

# Implementation of chlorine dioxide disinfection: Effects of the treatment change on drinking water quality in a full-scale distribution system

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**Abstract:** Drinking water utilities may have to consider changing disinfectant to improve water quality and meet more stringent disinfection regulations. The objective of this study was to evaluate the response of a full-scale drinking water distribution system to a change in disinfectant from chlorine to chlorine dioxide, in terms of its impact on microbiological stability and disinfection by-product formation. Chlorine dioxide residuals were consistently present above detection limits throughout the distribution system ( $>0.20$  mg/L). Over a study period of 3.5 months, no degradation of bacterial water quality occurred after implementing the new disinfectant. Chlorine dioxide maintained total bacteria (microscopic counts) and heterotrophic plate count levels below  $2 \times 10^5$  cells/mL and 1000 CFU/mL, respectively. The change in disinfectant from chlorine to chlorine dioxide led to an 85% reduction in trihalomethanes (i.e., from 30 to 5  $\mu\text{g/L}$ ) and a 60% reduction in haloacetic acids (i.e., from 20 to 8  $\mu\text{g/L}$ ). Chlorine dioxide represents a valuable tool to produce high quality water and is a strong alternative to chlorine for certain distribution systems.

*Key words:* distribution system, chlorine dioxide, drinking water quality, disinfection by-products.

**Résumé:** Certains distributeurs d'eau potable vont devoir considérer changer de désinfectant pour pouvoir respecter les nouvelles normes de désinfection. Cette étude avait pour but de déterminer les effets du passage du chlore au dioxyde de chlore sur la stabilité bactérienne et la formation des sous produits de désinfection dans un réseau d'eau potable. Après désinfection au dioxyde de chlore, il était possible de maintenir des résiduels de dioxyde de chlore détectables dans l'ensemble du réseau ( $>0,20$  mg/L). Durant la période d'étude de 3,5 mois, la qualité microbienne de l'eau ne s'est pas dégradée après passage au bioxyde de chlore. Les concentrations totales de bactéries restèrent inférieures à  $2 \times 10^5$  cellules/mL dans le réseau (comptage au microscope par épifluorescence), tandis que les denombrements de germes hétérotrophes ne dépassèrent pas 1000 UFC/mL. Après le changement de désinfectant, les concentrations de trihalométhane ont été réduites de 85 % (de 30  $\mu\text{g/L}$  à 5  $\mu\text{g/L}$ ). De même, les niveaux des acides haloacétiques ont diminué de 60 % (de 20  $\mu\text{g/L}$  à 8  $\mu\text{g/L}$ ). Le dioxyde de chlore peut s'avérer un outil valable pour remplacer le chlore dans certains réseaux de distribution.

*Mots clés:* réseau de distribution, dioxyde de chlore, qualité des eau potables, sous-produit de désinfection.

## Introduction

Drinking water distribution systems act as complex reactors in which water quality is governed by various physical, chemical, hydraulic, and operational parameters. The practice of primary disinfection and the maintenance of a disinfectant residual

within the distribution system are necessary to control microbial contaminants and restrain bacterial regrowth. It must be acknowledged, however, that disinfection by-products (DBPs) are formed that may have potential adverse health effects. To meet new regulations, water utilities need to evaluate different disinfectants that will be effective against a variety of microorganisms and produce limited amounts of DBPs.

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). 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 and LeChevallier 1997). However, when inactivation of resistant microorganisms such as *Giardia* and *Cryptosporidium* is considered, chloramines have a lower disinfection power than free chlorine and are typically not applied to provide primary disinfection. Chloramination can also lead to nitrification in the distribution systems (Wilczak et al. 1996). An alternative is chlorine dioxide, which is a more effective disinfectant. This strong oxidant can also be used for the control of iron, manganese,

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and taste and odor causing compounds. Chlorine dioxide has different reaction pathways than chlorine for the formation of DBPs. It has been shown, for example, that while it can react with naturally occurring organics to form organohalogenes, the concentrations are typically much lower than when using free chlorine, with little formation of trihalomethanes and haloacetic acids (Werdehoff and Singer 1987; Hofmann et al. 1999). Chlorine dioxide, however, forms inorganic by-products (chlorite and chlorate) upon its reaction with water constituents, and water utilities may need to implement additional treatment, depending on the concentrations of chlorite and chlorate and their regulatory requirements (Lykins 1992; Faust and Oly 1999).

