

## Effect of water hardness and pipe material on enhanced disinfection with UV light and chlorine

J. L. Rand, M. Sharafimasoleh and M. E. Walsh

### ABSTRACT

Limited studies have been conducted to evaluate synergistic benefits between disinfectants, although promise has been shown between ultraviolet (UV) light and chlorine-based disinfectants. This research aimed to determine drinking water quality factors that affect potential for enhanced removal of heterotrophic bacteria due to synergy between UV light and free chlorine. An additional goal was to determine the impact of pipe material on the formation of biofilm and the effectiveness of combined disinfection in controlling it. Annular reactors (ARs) containing polycarbonate or cast iron coupons were used to simulate typical drinking water distribution systems. Two experiments were conducted with untreated hard groundwater and groundwater treated with ion exchange to remove hardness, each comparing chlorine alone to chlorine with UV pre-treatment. Results show that enhanced removal of heterotrophic plate count (HPC) bacteria existed with hard water when UV preceded chlorine, but synergistic benefits were not consistently observed. Under softened water conditions, no enhanced reduction or synergy was observed. Cast iron ARs supported biofilm HPC bacteria that were unaffected by any disinfection scheme regardless of source water, while bulk HPC bacteria were more resistant to disinfection in polycarbonate than cast iron ARs. Results indicate that disinfection synergy between UV and chlorine is limited in groundwater sources.

**Key words** | chlorine, disinfection synergy, distribution system, heterotrophic bacteria, ultraviolet light, water quality

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### INTRODUCTION

In order to microbiologically monitor the water quality in distribution systems, heterotrophic plate count (HPC) constitutes a standard indicator that is widely used (Allen *et al.* 2004; Pavlov *et al.* 2004). High HPC may indicate some failure in the treatment process (Sartory 2004). A critical step in any drinking water treatment process is disinfection, to provide a level of microbial inactivation at the plant and residual concentration in the distribution system to limit regrowth. The most traditional and widely used disinfectant is chlorine (Cl<sub>2</sub>) because of its simplicity, economic benefits, and ability to provide a residual. Ultraviolet (UV) light is an increasingly common form of disinfectant that has the ability to inactivate pathogenic microorganisms that are chlorine-resistant and that may lower the required chemical disinfection dose (Cotton *et al.* 2001; Craik *et al.* 2001).

Various studies have indicated that synergistic benefits exist between UV light and other disinfectants in the treatment of water and wastewater. Wang *et al.* (2012) observed synergy between UV light and free chlorine in reducing microbial pathogens in reclaimed water. Dykstra *et al.* (2007) indicated UV pre-treatment followed by secondary chlorine-based disinfectants enhanced microbial control in a modelled distribution system and reduced chemical disinfectant dosage. Koivunen & Heinonen-Tanski (2005) showed synergistic benefits between UV light and peracetic acid (PAA) for the treatment of wastewater. This study presented a mode of evaluation for synergistic benefits, stating that synergy is when the 'efficiency of combined disinfection method is greater than the efficiency achieved when summing the effects of individual disinfectants'.

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Most enhanced disinfection studies focus on the potential for synergy between various disinfectants for the removal of pathogens, but few have related source water and operational characteristics to this potential. For groundwater sources, there are limited studies that have evaluated combined disinfection strategies. The purpose of this research was to identify water quality and design factors in drinking water distribution systems that could enable or hamper potential for disinfection synergy between UV light and free chlorine in suppressing heterotrophic bacteria. Untreated hard groundwater and groundwater softened with ion exchange (IX) were used as sources for two experiments to compare disinfection strategies in both types of water. Hard water contains an appreciable quantity of dissolved minerals (e.g., calcium and magnesium), but softened water often treated with IX technology involves the exchange of sodium ( $\text{Na}^+$ ) or potassium ( $\text{K}^+$ ) from the surface of the IX resin with hardness minerals from the water. This results in ionic variability between groundwater sources that have been treated or not for hardness removal. As noted in the review paper by Chowdhury (2012), significant research has been dedicated to understanding the roles pipe material and corrosion play in bacterial regrowth; therefore, the effects of two pipe materials, cast iron and polycarbonate, on HPC bacteria formation were also evaluated in this study.

