

# **Report for 2005ME40B: Do Natural Chloride Gradients Affect the Formation of Disinfection By-Products in Public Water Supplies**

## Publications

- There are no reported publications resulting from this project.

## Report Follows

# **DO NATURAL CHLORIDE GRADIENTS AFFECT THE FORMATION OF DISINFECTION BY-PRODUCTS IN PUBLIC WATER SUPPLIES?**

## **ABSTRACT**

This study assesses how chloride in source waters affects the formation of disinfection by-products (DBP). Preliminary analyses of DBPs in public water supplies in Maine showed a greater prevalence of trihalomethanes (THM) over haloacetic acids (HAA) with increasing distance from the ocean. Distance to the ocean could be related to a salt effect caused by deposition of marine aerosols. However, paired samples of raw and finished water show no correlation between the DBP ratio and chloride concentrations. While there is no linear correlation between total DBP concentrations and chloride concentrations; there is a decrease in both THM and HAA concentrations in water with high chloride concentrations. Since comparing multiple systems with diverse land uses and treatment procedures is complex, it is not yet possible to determine the process by which chloride affects DBP formation. Possibly, elevated concentrations of chloride in raw water are inhibiting the reactivity of hypochlorite with organic matter.

## **INTRODUCTION**

January 2004 marked the start of the compliance cycle for small drinking water systems serving <10,000 customers (40 CFR Parts 9, 141, 142). The majority of 2,200 water systems in Maine, and in many other states, fall in this size range. During 2004, these small systems started monitoring for disinfection by-products and in 2005 they needed to have instituted control measures. However, there are numerous variables that contribute to the formation of DBPs, and not all are well understood. The presence of natural factors that affect the formation and type of DBPs will have a significant effect on the types of management techniques employed. The understanding and control of DBPs is the single biggest compliance challenge facing water utilities in the near future. Public water utility managers in Maine have requested help to understand how DBPs are forming in their systems so that they can best manage their risks.

Disinfection by-products (DBPs) are chlorinated or brominated compounds; tri-halomethanes (THM) and haloacetic acids (HAA) that are formed by disinfection chemicals reacting with natural organic matter (NOM) in the water. The EPA has set maximum contaminant levels (MCLs) of 0.08 mg/L for THM and 0.06 mg/L for HAA (USEPA, 2001; Safe Drinking Water Act, *e.g.* 40CFR 141 and revisions). Water systems may exceed the current MCLs during some reporting quarters and at some locations within the system. Although these systems are commonly in compliance based on average THM and HAA concentrations, Stage 2 of USEPA's DBP Rule, which requires that all monitoring sites within a system be compliant, will force many of those systems to undertake costly changes. Public water utility managers need help to understand how DBPs are forming in their systems so that they can best manage associated risks.

Efforts have been made recently to understand the controls on DBP formation and the steps necessary to prevent DBP formation. Regression techniques have been used to identify parameters responsible for DBP formation. Predictive models of DBP formation include variables such as: temperature, pH, bromine concentrations, chlorine dose, and measures of the content and nature of natural organic matter (NOM), such as total organic carbon (TOC), dissolved organic carbon (DOC), ultraviolet absorbance at 254 nm ( $UV_{254}$ ), and specific  $UV_{254}$  absorbance ( $SUVA_{254}$ ), etc. (Sadiq and Rodriguez, 2004, Sohn et al., 2004). Although models developed using regression techniques can have high accuracy in laboratory studies or as defined through studies of single systems, accuracy decreases when models are applied across systems with differing water sources and treatment procedures.

TOC and the character of NOM are known factors in the formation of DBPs (Kitis et al., 2001; Leenheer et al., 2001; Charrois et al., 2004; Xie, 2004; Kim and Yu, 2005). In general, the higher the TOC content of the source water the greater the potential to form DBPs (Canale et al., 1997; Charrois et al., 2004).  $SUVA_{254}$  is used as a surrogate parameter for aromaticity of NOM, which is related to a high proportion of humic content (Traina et al., 1990).  $SUVA_{254}$  correlates positively with DBP formation potential (Kitis et al., 2001; Leenheer et al., 2001), and is used to predict DBP concentrations within a water system (Chow, 2006). The ratio of absorbance at 465 nm to 665 nm is also an indicator of the degree of humification and aromaticity of NOM (Stevenson and White, 1995; Chin et al., 1994). Korshin et al. (1997) have demonstrated that absorbance at 272 nm correlates with the formation of total organic halides (TOX) following chlorination.  $SUVA_{280}$  was found to be an accurate predictor of the production of DBPs from chlorine reactivity with NOM (Kitis et al., 2001)

