Association between haloacetic acid degradation and heterotrophic bacteria in water distribution systems

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ABSTRACT

The occurrences of trihalomethanes (THMs), haloacetic acids (HAAs) and heterotrophic bacteria were monitored in five small water systems over a nine-month period to investigate the association between HAA degradation and heterotrophic bacteria populations. The sampling sites were chosen to cover the entire distribution network for each system. An inverse association between heterotrophic bacteria and HAA concentrations was found at some locations where chlorine residuals were around or less than $0.3\,mg\,L^{-1}$. At other sample locations, where chlorine residuals were higher (over $0.7\,mg\,L^{-1}$), no HAA reduction was observed. A high heterotrophic bacteria count accompanied with a low chlorine residual could be used as an indicator for HAA degradation in distribution systems.

1. Introduction

Under the United States Environmental Protection Agency (USEPA) Stage 2 Disinfectants and Disinfection Byproducts Rule (D/DBPR)(USEPA, 2006), the locational running annual averages (LRAAs) are specified for maximum contaminant level compliance. The total concentration of five haloacetic acids (HAA5: monochloroacetic acid, ClAA; dichloroacetic acid, Cl_2AA; trichloroacetic acid, Cl_3AA; monobromoacetic acid, BrAA; dibromoacetic acid, Br_2AA) and four trihalomethanes (THM4: chloroform, bromochloromethane, dibromochloromethane, bromodichloromethane, bromoform) are regulated as 60 and $80\,\mu g\,L^{-1}$, respectively. For disinfection byproduct (DBP) monitoring, peak DBP concentration locations should be selected for each system. Water systems are required to identify peak DBP concentration locations throughout the distribution system via an Initial Distribution System Evaluation (IDSE) under this USEPA Stage 2 D/DBPR.

Several factors affect DBP concentrations in a distribution system. These factors include pH, water temperature, total organic carbon (TOC) concentrations, chlorine residuals, bromide concentration, and microbial activity (Krasner et al., 1989; Stevens et al., 1989; Summers et al., 1996). The concentration of DBPs within the distribution may also vary seasonally and spatially. Seasonal variations are due to changes in source water quality, such as natural organic matter (NOM) concentration, temperature, chlorine demand and pH. Spatial variations are due to long reaction time of chlorine and NOM, as well as degradation/formation of DBPs and intermediate byproducts in the distribution system.

In the presence of a chlorine residual in water, DBP formation continues during distribution and storage. In the distribution system chlorine, especially free chlorine, in the treated water could react with NOM and produce more DBPs. Therefore, DBP concentrations may be higher in the distribution system than in the plant effluent. This is generally true for trihalomethanes (THMs). Higher THM concentrations are often found in the distribution systems than in the plant effluents, especially at maximum residence time locations. THM concentrations may have a positive association with residence time or distance in the distribution system and/or the free chlorine consumption (Clark and Sivaganesan, 1998; Koch et al., 1991; Reckhow and Singer, 1990). However, the HAA concentrations do not follow this pattern. Because HAAs are easily biodegradable compounds, HAA concentrations may be lower at the locations where bioactivity is high. These locations are often associated with longer residence time (Malliarou et al., 2005; Williams et al., 1998) and low chlorine residual. Other studies have found a possible correlation between bioactivity and HAA degradation in distribution systems (Baribeau et al., 2005; Landmeyer et al., 2000; Meyer et al., 1993; Speight and Singer, 2005). With bioactivity, HAAs could be degraded within the distribution system or in storage tanks where longer residence time is observed (Chen and Weisel, 1998; Landmeyer et al., 2000; Singer et al., 1993; Thomas et al., 2000). Therefore, the concentration of HAAs in the distribution could be increased by continuous HAA formation in the presence of residual chlorine or decomposition from other chlorinated DBPs and be reduced by biodegradation or hydrolysis (Zhang and Minear, 2002). Most of the HAA degradation in distribution studies focused on large systems where longer residence time would be expected. Small systems, where water residence time might be shorter than large systems, whether the HAA degradation occurs in the distribution were unclear.