## MATERIALS AND METHODS

### Description of model distribution system

Annular reactors (ARs), widely used for drinking water research, (e.g., Camper 1996; Sharp *et al.* 2001; Ollos *et al.* 2003; Dykstra *et al.* 2007; Rand *et al.* 2007), were used to represent model distribution systems (Model 1120LS, BioSurface Technologies Corporation, Bozeman, MT). The ARs operated with a 2-hour retention time at a rotational speed of 50 rpm, which translates into a shear stress of  $0.25 \text{ N/m}^2$  (Camper 1995). All non-opaque exposed surfaces were covered with aluminium foil to reduce the potential of phototrophic growth. Before each experimental trial, all ARs and associated tubing were cleaned with antibacterial soap and disinfected using a 70% ethanol solution.

### Experimental design

The water feed to the ARs for the first study was an untreated aquifer water source with 192 mg/L hardness. The second study used a municipal groundwater source that was treated with a granular-activated carbon filter to remove residual chlorine followed by softening with ion exchange (IX) with a sodium chloride ( $\text{NaCl}$ ) resin to achieve 26 mg/L hardness. Both sources were monitored for various water quality parameters throughout the experiments (Table A1, available online at <http://www.iwaponline.com/jws/062/060.pdf>). Two sets of four ARs containing polycarbonate and cast iron coupons were used in each experiment. In each set, one AR acted as a raw water (RW) control and was not dosed with UV or chlorine throughout the experiment. A second AR acted as a control for UV disinfection (UV), and feed water was treated with UV but not dosed with chlorine. The final two ARs in each set were treated with chlorine, one that received raw water ( $\text{Cl}_2$ ) and one that received water pre-treated with UV (UV +  $\text{Cl}_2$ ). UV-treated water was dosed at  $40 \text{ mJ/cm}^2$  by a flow-through UV lamp (Trojan UVMax®, Trojan Technologies, London, ON). Concentrated free chlorine was produced using a 6% solution of analytical grade sodium hypochlorite ( $\text{NaOCl}$ ). The target free chlorine residual was 0.20 mg/L.

Sterile nutrient cocktail solutions were prepared as described by Dykstra *et al.* (2007) and pumped into each AR to achieve a total organic carbon (TOC) concentration of 2.0 mg/L and a C:N:P molar ratio of 100:20:5 in order to promote microbial growth. ARs receiving UV-treated feed water also received biodegradable organic matter cocktails that were pre-treated with UV light at a dose of  $40 \text{ mJ/cm}^2$ .

### Analytical methods

Sampling and testing protocols were carried out as described in the *Standard Methods for the Examination of Water and Wastewater*, 21st edn (American Public Health Association (APHA) *et al.* 2005). Free chlorine was measured three times per week using the DPD colorimetric method and a HACH2800 spectrophotometer. TOC, turbidity, iron, hardness and alkalinity were measured bi-weekly, while microbiological parameters were sampled weekly.

Coupons were removed from each AR in sequence to analyse for biofilm bacteria and prepared for plating as outlined by Rand *et al.* (2007). Suspended bacteria samples were taken from effluent water collected in sterile 50-mL test tubes containing 0.1% w/v sodium thiosulfate. All bacteria samples were enumerated with a standard spread-plate technique for HPCs on R2A agar (Difco Laboratories).

### Data analysis

Statistical procedures followed an analysis of variance (ANOVA) test, as described by Box *et al.* (1978), with a level of significance of  $\alpha = 0.05$ . All  $\pm$  values reported are one standard deviation from the mean. Error bars included in figures represent a 95% confidence interval.

## RESULTS AND DISCUSSION

### Acclimation/pre-disinfection period

In both studies, ARs were operated for a period of 4 weeks prior to the application of chlorine in order to establish pseudo-steady state biofilm on coupons. This is a common acclimation period observed by previous AR studies (Gagnon *et al.* 2005; Dykstra *et al.* 2007; Rand *et al.* 2007; Murphy *et al.* 2008). Steady state conditions over this biofilm acclimation period were determined through weekly HPC sampling.

All ARs were compared during acclimation for average HPC bacteria in bulk and biofilm phases and were found to be statistically similar except in two cases. In hard water, a significant difference existed between cast iron and polycarbonate ARs for bulk HPC ( $p = 0.015$ ), and for cast iron ARs, the average

bulk water HPC bacteria were found to be 1-log higher in softened water than hard water resulting in a statistically significant difference ( $p = 0.005$ ). HPC bacteria were also enumerated in influent streams throughout the experiment resulting in the following averages: hard water UV:  $1.80 \times 10^1$  CFU/mL; hard water RW:  $2.31 \times 10^2$  CFU/mL; softened water UV:  $3.63 \times 10^2$  CFU/mL; softened water RW:  $5.21 \times 10^2$  CFU/mL.

