Sequential Chlorination to Control Disinfection Byproducts and Meet Stringent Disinfection Requirements at the Kanapaha Water Reclamation Facility

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The Kanapaha Water Reclamation Facility in Gainesville is an advanced wastewater treatment plant operated by Gainesville Regional Utilities (GRU). The facility is permitted by the Florida Department of Environmental Protection (FDEP) to reclaim 14.9 million gallons per day (mgd) of domestic wastewater based on an annual average daily flow (AADF). Major treatment processes of the facility include pretreatment, advanced activated sludge processes for nitrogen removal, clarification, deep-bed filtration, and high-level disinfection. The facility is permitted to inject an annual average of 10 mgd final effluent into deep-injection wells and to provide an annual average of 12.58 mgd reclaimed water to an unrestricted public-access reuse system. The facility is also permitted to discharge final effluent to Lake Kanapaha during emergency conditions.

The facility disinfects filter effluent with sodium hypochlorite in two series-operated chlorine contact basins (CCBs). Chlorine reacts with organic matter in the effluent to produce disinfection byproducts (DBPs), including trihalomethanes (THMs) and haloacetic acids (HAAs). The FDEP operating permit requires the effluent directed to the deep wells to meet primary and secondary drinking water standards, including DBP standards for total THM (TTHM) and five regulated HAAs (HAA5). The annual average TTHM and HAA5 limits are 80 µg/L and 60 µg/L, respectively. At the same time, the facility's operating permit also has stringent effluent disinfection requirements. The monthly fecal coliform sampling results must be 75 percent non-detect with a single sample maximum of 4/100 mL.

The facility's effluent has consistently met the proposed TTHM compliance limit according to historical sampling results; however, historical data suggest that it will not consistently meet the HAA5 compliance limit using the current free chlorine disinfection method (CH2M HILL, 2006). The GRU investigated a number of alternative disinfection methods and completed several studies to reduce HAA5, including precursor removal using polyaluminum chloride. The studies indicate that the use of polyaluminum chloride reduces HAA formation, but creates unacceptable effluent turbidity levels. An ozone/ultraviolet (UV) system could meet the facility's strict effluent limits, but the system requires a very large capital expenditure and a significant increase in the operations and maintenance (O&M) costs.

A sequential chlorination disinfection method to control DBP formation and to meet the fecal coliform requirements at the facility was proposed. As shown in Figure 1, sequential chlorination involves free chlorination followed by chloramination. A sequential chlorination system will allow substantial capital and O&M cost savings by utilizing the existing chlorine storage and feed system, and requires only the addition of a new ammonia storage and feed system.

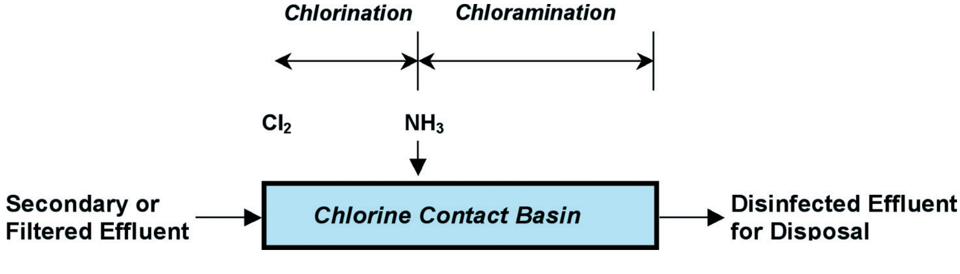

Figure 1. Sequential Chlorination Disinfection

Sequential Chlorination Technology

Chloramines generally form much fewer THMs and HAAs compared with free chlorine (Hua and Reckhow, 2008). Chloramination has been identified as a cost-effective technology to reduce wastewater DBP formation (Bober, 2007; Brandes et al, 2008; Erdal et al, 2008; Hua and Yeats, 2010; Maguin et al, 2009). Chloramines are weaker disinfectants than free chlorine; however, chloramines are more stable than free chlorine and will provide a longer lasting residual disinfectant. Many full-scale studies have shown that chloramination is an effective method to disinfect treated domestic wastewater (Erdal et al., 2008; Maguin et al., 2009).