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 project was to evaluate the short-term response of a full-scale distribution system to the introduction of chlorine dioxide in terms of subsequent impacts on water quality. An instantaneous switch in the disinfection treatment from free chlorine applied to chlorine dioxide applied was avoided by gradual replacement over a 2-week period. Distribution systems could respond unfavorably to sudden treatment changes, with a risk of sloughing of biofilm into the water, consumer complaints of taste, odor, and (or) water discoloration. Samples were taken from the distribution system before and after the conversion from free  $\text{Cl}_2$  to  $\text{ClO}_2$  and analyzed for disinfectant residuals, bacterial counts, and disinfectant by-products. After the trial, the system was converted back to free chlorine. Sampling was conducted over a 15-week period at five different locations along the water distribution system, representing a range of contact times.

## Material and methods

### Study site

The distribution system under study was located in Ontario, Canada. Raw water originating from Lake Huron is prechlorinated at the intake for disinfection and zebra mussel control. It is coagulated using polyaluminum chloride, flocculated, and filtered through sand–anthracite filters. Chlorine is added again post filtration to ensure that a residual is maintained. Average plant production is  $4600 \text{ m}^3/\text{d}$  (1.2 MGD). The treatment facility serves a population of 2300.

### Disinfectant dosing

Both pre- and post-chlorine-treatment steps were gradually replaced with chlorine dioxide over a 2-week period. The distribution system was then allowed to stabilize for 2.5 months. Finally, the addition of chlorine dioxide was terminated and chlorine was fed back into the distribution system. A generator model ERCO R101 (Sterling Pulp Chemicals Ltd, Toronto, Ont.) was used to produce  $\text{ClO}_2$  during the field trial. The generation process combines aqueous sodium chlorite electrochemical oxidation and gas membrane separation to provide a high

purity chlorine dioxide solution. The system delivered greater than 98% pure chlorine dioxide to the flow (6.5 kg/d), based on the chlorine mass ratio. The target plant effluent target chlorine and chlorine dioxide residuals were 0.7–1.0 and 0.5–0.6 mg/L, respectively.

### Sampling points and frequency

Samples were collected from five locations: (i) entry to the distribution system, (ii) elevated tank outlet (16 h retention time), (iii) one mid-point site (26 h detention time), and two sites at the end of the distribution system, (iv) 50 and (v) 72 h detention times. Sampling began 2 weeks before the initial “ramp up” period. Samples were collected twice or three times weekly during intervals of rapid expected change in drinking water quality (i.e., as chlorine dioxide was being ramped up or down) and once a week during periods of relative steady state. A total of 24 sampling campaigns were conducted over 4 months.

### Monitored parameters

Sampling and testing protocols were as described in the Standard Methods for the Examination of Water and Wastewater (American Public Health Association 1998) unless otherwise specified. Samples for bacteriological analyses were collected in sterile 250-mL polycarbonate bottles containing sodium thiosulfate. Total coliforms were tested using a membrane filtration technique on M-Endo Medium (BD, Franklin Lakes, N.J.) incubated at  $37^\circ\text{C}$  for 24 h. Heterotrophic plate counts (HPC) were performed using a spread plate technique on R2A agar (BD). R2A plates were incubated at  $20^\circ\text{C}$  for 7 d. Acridine orange direct counts (AODC) were used to enumerate all microbial cells. One millilitre of sample was mixed with 1 mL of a 0.1% (w/v) solution of acridine orange for 5 min. The mixture was filtered on black cellulose nitrate filters (Millipore, Bedford, Mass.). The filters were observed by epifluorescence microscopy (Olympus model BX-60, Melville, N.Y.) and enumerated using an image analysis system (Esprit™, Olympus).

Samples for chemical analyses were collected in acid-washed containers. Chlorine residuals were measured by the N,N-Diethyl-p-phenylene diamine (DPD) ferrous titrimetric method. A spectrophotometric method employing lissamine green was used to measure residual chlorine dioxide concentrations (Hofmann et al. 1998). Chlorite and chlorate were measured according to the U.S. Environmental Protection Agency (USEPA) Method 5510 using a Dionex 500 ion chromatograph® (Sunnyvale, Calif.). Trihalomethane (THM), and haloacetic acid (HAA) samples were collected in headspace-free in 40 mL amber glass vials and preserved with ammonium chloride. Trihalomethane samples were acidified to pH 4.5 prior to shipment to prevent any increase in chloroform formation. Trihalomethane levels were determined using USEPA Method 551. HAA<sub>9</sub> were measured according to USEPA Method 552.2, which involves derivatization of the acids with acidified methanol. HAA<sub>9</sub> standards were obtained as a mix from Supelco® (Bellafonte, Pa.). Gas chromatographic analyses of THMs and HAAs were performed using a Hewlett Packard gas chromatograph (Palo Alto, Calif.,

model 5890 Series II-Plus) equipped with a DB-5 column for primary analysis and a DB-1701 column for confirmation. In addition, temperature, pH, total organic carbon (TOC), and operational data were collected.