### Chlorine data

During the 12-week period following acclimation in each study, two cast iron and two polycarbonate ARs ( $\text{Cl}_2$  and UV +  $\text{Cl}_2$ ) were dosed with chlorine at a rate designed to yield 0.20 mg/L free chlorine residual in the effluents. A summary of free chlorine doses and residuals is presented in Table 1. As can be seen, UV pre-treatment did not necessarily result in lowered free chlorine dose to achieve the goal residual. In both studies, maintaining consistent free chlorine residual in cast iron ARs was difficult compared to polycarbonate ARs, and dosages were significantly higher in cast iron ARs. The coupons used in these ARs were corroded from previous use in experiments, which would add to the chlorine demand. This is consistent with several studies that have previously established cast iron exerts a demand on free chlorine (Frateur *et al.* 1999; Hallam *et al.* 2002; Mutoti *et al.* 2007).

### Post-disinfection HPC data

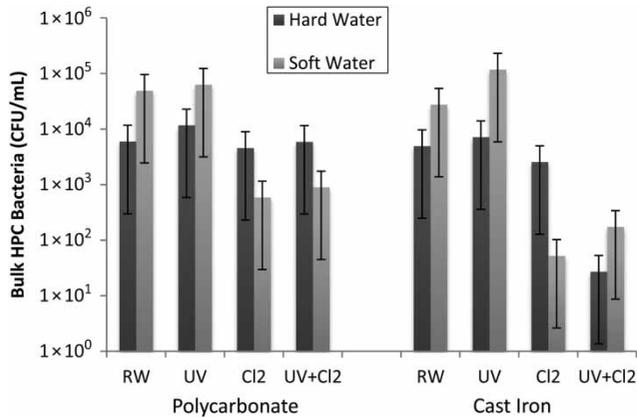
#### Comparison of hard and soft water on disinfection efficacy

The potential impact of water hardness on distribution system HPC bacteria in bulk and biofilm phases was investigated with UV,  $\text{Cl}_2$  and UV +  $\text{Cl}_2$  disinfection. Figure 1

**Table 1** | Free chlorine dosages and residuals during 12-week disinfection stage<sup>a</sup>

Experiment	Free $\text{Cl}_2$ (mg/L)	Polycarbonate ARs		Cast iron ARs	
		UV + $\text{Cl}_2$	$\text{Cl}_2$	UV + $\text{Cl}_2$	$\text{Cl}_2$
Hard water	Dose	$0.70 \pm 0.08$	$1.00 \pm 0.10$	$6.86 \pm 0.61$	$5.35 \pm 1.71$
	Residual	$0.20 \pm 0.05$	$0.20 \pm 0.05$	$0.24 \pm 0.15$	$0.20 \pm 0.20$
Softened water	Dose	$0.82 \pm 0.32$	$0.54 \pm 0.08$	$4.21 \pm 0.45$	$5.43 \pm 0.60$
	Residual	$0.19 \pm 0.10$	$0.22 \pm 0.08$	$0.23 \pm 0.15$	$0.22 \pm 0.17$

<sup>a</sup>Results demonstrate average chlorine concentration  $\pm 1$  standard deviation.

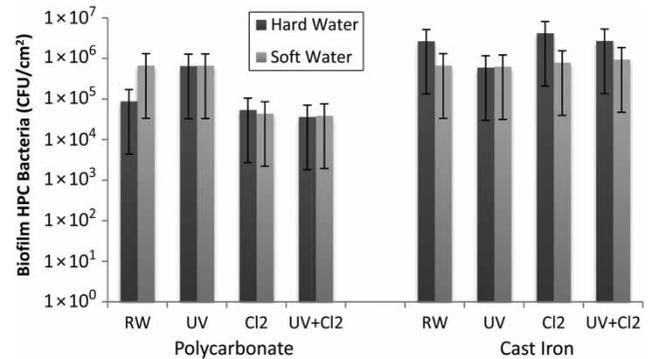


**Figure 1** | Average bulk heterotrophic bacteria in hard and softened water.

presents the bulk heterotrophic bacteria in the softened and hard water systems containing polycarbonate and cast iron coupons during the disinfection phase.

