

## Changes in microbiological quality in model distribution systems after switching from chlorine or chloramines to chlorine dioxide

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### ABSTRACT

North American drinking water utilities are currently re-evaluating their disinfection strategies for controlling microbiological growth in distribution systems. Most water systems in North America use free chlorine as a secondary disinfectant. Since chlorine is known to form potentially carcinogenic byproducts in drinking water, utilities are looking for an alternative to maintain a disinfectant residual in the distribution system. The objective of this study was to evaluate the response of model drinking water distribution systems to a change in disinfectant from either free chlorine or chloramines to chlorine dioxide, in terms of its impact on microbiological water quality (bulk water and biofilm). Switching from a chlorine residual of 0.5 mg/L to a chlorine dioxide residual of 0.25 mg/L did not impact (negatively or positively) microbial water quality as quantified by heterotrophic and total cell counts (sample size = 8 data points). Thus, on the basis of the mass of disinfectant applied, chlorine dioxide was more efficient than free chlorine at controlling microbiological growth in the model distribution system. Similarly, chlorine dioxide was more efficient than chloramines, as a chlorine dioxide residual of 0.25 mg/L inactivated 0.75–1 log more suspended organisms than 1.0 mg/L residual of chloramines. Therefore, under the tested conditions, chlorine showed similar or better disinfection efficiency than free chlorine and chloramines, respectively.

**Key words** | chloramines, chlorine, chlorine dioxide, disinfection, distribution systems, drinking water

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### INTRODUCTION

The water industry is actively pursuing new disinfection strategies for controlling microbial growth in distribution systems while mitigating the formation of disinfectant byproducts (DBPs). Biofilm control is becoming recognized as an important aspect of the operation of drinking water plants and distribution systems. Bacterial regrowth and coliform occurrence are dependent upon a complex interaction of drinking water characteristics, engineering and operational parameters (LeChevallier 1990; Geldreich 1996; LeChevallier *et al.* 1996). Free chlorine has been

applied to drinking water since the early 1900s. However, it is well established that free chlorine can react with natural organic matter to form harmful halogenated DBPs (Rooks 1977; Trussell 1999; van der Kooij *et al.* 1999). Consequently, the water industry has actively sought alternative disinfection strategies.

Under certain conditions, monochloramine can be a better biocide for controlling biofilm growth in distribution systems (Norton & LeChevallier 1997). However, monochloramine is considered a weaker disinfectant than free

chlorine because it is less effective against viruses and protozoa (van der Kooij *et al.* 1999). Chloramination has several benefits including lower trihalomethane (THM) formation, improved taste and odour, and it provides a more persistent disinfectant residual throughout the distribution system than free chlorine (Norton & LeChevallier 1997; Trussell 1999). Nevertheless, chloramination may also lead to nitrification in the distribution systems (Wilczak *et al.* 1996; Regan *et al.* 2002). This can cause a decrease in chloramines residual, increase heterotrophic bacteria and also increase nitrite concentrations (Norton & LeChevallier 1997).

Chlorine dioxide is a powerful oxidizing agent and has been used for the control of iron, manganese, and taste and odor causing compounds (White 1999). Because chlorine dioxide has different reaction pathways involving natural organic matter than free chlorine, the formation of organohalogens (e.g. trihalomethanes and haloacetic acids) is typically much lower in concentration than when using free chlorine (Werdehoff & Singer 1987; Hofmann *et al.* 1999). However, chlorine dioxide generation is known to form chlorite and chlorate which are potentially hazardous disinfection byproducts (Limoni & Teltsch 1985; Gordon *et al.* 1990). When applied to drinking water, a portion of the  $\text{ClO}_2$  will form  $\text{ClO}_2^-$  upon reaction with natural organic matter (NOM). Widely varying concentrations of  $\text{ClO}_2^-$  formed from  $\text{ClO}_2$  have been observed and reported in the literature, with proportions ranging between 30–70% (Gordon 1992; Baribeau *et al.* 2002). For systems using chloramines for secondary disinfection, there has been great interest in the use of chlorite, as McGuire *et al.* (2006) has found that nitrification can be acutely mitigated by supplementing chloramination with chlorite.