### Data analysis

Data were analyzed using a spreadsheet program (Lotus 123, Lotus Development Corp., Cambridge, Mass.), a graphing program (Harvard Graphics, Software Publishing Corp., Santa Clara, Calif.), and a statistical analysis program (Statgraphics, STSC, Rockville, Md.). Because most of the data were not normally distributed, nonparametric tests (comparison of medians) were performed to determine whether two data sets were statistically significant.

## Results and discussion

A total of 120 samples were analyzed for various physical, chemical, and microbiological parameters to evaluate the effect of the treatment change on disinfectant residuals, bacterial water quality, and disinfection by-product levels. Average plant effluent pH and turbidity were 8.0 (7.7–8.7) and 0.1 NTU (0.02–0.30 NTU), respectively. Plant effluent TOC levels ranged between 1.1 and 1.9 mg/L (average of 1.5 mg/L).

### Physical and chemical parameters

#### Temperature

Temperature is a critical parameter. It impacts many reactions, including the rate of disinfectant decay and by-product formation, the inactivation of microorganisms, and amplification of bacterial regrowth (Faust and Oly 1999; Geldreich 1996). During our study, water temperature varied from 11 to 20°C with a rapid and drastic increase from approximately 16 to 20°C at the end of July (Fig. 1). Such temperature changes are typical for the region.

#### Chlorine and chlorine dioxide

It was possible to maintain adequate disinfectant residuals ( $>0.2$  mg  $\text{Cl}_2/\text{L}$ ,  $>0.1$  mg  $\text{ClO}_2/\text{L}$ ) throughout the distribution network, either with chlorine or chloride dioxide. Prior to switching to chlorine dioxide, the total chlorine residual leaving the treatment plant averaged 0.68 mg/L and ranged between 0.43 and 0.78 mg/L. On average, free chlorine represented 70% of the total chlorine residuals. Disinfectant residual was stable in the distribution system. System chlorine concentration averaged 0.44 mg/L (range: 0.31–0.54 mg/L), corresponding to an average consumption of 36% (for temperatures averaging 13°C). Total disinfectant residual averaged 0.35 mg/L at the end of the distribution system. When plant effluent chlorine levels were progressively reduced to 0 mg/L at the end of June, the chlorine dioxide concentration was ramped up to 0.50–0.66 mg/L (average of 0.56 mg/L) (Fig. 1).  $\text{ClO}_2$  residuals averaged 0.27 mg/L in the distribution network and 0.20 mg/L at the end of the system. Chlorine dioxide consumption averaged 50% (for water temperatures of 16.1°C). As a comparison, lower

chlorine dioxide levels of 0.3 mg/L were measured at the point of entry of two different distribution systems (Gatel et al. 1995). However,  $\text{ClO}_2$  was reported to be completely dissipated at the end of these systems.  $\text{ClO}_2$  disappearance was also reported to be greater at high temperatures and organic matter concentrations. In another distribution network (Laval, Qué.), plant effluent residuals of  $\text{ClO}_2$  varied with temperature and TOC levels and ranged between 0.02 and 0.41 mg/L (Lafrance et al. 1992). As observed in the study by Gatel et al. (1995), the disinfectant residual disappeared within the network. After terminating the chlorine dioxide feed and switching back to chlorine, the total chlorine residuals averaged 0.71 and 0.32 mg/L in the plant effluent and distribution system, respectively (corresponding to a chlorine consumption of 55% for an average temperature of 17.3°C) (Fig. 1).