Although microbial degradation may be the predominant mechanism for HAA reduction in the distribution system, there is no reported association between the decrease of HAAs and microbial quantities or activities. The objective for this study was to investigate the association between DBPs, especially HAAs, and the heterotrophic bacteria quantities in the distribution systems. The association could be used to develop a water quality parameter surrogate for HAA degradation in distribution systems and to better control HAAs in drinking water.

2. Methods

2.1. Sampling sites

Five small systems using surface water as the source water and practicing conventional treatment processes were selected for this study. Free chlorine was used in all treatment plants as the disinfectant. Six samplings were performed in a nine-month period for each system. Three of the six sampling were performed in the cold season and the other three were in warm season. The cold season represents water temperature between 6 and 10 °C and the warm season represents water temperature between 10 and 20 °C at the time of sampling. Four to six points along the distribution mains, including the inlet and the maximum residence time location, were selected for each system. The sampling points for this study were chosen to obtain the diverse geographic and hydraulic sites. Table 1 lists the general information of the five small systems studied and the distance of the sampling sites from the distribution inlet. A numeric system identification (ID) along with a Roman numeral was given to each sample location. For the research purpose and to avoid interference from the different residential piping systems, samples for system LK were collected from fire hydrants along the main distribution line. All other samples were collected at residential or industrial buildings.

2.2. Sample analysis

General water quality parameters, such as temperature, pH, and chlorine residuals, were measured at the time of

Table 1 – Sampling site identification, distance from the distribution inlet (m), and general water treatment plant operation parameters for each system.					
System	GP	NO	JT	LY	LK
Site ID[a]					
I	0	0	0	0	0
II	457	1097	1097	396	152
III	762	1646	1646	762	701
IV	914	1737	2195	1219	1433
V	1067	1920		1676	2256
VI	1219	3292			3353
Plant flow rate ($m^3\, day^{-1}$)	270	4500	1700	1100	1500
Population	Seasonal	4300	1900	3200	2300
Disinfectant	NaClO	$Cl_{2(g)}$	$Cl_{2(g)}$	$Cl_{2(g)}$	$Cl_{2(g)}$
Average TOC ($mg\, L^{-1}$)[b]	2.4 ± 0.9	1.3 ± 0.2	1.3 ± 0.3	1.4 ± 0.2	1.2 ± 0.3
Average post free chlorine residual ($mg\, L^{-1}$)[b]	1.40 ± 0.80	1.81 ± 0.19	0.92 ± 0.14	1.3 ± 0.1	1.67 ± 0.56
Average post total chlorine residual ($mg\, L^{-1}$)[b]	1.64 ± 0.69	2.06 ± 0.13	0.99 ± 0.14	1.4 ± 0.1	1.91 ± 0.25

a The site ID I is the distribution inlet for each system and the highest ID number is the maximum residence time location.
b Values were obtained from the finished water during the sampling period.

sampling. The chlorine residual measurements were conducted with the N,N-diethyl-p-phenylenediamine (DPD) colorimetric method using an Hach DR890 handheld meter (Hach Company, Loveland, Colorado, USA). The total organic carbon (TOC) concentrations were analyzed using the persulfate oxidation method with a TOC analyzer (model 1010, OI Analytical, College Station, TX, USA). Modified EPA Methods 551.1 and 552.3 were used to measure total trihalomethanes (THM4, includes: chloroform, dibromochloromethane, bromodichloromethane, and bromoform) and six HAA (HAA6, HAA5 plus bromochloroacetic acid, BrClAA) concentrations. Heterotrophic plate count (HPC) was analyzed by the standard membrane filtration method with HPC media (Pall Corporation, East Hills, NY, USA) (AWWA, 1998; USEPA, 1995, 2003).

3. Results

Because chlorine residuals and organic concentrations vary depending on treatment facilities and distribution systems, individual analysis within each system was conducted. For easy comparison of temperature variations, all data points were separated into two groups: cold and warm seasons.

3.1. System GP

The system GP is a state park with 339 seasonal campsites (for camping vehicle or tents) and 13 year-round cottages/cabins. Lower water usage and longer water retention time are common during the winter months. High THM4 and HAA6 concentrations ($>100 \, \mu g \, L^{-1}$) were found in this system which is consistent with the high chlorine demand of the source water (approximately $5 \, mg \, L^{-1}$) (Fig. 1a). The THM4 concentrations in this system increased along the distribution pipelines (locations GP-I to GP-V, Fig. 1a), and slightly decreased near the end of the distribution systems (location GP-VI, Fig. 1a). DBP formation would continue if both precursors and residual chlorine were present. Many studies have observed the DBP formation in distribution systems, and increased THM concentrations along the distribution system were commonly reported (Baribeau et al., 2005; Chen and Weisel, 1998; Rodriguez and Serodes, 2005).