Wastewater chloramination may produce some emerging byproducts such as N-Nitrosodimethylamine (NDMA) and cyanogens. Sequential chlorination disinfection was developed to reduce NDMA formation in wastewater and is a two-step process, consisting of a free chlorine disinfection step followed by a chloramination step. In the first step, free chlorine is added to a fully nitrified effluent to inactivate pathogens and oxidize inorganic and organic compounds. Ammonia is added in the second step to form chloramines, which reduce the formation of DBPs and provide chloramine disinfection. Full-scale applications of sequential chlorination have shown that the prechlorination step can effectively oxidize precursors for some nitrogenous DBPs such as NDMA and cyanogens (Maguin et al., 2009). Full-scale studies have shown that sequential chlorination exhibits excellent inactivation of coliform bacteria and viruses in filtered wastewater (Maguin et al., 2009).

Scope of Work

In February 2010, a laboratory study was conducted to evaluate the disinfection and DBP formation of sequential chlorination using the facility's filter effluent. The results of the laboratory study indicate that sequential chlorination is a promising alternative to meet the TTHM and HAA5 limits and fecal coliform requirements (Jones Edmunds, 2010).

Based on the successful results of the laboratory study, a pilot-scale study was recommended to further investigate the performance of sequential chlorination at the pilot scale and to provide data for a full-scale design. The primary goals of this study were to further test and evaluate fecal coliform inactivation and THM and HAA formation of sequential chlorination. Several other important parameters were also analyzed during the pilot study for future regulatory considerations, including total coliforms, cyanide, viruses, *Giardia* cysts, and *Cryptosporidium* oocysts.

Materials and Methods

The sequential chlorination pilot study at the facility consisted of bench-scale tests, pilot-scale tests, and an evaluation of the impact of chloramines on the reclaimed water system. The following tasks were performed during the pilot study.

Sequential Chlorination Bench-Scale Tests

The sequential chlorination bench-scale tests were conducted to evaluate the impact of chlorine doses, free chlorine contact times, and temperatures on disinfection and DBP formation. Table 1 summarizes the bench-scale testing conditions. Chlorine dosages of 5 to 10 mg/L are typical for chlorination at the facility. A total contact time of 100 minutes simulates the total contact time of the two CCBs at the current average daily flow of 10 mgd. Ammonia doses were determined based on the chlorine residuals and a chlorine-to-ammonia mass ratio of 4:1.

Filter effluent samples used in the bench-scale tests were collected from the post-aeration basin. The bench-scale tests were

Continued on page 48

Table 1. Summary of Testing Conditions of the Bench-Scale Tests

Test	Cl_2 Dose (mg/L)	Free Cl_2 Contact Time (min)	Temperature (^oC)	Total Contact Time (min)
1. Free Chlorine Contact Time Test	9	5, 15, 20, 25, 30	23	100
	7	10, 20, 30		
2. Free Chlorine Dose Test	5, 7, 8, 10	0.5	23	100
3. Variable Temperature Test	9	10	15, 22, 30	100