With the promulgation of the Stage 1 Disinfectant/Disinfection By-Product Rule (D/DBPR) that requires lower maximum contaminant levels (MCLs) for TTHMs (USEPA 1998), a significant number of utilities are converting their systems from free chlorine to combined chlorine. Several examples exist in Florida, North Carolina, and the San Francisco Bay area (AWWA Research Foundation 2004). Some utilities also converted to chloramines to improve the taste and odour of treated water (Wilczak *et al.* 1996). Chlorine dioxide has been used as a secondary disinfectant in several European countries including Italy,

Germany, France and Switzerland (Dernat *et al.* 1995). On the other hand, its application in North America for disinfection purposes has been relatively limited. The primary objective of this study was to evaluate the effect of switching disinfectants on bacterial growth under controlled lab conditions. The research was conducted at bench scale using annular reactors to simulate drinking water distribution systems. The study investigated (i) the short-term impacts of switching from free chlorine or chloramines to chlorine dioxide on bacterial water quality (biofilm and suspended bacteria), and (ii) the impact of two pipe materials (i.e. polycarbonate and cast iron) on disinfection efficiency.

## MATERIALS AND METHODS

### Description of bench-scale system

Annular reactors (AR) (BioSurface Technologies Corp.) were used as model distribution systems. The annular reactor is comprised of a stationary outer glass cylinder that encompasses an inner rotating drum. The inner drum is controlled by a variable DC motor and can rotate between speeds of 25–430 rpm. The rotational speed of the inner drum is what simulates the shear stress encountered on a pipe wall in a distribution system. The ARs were operated at 50 rpm which translates to a shear stress of  $0.25 \text{ N/m}^2$  on the inner pipe wall. A shear stress of  $0.25 \text{ N/m}^2$  corresponds to a flow of approximately 0.30 m/s (1 fps) in a 100 mm (4 in) diameter smooth pipe which is similar to the shear conditions of other pilot and bench-scale investigations of drinking water distribution systems (e.g. Camper 1996; Ollos *et al.* 2003). Twenty removable cast-iron and polycarbonate coupons located at the surface of the inner cylinder allowed the evaluation of biofilm bacteria. Water retention times of 60 min were used in all experiments.

### Feed water

Tap water supplied by the Halifax Regional Water Commission was used as the primary process water. Surface water

from Pockwock Lake is conventionally treated at the J.D. Kline water treatment facility and is distributed with a free chlorine residual of 0.6 mg/L to the city of Halifax. On average, the treated water has an alkalinity of 15 mg/L as CaCO<sub>3</sub>, pH of 7.4 and total organic carbon (TOC) concentration of approximately 1.5 mg/L. Prior to entering an AR the tap water was passed through a filter containing fresh granular activated carbon (GAC) media to neutralize free chlorine present in the tap water. A second GAC filter operating biologically removed any biodegradable organic matter. Sterile influent cocktails of organic carbon were pumped into each AR to provide a constant source of organic nutrients. To ensure a carbon limiting system, ARs were also dosed with a separate cocktail of nitrogen (NaNO<sub>3</sub>) and phosphorus (K<sub>2</sub>HPO<sub>4</sub> and KH<sub>2</sub>PO<sub>4</sub>). The cocktail solution of biodegradable organic material (BOM) was composed of ethyl alcohol, propionaldehyde, oxalate, pyruvate and acetate to an organic carbon concentration of 100 µC/L. These compounds were selected because they represent major classes of organic compounds found in drinking water and they are relatively non-reactive with disinfectants (Camper 1996). The cocktails were pumped into the ARs to obtain a molar C:N:P ratio of 100:20:5 (Camper 1996). The influent mixture of biodegradable organic carbon, nitrogen and phosphate remained constant throughout the experimental campaign. Typically the influent dissolved organic carbon (DOC) concentration entering the ARs (i.e. feed water from GAC/BAC treatment and blended with BOM cocktail) ranged from 1.2–1.5 mg/L.

### Experimental setup

Eight reactors were used in parallel, 4 with polycarbonate coupons and 4 with cast-iron coupons (Figure 1). For the first three to four weeks of the study, all reactors were fed with water containing no disinfectant to identically colonize all reactors. After a pseudo-steady-state biofilm was established, either free chlorine or monochloramine was applied to both AR trains (Figure 1). The effluent residual concentration for free chlorine and monochloramine was 0.5 and 1.0 mg/L, respectively, representing typical values in full-scale distribution systems. Following an additional 12

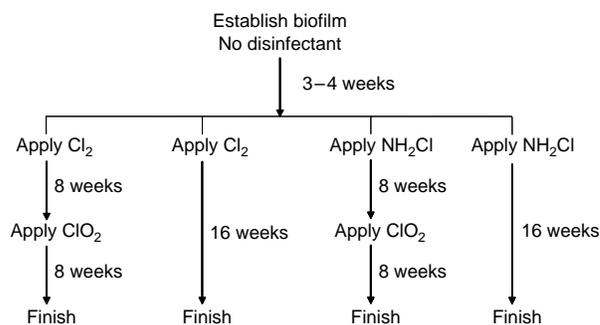


Figure 1 | Overview of experimental design for switching disinfectants.

weeks, chlorine dioxide was applied to four (test reactors) of the eight ARs at a concentration of 0.25 mg/L. The remaining four ARs continued to receive either free chlorine or monochloramine (control reactors). Disinfectants were prepared in sterile 4 litre amber bottles at various stock concentrations. The bottles were fed into the ARs at a prescribed flowrate in order to achieve the desired residuals. Disinfectant residuals were monitored daily.