Much has been learned about the formation of by-products, and the occurrence of byproduct precursors in NOM (Reckhow et al., 1999; Karimi and Singer, 1991; Singer, 1994; Westerhoff et al., 1998), but it is clear that engineered systems do not entirely mitigate the occurrence of by-products. Recent data show the function of watersheds in controlling NOM and DBP precursors (LeChavelier and Volk, 2002; Garvey and Tobiason, 2003). The type of NOM in water supplies is dependent on the biological activity in the source watershed. The export of NOM to lakes is dependent upon such factors as watershed land uses, geology, topography, climate, (Schiff et al., 1997; Cronan et al., 1999; David et al., 1999) and hydrology is the driving force for mass loading of NOM. Several studies in the northeast have documented the relationships between watershed compositions and DOC (Thurman, 1985; Cronan et al., 1999), although few have addressed the issue from the perspective of drinking water supplies (Reckhow, 2002). These relationships have also been measured with respect to the potential to form tri-halomethane (Amy et al., 1987; Arruda et al., 1989; Garvey and Tobiason, 2003). The study on the Quabbin reservoir in Massachusetts clearly indicated that inputs and internal processes affect the formation of DBPs and that there is a need to differentiate between allochthonous and autochthonous DOC (Garvey et al., 1998; Stepczuk et al., 1998) in an understanding of DBPs.

The scope of this project encompasses public water supplies using surface waters in a spatial gradient from adjacent to the coast to 200 kilometers inland (Figure 1). Data collected by us and other researchers at the University of Maine have established a gradient of chloride in surface and ground waters that decreases inland from ~100 ueq/L to <10 ueq/L. Preliminary data from public water suppliers have demonstrated that DBPs occur in Maine's drinking water, including

several systems near or above the MCLs for THM or HAA. Since road salt can be a significant source of chloride seasonally at all locations, road salt is expected to overprint a natural chloride gradient. Water supplies that bracket the range from undeveloped to developed source areas can be mapped across the chloride gradient.

Preliminary analyses of DBPs in public water supplies in Maine showed that there are some unusual relationships between the distance from the ocean and the occurrence of trihalomethanes (THM) and haloacetic acids (HAA). In particular, the relative ratio of HAA to THM (HAA-THM ratio), defined as:

$$(HAA - THM) / (HAA + THM) \quad (\text{equation 1}).$$

The HAA-THM ratio is positive if HAA is the dominant DBP and negative if THM is dominant. In Maine, the HAA-THM ratio is positive near the coast and negative inland (Figure 2). This variation exhibits the same trend as chloride in surface waters generated from research on lakes in Maine. Chloride in lakes varies from relative highs near the ocean to trace concentrations in extreme northern Maine (Kahl et al., 2004; see also National Atmospheric Deposition Program, <http://nadp.sws.uiuc.edu/>). Other halides in seawater such as bromide and iodide may be equally important. As can be seen in Figure 2, there is a correlation between proximity to the ocean and the dominant type of DBP formed. Our initial hypothesis was that background chloride concentrations were affecting DBP formation. If natural chloride from oceanic aerosols (along with bromide and iodide), or from road salt, is affecting DBP formation in the source, in addition to disinfection processes, then very different control techniques will be needed. This research investigated the relationship between chloride concentrations and DBP formation.

The sites investigated in this report are public water supplies that utilize surface water throughout the state of Maine. The sites are located from 2 to 283 km (1.2 to 176 miles) from the ocean. The water sources for these utilities include lakes, ponds, rivers, streams, and reservoirs. Watershed hydrologic characteristics and land development for these sources vary considerably. Land use ranges from highly developed (urban to suburban) to undisturbed (forested), making a large gradient of influences for the delivery of NOM to surface waters.

## **METHODS**

Two rounds of data were collected. In the first round, raw- and finished-water samples from 30 public water systems were analyzed for major cations, anions, acid neutralizing capacity (ANC), pH, conductivity, dissolved organic carbon (DOC), total organic carbon (TOC), and specific ultraviolet absorbance at 254 nm (SUVA<sub>254</sub>). THM and HAA concentrations in finished water for round one were obtained from the water districts records for a date closest to the sample date for this project. In the second round, the same data were obtained, but THM and HAA concentrations were measured from samples collected at the same time.

Eight representative sites were selected from the first round sites for a second sampling round. The second round sites were sampled and analyzed as described above. Additionally, the major THMs (chloroform, bromodichloromethane, dibromochloromethane, and bromoform) and HAAs (monochloroacetic acid, monobromoacetic acid, dichloroacetic acid, trichloroacetic acid, and dibromoacetic acid) were measured in raw and finished water that was collected at the same time.

Sample analysis was performed at the University of Maine for pH, specific conductance, acid neutralizing capacity, anions, major cations, DOC, and UV absorbance and at a commercial laboratory for DBPs. Analytical methods for tri-halomethanes followed EPA Method 524.2 and Standard Method 6251B for halo-acetic acids.