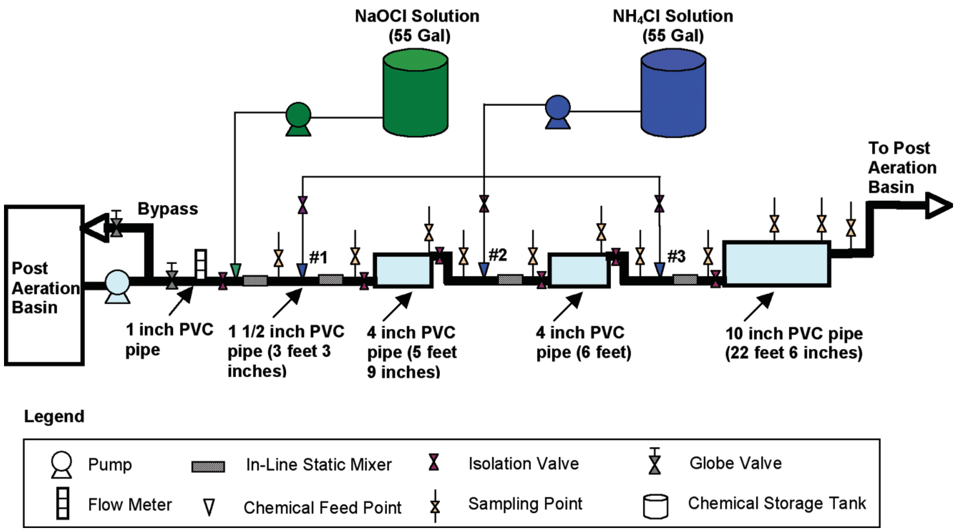

Figure 2. Facility Sequential Chlorination Pilot Plant Schematic

Figure 3. Sequential Chlorination Pilot Plant

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performed using a jar tester apparatus equipped with six variable-speed stirs and six 2000-mL containers. Water incubators were used to maintain the sample temperatures during the variable temperature tests. Each of the three bench-scale tests was repeated twice to investigate the impact of different effluent quality on the test results.

Sequential Chlorination Pilot-Scale Tests

The facility operates two CCBs in series (the North CCB followed by the South CCB). Sodium hypochlorite is injected into a 36-in. diameter pipe in the final effluent flow meter vault upstream of the North CCB, which has a mixing chamber with two low-speed turbine mixers to mix chlorine with the filter effluent. A pilot plant was designed and constructed to simulate the contact times of the facility's CCBs. Three points at the North CCB were identified as potential ammonia injection points for the proposed full-scale sequential chlorination operation. The three injection points include the mixing chamber, the end of the first channel, and the end of the second channel of the North CCB. The average hydraulic retention times after chlorine injection at the current average flow for the three points are 0.5, 4.5, and nine minutes, respectively. Dye-tracer tests were conducted to evaluate the hydraulic conditions of the CCBs. The results suggest that the facility's CCBs performed similarly to an ideal plug-flow reactor. Therefore, the theoretical retention times of the CCBs were used to select the design conditions of the pilot plant to simulate the full-scale operating conditions.

The pilot plant consists of one feed assembly, four pipe reactors, chlorine and ammonia storage and feed system, and piping and accessories. Figure 2 is a schematic of the pilot plant. Filter effluent was continuously pumped to the pilot plant from the post-aeration basin. The flow to the pilot plant was controlled by the openings of the two globe valves for the bypass line and the pilot feed line. The pilot plant flow rate was monitored using a flow meter. The pilot plant flow rate was controlled at 0.5 or 1.0 gallons per minute (gpm), depending on the operating conditions.

The pilot plant has one sodium hypochlorite injection point and three ammonia injection points. A static mixer was used after each chemical injection to mix the chemicals with the water. Four polyvinyl chloride (PVC) pipe reactors with different diameters were used to simulate the hydraulic retention times of the facility's CCBs. The pilot plant ammonia injection points were designed to simulate the contact times of three potential full-scale ammonia injection points at the North CCB. The total contact time of the pilot plant at the flow rate of 1 gpm was 100 minutes. The final effluent of the pilot plant was discharged to the post-aeration basin.

The chemical feed systems of the pilot plant consisted of one sodium hypochlorite storage tank, one ammonium chloride storage tank, and two variable-speed metering pumps for chlorine and ammonia injection. The metering pumps are low-flow peristaltic pumps. Two 55-gallon polyethylene tanks stored the sodium hypochlorite and ammonium chloride solutions during the pilot tests. The sodium hypochlorite solution was prepared by diluting the bulk chlorine solution used at the facility. The ammonium chloride solution was prepared by dissolving ammonium chloride into 50 gallons of deionized water. The flow rates of the two chemical metering pumps were calibrated before each new operating condition.

