *S. sonnei*, f2, and poliovirus 1). At time zero, predetermined amounts of seeded sewage, according to test protocol, were added to the tap water and mixed. Samples for determining microbial survival were withdrawn into tubes containing an excess of sodium thiosulfate at 2, 30, 60 and 120 min contact time. Chloroform (2–3 drops) was added to the sample before viral analysis to eliminate interference from bacteria. Analysis for chlorine residual was performed in all trials at 2 and 120 min after sewage was added.

Reservoir Studies. Reservoir studies were similar to the holding tank studies described above except that the sample volume withdrawn was replaced with an equal volume of fresh chlorinated water; 120-L (30-gal) tanks were used; raw (unautoclaved) sewage was used as the contaminant; and the sewage was not seeded with bacteria, and naturally occurring coliform were assayed.

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. A schematic is shown in Figure 2. 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-m^3 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 adding 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 10^6 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
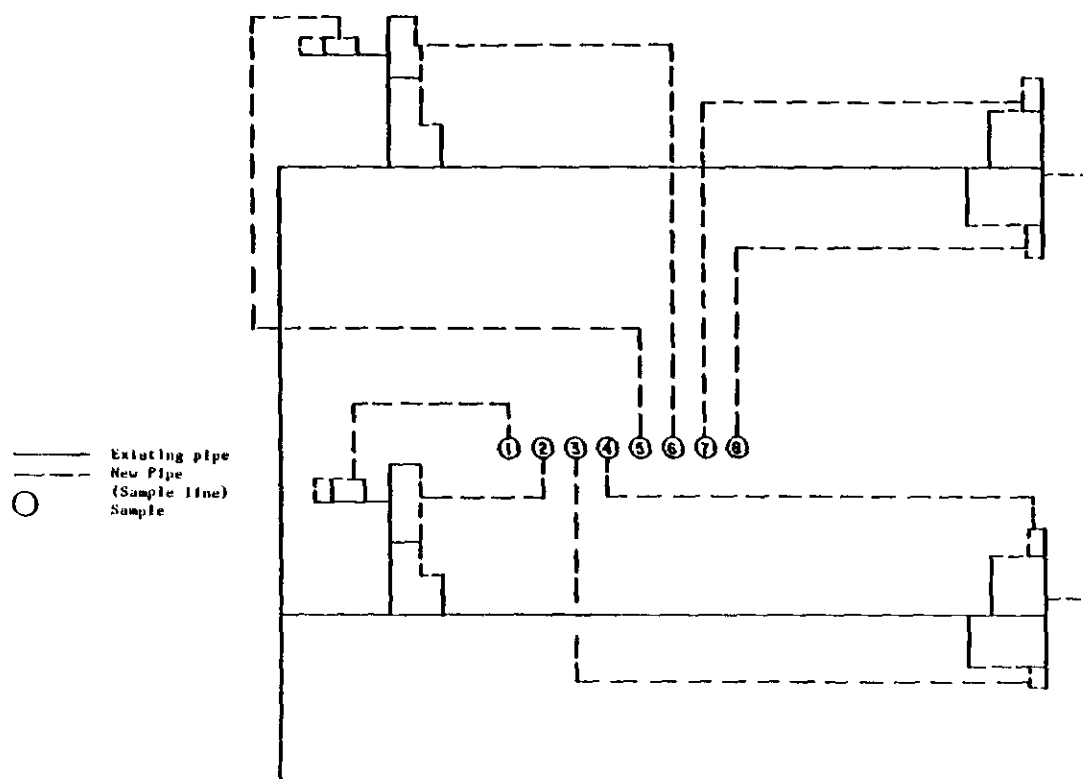

Existing pipe
New Pipe
(Sample line)
Sample

FIGURE 2. Schematic of water distribution system, Fort Meade buildings 152 and 162.

concentration. The distribution system was contaminated by forcing the sewage slug into a tee at the head of the system using air pressure.

The studies in the test distribution system were divided into four sections: single-tap, short-term test; multi-tap, short-term test; multi-tap, long-term, constant flow test; and multi-tap, long-term, variable flow test. 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 at least 24 hr to clean and disinfect the system.

Results

Holding Tank Trials

The inactivation curves of the coliform, f2, and polio 1 microorganisms under varying conditions of initial free or combined chlorine residuals and sewage levels are shown in Figures 3 and 4. Results shown are mean values for four trials. The test temperature was 20°C for different batches of sewage. For clarity of presentation, the results for *S. typhimurium* and *S. sonnei* were omitted from the figure, since these organisms behaved similarly to the coliform organism (shown above). Figure 3 shows the inactivation of the coliform, f2, and polio 1 microorganisms at pH 8 in the presence of 1%, 5%, and 10% added sewage, with an initial free or combined chlorine residual of approximately 1 mg/L. The results are plotted as log N/N_0, where N_0 is the number of microorganisms at time zero and N is the number of microorganisms at any time t. An initial free chlorine residual was more effective than an initial combined chlorine residual for 1% sewage. Greater than 2.7 log inactivation of the coliform occurred in 30 min with the free residual and 2.0 log inactivation occurred in 120 min with the initial combined residual. The free residual was also more effective against f2 and polio at 1% sewage. At the higher sewage levels, the initial free and combined residuals were both ineffective against the introduced microorganisms, with 0.5 log or less difference in the inactivation after 2 hr contact time. Both residuals decreased in effectiveness as the level of sewage was increased. At 10% sewage, less than 1.0 log bacterial inactivation and almost no f2 inactivation were observed. (Poliovirus was not included in the 10% sewage test.)

Inactivation curves for 0.1% added sewage with an initial 0.3 mg/L free or combined chlorine residual at pH 6 and 8 are shown in Figure 4 and demonstrate the superiority of the initial free residual. The difference in inactivation is particularly evident at pH 6, where coliform levels were reduced to the sensitivity limit of the assay within 2 min with an initial free chlorine residual, whereas equivalent reductions with an initial combined residual required 2 hr.

Similar experiments were conducted with chlorine dioxide as the disinfectant. The results shown in Figure 5 show that reductions of *E. coli* B and f2 virus to the
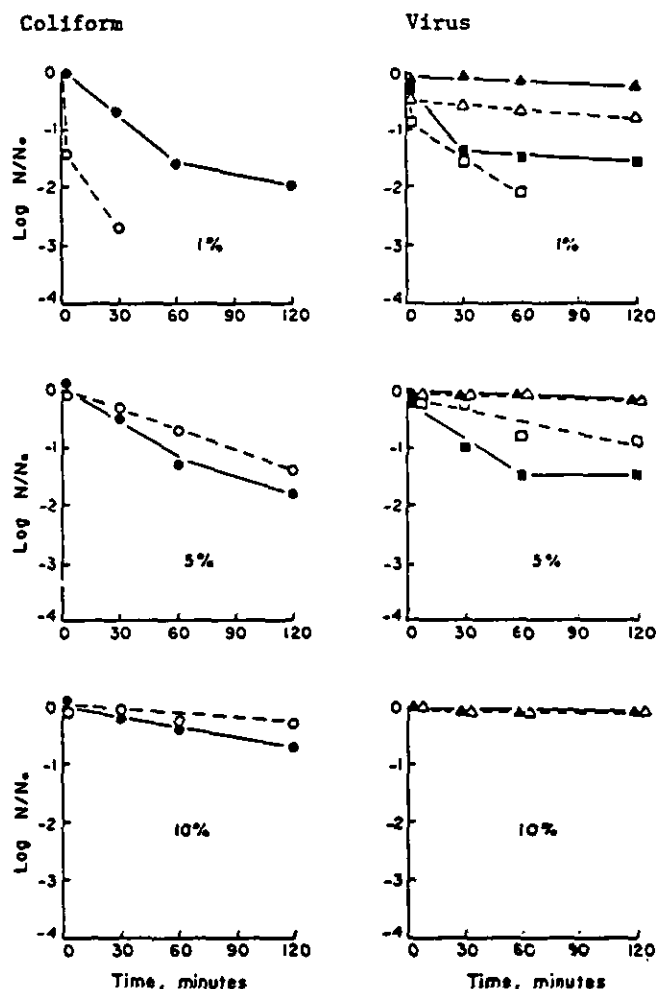

FIGURE 3. Inactivation of the coliform (O), f2 (△), and poliovirus 1 (□) by an approximate initial chlorine residual of 1 mg/L the presence of 1%, 5%, and 10% sewage at pH 8.0. Open symbols: initial free chlorine residual, average of four trials; closed symbols: initial combined chlorine residual, average of seven trials.

lower sensitivity limit of the assay occurred within 5 min for initial chlorine dioxide residuals of 0.75 and 0.55 mg/L at 1% sewage. Levels of 0.17 mg/L chlorine dioxide were not effective in inactivating the microorganisms. The 0.17 mg/L chlorine dioxide residual was reduced to zero within 10 min, accounting for the poor inactivation obtained. Residuals of 0.3 mg/L chlorine dioxide remained at the end of 60 min when the higher initial residuals were used.