HAA6 concentrations increased slightly from GP-I to GP-II locations and decreased at locations GP-III to GP-VI where high HPC numbers were detected (Fig. 1a, b). Lower chlorine residuals (from 0 to $0.25 \, mg \, L^{-1}$) were also observed at locations GP-III to GP-VI. Under these low chlorine conditions, HAA degrading microorganisms may be highly active and result in substantial HAA reduction.

Fig. 1c shows the average HAA6 concentration at different season. The HAA6 concentrations were higher in the warm season and lower in the cold season. For both seasons, HAA6 concentrations were reduced at locations with high HPC numbers (GP-III to GP-VI for the warm season and GP-IV to GP-VI for the cold season). No samples were collected from locations GP-II and GP-III during the cold season because those locations were closed for the winter. At location GP-IV, a 28% reduction in HAA concentration was observed (in comparison to HAAs in finished water) for both warm and cold seasons. A much lower HPC value was obtained in the cold
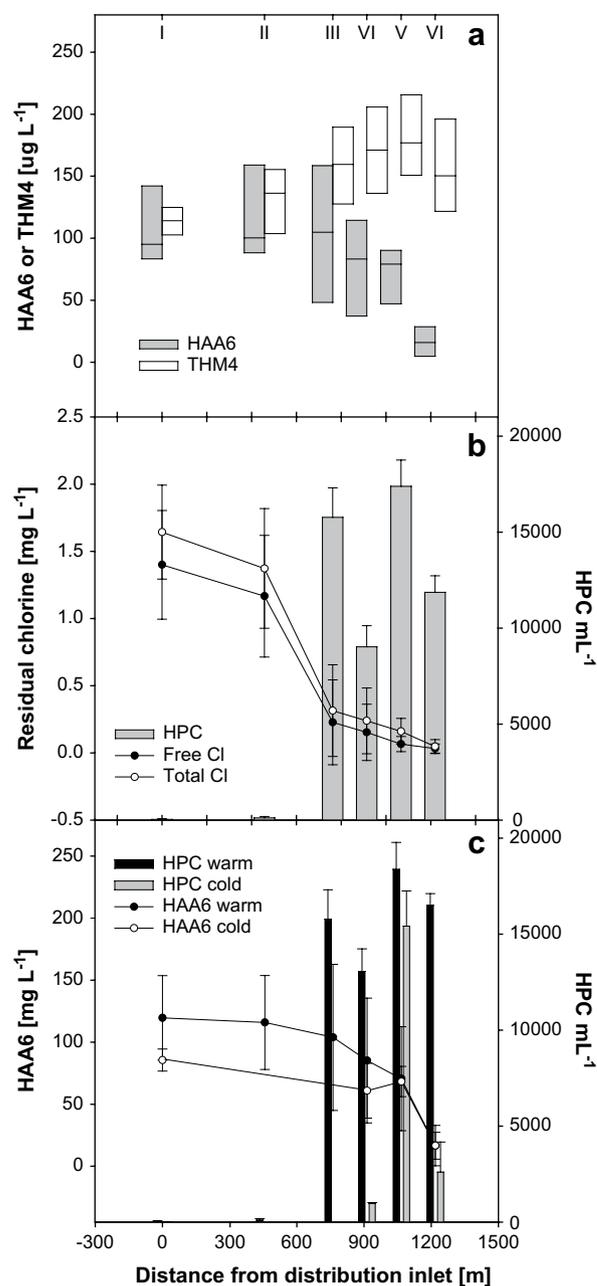
Fig. 1 – System GP. The Roman numerals labeled at the top x-axis indicate the location identification. (a) THM4 and HAA6 concentrations. The bars were constructed by 90th percentile, median, and 10th percentile of the six samplings taken from the experimental period. (b) Average chlorine residuals and HPC. (c) Average HAA6 concentrations and HPC at each temperature range.

season, indicating the impact of temperature on microbial population. HAA reductions at 86% and 81% were observed at location GP-VI (the maximum residence time location) in the warm and cold seasons, respectively. At location GP-V where HPC values were similar for both seasons, different HAA reductions were observed. The reduction was 41% for the warm season and was only 20% for the cold season. This indicates that the reduction of HAAs was not proportional to the amount of heterotrophic bacteria suspended in the water.