The GRU personnel constructed the pilot plant. The pilot plant and the chemical feed systems were located adjacent to the post-aeration basin, as shown in Figure 3. The PVC pipe reactors and piping of the pilot plant were placed on the ground and were secured by plastic or wood supports. The chemical feed systems were placed in a metal shed.

Table 2. KWRF Sequential Chlorination Pilot-Scale Test Schedule

Pilot Test	Date	Flow Rate (gpm)	Chlorine Dose (mg/L)	Ammonia Injection Point	Free Cl_2 Contact Time (min)	Ammonia Dose (mg N/L)	Total Contact Time (min)
1	Oct. 25-28	1.0	9	#1	0.5	2.1	100
2	Nov. 01-04	1.0	7	#1	0.5	1.6	100
3	Nov. 08-11	1.0	9	#2	4.5	1.9	100
4	Nov. 15-18	1.0	7	#2	4.5	1.4	100
5	Nov. 22-25	1.0	7	#3	9.0	1.2	100
6	Nov. 29-Dec. 02	0.5	7	#1	1.0	1.4	100
7	Dec. 13-16	1.0	5	#1	0.5	1.1	100
8	Dec. 20-27	1.0	5	Before Cl_2	0	1.3	100

The pilot plant was continuously operated from October to December 2010. Table 2 summarizes the pilot-scale test schedule. Eight pilot operating conditions were performed during the pilot study. Free chlorine contact times between 0.5 and nine minutes were tested to simulate the three potential ammonia injection points at the North CCB. The selected ammonia doses were based on the measured chlorine residuals at the ammonia injection points and a chlorine-to-ammonia ratio of 4:1. A total contact time of 100 minutes was used for all operating conditions. Pilot Tests 1–7 were performed to investigate sequential chlorination disinfection in which chlorine was injected into the filter effluent before ammonia addition. Pilot Test 8 was performed to investigate pure chloramination disinfection (i.e., no free chlorine contact time). Ammonia was added to the filter effluent before chlorine was added for this test condition.

Total coliform and fecal coliform samples were collected daily during the pilot tests. Total coliform and fecal coliform samples were analyzed by GRU's certified laboratory. TTHM, HAA5, and cyanide samples were collected on a daily basis and stored at 4oC. The DBP samples were sent to a laboratory in Savannah, Ga., once per each pilot test. Enterovirus samples were collected once per each pilot test and analyzed as per EPA (Environmental Protection Agency) 600/R-95/178. *Cryptosporidium*, and *Giardia* were collected during three selected pilot tests and analyzed as per EPA 1623. These samples were analyzed by a laboratory in Gainesville.

Chlorine and Chloramines Residual Decay Test

The Kanapaha Botanical Gardens and the University of Florida TREEO (Training, Research, and Education for Environmental Occupations) Center use reclaimed water from the facility for their aesthetic water features. Bench tests were conducted to evaluate the free chlorine and chloramine decay kinetics of the mixture of treated facility effluent and water from the university and gardens water features. The objective of this test was to evaluate if a change to sequential chlorination will cause noticeable changes to the aesthetic water features connected to the facility's reclaimed water system.

The facility's filter effluent samples were collected; one sample was chlorinated with a chlorine dose of 9 mg/L and a second sample was chlorinated with a chlorine dose of 7 mg/L. At a 10-minute free chlorine contact time, ammonium chloride was added to the sample chlorinated at 7 mg/L; no ammonia was added to the other sample. At a total contact time of 100 minutes, the treated samples were mixed with the water collected from the university and the gardens based on a ratio of 1:9. This ratio was used in the chlorine residual decay test to conservatively simulate the conditions of adding reclaimed water from the facility to the gardens and university water features.

Results and Discussion

Sequential Chlorination Bench-Scale Test Results

Table 3 presents the facility's bench-scale test results for sequential chlorination of the filter effluent with chlorine doses of 7 and 9 mg/L and free chlorine contact times of five to 10 minutes. The final total residual chlorine (TRC) concentrations gradually decreased as the free chlorine contact increased. Filter effluent chlorine demands of 3 to 4 mg/L were observed for these tests. Total and fecal coliforms were not detected in treated samples. The TTHM concentrations ranged from 14 to 22 µg/L, which are well below the compliance limit of 80 µg/L.