The pH and specific conductance were analyzed in accordance with standard methods. Acid neutralizing capacity was analyzed using an ARAS and TIM900 titrator system. Cations were analyzed using an Inductively Coupled Plasma-Optical Emission Spectroscopy (ICP-OES). Anions were analyzed by ion chromatography. UV absorbance was measured using a UV-VIS spectrophotometer at 254 nm. TOC and DOC were analyzed automated by persulfate digestion followed by infra-red detection of carbon dioxide.

## RESULTS

**First Round.** Total trihalomethanes ranged from 3.3 to 224 µg/L and total halo-acetic acids ranged from 5.2 to 130 µg/L. Background chloride concentrations ranged from 14.8 to 1094 µeq/L (525 to 39,000 µg/L). TOC concentrations in raw water ranged from 1.8 to 18 mg/L. The HAA-THM ratio ranges from -1 to +0.6. Neither HAAs nor THMs consistently dominate the total DBP concentrations, although THM concentration is about 36 times HAA concentration at two sites. There is no pattern in THM or HAA dominance with source type. The HAA-THM ratio from the finished water decreased linearly ( $R^2 = 0.30$ ) with distance from the ocean (Figure 3), indicating an increase in THM dominance away from the coast. Chloride concentrations decrease with distance from the ocean, but the correlation with the HAA-THM ratio is very weak ( $R^2 = 0.10$ ). In spite of similar relationships of HAA-THM ratio and chloride concentrations with distance from the ocean, no correlation exists between the HAA-THM ratio in the finished water and chloride concentrations in the raw water (Figure 4). Total THM and HAA concentrations in finished water are lowest at highest raw-water chloride concentrations, but there are no linear correlations (Figure 5). Total THM concentrations decrease more sharply than total HAA with increasing chloride concentrations.

Chloride concentrations are higher in the finished water than in the raw water for all but one site. THM and HAA concentrations are highest at the greatest relative difference of chloride:

$$(Cl_{\text{finished}} - Cl_{\text{raw}}) / Cl_{\text{finished}} * 100 \quad (\text{equation 2}).$$

There is a weak linear correlation ( $R^2 = 0.26$ ) for the THM data (Figure 6) and insignificant correlation ( $R^2 < 0.02$ ) for the HAA data .

Typically, DOC concentrations are within analytical error of the TOC concentrations (i.e. there is no significant difference between TOC and DOC for the same sample). All but one site had higher TOC concentrations in the raw water than the finished water. THM concentrations of finished-water samples correlate weakly with TOC of the raw water, but HAA concentrations show no correlation (Figure 7). No correlations were found between either ANC or pH and DBP concentrations.

Although correlations between  $SUVA_{254}$  and both THM and HAA concentrations in finished water are not significant, the DBP concentrations are highest at highest  $SUVA_{254}$  (Figure 8). Utilities with a river source have higher  $SUVA_{254}$  than those with lake sources, but no correlation exists between DBP concentrations and  $SUVA_{254}$  in those data (Figure 8).

**Second Round.** THM and HAA concentrations were measured for both raw and finished water, and only one sample had measurable DBP (1.6 µg/L trichloroacetic acid) in the raw water. Most of the finished water had HAAs with similar dichloroacetic acid and trichloroacetic acid concentrations, but two lake waters had considerably more trichloroacetic acid than dichloroacetic acid. Two river waters had minor monochloroacetic acid concentrations. Most of the total THMs in the finished water were composed of chloroform, with minor bromodichloromethane. Two river waters had minor dibromochloromethane.

Concentrations of both THM and HAA correlate positively with TOC (Figures 9 and 10) and with SUVA<sub>254</sub> (Figure 11). The lowest THM and HAA concentrations occur with the highest chloride concentrations.

## DISCUSSION

There is a linear trend of decreasing HAA-THM ratio with distance from the ocean (Figure 3), indicating greater prevalence of THM concentrations inland. The initial hypothesis that this trend is due to a chloride gradient across the area is not supported by the data, which show no relationship between HAA-THM ratios and chloride concentrations (Figure 4). Distance from the ocean is associated with other changes, such as climatic gradients and land uses that affect NOM. The nature of NOM controls the formation of DBPs (Korshin et al., 1999; Kitis et al., 2001; Leenheer et al., 2001). THMs are more influenced by the hydrophobic fraction and HAAs are more influenced by the hydrophilic fraction (Kim and Yu, 2005). It is possible, therefore, that inland decreases in HAA-THM ratios may be controlled by differences in organic matter arising from other factors. SUVA<sub>254</sub> values are variable (73 to 238 L/mg-m) near the ocean, but all are above 180 L/mg-m at distances greater than 100 km from the ocean, suggestive of a spatial control on the humic factor of NOM.