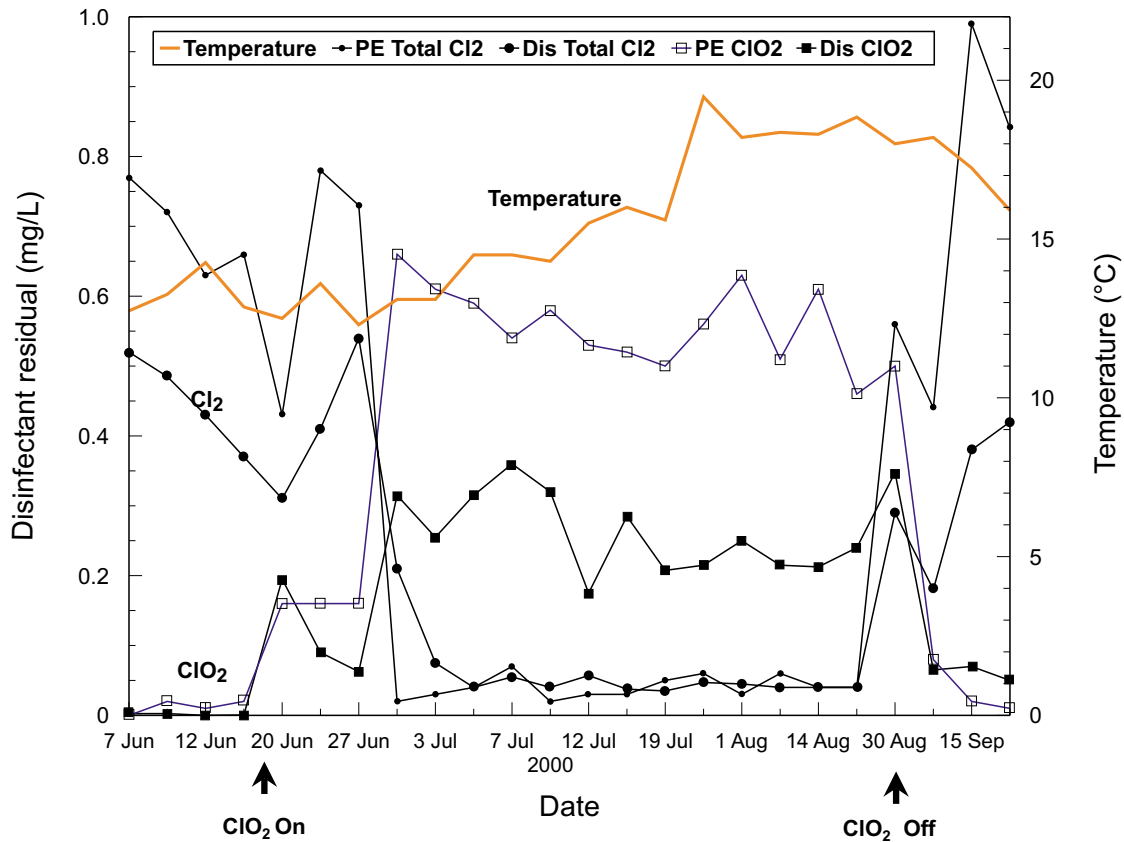
### Microbiological water quality

#### Effect of disinfectant type

Microbiological monitoring included total coliforms, heterotrophic plate counts, and acridine orange direct counts. The treatment change did not lead to detectable coliform occurrences or bacteria release from the biofilm during the transition period. Among the 120 samples collected over the entire study, three samples (2.5%) were positive for total coliform. Two of the three positive samples (5.7% of the chlorinated samples) were collected prior to initiating  $\text{ClO}_2$ , while 1.5% of the samples were coliform positive with  $\text{ClO}_2$  (no statistical difference between the two percentages,  $p > 0.05$ ). The occasional occurrence of coliforms is typical of surface water distribution systems and is related to various physical, chemical, and operational parameters (Geldreich 1996; LeChevallier et al. 1996; Volk and LeChevallier 2000). Two other monitoring studies that were conducted in distribution systems using chlorine dioxide showed no coliform occurrences with  $\text{ClO}_2$  (Lafrance et al. 1992; Gatel et al. 1995) after switching disinfectant. Norton and LeChevallier (1997) monitored the changes in bacterial water quality in two systems with coliform problems when the disinfectant was switched from chlorine to chloramines. After the disinfectant switch, coliform densities were drastically lowered, but HPCs data were only marginally affected.