The HAA5 varied from 57 to 80 µg/L for samples chlorinated at a dose of 9 mg/L. The HAA5 concentration was 57 µg/L when the free chlorine contact time was five minutes. The HAA5 exceeded the compliance limit of 60 µg/L when the free chlorine time was longer than 10 minutes for a chlorine dose of 9 mg/L. The HAA5 ranged from 56 to 72 µg/L for samples chlorinated at a dose of 7 mg/L. The HAA5 concentration was 56 µg/L when the free chlorine contact time was 10 minutes. However, the HAA5 exceeded the compliance limit when the free chlorine contact time was longer than 20 minutes for a chlorine dose of 7 mg/L.

Table 4 presents the results for sequential chlorination with chlorine doses of 5 to 10 mg/L and a short free chlorine contact time of 0.5 minutes. The average chlorine demand for these conditions was 2.4 mg/L. The total and fecal coliforms were not detected for these tests. The two Cl_2/NH_3 dosage ratios used in these tests did not affect the disinfection efficacy of sequential chlorination. The TTHM concentrations varied from 8 to 19 g/L and the HAA5 concentrations ranged from 32 to 45 µg/L. These DBP levels are below the facility's compliance limits.

Table 5 shows the results of the variable temperature tests. No fecal and total coliform positive samples were identified for the range of wastewater temperatures (15-30°C) tested. Higher temperatures resulted in higher chlorine demand due to the increased reactivity between chlorine and compounds in the wastewater. The formation of TTHM increased as the temperature increased; for example, the TTHM increased by 65 percent when the temperature increased from 15°C to 30°C. However, no consistent temperature effect was observed for the formation of HAA5.

All disinfection scenarios of the bench-scale tests exhibited excellent disinfection efficacy. Based on the bench-scale test results, the free-chlorine contact time should be limited to five minutes for a chlorine dose of 9 mg/L to reduce the formation of HAA5. The free chlorine contact time may be increased to 10 minutes when a chlorine dose of 7 mg/L is used.

Sequential Chlorination Pilot-Scale Test Results

Table 6 presents the DBP results of the pilot-scale tests. In general, the TTHM and HAA5 concentrations increased when free chlorine contact time increased. The pure chloramination test (Test 8) produced the lowest TTHM and HAA5 concentrations among the eight pilot tests; this indicates chloramines effectively reduce the DBP formation. The average TTHM concentrations of the eight pilot tests ranged from 5 to 40 µg/L, which are much lower than the compliance limit of 80 µg/L. The average HAA5 concentrations ranged from 12 to 37 µg/L, and are also well below the compliance limit of 60 µg/L.

The primary drinking water standards set a maximum contaminant level at 200 µg/L for cyanide. Fourteen effluent samples were analyzed for cyanide during the pilot tests; only two samples had cyanide concentrations above the laboratory detection limit of 2.5 µg/L. This suggests that sequential chlorination generated insignificant amounts of cyanide during the pilot tests.

Continued on page 50

Table 3. Sequential Chlorination Bench-Scale Test Results—Free Cl_2 Contact 5 to 30 min.

Chlorine Dose (mg/L)	Free Cl_2 Contact Time (min)	Total Contact Time (min)	TRC (mg/L)	Fecal Coliform (#/100 mL)	Total Coliform (#/100 mL)	TTHM (µg/L)	HAA5 (µg/L)
9	5	100	6.3	0	0	14	57
9	10	100	5.7	0	0	16	62
9	15	100	5.4	0	0	18	72
9	20	100	5.3	0	0	20	75
9	25	100	5.1	0	0	21	74
9	30	100	5.0	0	0	21	80
7	10	100	4.2	0	0	18	56
7	20	100	4.0	0	0	22	67
7	30	100	3.8	0	0	25	72

Table 4. Sequential Chlorination Bench-Scale Test Results—Free Cl_2 Contact 0.5 min.