### Disinfectant generation

Chlorine dioxide was produced according to the *Standard Methods for the Examination of Water and Wastewater* (APHA, AWWA & WEF 1998). The method uses sodium chlorite addition to strong acid to generate chlorine dioxide. A gas-washing bottle contained in an ice bath is used to collect the chlorine dioxide. This system generates a stock solution of approximately 2.0 g/L and is essentially free of all other chlorine species other than chlorine dioxide. The concentration of the ClO<sub>2</sub> stock was relatively constant over time. Changes in concentration were negligible as concentrated stock was kept refrigerated in an amber bottle. In addition, ClO<sub>2</sub> feed-bottles for the ARs were prepared weekly in amber bottles to prevent degradation due to light exposure. Free chlorine stock was prepared by diluting reagent grade sodium hypochlorite in phosphate buffered saline (pH 7.4). Monochloramine was prepared by combining equivalent amounts (molar basis) of ammonium chloride and sodium hypochlorite in phosphate buffered saline (pH 9.5) and diluting to achieve a final pH to 7.4. Monochloramine and free chlorine disinfectant residuals were measured using the DPD ferrous titrimetric method

(APHA, AWWA & WEF 1998). Chlorine dioxide was measured using the Lissamine Green B spectrophotometric method (Chiswell & O'Halloran 1991). Disinfectant residuals were monitored daily and pH was monitored weekly to ensure that the addition of disinfectant had negligible effects on pH with the reactors.

### Measured water quality parameters

Sampling and testing protocols were as described in the *Standard Methods* (APHA, AWWA & WEF 1998). Sampling was performed on a weekly basis. Chemical water quality parameters monitored throughout the experiments were Total Organic Carbon (TOC), Dissolved Organic Carbon (DOC), pH and temperature. Temperature was maintained at 20°C while the pH was on average 7.4. Heterotrophic plate counts (HPC) were performed using a spread plate technique on R2A agar (BD-Difco, Sparks, MD). R2A plates were incubated at 20°C and the colonies were counted after 7 days. Acridine orange direct counts (AODC) were used to obtain total microbial cell counts. One mL of sample was mixed with 1 mL of a 0.1% (w/v) solution of acridine orange for five minutes. The mixture was filtered on black cellulose nitrate filters (Millipore, Bedford, MA). The filters were observed by epifluorescence microscopy (Olympus model BX-60, Melville, NY), and enumerated using an image analysis system (Esprit<sup>™</sup>, Olympus). Fluorescent (orange to green) bacterial cells were counted in 25 different randomly chosen fields. The counting was automated using an image analysis system and a digital CCD camera.

Removable coupons were sampled to evaluate biofilm. Polycarbonate and cast-iron coupons were aseptically transferred into 150 mL sterile test tubes containing 25 mL phosphate buffered saline (PBS) and 25 µL of 10% (w/v) sterile sodium thiosulfate. Cells attached to polycarbonate coupons were immediately removed with a stomacher as described by Gagnon & Slawson (1999). Biofilm was removed from the polycarbonate coupon surface using a MIX 2 Stomacher (AES Laboratories). Attached cells were removed from the cast-iron coupons by manually scraping the coupon surface into a sterile stomacher bag (177 mm × 302 mm). The 50 mL of PBS solution was used to rinse the remaining biofilm off the coupon and into the

stomacher bag. Cast-iron biofilm samples were homogenized (Brinkmann homogenizer PT 1200C, Switzerland). Typically, more than 95% of the heterotrophic bacteria were recovered from the polycarbonate coupon while the recovery from cast-iron coupons was less (80–90%). Viable cells were enumerated by plating on R2A agar and total microbial cell counts were determined by epifluorescence. AODC could not be performed for the cast-iron coupons because the corrosion related particulate material would fluoresce along with the bacteria. The log inactivation is derived from the logarithm of the ratio of the bacterial levels after and before disinfection ( $\log \text{inactivation} = -\log C/C_0$ ). Because most of the data were not normally distributed, geometric means were calculated. Non-parametric tests were performed to determine whether the two data sets were statistically different (comparison of medians). The level of significance ( $\alpha$ ) for these tests was 5%.