Although there is no correlation between HAA-THM ratio and chloride concentrations, there is a relationship between total DBPs and chloride concentrations. In the first sampling round, the greatest DBP formation occurs in water with the lowest chloride concentrations, although the correlation is very weak (Figure 4). DBP and chloride concentrations in the second round of sampling show a similar decrease in both THM and HAA concentrations in water with high chloride concentrations, and have much better correlations (Figure 12).

TOC and SUVA<sub>254</sub> are known factors in the formation of DBPs (Kitis et al., 2001; Leenheer et al., 2001; Charrois et al., 2004; Xie, 2004; Kim and Yu, 2005). The data in the first sampling round show a poor correlation between DBP concentrations and TOC and SUVA<sub>254</sub>. This is probably because of the time differential between data collection of the DBP concentrations and the other parameters in the first dataset. There is a better correlation of DBP concentrations and TOC and SUVA<sub>254</sub> in the second round of sampling than in the first, reflecting the simultaneous collection of all the data. Correlations between DBP concentrations and other parameters are compromised by the disconnect in sampling of DBP concentrations and other parameters in the first dataset, but the larger size of the dataset makes it useful, especially if similar relationships are detected in both datasets. General relationships, such as increases and decreases, show up in both datasets. Of particular importance is the occurrence of lowest DBP concentrations at the highest background chloride concentrations.

Assuming that a direct link exists between chloride and DBP concentrations, and chloride is not acting as a proxy, an explanation is needed for this relationship. Chloride is assumed to be a conservative element and is unlikely to participate in chemical reactions. Chloride does adsorb weakly, however, and can interfere with some surface reactions. For example, chloride, unlike other anions, acts as a linear inhibitor of NADH:nitrate reductase activity (Barber et al., 1989), and chloride adsorption onto iron filings inhibits perchlorate reduction (Moore and Young, 2005). Chloride adsorbs onto iron surfaces more at low pH (Moore and Young, 2005). THM concentrations differ considerably from low to high chloride concentrations, but HAA concentrations do not (Figure 5), suggesting that THM is more affected by chloride concentrations. It is known that THM formation increases with increasing pH, but HAA and total organic halide formation decreases with increasing pH (Reckhow and Singer, 1985; Xie, 2004; Malliarou et al., 2005). It may be that greater chloride interference at low pH prevents THM formation. There is, however, no trend in DBP concentrations with pH. Because of the complexity inherent in comparing multiple systems with diverse sources, source types, and treatment procedures, it is difficult to make conclusions as to the nature of the relationship between chloride concentrations and DBP formation. Possibly, elevated concentrations of chloride are inhibiting the activity of hypochlorite. Laboratory studies are being designed to investigate the nature of the effect of chloride on THM and HAA formation.

## **CONCLUSIONS**

HAA dominance of total DBP concentrations decreases linearly with distance from the ocean. This relationship is not due to a chloride gradient, but may be related to the character of the NOM. Chloride concentrations in source water do appear to affect the total amount of DBP formed. DBP concentrations (particularly THM concentrations) are lowest at high chloride concentrations, implying potential chloride interference in DBP formation.

## **ACKNOWLEDGEMENTS**

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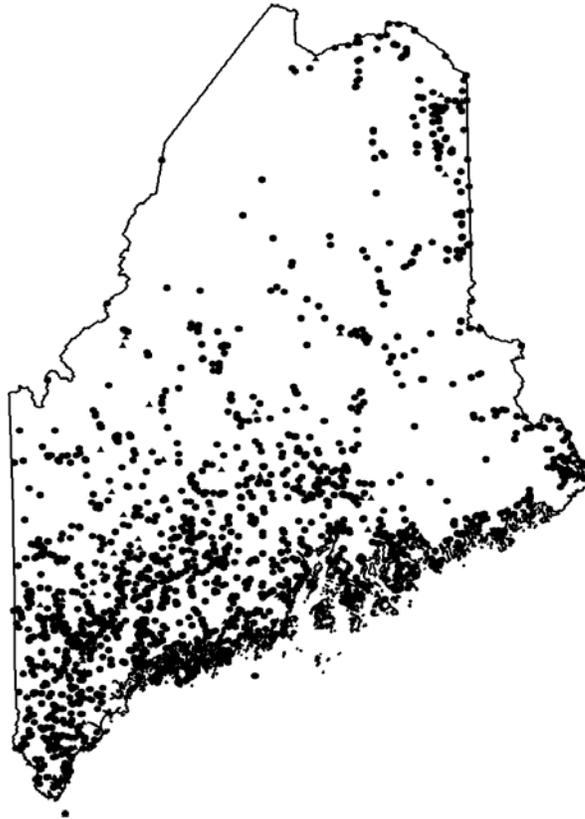
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## FIGURES