The mean time zero, 2-min, and 120-min free and total chlorine residual concentrations accompanying the experimental trials shown in the previous figures are presented in Table 2. A total chlorine residual was always detected 120 min after sewage addition under all the conditions tested. This total residual was generally in the combined chlorine form, with traces of free chlorine detectable only under conditions of low contaminant levels and pH 6. The total chlorine residual was always
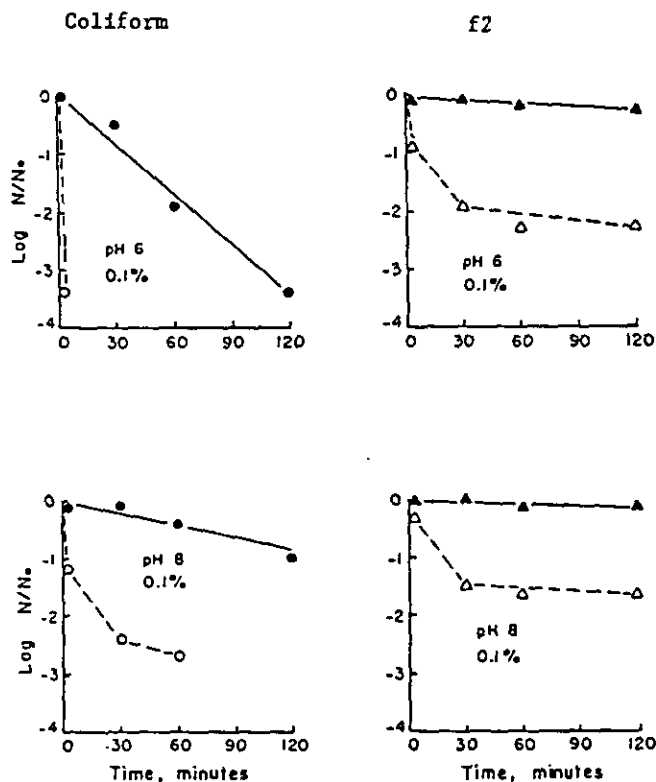
FIGURE 4. Inactivation of coliform (O) and f2 (△) in the presence of 0.1% sewage with an approximate initial chlorine residual of 0.3 mg/L at pH 6.0 and 8.0. Open symbols: initial free chlorine residual, average of four trials; closed symbols: initial combined chlorine residual, average of five trials.

FIGURE 5. Inactivation of f2 virus (solid symbols) and E. coli B (open symbols) by 0.17 (△), 0.55 (□) and 0.75 (O) initial chlorine dioxide (ClO₂) residual as available chlorine in the presence of 1% autoclaved raw sewage. All of pH 7.0, 25°C.

larger when an initial combined residual, rather than an initial free residual, was used, even though the mean initial concentrations were higher for free chlorine.

Reservoir Trials

Reservoir studies were originally designed to be performed without mixing to simulate the manner in which a contaminant enters a large tank or reservoir in the distribution system. However, since tanks become thoroughly mixed when samples are withdrawn and during refilling, the tanks were mixed after contaminant addition to ensure reproducible conditions.

Figure 6 shows the inactivation curves of coliform and f2 virus in the presence of 1%, 5%, and 10% sewage with an initial 0.38 to 0.52 mg/L free chlorine residual, (0.85–0.93 mg/L total chlorine) at pH 8.0 to 8.4 and 28 to 29°C. Biological data were corrected for dilution, so the curves indicate the actual inactivation observed (See Fig. 7). Table 3 gives the chemical data for this experimental run. Three-log inactivation of coliform was observed after 120 min contact time with 1% sewage, whereas between 1- and 2-log removal was observed with the higher percentages of sewage. The bacterial virus, f2, was more resistant than coliform, with a max-
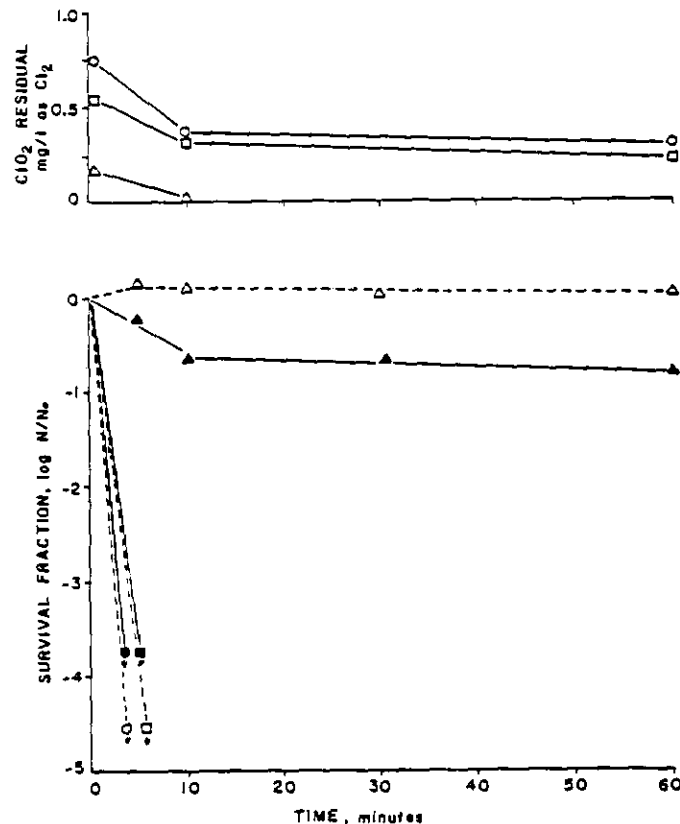
imum inactivation of 2 log with 1% sewage. Chlorine residual data showed no free chlorine present after adding the contaminant. The total chlorine residual remained fairly constant after the initial decrease caused by adding sewage. Results obtained under similar conditions in the holding tank experiments are presented in Figure 3. In the holding tank studies, the contaminant was seeded, autoclaved raw sewage instead of the raw sewage with natural coliform populations being used in the reservoir studies. The studies also differed in that, in reservoir studies, the withdrawn sample was replaced with fresh water containing chlorine, but this procedure was not followed in the holding tank studies. Greater inactivation was observed in the reservoir studies at higher sewage levels, whereas greater inactivation was observed in the holding tank studies at lower sewage levels.

Test Distribution System

Single-Tap, Short-Term Tests. A control experiment with no disinfectant residual is shown in Figure 7. The lower panel shows a typical curve obtained for the sewage slug, measured by fluorescent dye, as it leaves the test distribution system at the tap. The upper

Table 2. Mean chlorine concentrations after addition of varying amounts of sewage at pH 6 and 8.