## RESULTS AND DISCUSSION

### Acclimation of annular reactors

The reactor surface became quickly colonized when supplied with dechlorinated tap water. A steady-state biofilm, as defined by having a consistent number of HPC bacteria with respect to time, was established within three–four weeks for both cast-iron and polycarbonate coupons, which is consistent with previous biofilm studies (e.g. Volk & LeChevallier 1999; Gagnon *et al.* 2004). Ollos *et al.* (2003) showed in bench-scale studies using annular reactors that pseudo-steady-state biofilm HPC numbers were typically established within 10 d after starting an experiment. It has been reported that, depending on the conditions, biofilm levels at steady state in pilot or full-scale distribution systems generally range between  $10^5$ – $10^7$  bacteria/cm<sup>2</sup> (e.g. Block *et al.* 1993; Volk & LeChevallier 1999). In this study, biofilm HPCs were monitored weekly and once they were constant for a period of 10 d it was assumed that pseudo-steady-state had been reached. Biofilm HPC levels at steady state were  $10^7$  CFU/cm<sup>2</sup> on polycarbonate and cast-iron coupons (Figures 5 and 6). Fixed bacteria AODC levels ranged from  $10^6$ – $10^7$  cells/cm<sup>2</sup> on polycarbonate coupons (no statistical differences were observed between paired

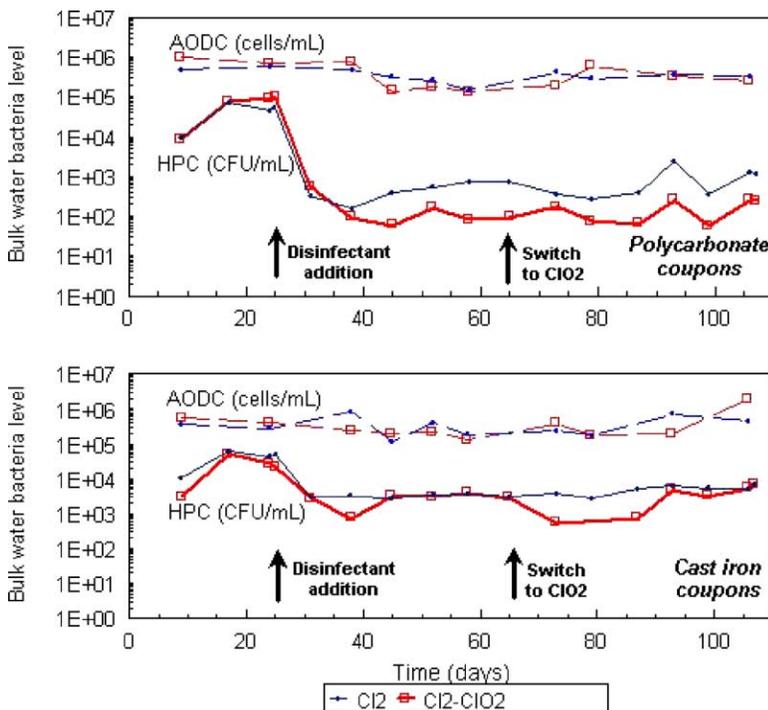
annular reactors during the colonization period,  $P > 0.3$ ). Under acclimation conditions, biofilm HPC densities on cast-iron coupons (Figure 5) were approximately 0.5-log greater than the number of HPCs on the polycarbonate coupons (Figure 6). The difference in HPC bacteria on the two surfaces was statistically significant ( $P = 0.001$ ). Iron pipe surfaces have been shown to stimulate bacterial growth (LeChevallier *et al.* 1993, 1998; Gagnon *et al.* 2004). Camper (1996) found that more heterotrophic bacteria grew on mild steel surfaces than on polycarbonate: mild steel surfaces contained 10 times more heterotrophic bacteria and 2- to 10-fold more coliform bacteria. Suspended heterotrophic bacteria concentrations were between  $10^4$ – $10^5$  CFU/mL (Figures 3 and 4), while suspended bacteria AODC levels were in the  $10^5$  cells/mL range for both cast-iron and polycarbonate coupons (Figure 2).