3.2. System NO

The average chlorine residual decreased slightly with distance from the distribution inlet, except at location NO-II (Fig. 2b) where a substantial drop of chlorine residual was observed. Location NO-II is an old public building and consistently has low chlorine residuals for both free and total chlorine. THM4 concentrations were higher at this point and a longer water retention time was suspected. NO-II also had the highest HPC numbers but HAA reduction was not observed. There are two potential reasons. First, the HPCs were under 40 counts mL^{-1}; thus, the bioactivity inside the pipe was insignificant in comparison to HAA formation. Second, the overall distribution system chlorine residuals were above 0.7 mg L^{-1} for all locations. Although bacteria could be viable under these high chlorine conditions, they might not be metabolically active regarding HAA biodegradation. Therefore, the HAA degradation might be inhibited by the high chlorine residuals in this distribution system.

HAA6 and HPC concentrations were higher when water temperatures were elevated at the same locations (Fig. 2c). Warm water results in higher HAA formation and higher heterotrophic bacteria growth. However, as shown in Fig. 2a, c, little or no HAA biodegradation was observed at system NO even in the warm season which might be due to high chlorine residuals.

3.3. System JT

System JT is a small system with a relatively linear distribution system and only four sampling locations were selected along the main distribution line. Fig. 3a shows that both the THM4 and HAA6 concentrations remained similar throughout the distribution system. Fig. 3b shows that both free and total chlorine decreased only slightly at the end of distribution system. The THM4 concentrations were slightly higher at the end of the system which might be the result of THM formation. The HAA6 concentrations remain similar throughout the system (Fig. 3a). Fig. 3c shows that HAA6 concentrations were higher in the warm weather. There was no substantial HAA degradation observed in the distribution system. This might be due to high chlorine residuals. This system maintains both free and total chlorine residuals over 0.7 mg L^{-1} throughout the distribution which might limit the bioactivity.