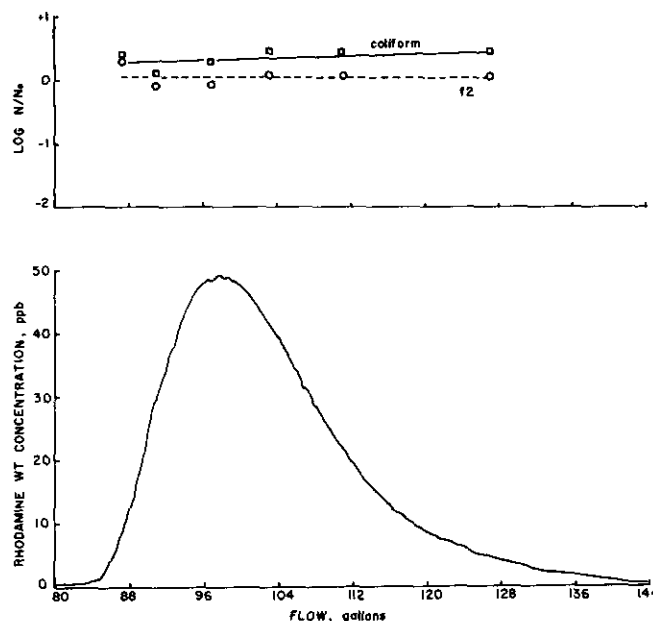| pH	Sewage added, %	Mean initial chlorine concentration (standard deviation)		2-Minute mean chlorine concentration (standard deviation)		120-Minute mean chlorine concentration (standard deviation)	
		Free	Total	Free	Total	Free	Total
8	1.0	1.02 (0.26)	1.22 (0.25)	0.03 (0.01)	0.81 (0.11)	0.01 (0.01)	0.76 (0.11)
	1.0	0	1.07 (0.14)	0	0.90 (0.12)	0	0.85 (0.12)
	5.0	1.02 (0.26)	1.22 (0.25)	0.01 (0.01)	0.47 (0.11)	0 (0.005)	0.46 (0.11)
	5.0	0	1.07 (0.14)	0	0.64 (0.13)	0	0.55 (0.16)
	10.0	1.02 (0.26)	1.22 (0.125)	0 (0)	0.28 (0.16)	0 (0)	0.23 (0.16)
	10.0	0	1.07 (0.14)	0	0.38 (0.16)	0	0.34 (0.17)
6	1.0	1.02 (0.23)	1.21 (0.24)	0.06 (0.02)	0.83 (0.14)	0.02 (0.02)	0.77 (0.13)
	1.0	0	1.07 (0.14)	0	0.97 (0.90)	0	0.84 (0.13)
	5.0	1.02 (0.23)	1.21 (0.24)	0.01 (0.01)	0.50 (0.10)	0 (0)	0.41 (0.08)
	5.0	0	1.07 (0.14)	0	0.67 (0.14)	0	0.53 (0.14)
	10.0	1.02 (0.23)	1.21 (0.24)	0 (0)	0.24 (0.11)	0 (0)	0.16 (0.09)
	10.0	0	1.07 (0.14)	0	0.38 (0.21)	0	0.27 (0.19)
8	0.1	0.24 (0.02)	0.36 (0.02)	0.04 (0.01)	0.23 (0.01)	0.02 (0.01)	0.21 (0.01)
	0.1	0	0.31 (0.06)	0	0.27 (0.05)	0	0.26 (0.06)
6	0.1	0.25 (0.5)	0.36 (0.05)	0.07 (0.03)	0.26 (0.03)	0.04 (0.01)	0.23 (0.05)
	0.1	0	0.30 (0.06)	0	0.29 (0.06)	0	0.27 (0.07)
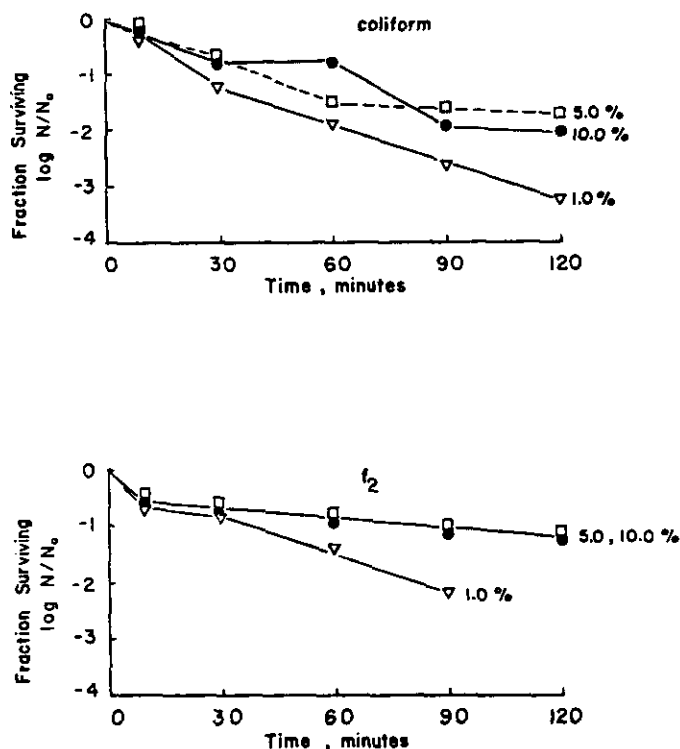
FIGURE 6.　Inactivation of natural populations of coliform and seeded f2 virus contained in sewage after addition of indicated percentage of sewage to tap water in the reservoir with 0.38–0.52 mg/L free chlorine (0.85–0.93 mg/L total chlorine) at pH 8.0–8.4, 28–29°C.

FIGURE 7.　Recovery of sewage containing f2 and coliform at sample station 162 LS for dechlorinated water (control). The flow rate was 7.6 L/min and the sewage slug was 940 mL: (bottom panel) the tracer dye concentration; (upper panel) the recovery of coliform and f2. N_o calculated from the level of microorganisms in the sewage slug and the dye solution.

panel shows the log of the survival fraction corrected for dilution by the dye measurement data determined by Eq. (1):

(dye sewage/dye sample) × microbial density sample =
　　　　　　　　　　corrected microbial density　　　(1)

Over the course of the experiment, little change in the corrected level of coliform and f2 in the control experiment was observed, which demonstrated the validity of the dye correction procedure for subsequent tests.

The results shown in Figure 8 indicate that a 0.90 mg/L initial free chlorine residual was effective in inactivating the microorganisms found in a 450 mL sewage challenge (right panel). Reduction of f2 and coliform to the lower sensitivity limit of the assay occurred in all samples. Figure 8 (left panel) shows the results for an identical challenge with a 0.18 mg/L initial free chlorine residual.

Table 3. Chemical data after the addition of raw sewage to tap water from the Fort Meade, MD, water distribution system with free chlorine at 28–29°C.

Sample time, min	Sewage added, %	Chlorine residual, mg/L		Turbidity, NTU	pH
		Free	Total		
0	1.0	0.41	0.85		8.2
10		0	0.62	2.5	8.4
30		0	0.67	1.5	8.4
60		0	0.67	1.5	8.4
90		0	0.72	1.5	8.4
120		0	0.70	1.2	8.3
0	5.0	0.38	0.74		8.2
10		0	0.35	3.3	8.1
30		0	0.37	3.4	8.2
60		0	0.48	2.9	8.2
90		0	0.74	2.5	8.1
120		0	0.62	2.0	8.2
0	10.0	0.52	0.93		8.2
10		0	0.14	5.3	8.1
30		0	0.21	5.1	8.0
60		0	0.39	4.2	8.1
90		0	0.49	3.1	8.1
120		0	0.55	2.8	8.1

The action of combined chlorine, free chlorine, and chlorine dioxide under similar conditions of sewage challenge and disinfectant residual is shown in Figure 9. The sewage challenge was 225 mL for combined chlorine (top panel) and 450 mL for free chlorine and chlorine dioxide. A free chlorine residual was the most effective in inactivating the microorganisms, with reductions to the sensitivity limit of the assay in all samples. Chlorine dioxide was equally effective against the virus, but coliform inactivation was less than 1 log for all but the most dilute sample. It should be noted that the initial chlorine dioxide residual used was lower than the initial free chlorine residual and that the contact time was different as well. A combined chlorine residual was in-
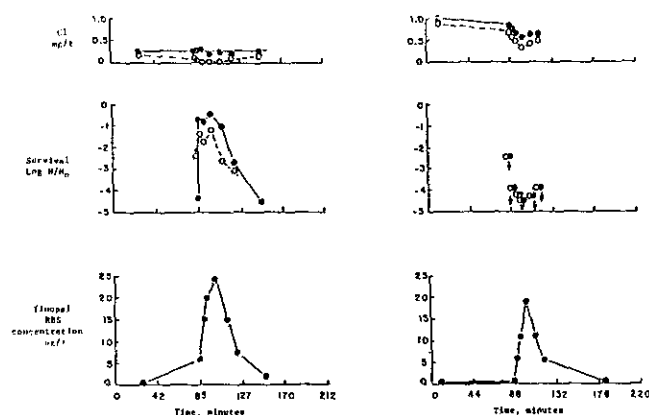

FIGURE 8. Free (O) and combined (●) chlorine residuals, f2 (●) and coliform (O) survival and Tinopal RBS concentration for a 450-mL sewage challenge added at the head of the test distribution system for two different initial free chlorine residuals: (left panel) pH 7.8; (right panel) pH 7.6. Temperature 14°C.

effective in inactivating the virus but did provide a 2-log reduction in coliform.