### Disinfectant implementation

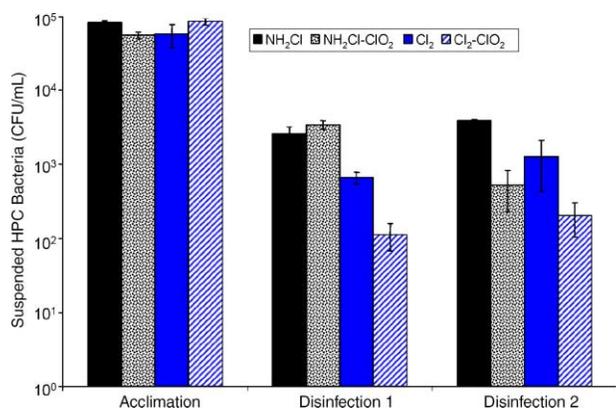
Following the initial colonization period, the reactors received either free chlorine or chloramines as an oxidant.

The effect of disinfectants on heterotrophic bacteria was clearly quantifiable (Figures 2–6). An initial microbial inactivation period was quickly observed after the application of disinfectant. This period lasted about three weeks at which point the bacteria levels stabilized to a new steady state. For suspended HPC bacteria, under disinfection conditions the number of heterotrophic bacteria ranged from  $10^2$ – $10^3$  CFU/mL for polycarbonate coupons and  $10^3$ – $10^4$  CFU/mL for cast-iron coupons (Figures 3 and 4). As shown in Figure 2, the number of suspended HPCs varied to a certain degree during disinfection. This variability was largely in response to changing influent water quality conditions. Accordingly the system steady state was defined by the number of biofilm bacteria, which were more consistent between sampling points. For biofilm HPC bacteria, the disinfection steady-state values were  $10^4$ – $10^5$  CFU/cm<sup>2</sup> and  $10^6$  CFU/cm<sup>2</sup> for polycarbonate and cast-iron coupons, respectively.

A significant decrease in bulk water HPC levels was observed after the application of the first disinfectant ( $P < 0.001$ ) (Figures 3 and 4). The log reduction achieved with chlorine for suspended heterotrophic bacteria was 1.2

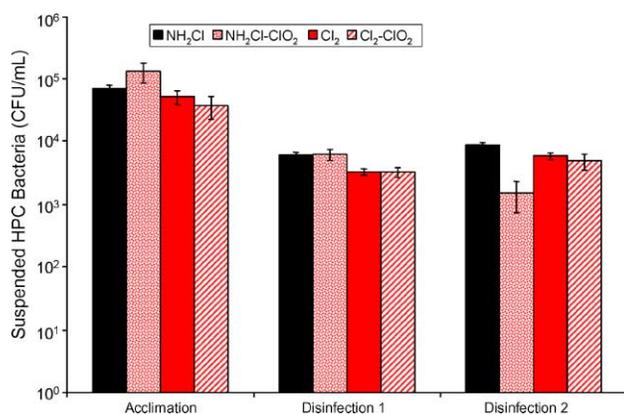


**Figure 2** | Example of the evolution of suspended AODC and heterotrophic bacteria levels in polycarbonate and cast-iron coupon reactors for chlorine to chlorine dioxide disinfectant switch.

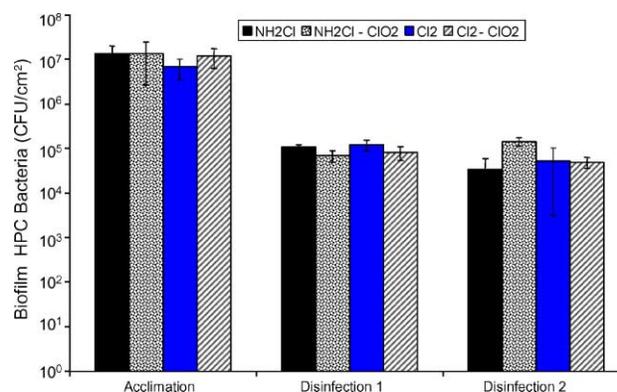


**Figure 3** | Effects of disinfectants ( $\text{NH}_2\text{Cl}-\text{ClO}_2$  and  $\text{Cl}_2-\text{ClO}_2$ ) on suspended heterotrophic bacteria levels in polycarbonate coupon reactors.

and between 2.1–2.7 for cast-iron and polycarbonate ARs, respectively (Table 1). The log reduction for biofilm heterotrophic bacteria under chlorine disinfection was 1.1 for cast-iron and between 1.5–1.9 for polycarbonate ARs (Table 1). Chlorine was more effective on suspended bacteria than on biofilm bacteria in both polycarbonate and cast-iron reactors. Adhesion has been described as a factor of protection of attached bacteria (Camper 1997). The partial penetration of biofilm by disinfectants can explain the reduced efficacy of such agents against biofilm bacteria. With chlorine, inactivation levels were also related to the pipe material with higher disinfection efficiency in polycarbonate coupon reactors. It has been shown that pipe material and conditions (such as corrosion, pitting and tuberculation) affect bacterial water