Chlorine Dose (mg/L)	Free Cl_2 Contact Time (min)	Cl_2: NH_3-N	Total Contact Time (min)	TRC (mg/L)	Fecal Coliform (#/100 mL)	Total Coliform (#/100 mL)	TTHM (µg/L)	HAA5 (µg/L)
5	0.5	5:1	100	3.1	0	0	8	40
7	0.5	5:1	100	5.3	0	0	10	44
8	0.5	5:1	100	5.7	0	0	12	45
10	0.5	5:1	100	6.9	0	0	14	44
5	0.5	6:1	100	2.8	0	0	15	32
8	0.5	6:1	100	4.6	0	0	19	36

Table 5. Sequential Chlorination Bench-Scale Test Results—Variable Temperature Test

Chlorine Dose (mg/L)	Free Cl_2 Contact Time (min)	Temp (°C)	Total Contact Time (min)	TRC (mg/L)	Fecal Coliform (#/100 mL)	Total Coliform (#/100 mL)	TTHM (µg/L)	HAA5 (µg/L)
9	10	15	100	6.6	0	0	23	69
9	10	22	100	6.0	0	0	24	58
9	10	30	100	5.1	0	0	38	70

Table 6. Sequential Chlorination Pilot-Scale Test Results—DBPs

Pilot Test	Cl_2 Dose (mg/L)	Free Cl_2 Contact Time (min)	TRC (mg/L)	TTHM (µg/L)	HAA5 (µg/L)	Cyanide (µg/L)
KWRF Permit Limit				*80*	*60*	*200*
1	9	0.5	7.1	24	29	<2.5
2	7	0.5	5.8	22	28	<2.5
3	9	4.5	6.6	28	35	<3.8
4	7	4.5	5.0	31	37	<2.5
5	7	9.0	4.0	40	37	<2.5
6	7	1.0	5.3	25	34	<2.5
7	5	0.5	3.8	17	23	<2.5
8	5	0	4.2	5	12	4.6