Multi-Tap, Short-Term Tests. The multi-tap short-term experiments were designed to evaluate the effectiveness of the disinfectant residuals after challenging with sewage for contact times of less than 4 hr. The sewage challenge consisted of 1800 mL raw sewage plus 200 mL of tracer dye solution (100 mg/L tinopal CBS) and seeded f2 virus. The actual sewage-dye volume introduced into the test system varied between 1920 mL and 1970 mL for the 12 trials. The temperature ranged between 13°C and 17°C, and the pH varied between 7.3 and 7.7. The flow rate through the pipe network was 3.8 L/min (1 gal/min).

The inactivation of microorganisms contained in contaminating material in the water distribution system was heavily dependent on disinfectant residual, contact time, and temperature in the test distribution system. Figure 10 shows the comparative efficiency of the residual free chlorine, combined chlorine, and chlorine dioxide to inactivate natural populations of coliform from sewage for varying contact times. The upper left panel shows the levels at the tap with no disinfectant and represents the reduction in levels of coliform caused by dilution. Natural die-away over the 240-min period was insignificant. The presence of any disinfectant residual dramatically reduced the level and frequency of coliform recovery at the tap. The residual disinfectant inactivated natural coliform in the sewage challenge. Free chlorine (upper right panel) appeared to be the most effective residual disinfectant for short contact times and consistently yielded the lowest level and frequency of coliform recovery. The levels of free chlorine appeared to "flag" the sewage slug. For short contact times, combined chlorine (lower left panel) residuals decreased the density of the coliforms at the tap, but the frequency of coliform recovery (80%) was almost as high as that observed in the absence of residual disinfectant. The levels of combined chlorine residual were high throughout the system. Coliform were recovered at combined chlorine residuals between 0.7 and 1.0 mg/L. Little suggestion of contamination was indicated by the level of combined chlorine. Chlorine dioxide residual dioxide residuals (lower right panel) for short contact times, yielded low levels of coliform 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 is shown for seeded f2 virus in Figure 11. Except for combined chlorine, the disinfectant residuals showed activity against viruses. Chlorine dioxide residuals consistently yielded f2-free water at the tap.

The previous figures presented the levels of microorganisms observed at the tap. Table 4 shows the same data presented as the log of the coliform and f2 survival fraction. The N_0 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 ≤ -4.4 for f2 virus but only ≤ -2.8 for
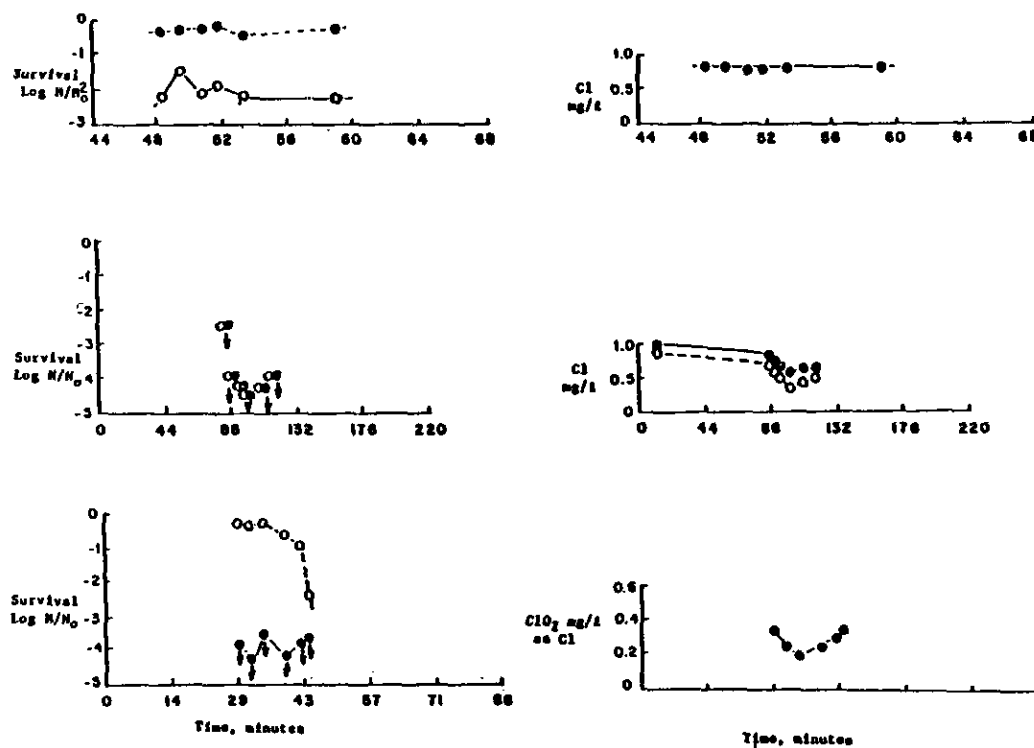
FIGURE 9. Survival of the microorganisms in the raw sewage challenge and different disinfectant residuals. For initial combined chlorine: (O) coliform; (●) f2. temperature approximately 15-20°C, pH 8.2 (top panel); for free chlorine, temperature 14°C, pH 7.6 (middle panel); and for chlorine dioxide, temperature 10°C, pH 7.5 (bottom panel).

coliform. For combined chlorine, the mean log survival for coliform was ≤ -3.2, but only -0.9 for f2 virus. Free chlorine and chlorine dioxide were effective against coliform and test virus during short-term trials (240 min). Combined chlorine residuals, however, were an effective bactericide but a relatively poor viricide.

Multi-Tap, Long-Term Tests. The multi-tap long-term experiments were intended to provide information on the efficacy of residual disinfectants given a long contact time in the pipe network. Flow in the test distribution system was reduced to 0.38 L/min (0.1 gal/min), and composite samples were collected over 50-min periods for 3 days. The sewage slug consisted of 1800 mL sewage with 200 mL of tracer dye, 100 mg/L (Tinopal CBS). Trials were performed at temperatures ranging from approximately 10°C to approximately 20°C, and pH ranged from 7.3 to 8.5.

The extended-contact-time trials emphasize several important factors about the ability of the disinfectant to respond to a challenge in the pipe network. Figure 12 compares the levels of coliform at the tap found for long contact times (72 hr). Similar to the short-term trials (240 min), high levels of coliform were consistently recovered in the absence of a disinfectant residual (upper left panel). The presence of any free, combined, or chlorine dioxide residual dramatically reduced the density and frequency of coliform recovery at the tap. Combined chlorine (lower left panel) performed most effectively against coliform, and only three samples of 28

were positive for coliform at low levels. Combined chlorine residuals were effective bactericides given an adequate contact time. Under the conditions of this experiment, free chlorine (upper right panel) and chlorine dioxide (lower right panel) were not as effective as combined chlorine. Coliform were frequently recovered during the chlorine dioxide trials and consistently recovered during the free chlorine trial. The levels of coliform were markedly reduced, and most of the samples collected for the trial that were designated free chlorine 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 chlorammoniation process in the 1930s and for the continued preference for using chloramine residuals by a segment of water plant operators. A situation similar to that of free chlorine existed for chlorine dioxide.