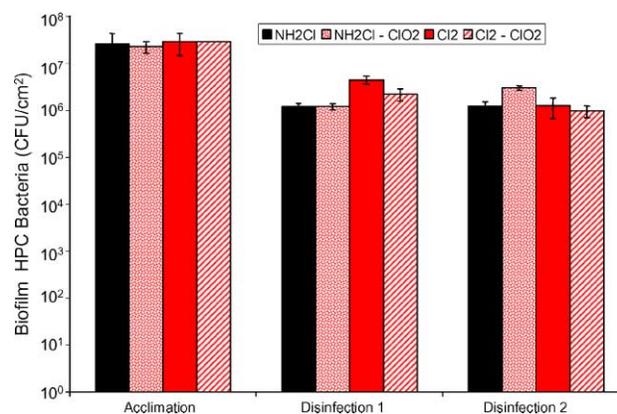
**Figure 4** | Effects of disinfectants ( $\text{NH}_2\text{Cl}-\text{ClO}_2$  and  $\text{Cl}_2-\text{ClO}_2$ ) on suspended heterotrophic bacteria levels in cast-iron coupon reactors.



**Figure 5** | Effects of disinfectants ( $\text{NH}_2\text{Cl}-\text{ClO}_2$  and  $\text{Cl}_2-\text{ClO}_2$ ) on biofilm heterotrophic bacteria levels in polycarbonate coupon reactors.

quality, and indirectly impact biofilm bacteria by affecting disinfection efficiency, especially with free chlorine. Rompre *et al.* (1997) reported that free chlorine produced a rapid decrease in biofilm density ( $>1$  log) in polycarbonate reactors, whereas chlorine did not affect the biofilm in grey iron reactors.

The trend was different with chloramines; the average log inactivation of suspended heterotrophic bacteria were similar in cast-iron and polycarbonate coupon reactors (removal of 1.3 log) (Figures 3 and 4, Table 1). The log inactivation for biofilm heterotrophic bacteria under chloramine disinfection was 1.2 for cast-iron and between 1.6–1.7 for polycarbonate ARs (Table 1). Consequently, chloramines provided similar log inactivation in both suspended and biofilm heterotrophic bacteria.



**Figure 6** | Effects of disinfectants ( $\text{NH}_2\text{Cl}-\text{ClO}_2$  and  $\text{Cl}_2-\text{ClO}_2$ ) on biofilm heterotrophic bacteria levels in cast-iron coupon reactors.

**Table 1** | Log removal of bulk water and biofilm bacteria (HPC and AODC) in annular reactors submitted to various disinfectant conditions

Coupon type	Experimental change	Suspended HPC Log removal	Biofilm HPC Log removal	Suspended AODC Log removal	Biofilm AODC Log removal
PC	Acclimation – NH <sub>2</sub> Cl	1.4	1.7	0.5	0.6
CI	Acclimation – NH <sub>2</sub> Cl	1.2	1.2	0.0	–
PC	Acclimation – NH <sub>2</sub> Cl	1.3	1.6	0.0	0.6
CI	Acclimation – NH <sub>2</sub> Cl	1.3	1.2	0.1	–
PC	NH <sub>2</sub> Cl–NH <sub>2</sub> Cl*	–0.2	0.3	–0.4	–0.2
CI	NH <sub>2</sub> Cl–NH <sub>2</sub> Cl*	–0.2	0.2	–0.3	–
PC	NH <sub>2</sub> Cl–ClO <sub>2</sub>	1.0	–0.2	–0.1	–0.1
CI	NH <sub>2</sub> Cl–ClO <sub>2</sub>	0.9	0.3	–0.1	–
PC	Acclimation – Cl <sub>2</sub>	2.1	1.5	0.3	0.1
CI	Acclimation – Cl <sub>2</sub>	1.2	1.1	0.1	–
PC	Acclimation – Cl <sub>2</sub>	2.7	1.9	0.6	0.7
CI	Acclimation – Cl <sub>2</sub>	1.2	1.1	0.4	–
PC	Cl <sub>2</sub> –Cl <sub>2</sub> *	–0.2	0.3	–0.1	0.1
CI	Cl <sub>2</sub> –Cl <sub>2</sub> *	–0.2	0.1	–0.1	–
PC	Cl <sub>2</sub> –ClO <sub>2</sub>	–0.2	0.2	–0.3	–0.2
CI	Cl <sub>2</sub> –ClO <sub>2</sub>	–0.2	0.2	–0.3	–

PC: polycarbonate, CI: cast iron.