The results show that bulk HPC bacteria in polycarbonate ARs were more resistant to both Cl<sub>2</sub> and UV + Cl<sub>2</sub> disinfection in hard water than in softened water. There was a significant difference between hard and softened water bulk bacteria for both Cl<sub>2</sub> ( $p = 0.021$ ) and UV + Cl<sub>2</sub> ( $p = 0.03$ ) disinfection in polycarbonate ARs. Similarly, in cast iron ARs with Cl<sub>2</sub> disinfection, there were significantly more bulk bacteria in hard water than softened water ( $p = 0.00097$ ). In contrast, there were more bulk bacteria in softened water than hard water with UV + Cl<sub>2</sub> disinfection in cast iron ARs, although there was no statistically significant difference observed ( $p = 0.06$ ). Positive correlations have been found between turbidity and HPC bacteria (Power & Nagy 1999; Lehtola *et al.* 2007). Turbidity decreased in softened water but increased in hard water post-chlorination in cast iron and polycarbonate systems. This increase could relate to resistance of HPC bacteria in hard water systems through harbouring of bacteria or increased demand on chlorine.

Figure 2 presents the biofilm heterotrophic bacteria data and shows there was no significant difference found between biofilm bacteria in hard and softened water in both Cl<sub>2</sub> ( $p = 0.23$ ) and UV + Cl<sub>2</sub> ( $p = 0.3$ ) disinfection trials for polycarbonate coupons. However, there were statistically significant differences between biofilm bacteria levels in hard and softened water in both Cl<sub>2</sub> ( $p = 0.008$ ) and UV + Cl<sub>2</sub> ( $p = 0.0002$ ) disinfection trials for cast iron coupons, with hard water showing higher counts.

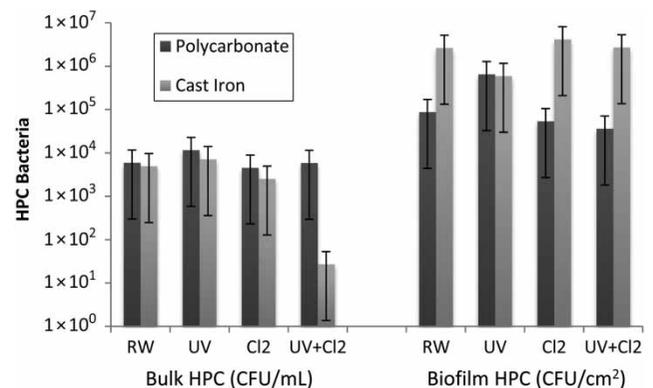


**Figure 2** | Average biofilm heterotrophic bacteria in hard and softened water.

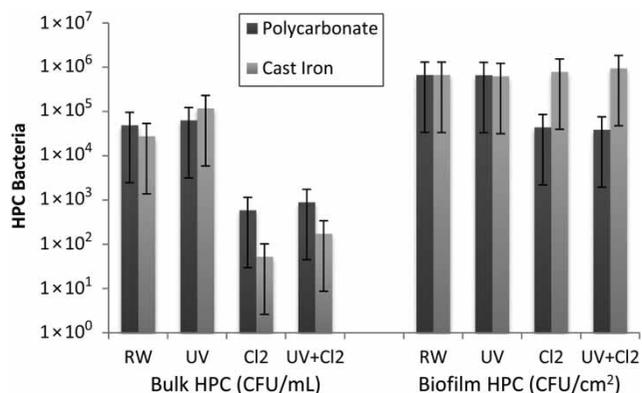
### Comparison of pipe material on disinfection efficacy

Figure 3 shows average bulk and biofilm heterotrophic bacteria collected in ARs containing polycarbonate and cast iron coupons in the hard water study. Bulk bacteria in polycarbonate ARs were more resistant to UV + Cl<sub>2</sub> disinfection than in cast iron, with a highly significant difference observed ( $p = 0$ ). In contrast, there was no significant difference between cast iron and polycarbonate ARs for bulk bacteria with Cl<sub>2</sub> disinfection ( $p = 0.29$ ). The opposite trend was observed for biofilm bacteria, where disinfection schemes performed better in polycarbonate ARs than in cast iron, with a highly significant difference observed with Cl<sub>2</sub> ( $p = 0$ ) and UV + Cl<sub>2</sub> ( $p = 0$ ) disinfection. It is important to note that neither disinfection scheme was found to be effective in removing HPC biofilm bacteria from cast iron ARs.