Figure 13 shows the level of f2 virus at the tap for the multi-tap long term trials. The bacterial virus, f2, was recovered at high densities at the tap in the test system, even after 72 hr when no disinfectant residual was present. As they were for coliform in the longer-term trials, combined chlorine (lower left panel) residuals were effective against f2. Free chlorine (upper right panel) and chlorine dioxide (lower right panel) were less effective. However, the free chlorine and
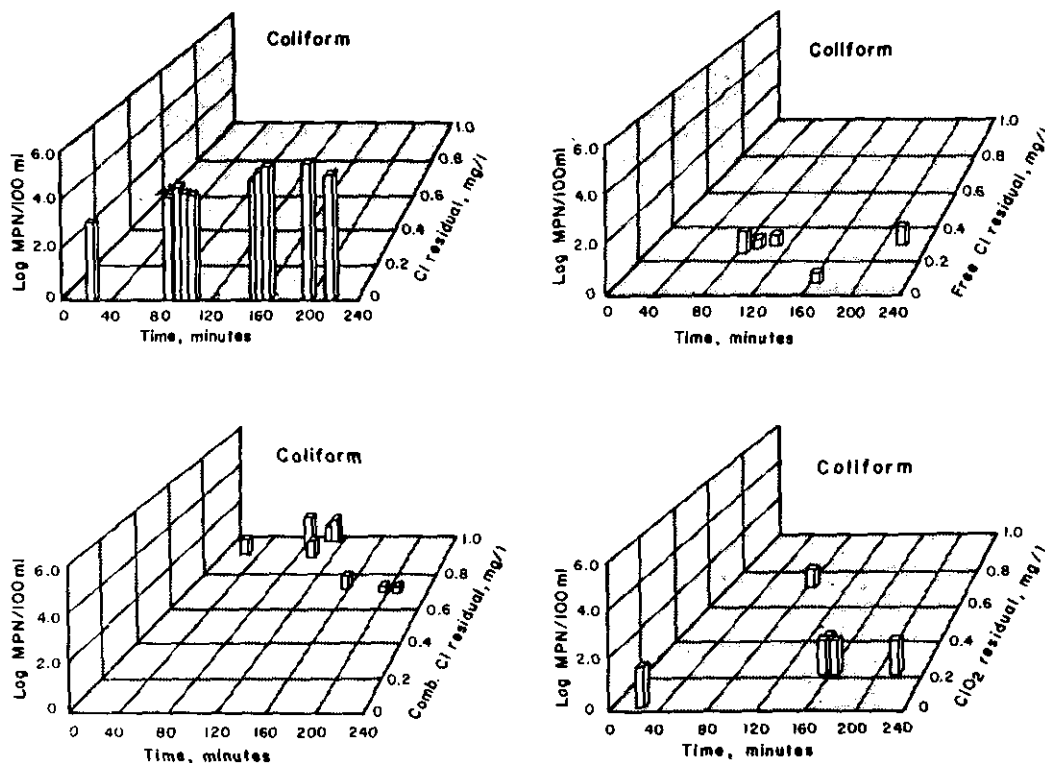
FIGURE 10. Level of coliform at the tap in the test distribution system with no residual disinfectant and an initial 1.04 mg/L free chlorine, 1.08 mg/L combined chlorine and 0.95 mg/L chlorine dioxide residual. Temperature 13–17°C; pH 7.3–7.7.

chlorine dioxide were consumed in the distribution system, and little or no residual was observed.

Figure 14 compares the level of coliform and f2 for long-term (72 hr) trials with 1 mg/L initial free chlorine residual at 19°C (left panel) and at 10°C (right panel). At 10°C, the free chlorine was considerably more stable. Although the level of free chlorine decreased, only seven of 28 samples did not have a free chlorine residual. At 19°C, free chlorine was absent in 21 of 28 samples and effectively does not 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. However, during long-term trials, the stability of the disinfectant became an important factor. Free chlorine, when present, was the superior residual disinfectant. Alternately, combined chlorine, given a sufficient contact time, was able to provide water at the tap with low coliform and f2 virus levels.

Disinfectant Stability

Disinfectant stability was evaluated under static conditions. The test distribution system was flushed at maximum flow until the disinfectant residual level throughout the system was equal to the level of disinfectant introduced at the beginning of the pipe network.

All taps were closed, and small volumes were sampled (< 500 mL) at various times for microbial analysis or residual disinfectant.

The stability of free chlorine at three points in the test distribution system at 20°C and pH 7.7 is shown in Figure 15. The upper panel shows the level of free chlorine vs. time in hours. The free chlorine residual dropped rapidly from 1.3 mg/L to 0.5 mg/L in less than 4 hr. The lower panel shows the same data plotted as the logarithm of the fraction of free chlorine remaining (C/C_0) vs. time in hours. The consumption of free chlorine in the distribution system appears to follow first-order kinetics. The time for half the free chlorine to react was 140 min. At 20°C, the free chlorine residual was unstable and was rapidly consumed in the test pipe network.

Figure 16 shows a similar trial with chlorine dioxide at 22°C. The chlorine dioxide residual (upper panel) decreased from an initial residual of 1.5 to 0.5 mg/L in 3 hr. A plot of the logarithm of the fraction chlorine dioxide remaining yielded a straight line (bottom panel) and indicated that the loss of residual chlorine dioxide follows first-order kinetics. The half-life for chlorine dioxide in the static test distribution system was 93 min. Chlorine dioxide was unstable and was consumed more rapidly than free chlorine in the test pipe network.

The stability of combined chlorine at four stations in the test distribution at 20°C and pH 7.7 is shown in Figure 17. Little change in the combined chlorine residual was observed over 6 hr (upper panel). The log-
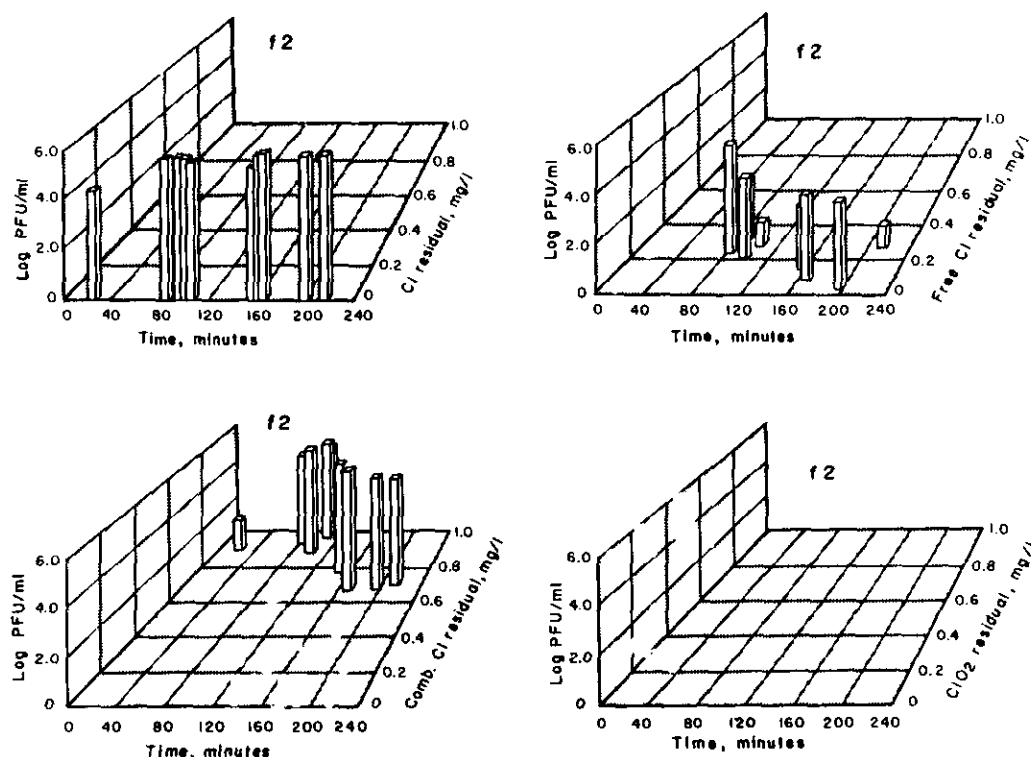
FIGURE 11. Level of f2 virus at the tap in the test distribution system with no residual disinfectant and an initial 1.04 mg/L free chlorine, 1.08 mg/L combined chlorine and 0.95 mg/L chlorine dioxide residual. Temperature, 13–17°C; pH 7.3–7.7.