**Figure 1.** Locations of Surface Water Intakes and Supply Wells for Public Water Supplies in Maine.



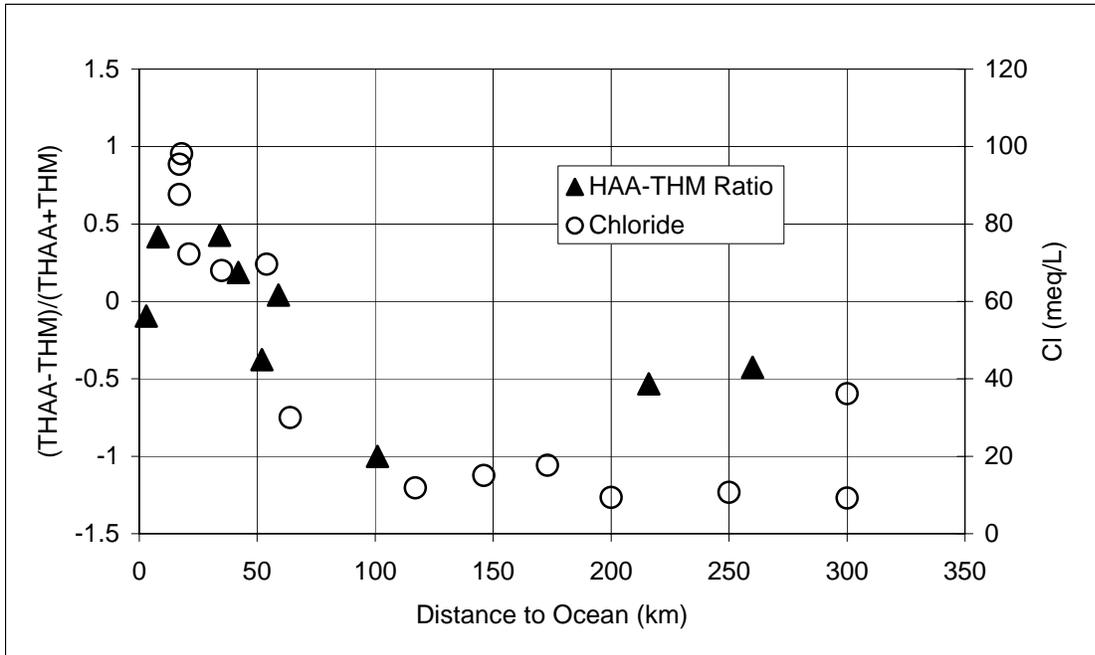


Figure 2. HAA-THM ratio in finished water (left axis) and Cl concentrations in raw water (right axis) versus distance from the ocean, preliminary data.

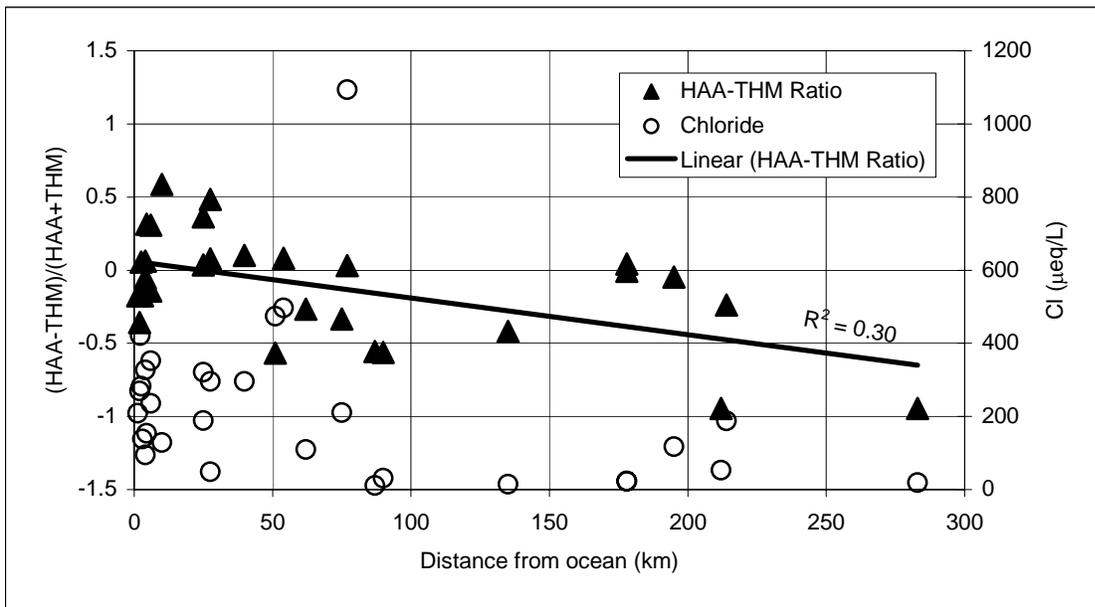


Figure 3. HAA-THM ratio in finished water (left axis) and Cl concentrations in raw water (right axis) versus distance from the ocean, first-round samples.