3.4. System LY

Fig. 4a, b shows the average chlorine residuals, HPC, HAA6 and THM4 concentrations. Similar to some of the other systems described previously, both the free and total chlorine residuals reduced as the distance from the distribution inlet increased. The decrease of chlorine residuals from location LY-II to LY-V was accompanied by an increase in THM4 and HAA6 concentrations. Fig. 4c shows that in the warm season, HAA6 concentrations were higher than those in the cold season. The HPC numbers were higher in locations LY-III to LY-V, but still in a low range (<40 counts mL^{-1}) in comparison to system GP. Due to the low heterotrophic bacteria concentrations and/or higher chlorine residuals (>0.7 mg L^{-1}) which might indicate lower bioactivities inside the pipe, no substantial HAA biodegradation was observed in this system.
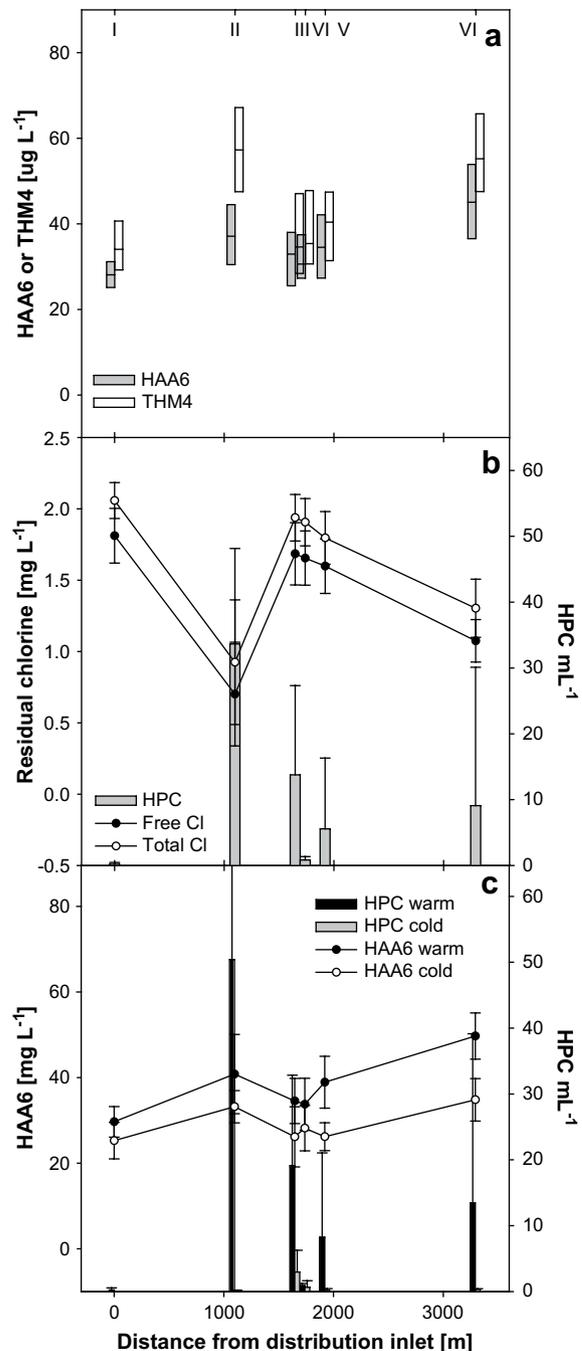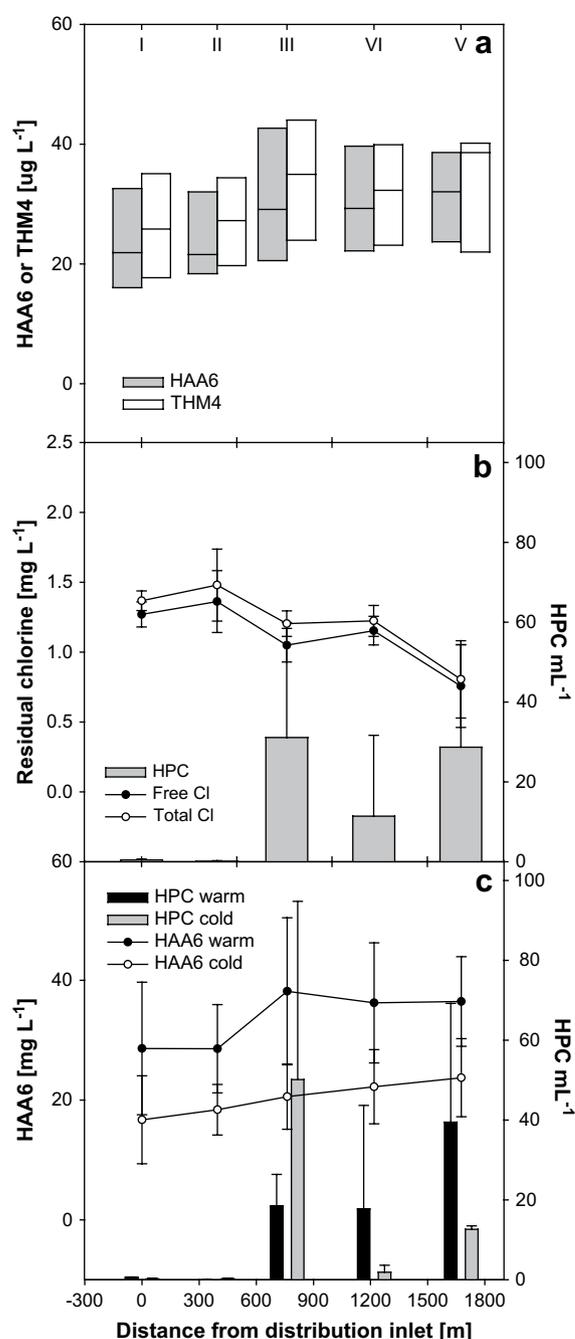

Fig. 2 – System NO. The Roman numerals labeled at the top *x*-axis indicate the location identification. (a) THM4 and HAA6 concentrations. The bars were constructed by 90th percentile, median, and 10th percentile of the six samplings taken from the experimental period. (b) Average chlorine residuals and HPC. (c) Average HAA6 concentrations and HPC at each temperature range.