Figure 4 shows the average bulk and biofilm heterotrophic bacteria collected in the softened water study. Bulk



**Figure 3** | Average bulk and biofilm HPC bacteria in polycarbonate and cast iron ARs (hard water system).



**Figure 4** | Average bulk and biofilm HPC bacteria in polycarbonate and cast iron ARs (softened water system).

bacteria in polycarbonate ARs were much more resistant to Cl<sub>2</sub> and UV + Cl<sub>2</sub> disinfection than in cast iron, similar to the hard water experiment, with a highly significant difference observed with Cl<sub>2</sub> ( $p=0$ ) and UV + Cl<sub>2</sub> ( $p=0.001$ ) disinfection. Again, similar to the hard water study, the trends were opposite for attached bacteria, where disinfection was more effective at removing biofilm bacteria in ARs containing polycarbonate coupons. There was a highly significant difference between cast iron and polycarbonate coupons in terms of biofilm bacteria in Cl<sub>2</sub> ( $p=0$ ) and UV + Cl<sub>2</sub> ( $p=0$ ) disinfection.

Previous studies have indicated corroding iron surfaces support higher HPC bacteria growth than plastic surfaces (Camper *et al.* 2003), although similar studies have also shown that disinfectant residual is a more important factor in reducing attached HPC bacteria than pipe material (Ollos *et al.* 2003; Murphy *et al.* 2008). In the current

study, the inability of chlorine to reduce biofilm bacteria in the cast iron ARs may be caused by the promotion of biofilm growth due to the iron surface, and/or the demand on chlorine at iron surfaces.

### Disinfection synergy calculations

Koivunen & Heinonen-Tanski (2005) presented a model to calculate disinfection synergy for pathogen removal (Equation (1)).

*Equation (1): Synergy* (Koivunen & Heinonen-Tanski 2005)

#### Synergy (log units)

$$= \log \text{ reduction by combined chemical/UV disinfection} \\ - (\log \text{ reduction by UV disinfection} + \log \text{ reduction by} \\ \times \text{ chemical disinfection}) \quad (1)$$

This equation was applied to the collected data to evaluate synergy between UV light and Cl<sub>2</sub> when combined as a treatment (UV + Cl<sub>2</sub>) for the removal of HPC bacteria (Table 2). Log reductions were calculated using average bulk and biofilm HPC data from the acclimation and disinfection phases in each AR.

The UV ARs acted as controls and did not have a change in disinfection throughout the experiment. However, slight reductions in average bulk bacteria was observed between acclimation and disinfection stages in both UV and RW control ARs, which was most likely due to changes in bacteria levels in source water. For both water sources and pipe

**Table 2** | Bulk/biofilm HPC log reductions and synergy calculations

Water source	Disinfection type	Bulk bacteria		Biofilm bacteria	
		Polycarbonate	Cast iron	Polycarbonate	Cast iron
Hard water	UV	0.87	0.87	0.56	0.35
	Cl <sub>2</sub>	1.30	1.17	1.32	-0.076
	UV + Cl <sub>2</sub>	2.00	3.74	1.68	-0.24
	Synergy	-0.17	+1.7	-0.2	-0.51
Softened water	UV	0.43	0.23	-0.14	0.43
	Cl <sub>2</sub>	2.65	3.65	1.24	-0.043
	UV + Cl <sub>2</sub>	2.22	3.72	1.38	0.30
	Synergy	-0.86	-0.16	+0.28	-0.09

materials, disinfection with chlorine or UV + Cl<sub>2</sub> resulted in higher reductions of bulk HPCs than UV alone. For biofilm HPCs, Cl<sub>2</sub> and UV + Cl<sub>2</sub> disinfection in polycarbonate systems was also more effective compared to the control UV disinfection. However, disinfection with chlorine or combined UV + Cl<sub>2</sub> was not effective in cast iron ARs for the removal of attached bacteria.

In hard water, the combined UV + Cl<sub>2</sub> treatment was found to be more effective in the cast iron ARs than the polycarbonate ARs for deactivating bulk HPC bacteria. Log removal of bulk HPC bacteria in the polycarbonate ARs during the UV + Cl<sub>2</sub> disinfection was not significantly higher ( $p = 0.5$ ) than that achieved with Cl<sub>2</sub> disinfection alone. However, in the cast iron ARs, log removal of bulk HPC bacteria with UV + Cl<sub>2</sub> disinfection was found to be significantly higher ( $p = 0.00025$ ) than Cl<sub>2</sub> disinfection. Log removal of biofilm HPC bacteria in the polycarbonate ARs with UV + Cl<sub>2</sub> disinfection was not found to be significantly higher ( $p = 0.82$ ) than log removal of biofilm HPC with Cl<sub>2</sub> disinfection. In the cast iron ARs, no log removal of biofilm HPCs was observed with either Cl<sub>2</sub> or UV + Cl<sub>2</sub> disinfection.