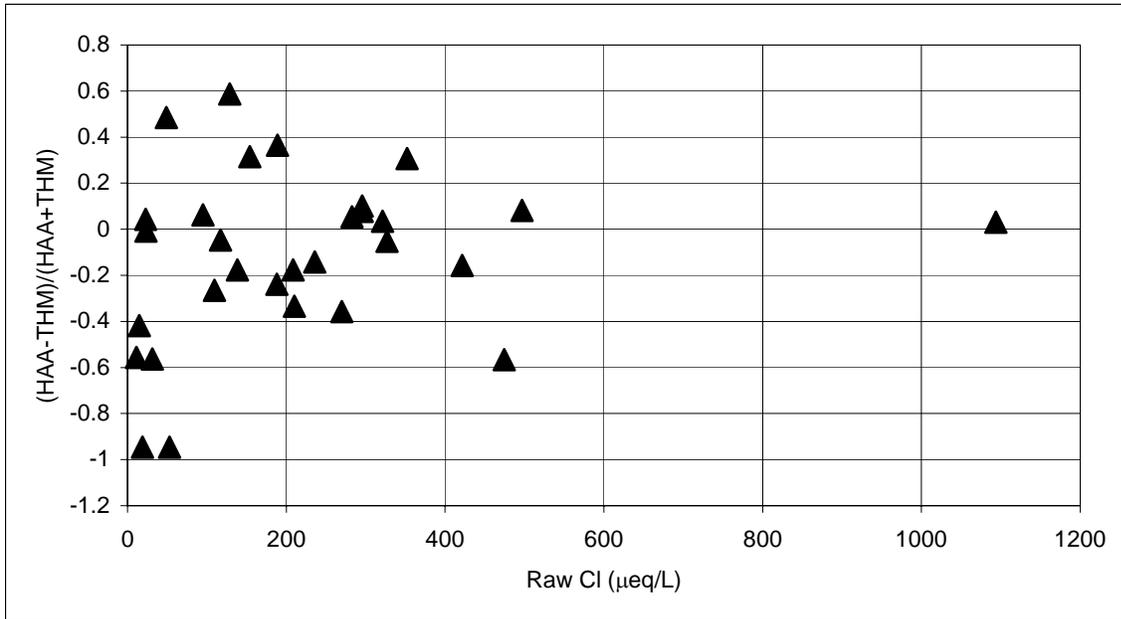


Figure 4. HAA-THM ratio in finished water versus Cl concentrations in raw water, first-round samples.

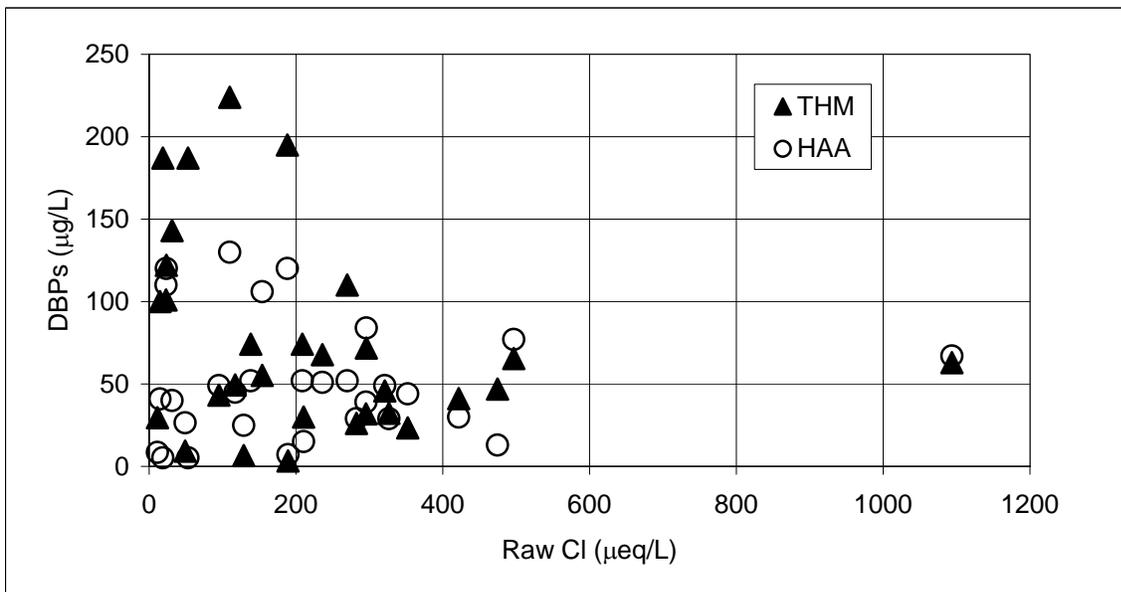


Figure 5. THM and HAA concentrations in finished water versus Cl concentrations in raw water, first-round samples.