Figure 2 displays total bacterial (AODC) concentrations and HPC levels in the plant effluent and distributed waters. There was no strong effect of the disinfectant switch on total or cultivable bacteria counts. At first, the AODC data showed a 50% decrease in total bacteria levels in the plant effluent and distribution system following chlorine dioxide implementation (from  $1 \times 10^6 - 2 \times 10^6$  cells/mL to  $5 \times 10^5 - 8 \times 10^5$  cell/mL, significant differences,  $p < 0.01$ ; June with free chlorine, vs. July with chlorine dioxide, with temperatures  $<15^\circ\text{C}$ ). Lower bacterial counts might be due to the fact that  $\text{ClO}_2$  is a stronger oxidant than chlorine and leads to the destruction of more bacterial cells. Chlorine dioxide is highly effective for disinfection, possibly because it can very easily permeate a bacteria cell wall (Simpson 1995). Its affinity for reduced sulfur compounds, including the sulfhydryl groups, has been suggested as

**Fig. 1.** Water temperature, chlorine and chlorine dioxide concentrations in the plant effluent and distribution system. (PE: plant effluent, Dis: mean values of the four distribution sampling locations).



one mode of bacterial disinfection (Ingols and Ridenour 1948; Simpson 1995). ClO<sub>2</sub> did not impact negatively bacterial water quality in the distribution system. Plant effluent HPC levels remained low (<10 CFU/mL) prior to and after implementing chlorine dioxide, while HPC data were <100 CFU/mL in the distribution system for the same period (no statistical differences,  $p > 0.05$ , June with Cl<sub>2</sub> vs. July with ClO<sub>2</sub>, water temperatures <15°C) (Fig. 2). Although chlorine dioxide is a stronger disinfectant than either monochloramine or free chlorine, relatively few studies have examined its ability to control bacterial regrowth in distribution systems. Limoni and Teltsch (1985) reported low suspended heterotrophic plate counts during the distribution of drinking water that had finished a chlorine dioxide residual concentration of approximately 0.2 mg/L. The Laval distribution system, which used ClO<sub>2</sub> 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.

In the full-scale system of Toulouse, France, using chlorine dioxide disinfection (Gatel et al. 1995), AODC levels were approximately 10<sup>5</sup> cells/mL in the treatment plant effluent and 5 × 10<sup>5</sup> cells/mL in the distribution system. Interestingly, the study showed that the proportion of viable bacteria (R2A at 20°C) stabilized at 0.2% of the total bacteria (AODC). Gatel et al. (1995) also reported that, in the presence of chlorine dioxide, the biodegradable organic matter did not promote bacterial

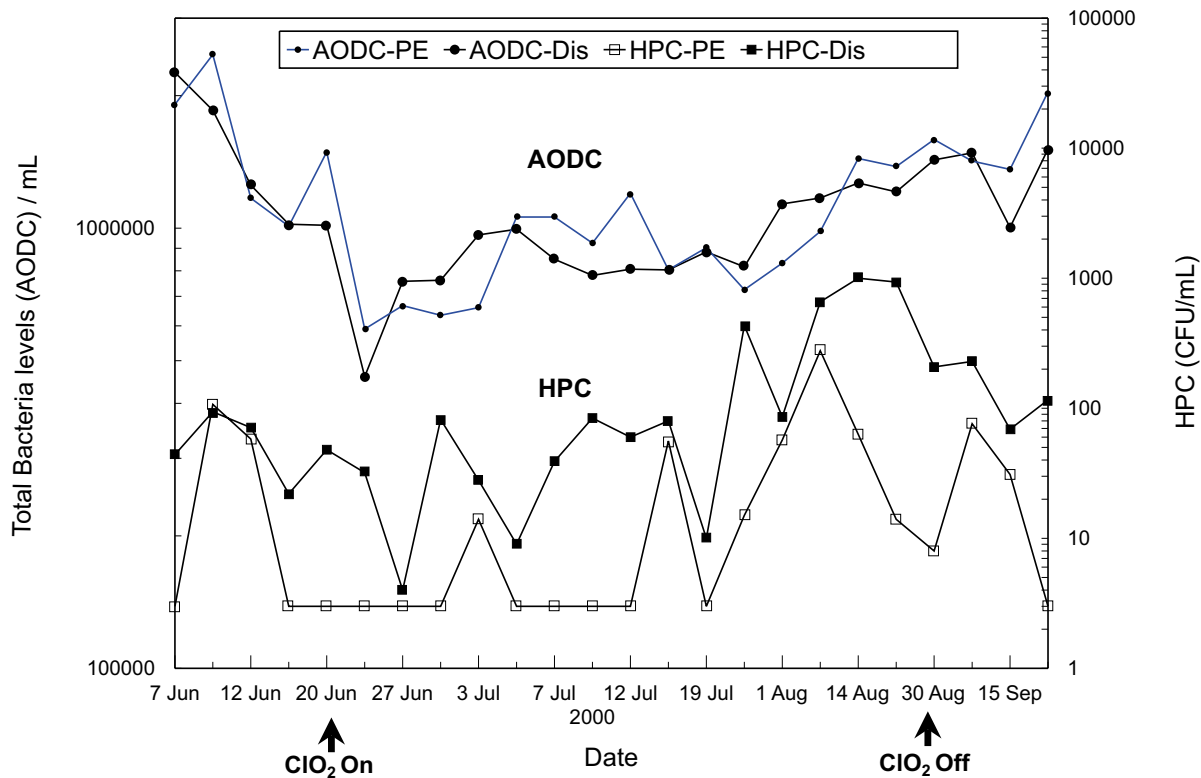
regrowth as much as in other distribution system using chlorine. For the Ontario system, the HPC/AODC ratio was lower: HPC represented 0.01–0.05% of the total bacteria concentration. In another study, Block (1995) found that 1% of the bacteria (AODC measurement) was cultivable on R2A agar for a pilot distribution system with free chlorine.