3.5. System LK

For the research purpose and to avoid the interference of different plumbing system at each sampling location, fire hydrants from one water main (two directions outward from the treatment plant) were used as the sampling locations.
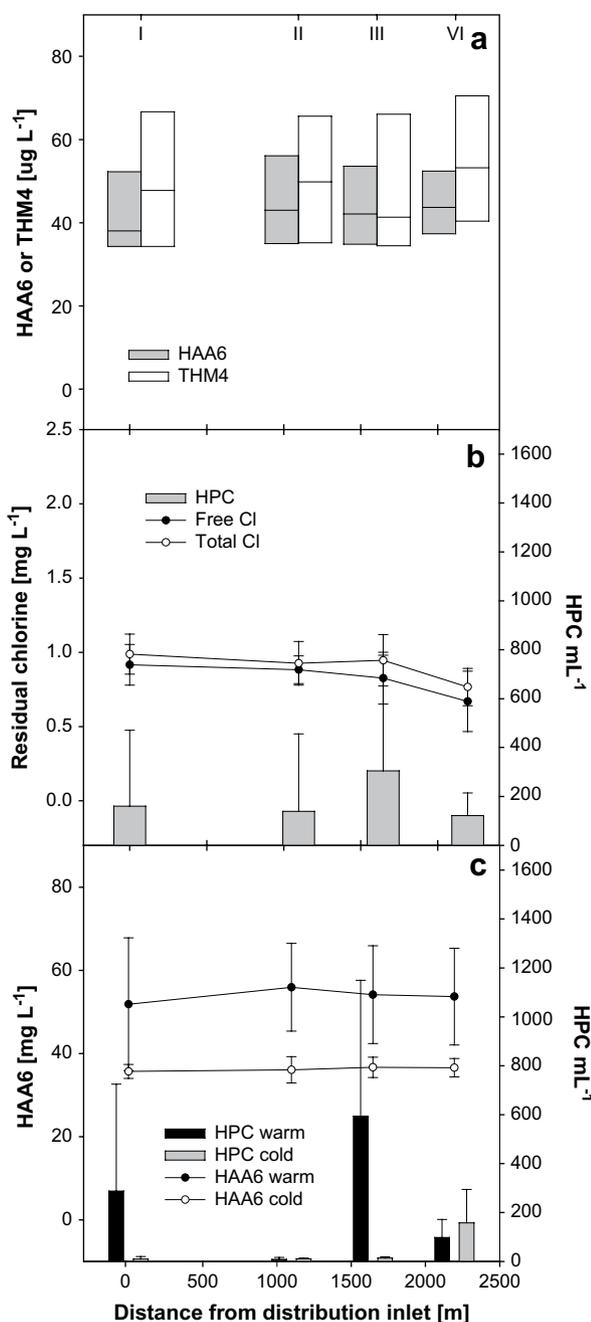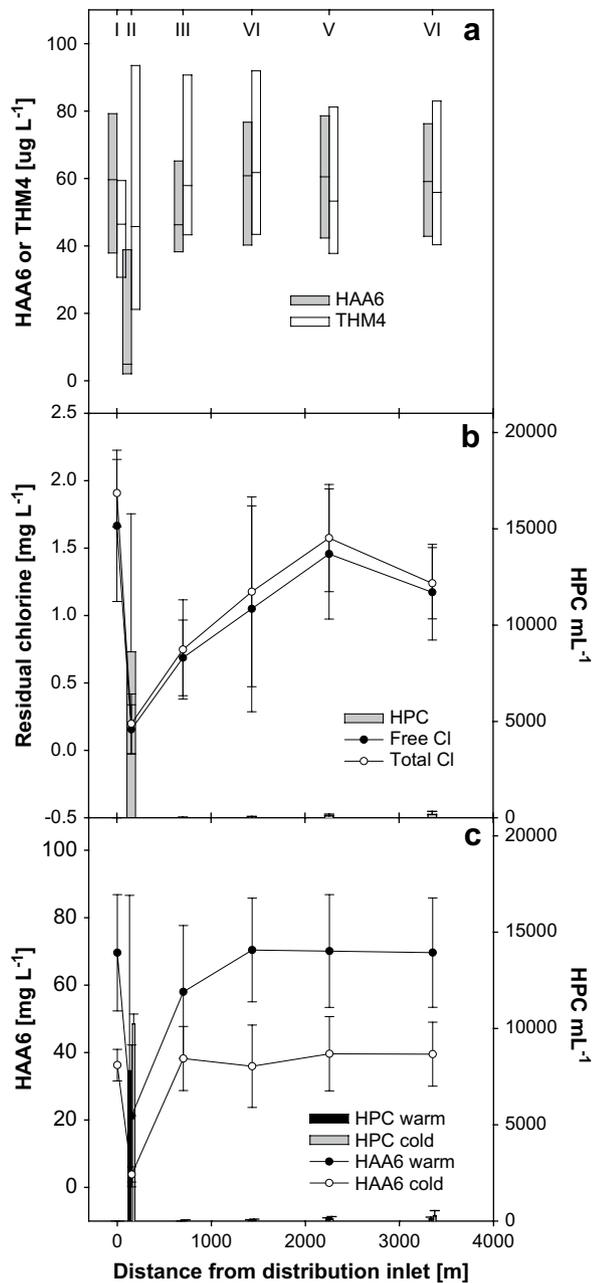
Fig. 3 – System JT. The Roman numerals labeled at the top x-axis indicate the location identification. (a) THM4 and HAA6 concentrations. The bars were constructed by 90th percentile, median, and 10th percentile of the six samplings taken from the experimental period. (b) Average chlorine residuals and HPC. (c) Average HAA6 concentrations and HPC at each temperature range.