Table 4. Log coliform and f2 virus survival fraction at the tap in the test distribution system during multi-tap, short-term trials.[a]

Disinfectant	Disinfectant residual		Coliform, log N/N_0		f2 Virus, log N/N_0	
	Initial, mg/L	Tap, mg/L	Mean $\pm$ σ	Range	Mean $\pm$ σ	Range
Free chlorine	1.00–1.20	0.49–0.95	$\leqslant - 3.8 \pm 0.5$	$- 2.6$ to $\leqslant 4.5$	$\leqslant - 3.0 \pm 1.3$	$- 1.0$ to $- 5.1$
Chlorine dioxide	0.85–0.95	trace–0.94	$\leqslant - 2.8 \pm 0.8$	$- 1.4$ to $\leqslant - 4.7$	$\leqslant - 4.4 \pm 0.5$	$- 3.7$ to $\leqslant - 5.3$
Combined chlorine	0.81–1.08	0.49–1.08	$\leqslant - 3.2 \pm 1.1$	$- 1.6$ to $\leqslant - 5.3$	$- 0.9 \pm 0.6$	$- 0.04$ to $- 2.6$

[a] Temperature, 14–17°C; pH, 7.3–7.7; flow, 2 gal/min.

arithm of the remaining fraction (C/C_0) plotted against time in hours (lower panel) yielded a straight line with a low negative slope. The loss of combined chlorine residual also appeared to follow first-order kinetics. The half-life for the combined chlorine was 1680 min (28 hr), more than 10-fold greater than the half-lives of free chlorine and chlorine dioxide. The combined chlorine appeared reasonably stable and was consumed slowly in the static test distribution system.

Water Distribution System Simulation

Two timer motors and solenoid valves on each tap were used to vary the flow in the system from 1.9 L/min to 11.4 L/min (0.5 gal/min to 3.0 gal/min). A 2-L sewage slug was injected at time zero, and samples were taken over a 10-day period to evaluate the recovery of the system from the contaminant. Figure 18 shows the emergence pattern of the dye for replicate trials using

free chlorine. All the contaminant was washed out of the system within 500 min. The peak first appearing is the sewage slug in building 152, and the second peak came from building 162. A total of 30 samples (15 grab and 15 composite) was taken over the 10-day period. Only one sample positive for coliform (Table 5) was obtained in replicate trials with free chlorine at approximately 1.0 mg/L initial residual. Out of 60 samples, four were positive for f2. Positive samples at 2 and 3 hr were contaminated with sewage slug as indicated by the presence of tracer dye. The samples at 21 and 27 hr contained no dye.

Out of 60 samples, one sample was positive for f2 and five samples were positive for coliform when chlorine dioxide was the residual disinfectant. High coliform and f2 levels were obtained at the early times while the sewage contaminated was present. Sporadic lower levels of coliform were found at the intermediate times.

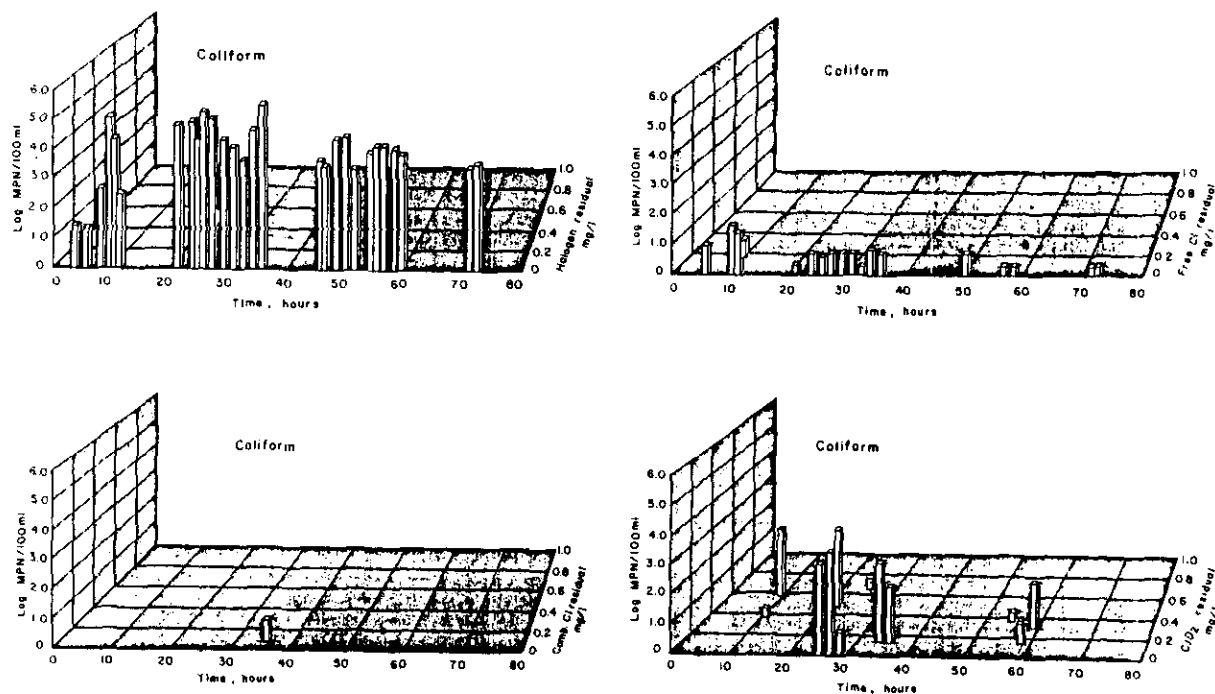The use of an initial combined chlorine residual re-