\*For the control reactors, the log inactivation calculated between the first 12 week period and the second 12 week period was used to determine if unknown parameters could affect bacterial water quality and evaluate the impact of chlorine dioxide during the same period of time.

When comparing chlorine at a *Ct* of 30 mg/L min., and chloramines at a *Ct* of 60 mg/L min, chlorine outperformed chloramines for controlling bulk water HPCs in polycarbonate coupon pipes (2.1–2.7 log inactivation for chlorine vs. 1.3–1.4 log inactivation for chloramines,  $P < 0.02$ ) (Figure 3, Table 1), while both disinfectants lead to similar results in CI coupon reactors (1.2–1.3 log removal) (Figure 4, Table 1). Regarding biofilm removal, both disinfectants led to comparable biofilm inactivation on polycarbonate (1.5–1.9 log removal,  $P = 0.20$ ) and cast-iron coupons (1.1–1.2 log removal,  $P = 0.18$ ) (Table 1, Figures 5 and 6). There are conflicting results on the ability of disinfectants such as

chlorine or chloramines to limit bacteria proliferation (Camper *et al.* 1997). In general, results have been found to be system specific and showed the complexity of a biofilm response to an interrelated set of parameters. Pilot studies (LeChevallier *et al.* 1990) have shown that low levels of chlorine and chloramines (1 mg/L) could reduce heterotrophic bacteria levels more than a hundredfold (2 log) for biofilm grown on galvanized and plastic pipes. However, chlorine residuals of 3–4 mg/L were ineffective on iron pipes, while biofilm treated with 2 mg/L of chloramines exhibited a more than 3 log die-off. It has been suggested that monochloramine is better able to penetrate the biofilm, while free

chlorine reacts with various constituents (corrosion products) and is consumed prior to penetrating the biofilm (LeChevallier *et al.* 1990).

This study showed a marginal decrease in AODC levels after disinfection (Figure 2, Table 1). The log reduction of AODC bacteria was 0–0.6 and 0.1–0.7 for suspended ( $->0.05$ ) and fixed ( $P<0.05$ ) bacteria, respectively. The bulk water AODC levels were not affected by disinfection ( $P>0.3$ ) (Figure 2, Table 1). AODC measurement yields total cell counts (dead and living microorganisms), but it cannot be used alone to assess the effects of disinfection, unless there is complete cell destruction after treatment. Experiments on biofilms established on stainless steel surfaces treated with 2 mg/L of chloramines showed relatively little biofilm removal as evidenced by total cell direct counts, while plate counts indicated an average 1.3 log decrease (Huang *et al.* 1995). AODC data were used in conjunction with HPC levels to determine a HPC/AODC ratio. It showed that the proportion of cultivable bacteria decreased drastically following disinfection. In the absence of a disinfectant, the bulk water HPC/AODC ratio stabilized at 5.3% (2.0–10.1%), while it was 0.6% with chlorine and 1.2% with chloramine disinfection (Table 2).

### Switching disinfectant

One of the two paired reactors fed either with chlorine or chloramines was switched to chlorine dioxide (test reactor,  $Ct$  of 15 mg/L.min.), while the second reactor continued to receive the same disinfectant (control reactor fed with chlorine or chloramines). Overall, after the switch, no negative short-term effects (such as biofilm sloughing and high HPC levels) were observed (Figure 2). Switching from free chlorine to chlorine dioxide did not lead to changes in bulk water or biofilm heterotrophic bacteria levels, for either polycarbonate or cast-iron coupon pipes (Figures 3–6, Table 1) ( $P>0.05$ ). After switching from chlorine to chlorine dioxide, the log inactivation of suspended HPC bacteria in cast-iron and polycarbonate ARs was  $-0.2$  log while the biofilm HPC bacteria log removal was 0.2 for both pipe materials (Table 1). In the control chlorine reactor, suspended HPCs also achieved a  $-0.2$  log removal in both cast-iron and polycarbonate systems and biofilm HPC bacteria levels changed by 0.1–0.3 log (Table 1). However, chlorine dioxide led to approximately a 10-fold reduction in suspended bacteria levels in the chloraminated reactor (Table 1, Figures 3 and 4), while biofilm HPC counts were not significantly affected (0.2–0.3 log removal)