In addition to chlorine dioxide, chlorite (ClO<sub>2</sub><sup>-</sup>), one of its by-products, could also influence bacterial levels. Masschelein (1991) noted that chlorite had bacteriostatic properties and retarded the rate of bacterial rebound in drinking water systems. Moreover, in a recent field investigation involving five water utilities, McGuire et al. (1999) demonstrated that the presence of ClO<sub>2</sub><sup>-</sup> in the distribution system (levels of 0.05–0.9 mg/L) resulted in significant inactivation of ammonia oxidizing bacteria (AOB), thus resulting in less nitrification.

#### Effect of temperature

In the present study, bacterial concentrations were related to water temperature. Heterotrophic plate counts and AODC levels in the plant effluent and distribution system increased by approximately 60% when temperature increased dramatically in August (significant difference,  $p < 0.01$ ) (Figs. 1 and 2). Field experiments have shown that a variety of parameters influence biofilm growth in distribution systems. In addition to the disinfectant type and concentrations, other parameters including water temperature, source water quality, biodegradable organic

**Fig. 2.** Acridine orange direct count (AODC) and HPC levels in the plant effluent and distribution system. (PE: plant effluent, Dis: mean values of the four distribution sampling locations).



matter levels, plant operations, the condition and composition of the distribution system, corrosion control, and hydraulics may be more influential than disinfectant for regulating biological activity of the biofilm (LeChevallier et al. 1993; Camper 1995; Volk and LeChevallier 1999). Therefore, it is difficult to relate the changes in bacterial water quality to one specific parameter.