FIGURE 12. Level of coliform at the tap in the test water distribution system with no residual disinfectant and an initial 1.0 mg/L free chlorine, combined chlorine, and chlorine dioxide residual. Temperature 19–22°C; pH 7.3–8.2.
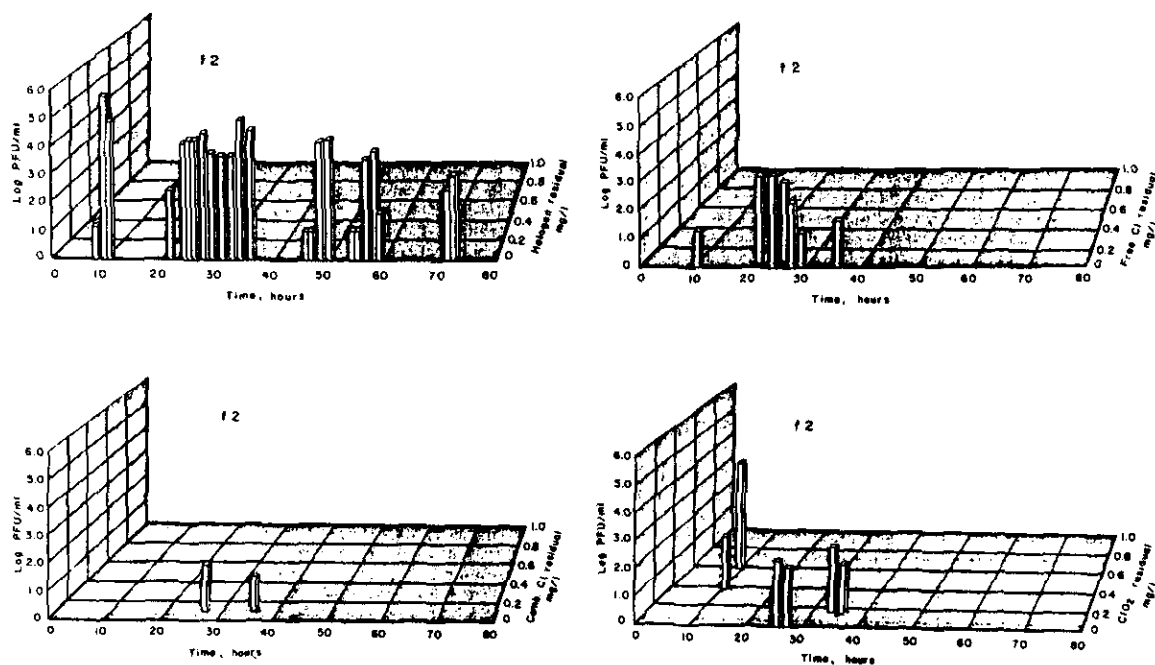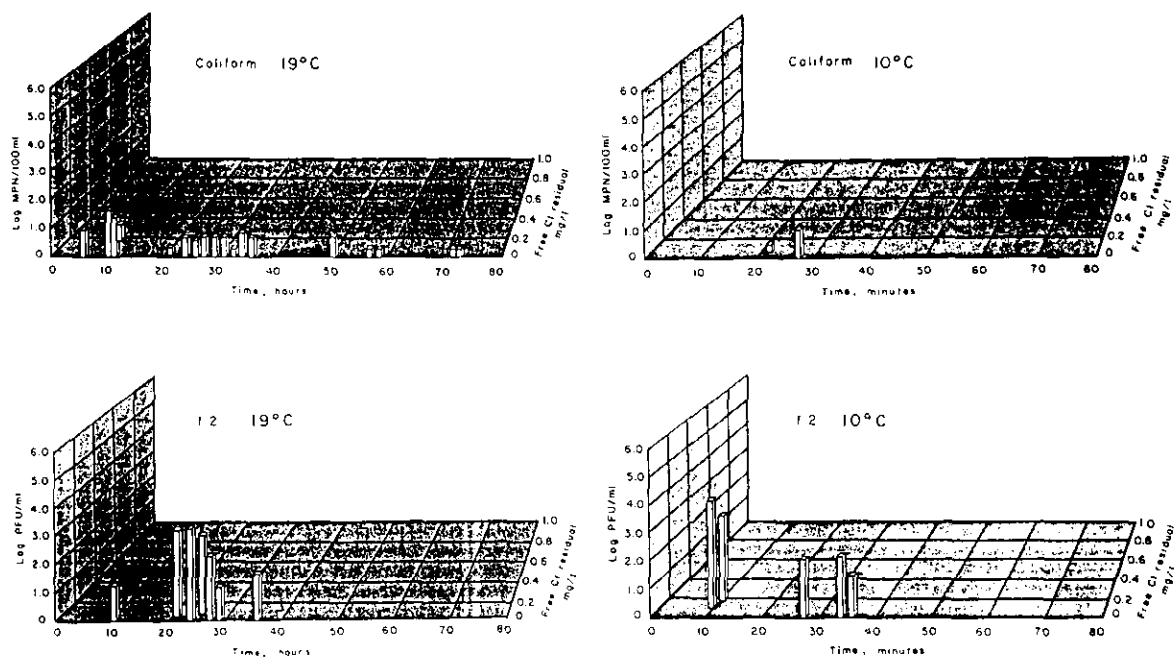

FIGURE 13. Level of f2 virus at the tap in the test water distribution system with no residual disinfectant and an initial 1.0 mg/L free chlorine, combined chlorine, and chlorine dioxide residual. Temperature 10–22°C; pH 7.3–8.2.

FIGURE 14.   Level of coliform and f2 virus at the tap in the test water distribution system with an initial 1.0 mg/L free chlorine residual. Temperature 19°C and pH 7.7–8.0; or temperature 10°C and pH 8.05–8.5.
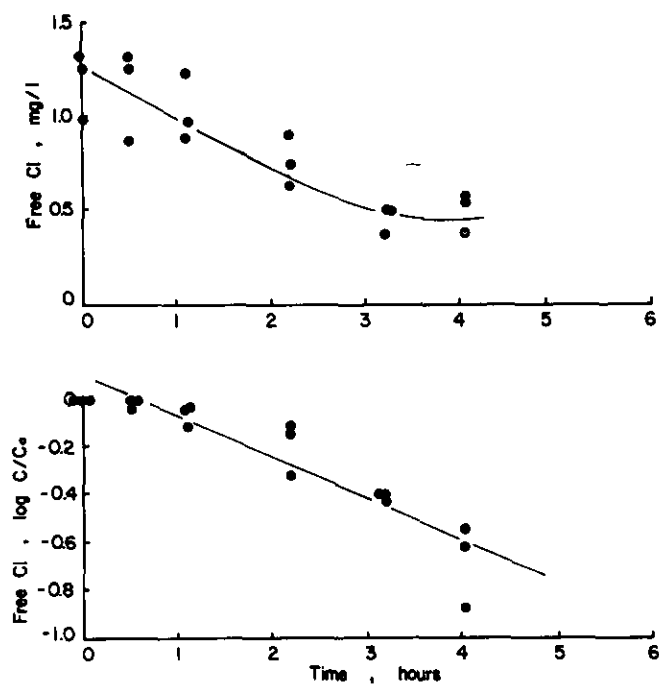

FIGURE 15.   Stability of free chlorine at three sample stations in the test distribution system under static conditions. Temperature 20°C; pH 7.7.
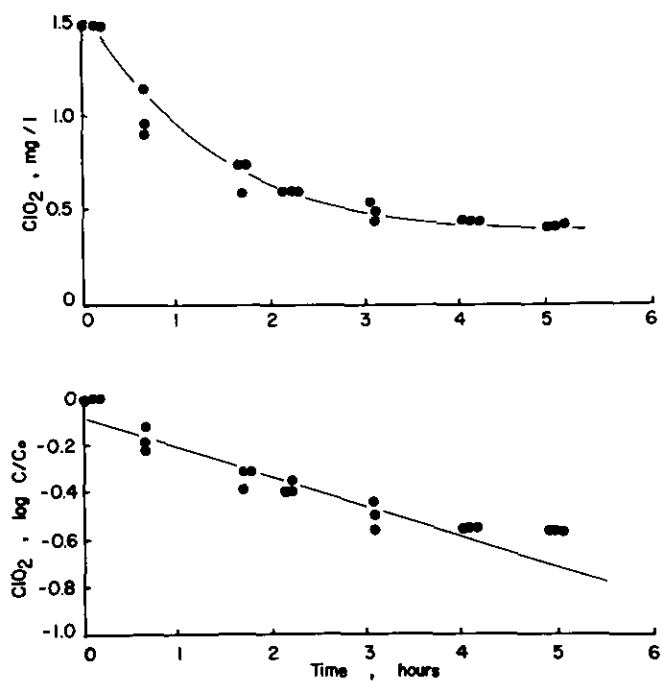
FIGURE 16.   Stability of chlorine dioxide at three stations in the test distribution system under static conditions. Temperature 22°C; pH 7.7.
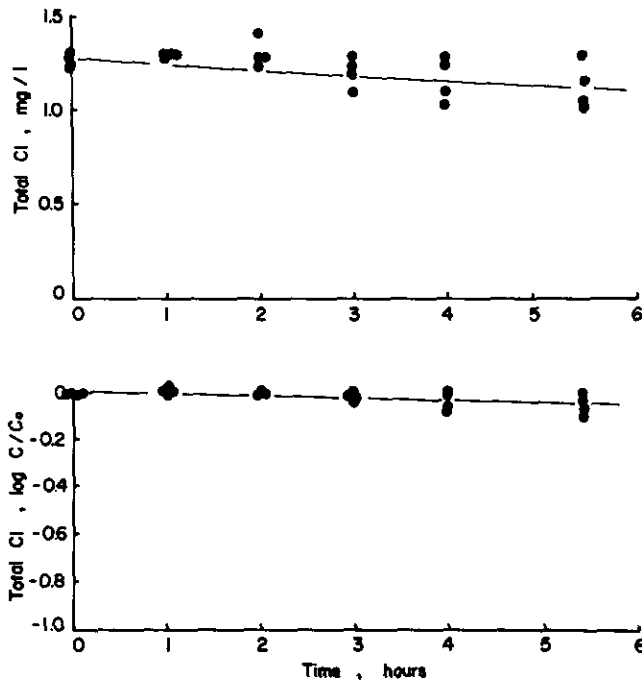
FIGURE 17. Stability of combined chlorine at four sample stations in the test distribution system under static conditions. Temperature 20°C; pH 7.7.

sulted in 2/60 samples positive for coliform and 1/60 positive for f2 (Table 5).