Similar to hard water, UV + Cl<sub>2</sub> treatment was found to be more effective in the cast iron ARs than the polycarbonate ARs for deactivating bulk HPC bacteria in softened water. However, log removal of bulk HPC bacteria with combined UV + Cl<sub>2</sub> disinfection was similar to that achieved with Cl<sub>2</sub> disinfection alone in both polycarbonate ( $p = 0.109$ ) and cast iron ( $p = 0.38$ ) ARs. Log removal of biofilm HPC bacteria with UV + Cl<sub>2</sub> disinfection was not found to be significantly different from Cl<sub>2</sub> disinfection in polycarbonate ( $p = 0.5$ ) or cast iron ( $p = 0.865$ ) ARs.

Although the combined UV + Cl<sub>2</sub> treatment resulted in higher log reductions for both bulk and biofilm bacteria compared to treatment with Cl<sub>2</sub> alone (except for polycarbonate bulk bacteria in softened water), positive synergistic benefits were only observed in two cases (bulk bacteria in hard water and biofilm bacteria in softened water, both in polycarbonate systems). Previous findings have varied in terms of observing synergy between UV light and chlorine. Wang *et al.* (2012) found synergistic benefits between UV light and free chlorine for removal of pathogenic microorganisms in reclaimed water, where the disinfectants acting alone (UV and free Cl<sub>2</sub>) were not effective against these pathogens. Cho *et al.* (2011) found that no synergy was observed between UV pre-treatment

and free chlorine for the inactivation of MS-2 bacteriophage and *Bacillus subtilis* spores unless hydrogen peroxide was added in the primary disinfection step. Dykstra *et al.* (2007) showed synergistic benefits between UV and Cl<sub>2</sub> at low dosages for removal of bulk and biofilm HPC bacteria in polycarbonate ARs; however, that study used municipal surface water for source water and had a goal residual free chlorine concentration of 0.5 mg/L. In that same study, synergistic benefits were not observed when the residual chemical concentration increased to 1.0 mg/L. Another surface water study by Rand *et al.* (2007) showed increased removal of bulk and biofilm bacteria when UV light preceded chlorine disinfection, but there was no statistically significant difference between the combined treatment and treatment with chlorine alone. Results from this current study indicate that although there is potential for improved reduction of bulk water HPC bacteria in groundwater with a combined UV + Cl<sub>2</sub> treatment, synergistic benefits are not observed consistently. This may suggest that it is selective disinfection rather than disinfection synergy that leads to improved removal of HPC bacteria with sequential disinfection. Results suggest groundwater sources are less likely to consistently show enhanced reductions with combined treatments compared to surface water sources, or that the addition of a catalyst may be required to achieve consistent synergy between UV light and free chlorine regardless of water source.

## CONCLUSIONS

It was found in this study that synergistic benefits with combined UV light and free chlorine disinfection for the control of HPC bacteria in drinking water distribution systems was not observed in either hard or softened groundwater. Results showed that greater removal was more often achieved with integrated disinfection of UV + Cl<sub>2</sub> compared to disinfectants acting alone, but positive synergistic calculations were not consistently observed, indicating that increased removal may simply be due to selective disinfection.

It was found in general that suspended HPC bacteria were more resistant to disinfection in hard water than in softened water, and also in polycarbonate ARs compared to cast iron ARs. Biofilm bacteria showed similar response to the disinfectants evaluated in hard and softened water

when the coupons were polycarbonate. In the cast iron ARs, no disinfectant scheme was able to significantly reduce biofilm growth on the coupons, indicating UV light and/or free chlorine with a residual concentration of 0.20 mg/L is ineffective at controlling microbial regrowth in cast iron distribution systems. Cast iron ARs required significantly higher doses of chlorine than polycarbonate ARs to achieve the goal residual concentration. Required dosages to achieve the goal residual for chlorine were not always reduced with UV pre-treatment and total trihalomethane formation remained similar regardless of the disinfection scheme.

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