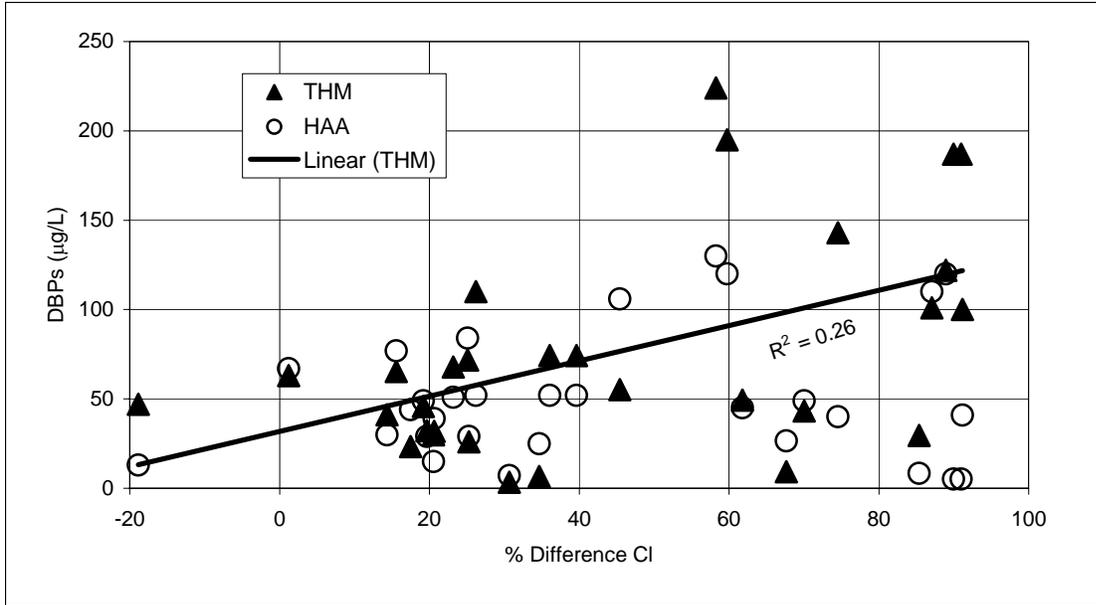


Figure 6. THM and HAA concentrations in finished water versus % difference Cl.

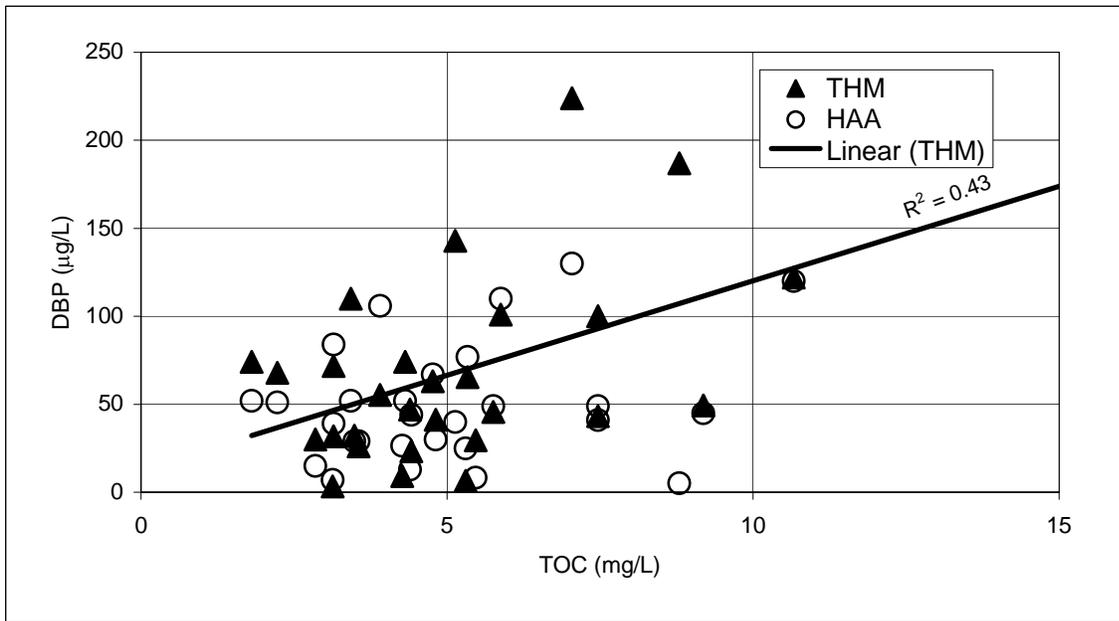


Figure 7. THM and HAA concentrations in finished water versus TOC in raw water, and the trend line for the THM data.