### Disinfection by-products

#### *Trihalomethanes and haloacetic acids*

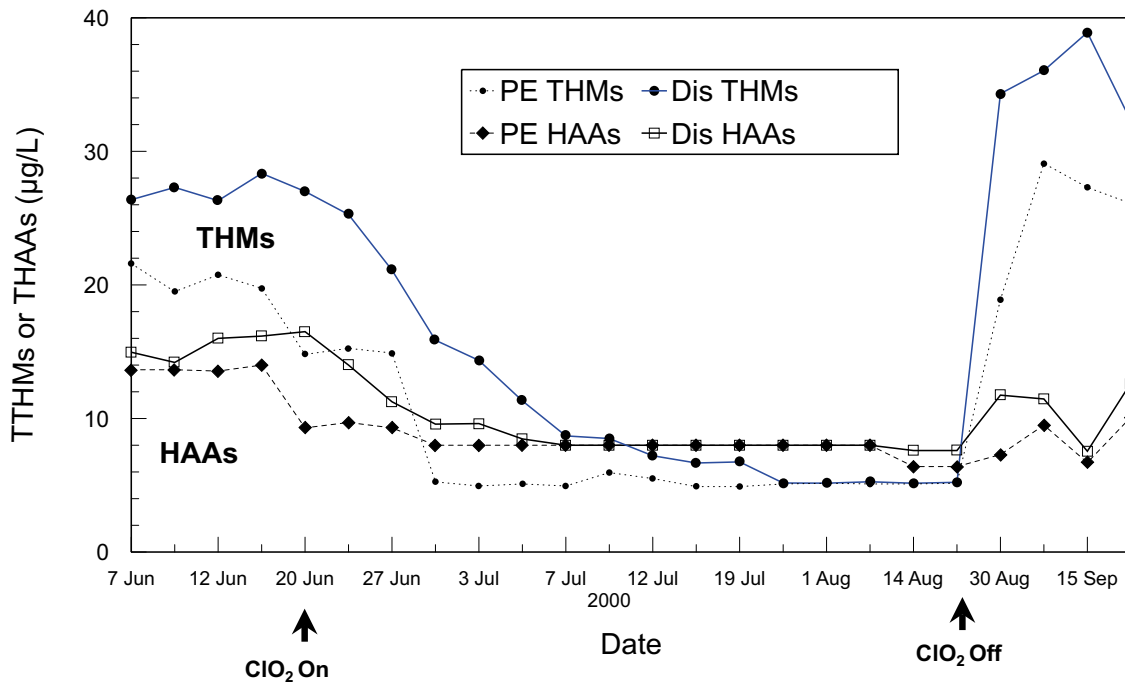
A dramatic decrease in THM levels was observed after the implementation of chlorine dioxide. Trihalomethane concentrations were approximately  $26 \mu\text{g/L}$  in the chlorinated distribution system (Fig. 3). Trihalomethane levels would have increased if chlorine had been fed during the summer months, because of elevated temperatures. Upon the introduction of chlorine dioxide and the phasing-out of chlorine, THM concentrations decreased drastically, ultimately stabilizing at approximately  $5 \mu\text{g/L}$  by mid-summer (Fig. 3). The replacement of chlorine by chlorine dioxide corresponded to an 81% reduction in THM concentrations. After the  $\text{ClO}_2$  trial, when chlorine was reintroduced in the system, THM levels increased back to  $35\text{--}40 \mu\text{g/L}$ . In comparison, Grubbs (1995) reported that total organic halides (TOX) formation was generally 1–25% of that observed with chlorine under the same conditions. Lafrance et al. (1992) found that THMs and other chlorinated products were below the detection level ( $<2 \mu\text{g/L}$ ) with chlorine dioxide, while the THM formation potential was  $100\text{--}200 \mu\text{g/L}$  with chlorine. Haloacetic acid levels were also reduced af-

ter  $\text{ClO}_2$  implementation. Haloacetic acid concentrations were approximately  $15 \mu\text{g/L}$  during the initial phase with chlorine (Fig. 3). Haloacetic acid concentrations decreased and eventually reached the detection limit once all of the chlorine present in the distribution system was replaced with chlorine dioxide. These results are in agreement with another study that suggested that chlorine dioxide does not form significant amounts of HAAs under most conditions, although some dichloroacetic acid formation has been observed in isolated tests (Korn 1998).