Discussion

Should post-treatment contamination ever occur, all evidence from the control runs suggests profound human health effects in systems lacking any residual dis-

infectant. The level of pathogenic microorganisms that reaches the consumer's tap during cross connection and back-siphoning 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 characterisitics of the pipe network and the flow of water in the local area at the point of perturbation. Because of the infinite combinations and permutations of water flows, the degree of dilution cannot be predicted even in well-defined systems. An aspect related to flow as a mechanism for removing contamination from a water distribution system is wash-out. With continued use and consumption of water, the contaminant slug will be eventually purged from the system in a relatively short time. However, dilution and wash-out cannot be depended on to provide safe water or good quality water at the tap. The enteric microorganisms responsible for the transmission of waterborne diseases tend to die when introduced into the aquatic environment. Fortunately, multiplication of these microorganisms in pipe systems has generally not been observed. However, rates of die-away are relatively slow (7), and sufficient numbers of microorganisms can be expected to survive for the duration water is held in most water distribution systems.

The expected time that the slug of contaminant will spend in the water distribution system appears to be too short to provide pathogen reduction by natural die-away. The residual disinfectant therefore appears to offer the only reliable mechanism for reducing the density of pathogenic microorganism that may enter the public water system. Surprisingly, little data have been available to support this statement, although emergency disinfection during times of disasters such as flood or tornadoes has prevented outbreaks of waterborne
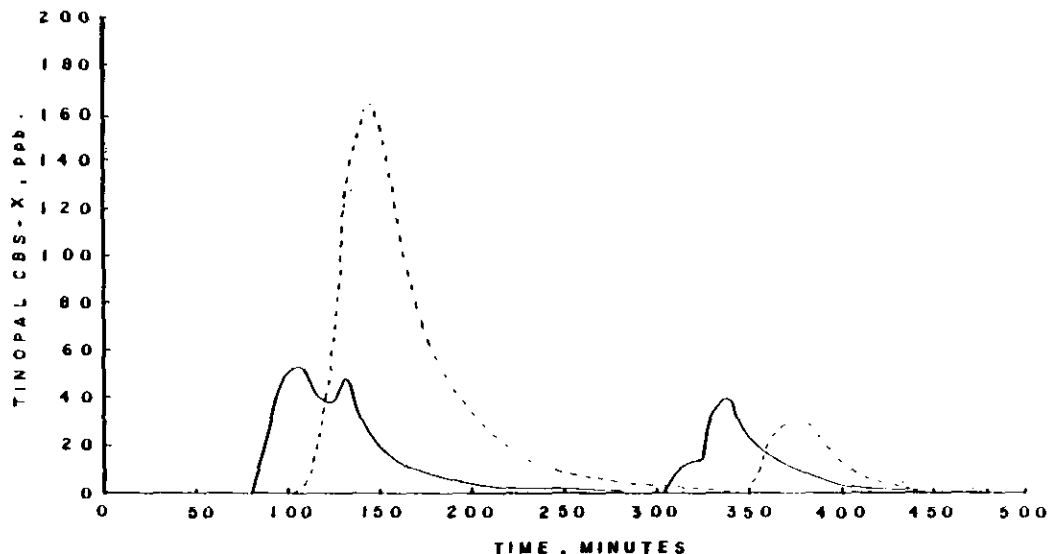

FIGURE 18. Tracer dye profiles for replicate combined chlorine trials for the long-term, variable-flow experiment.

Table 5. Levels of coliform and f2 in long-term, variable-flow trials for an initial free chlorine, combined chlorine, and chlorine dioxide residual.

Disinfectant residual	Time, hr	Coliform, MPN[a]/100mL	f2 Phage, PFU/mL
Free chlorine	2	4.5×10^0	3.5×10^4
	3	LSL[b]	1.0×10^0
	21	LSL	3.0×10^0
	27	LSL	1.0×10^0
Combined chlorine	2	LSL	3.5×10^2
	21	4.5×10^0	LSL
	165	2.0×10^0	LSL
Chlorine dioxide	2	7.9×10^4	7.2×10^4
	51	2.0×10^0	LSL
	141	2.3×10^1	LSL
	147	7.8×10^0	LSL
	165	4.9×10^1	LSL

[a]MPN = Most probable number.
[b]SL = lower sensitivity limit. No coliforms detected.

disease. It is plausible that the current standard practice of maintaining a halogen residual has done much more to reduce the number of cases of enteric disease outbreaks to the point that they often go unreported. The record shows that waterborne disease in recent years occurs in public systems only when there is a breakdown in standard practice or an abnormal situation occurs in the distribution system. The results of the laboratory simulations of sewage contamination of drinking water (8) showed that even low concentrations of residual disinfectants provided significant inactivation of seeded coliform and f2 virus. In the uncovered reservoir model, chlorine dioxide was found to be the most effective residual disinfectant, followed by free chlorine, and then combined chlorine. The superiority of chlorine dioxide can be attributed to its relative lack of reaction with nitrogenous compounds compared to that of free chlorine.

In the test distribution system, the inactivation of microorganisms in sewage slugs was heavily dependent on disinfectant residual, contact time, and temperature. The results for the single household and neighborhood contamination showed that the presence of any disinfectant residual reduced the level and frequency of occurrence of microorganisms at the tap. Free chlorine residuals effectively reduced levels of virus by several order of magnitude (99.94% pathogen destruction). The f2 virus was seeded at very high densities (10^6 PFU/mL in the added sewage) to permit recovery and evaluation of virus inactivation. Densities of natural population of human enteric viruses in sewage generally range from less than 1 to 10 PFU/mL (9). Therefore, the data for f2 in the test system exaggerate the virus survival compared to situations encountered in the real world. The free chlorine residual had been almost completely consumed in each case when virus was recovered. In the presence of combined chlorine residual, f2 virus was consistently recovered at relatively high densities. Chlorine dioxide residuals effectively eliminated the virus. In evaluating these trials in the test distribution system, the difference between seed virus

titer and natural human enteric virus densities should also be noted. The bacterial virus also exaggerates the poor viricidal performance of combined chlorine since f2 is more resistant to combined chlorine than human enteric virus (10).

Levels of free chlorine appeared to "flag" the sewage slug. The free chlorine residual can serve as a marker for contamination since free chlorine reacts rapidly with nitrogenous and organic materials. In a system where free residual chlorine is normally maintained, its absence is evidence that chlorine-demanding substances may have entered the system. Chemical results obtained in these experiments indicate that a total chlorine residual is present even after sizeable amounts of contaminant have been added and that the detection of a combined chlorine residual does not ensure water potability. Positive samples were obtained less frequently when a free chlorine residual was present at the tap than when a chloramine or chlorine dioxide residual was detected.

The extended contact time trials emphasize several important factors in the ability of the disinfectant to respond to a challenge in the pipe network. Under the low-flow conditions used, the rate of influx of free chlorine was apparently less than the rate of consumption by the pipes. The free chlorine was consumed during extended contact in a static distribution system. A similar situation of consumption by the pipe network existed with chlorine dioxide. As in the short-term trials, the disinfectant residual was the primary barrier against the sewage challenge. However, the stability of the disinfectant becomes an important factor. When conditions favored the stability of free chlorine in the distribution system (decreased temperature) the residual functioned when challenged. Alternatively, combined chlorine, given a sufficient contact time, was able to provide water at the tap with low coliform and f2 virus levels.

Clearly, the free 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 (8). It should be recognized that the residual disinfectant will have little effect on the levels of microorganisms contained in a massive intrusion into the water distribution system.

It is believed that the organoleptic and visual controls would protect against such gross pollution in drinking water (note turbidity in Table 3). It is the unsuspected contamination that may offer the greatest threat and may be best prevented by a residual disinfectant.

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