Fig. 4 – System LY. The Roman numerals labeled at the top x-axis indicate the location identification. (a) THM4 and HAA6 concentrations. The bars were constructed by 90th percentile, median, and 10th percentile of the six samplings taken from the experimental period. (b) Average chlorine residuals and HPC. (c) Average HAA6 concentrations and HPC at each temperature range.

Fig. 5a, b shows the average chlorine residuals, HPC, HAA6 and THM4 concentrations for this system. Except for location LK-II, the chlorine residuals were lower than the distribution inlet but were all above 0.7 mg L^{-1}. At location LK-II, which the chlorine residuals dropped substantially to less than 0.1 mg L^{-1}, was only 152 m away from the inlet. The highest HPC concentration was observed and substantial reductions

in HAA concentrations were also observed at this location. The low chlorine residual and high HPC at this location may be due to excess biofilm inside the pipe feeding this fire hydrant since excess biofilm could result in a greater chlorine demand. The other possibility was that the location LK-II might be a dead-end or hydraulically isolated from the distribution which resulted in longer water residence time. The low

Fig. 5 – System LK. The Roman numerals labeled at the top *x*-axis indicate the location identification. (a) THM4 and HAA6 concentrations. The bars were constructed by 90th percentile, median, and 10th percentile of the six samplings taken from the experimental period. (b) Average chlorine residuals and HPC. (c) Average HAA6 concentrations and HPC at each temperature range.

chlorine residuals ($<0.1 \, mg \, L^{-1}$) and high HPC ($>10,000 \, mL^{-1}$) results were only found in four out of six total samples for the location LK-II. For these four samplings (included both warm and cold samples), HAA concentrations were close to zero. However, for the other two samplings, the chlorine residuals were higher (around $0.5 \, mg \, L^{-1}$) and the HPC were much lower ($<20 \, mL^{-1}$). For these two samplings, the HAA concentrations were higher (around $40 \, \mu g \, L^{-1}$) and were similar to other locations. This indicated that the low chlorine residual

concentrations might possibly associate with extremely high HPC in the water and resulted in the low HAA concentrations. Exclude location LK-II, HAA6 concentrations remained similar throughout the distribution system (Fig. 1a). HPC values increased slightly with the increase of the distance from the distribution inlet (from location LK-III to LK-VI). The increase of HPC in the distribution systems did not result in HAA reduction. This might be due to the higher average chlorine residuals. At locations LK-I and LK-III to LK-VI, chlorine residuals were all above $0.7 \, mg \, L^{-1}$.