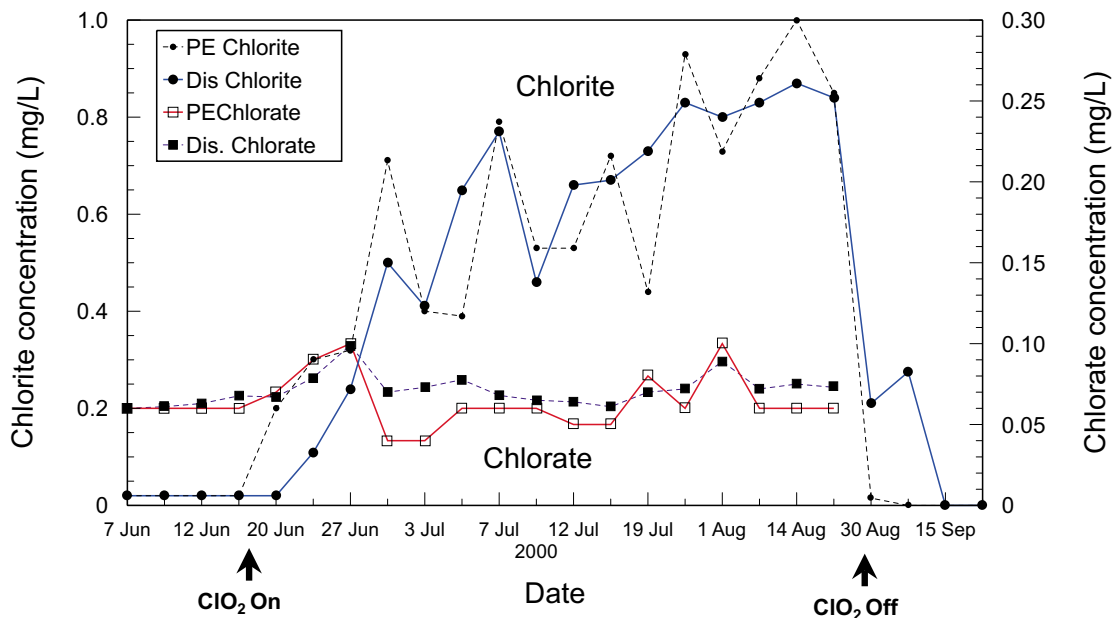
#### *Chlorite and chlorate*

When chlorine dioxide reacts and decays, chlorite and chlorate are formed. Previous studies have shown that approximately 70% of the applied chlorine dioxide will eventually form chlorite, while about 10% will form chlorate (Korn 1998). Chlorite levels averaged  $0.68 \text{ mg/L}$  ( $0.39\text{--}1.00$ ) and  $0.69 \text{ mg/L}$  ( $0.33\text{--}0.90$ ) in the plant effluent and distribution system, respectively. Importantly, chlorite levels were below the recommended levels of  $1.0 \text{ mg/L}$  in the United States (USEPA 1998). The maximum daily average chlorite concentration measured in the distribution system was approximately  $0.9 \text{ mg/L}$  (14 August), with a highest overall measurement of  $1.0 \text{ mg/L}$  on that same day at the water treatment plant (Fig. 4). It should be noted that during the study period the average chlorine dioxide residual concentration was  $0.27 \text{ mg/L}$ . It is anticipated that this value could be trimmed over time, and the associated chlorite concentration also lowered. No drastic changes in chlorite levels were observed as a function of the detention time (data not

**Fig. 3.** Total THM and HAA concentrations in the plant effluent and distribution system. (PE: plant effluent, Dis: mean values of the four distribution sampling locations).



**Fig. 4.** Chlorite and chlorate concentrations in the plant effluent and distribution system (Chlorate testing was not conducted after 8/18/2000). (PE: plant effluent, Dis: mean values of the four distribution sampling locations).



shown). This observation could be related to the fact that the distribution system was too small (short detention times) to lead to drastic changes in the disinfectant species. A different trend was observed for a larger system. In Laval, ClO<sub>2</sub> disappeared rapidly in the distribution network, and only residual chlorite was found at the extremities of the system. The chlorite disappeared entirely in dead end points (small pipes with low flows) (Lafrance et al. 1992).

Chlorate concentrations remained below 0.1 mg/L during the entire study (average of 0.060 and 0.071 mg/L in the plant effluent and distribution system, respectively) (Fig. 4), which is a very encouraging observation. While chlorate formed through the reaction or decay of chlorine dioxide is generally minimal, past surveys of water treated with chlorine dioxide often showed high chlorate concentrations (Gordon and Bubnis 1995). High chlorate concentrations were related not to chlorine dioxide re-

action and decay but to inefficient chlorine dioxide production. The low chlorate concentrations measured during this project confirm that the ERCO R101 generator technology does not contribute significantly to the overall chlorate concentration in the water. For the Toulouse system (Gatel et al. 1995), chlorate levels also remained stable throughout the system, while chlorite tended to decay with high retention times.

### Other data

It was also critical to monitor consumer complaints, since specific taste and odor problems may be associated with chlorine dioxide (Lafrance et al. 1992; Hoehn et al. 1990). No customer taste and odor complaints were reported during the study period. For other drinking water systems, various complaints could be observed when chlorine dioxide was used (Lafrance et al. 1992).

### Conclusions

This study documented water quality changes in a full-scale distribution system following chlorine dioxide implementation. The research work showed the following:

- Both chlorine and chlorine dioxide were capable of maintaining a measurable residual in the distribution system.
- Overall, the microbiological quality of the water was not adversely affected by chlorine dioxide during the transition period following the disinfectant switch. However, in addition to disinfectants, other parameters controlled the development of biofilm bacteria and subsequent bacterial water quality in distribution systems. Therefore, it is difficult to predict the effects of a treatment change at a specific site, which depends on the weight of the different factors regulating bacterial growth.
- THM and HAA levels approached detection limits when  $\text{ClO}_2$  was used. Chlorite concentrations remained below the USEPA regulatory levels, and the generator used in the study allowed chlorate concentrations to be maintained at a much lower level ( $<0.1$  mg/L) than anticipated when using traditional generation techniques.

Based on these data collected from a specific site, it was demonstrated that chlorine dioxide could represent a viable alternative to chlorine in systems that need to replace chlorine in the treatment train to control DBP levels.

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