**Table 2** | HPC/AODC ratios (%) in annular reactors during all three experiment phases under various disinfectant conditions

Reactor ID	Effluent			Biofilm		
	Acclimation	Disinfection 1	Disinfection 2	Acclimation	Disinfection 1	Disinfection 2
PC NH <sub>2</sub> Cl control	14.16	1.80	0.57	51.08	0.37	0.46
CI NH <sub>2</sub> Cl control	19.08	1.77	1.00	–	–	–
PC NH <sub>2</sub> Cl–ClO <sub>2</sub>	8.82	2.04	0.15	153.24	0.92	1.07
CI NH <sub>2</sub> Cl–ClO <sub>2</sub>	32.18	1.71	0.52	–	–	–
PC Cl <sub>2</sub> control	10.85	0.22	0.36	65.91	0.53	3.87
CI Cl <sub>2</sub> control	16.18	0.89	1.52	–	–	–
PC Cl <sub>2</sub> –ClO <sub>2</sub>	21.01	0.04	0.02	65.91	0.53	3.87
CI Cl <sub>2</sub> –ClO <sub>2</sub>	8.00	1.70	0.76	–	–	–

(Figures 5 and 6). For suspended bacteria, the HPC/AODC ratio dropped from 0.41% (PC) and 1.65% (CI) with chloramines to 0.04% (PC) and 0.21% (CI) with chlorine dioxide (Table 2).

The effects of disinfectant switch on bacterial water quality are variable. Switching from free chlorine to chloramines in various full-scale distribution systems could lead to positive or negative impacts on water quality. The disinfectant switch showed a decrease in coliform bacteria, HPC and disinfection byproduct levels in a medium size distribution system (Muncie, IN) while it led to an increase in coliform levels in a system in Washington, DC (in addition to an increase in lead levels) and Florida (AWWA Research Foundation 2004). Although chlorine dioxide is known to be a strong disinfectant, relatively few studies have examined its ability to control bacterial regrowth in distribution systems. Limoni & Teltsch (1985) reported low suspended heterotrophic plate counts during the distribution of drinking water that had finished with a chlorine dioxide residual concentration of approximately 0.2 mg/L. A Canadian system (Laval distribution system), which had used  $\text{ClO}_2$  disinfection since 1984, did not experience bacteriological problems (Lafrance *et al.* 1992). From 1987 to 1991, HPC levels (on R2A, 20°C, 7 d incubation) were less than 10 CFU/100 mL. Similarly, Volk *et al.* (2002) evaluated the response of a full-scale drinking water distribution system to disinfectant change from chlorine to chlorine dioxide in terms of its impact on microbiological stability, and disinfection byproduct formation. In that study, chlorine dioxide residuals were consistently present above detection limits throughout the distribution system. No degradation of bacterial water quality occurred after implementing the new disinfectant. Average HPC levels in the system were lower than 100 CFU/mL with an average chlorine residual of 0.45 mg/L (plant effluent chlorine of 0.6–0.8 mg/L), and temperatures below 15°C. When the system switched to chlorine dioxide, the average  $\text{ClO}_2$  in the system was 0.2–0.3 mg/L (for a plant effluent residual of 0.5 mg/L). Following the treatment change the HPC levels remained below 100 CFU/mL as water temperature was below 15°C. The effect of the disinfectant switch was tempered by temperature; when the temperature increased during

summer time, HPC levels increased. Chlorine dioxide maintained the total bacteria (microscopic counts) and heterotrophic plate count levels below  $2.0 \times 10^5$  bacteria/mL and  $1.0 \times 10^3$  CFU/mL, respectively.

## CONCLUSIONS

This study examined the impact on bacterial growth after switching from free chlorine or monochloramine to chlorine dioxide in a model drinking water distribution system. The data presented herein demonstrates the potential benefits of switching from monochloramine or free chlorine to chlorine dioxide. In each case the dosage of chlorine dioxide was two (free chlorine) to four (monochloramine) times lower than the original disinfectant and the microbiological quality of the water was at least maintained or improved. However the most noticeable impact in bacterial counts came from switching the AR treated with chloramines to chlorine dioxide, approximately a 10-fold reduction in suspended bacteria, which is consistent with observations by McGuire *et al.* (1999) for mitigating nitrification chlorine dioxide or chlorite. From a practical perspective, these findings could hold particular benefits for utilities that require acute mitigation of unmanageable microbiological growth in a distribution system or reservoir. It is anticipated that the time required to mitigate the regrowth episode could be relatively short (e.g., several weeks), although the specific operational requirements and procedures for field-scale implementation require further investigation at the pilot-scale level.

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