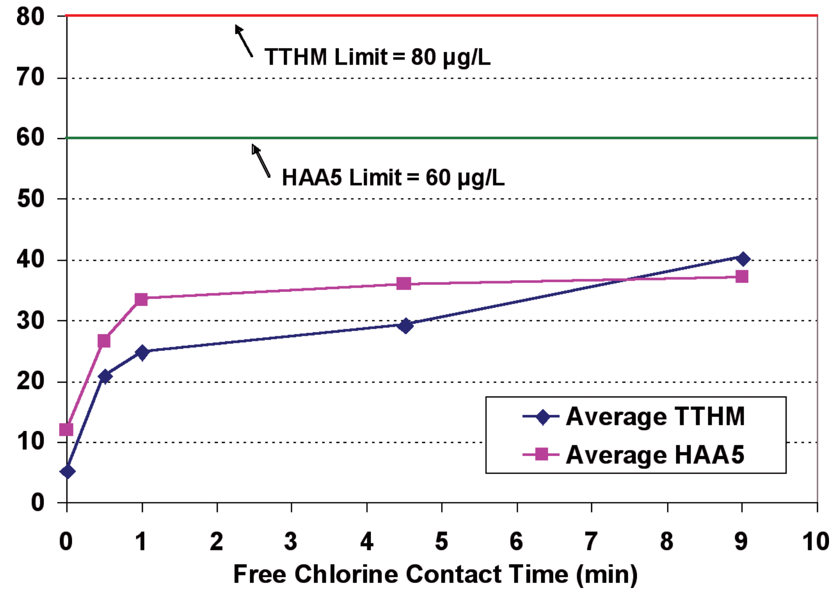

Figure 4. Effect of Free Chlorine Contact Time on DBP Formation

Table 7. Sequential Chlorination Pilot-Scale Test Results—Disinfection Efficacy

Pilot Test	Cl_2 Dose (mg/L)	Free Cl_2 Contact Time (min)	Fecal Coliform (#/100mL)	Total Coliform (#/100mL)	Virus (pfu/100 L)	*Cryptosporidium* (oocysts/100L)	*Giardia* (cysts/100L)
KWRF Permit Limit			*4*	*None*	*Report (0.165)**	*Report (22)**	*Report (5.0)**
1	9	0.5	0	0	<0.3	<0.6	1.8
2	7	0.5	0	0	<0.4		
3	9	4.5	0	<1	<0.3	<1.1	<0.9
4	7	4.5	0	0	<0.2		
5	7	9.0	0	0	<0.3	<1.0	<1.0
6	7	1.0	0	0	<0.3		
7	5	0.5	0	0	<0.3		
8	5	0	0	0	<0.3		

*Numbers given between parentheses are the FDEP reuse water pathogen guidelines.

Continued from page 49

Figure 4 presents the effect of free chlorine contact time on the DBP formation. The TTHM and HAA5 concentrations shown in this figure are the averages of the pilot tests with the same free chlorine contact time. This figure shows that the TTHM and HAA5 concentrations increased when the free chlorine contact time increased from 0 to nine minutes. This suggests that the majority of the DBPs during sequential chlorination are formed by the free chlorination step. The contribution of the chloramination step to the DBP formation is insignificant.

Table 7 presents the disinfection results of the pilot tests. The pilot plant influent fecal coliforms ranged from 100 to 1,700/100 mL, with an average of 538/100 mL during the pilot test. The pilot plant influent total coliforms ranged from 850 to 6,000/100 mL, with an average of 2,925/100 mL. For all test conditions, no fecal coliforms were detected in any of the pilot effluent samples. In Test 3, only one total coliform positive sample was detected (1/100 mL), but no total coliforms were detected in the duplicate sample. In all the other pilot test effluent samples, no total coliforms were detected. The pilot test coliform results confirmed that sequential chlorination with a short free chlorine contact time (0.5 to nine minutes) is highly effective at inactivating coliform bacteria in the facility's filter effluent. Pure chloramination with a chloramine dose of 5 mg/L also completely inactivated the coliform bacteria in the filter effluent. This suggests chloramination can be used as an alternative operating mode to sequential chlorination in the full-scale application.

Eight pilot effluent samples were collected and analyzed for viable infectious enteroviruses during the pilot tests. The pilot plant influents had viruses at levels of 0.3-1.3 pfu/100 L. No infectious viruses in the pilot plant effluents were detected, which suggests that sequential chlorination is effective at inactivating enterovirus. Three pilot effluent samples were collected for the analysis of *Giardia* cysts and *Cryptosporidium* oocysts. The total and potentially viable *Giardia* and *Cryptosporidium* levels in these samples were below the proposed FDEP reuse water guidelines (York et al., 2003). These results indicate the deep-bed filters effectively remove Giardia and Cryptosporidium from the facility's secondary effluent.

Chlorine and Chloramines Residual Decay Test Results

Figures 5 and 6 show the results of the free chlorine and chloramines residual decay tests for the treated facility filter effluent mixed with waters collected from the university and the gardens. Free chlorine residuals showed a fast decay for both samples. The chloramines residuals in the reclaimed water also decayed rapidly. The aesthetic water features at the university and the gardens rely on the reclaimed water at the facility to maintain water levels. The natural organic content of these water features is expected to rapidly consume the total chlorine residual in the reuse water supplied to these locations. Based on the chlorine residual decay tests and field observation, sequential chlorination should not impose a significant risk to the aesthetic water features at the university and the gardens.

Conclusions

The results of this study showed that sequential chlorination with a short free chlorine contact time (0.5 to nine minutes) completely inactivated fecal and total coliforms in the facility's filter effluent. The average TTHM concentrations of the pilot-scale tests ranged from 5 to 40 µg/L and the average HAA5 concentrations ranged from 12 to 37 µg/L. These TTHM and HAA5 levels are well below the compliance limits of 80 and 60 µg/L, respectively. Based on the chlorine residual decay tests and field observation, sequential chlorination should not create a significant risk to the aesthetic use of the reclaimed water.