HAA concentrations at system LK were higher in the warm season and lower in the cold season (Fig. 5c). However, the HPC values were higher in the cold season and lower in the warm season. The number of heterotrophic bacteria could be affected by many factors, such as temperature, treatment process, flushing intervals, organic concentrations and chlorine residuals. The actual cause of higher HPC in cold season could not be determined by the data obtained.

4.   Discussion

In this study, the THM4 concentrations were closely associated with chlorine residuals. Higher THM4 concentrations were detected at low chlorine residual locations in all the distribution systems. This is in agreement with other findings (Clark and Sivaganesan, 1998; Koch et al., 1991; Reckhow and Singer, 1990). At the sample locations where free chlorine residuals were lower than those at the distribution inlet, the THM4 concentrations were higher because of the continuous THM formation. For the same reason, decreasing chlorine residuals should increase HAA concentrations in the absence of HAA degradation.

There are many factors which affect the HAA concentration in distribution systems. HAA formation and HAA biodegradation are the two major processes which could change the HAA concentrations. Since most USEPA regulated HAAs are easily biodegradable compounds, the decrease of HAA concentration is expected at distribution system locations high in bioactivity. Higher halogen substitution HAA species such as Cl_3AA were not easy to be biodegraded (Baribeau et al., 2000; Singer et al., 1999). In this study, the major species of HAAs for all the samples analyzed were di- and trichloroacetic acids (approximately 1:1 wt/wt). At the locations where chlorine residuals were low, both di- and trichloroacetic acids were degraded to lower concentrations with similar ratio. This was inconsistent with other study in which Cl_3AA was not degraded in cold season (Baribeau et al., 2005). This might be due to different experimental condition. The data in this study were collected from the field where biofilm in the distribution system might already been adapted to the lower water temperature and be able to degrade Cl_3AA in both cold and warm seasons. The systems GP and LK, HAA concentrations were lower at locations where chlorine residuals were around or less than $0.3 \, mg \, L^{-1}$ and HPCs were high. This threshold chlorine residual value as $0.3 \, mg \, L^{-1}$ was determined by field data and may not apply to all systems. In systems with high chlorine residuals ($>0.7 \, mg \, L^{-1}$), such as systems NO, JT, and LY in this study, HAA degradation was not observed even in the presence of high heterotrophic bacteria populations. Although the

bacteria could survive under high chlorine conditions, the biofilm activity might be limited. A further exploration of the microbial degradation kinetics under high chlorine conditions is needed to verify the inhibition mechanisms.

Although HPC concentration in the water alone might not represent the actual microbial density inside the pipe biofilm, the use of HPC in this study was based on an assumption that higher HPC in the effluent water was the result of higher biofilm activity inside the pipe. A paired t-test with 95% confidence interval was calculated using the reactor data from Baribeau et al. (2005). The result showed that the difference between suspended HPC concentrations in the reactor effluent water and the HPC concentration on the reactor biofilm was not statistically significant ($p = 0.1206$). Another assumption that the biofilm at the systems studied was mature and produced constant growing and shedding rates. The biofilm could occasionally shed and produce a "peak" microbial concentration in the effluent (Zacheus et al., 2000). By sampling the same location continuously over a long period of time, the biofilm "shedding" effects could be reduced.

As reported in other studies (Nikolaou et al., 2004; Shimazu et al., 2005), THM4 and HAA6 concentrations were higher in the warm seasons than in the cold seasons for all five systems studied. Higher NOM concentrations, water temperature, chlorine dosages or reaction rates for DBP formation might be the main causes for higher DBPs in warm seasons. Biodegradation rates should also be higher in the warm seasons than in the cold seasons. However, this increased biodegradation during warm weather was not observed in this study.

5.　Conclusions

To comply with the Stage 2 D/DBP rule, water systems are required to identify distribution system locations with peak DBP concentrations. A thorough investigation of the distribution system could provide a better view of the spatial variation of DBPs. The results from this study suggested that HAA degradation would occur when chlorine residuals are lower than $0.3\,mg\,L^{-1}$ and heterotrophic bacteria are high ($>10,000$ HPC mL^{-1}). This might be applicable to other biodegradable DBPs as well. These findings could help water professionals to select proper locations best representing the peak DBP concentrations in their distribution systems.

Acknowledgements

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