The results of this study suggest that sequential chlorination is a highly effective method for wastewater disinfection and DBP control, and can be utilized at the facility to meet FDEP disinfection and DBP permit requirements. Sequential chlorination disinfection also presents substantial capital and annual operating costs savings compared to other disinfection alternatives such as UV and ozone.

Acknowledgements

This sequential chlorination disinfection study was funded by GRU. The authors gratefully acknowledge the help from the GRU laboratory, operations, and maintenance staff throughout this study.

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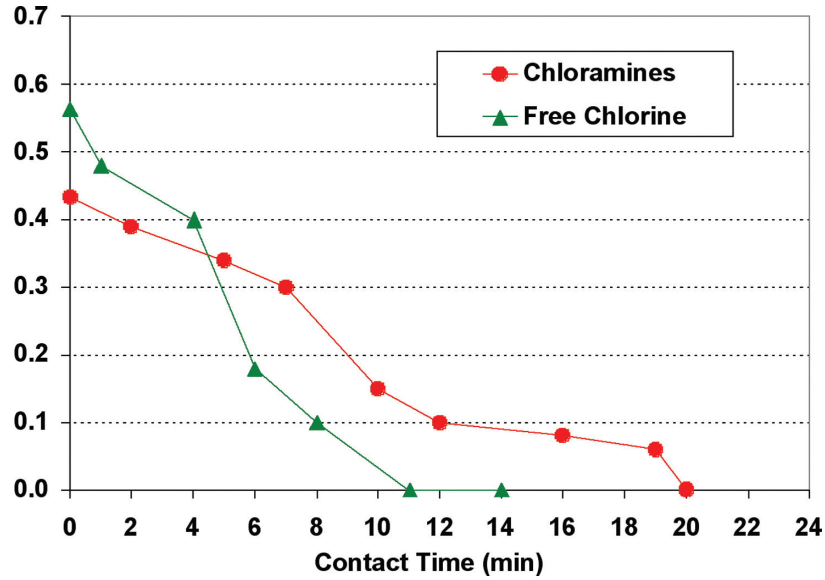

Figure 5. Total Residual Chlorine Decay Test Results—TREEO Sample

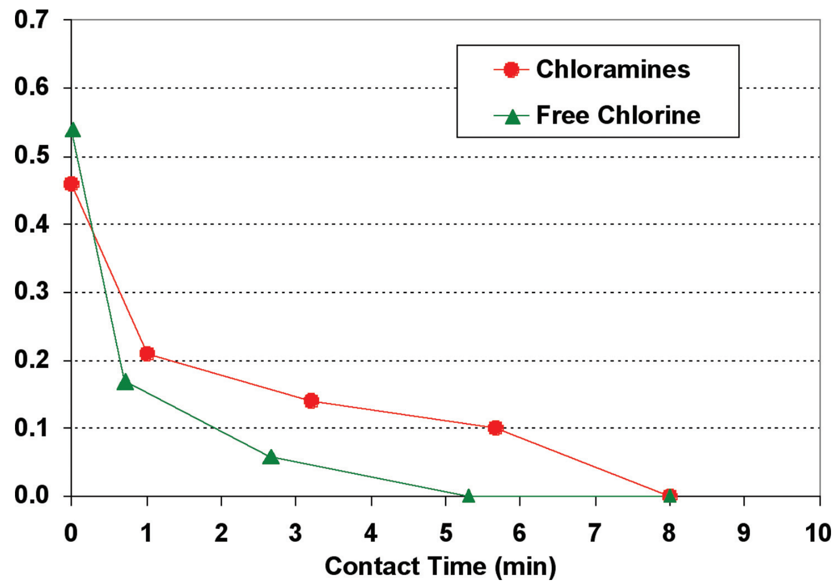

Figure 6. Total Residual Chlorine Decay Test Results—Botanical Gardens Sample