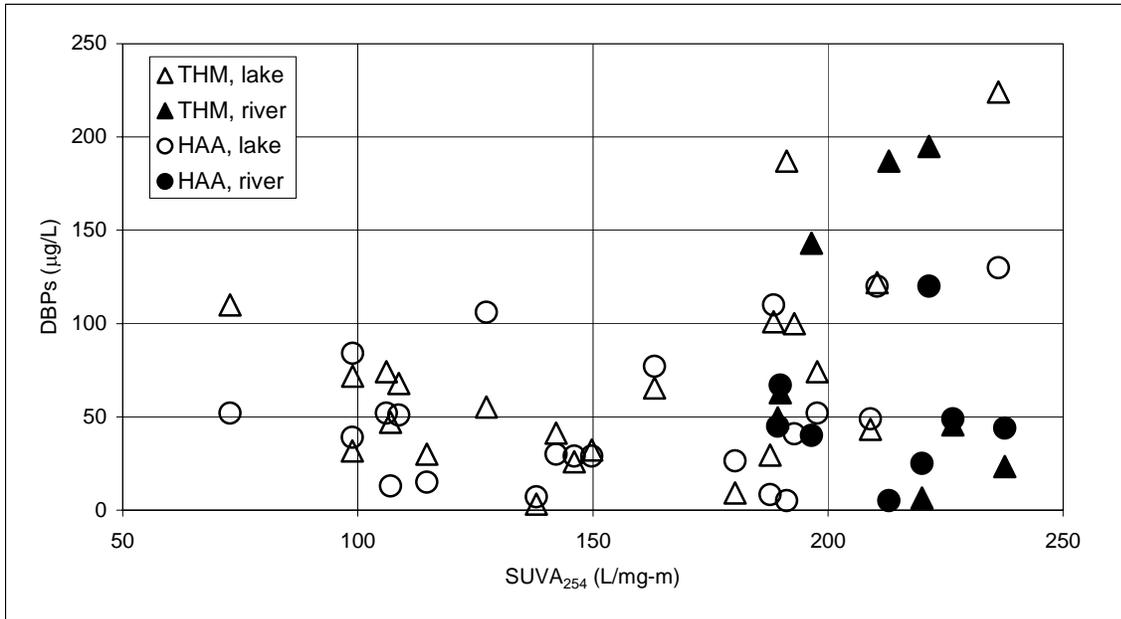


Figure 8. THM and HAA concentrations in finished water versus SUVA in raw water.

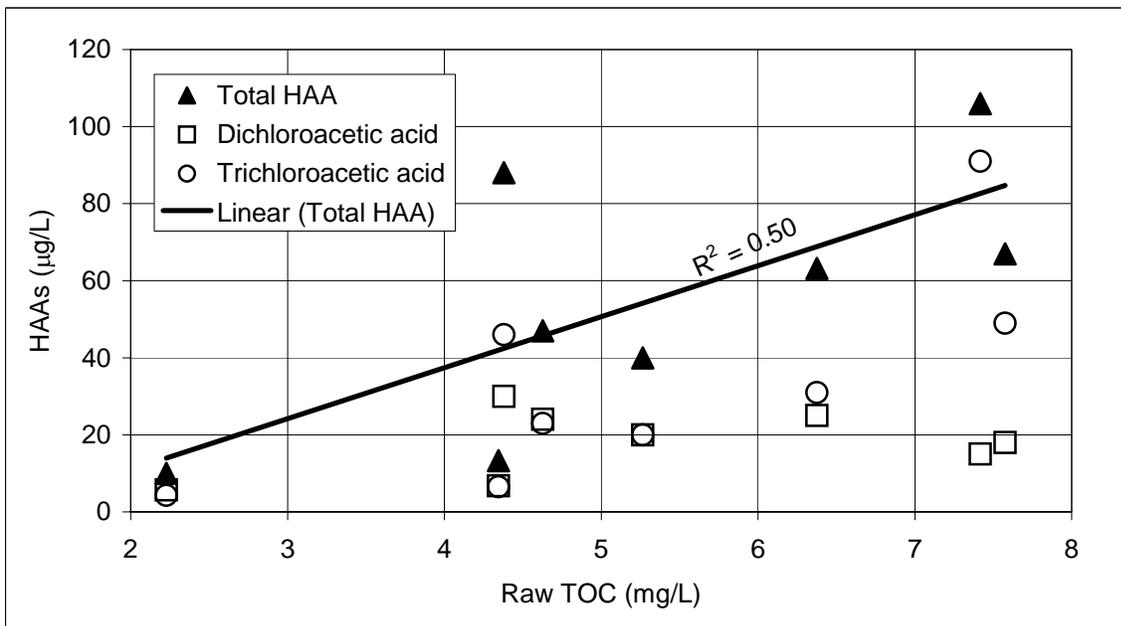


Figure 9. HAA components and total HAA concentration of the finished water versus TOC concentrations of the raw water, second sample round.

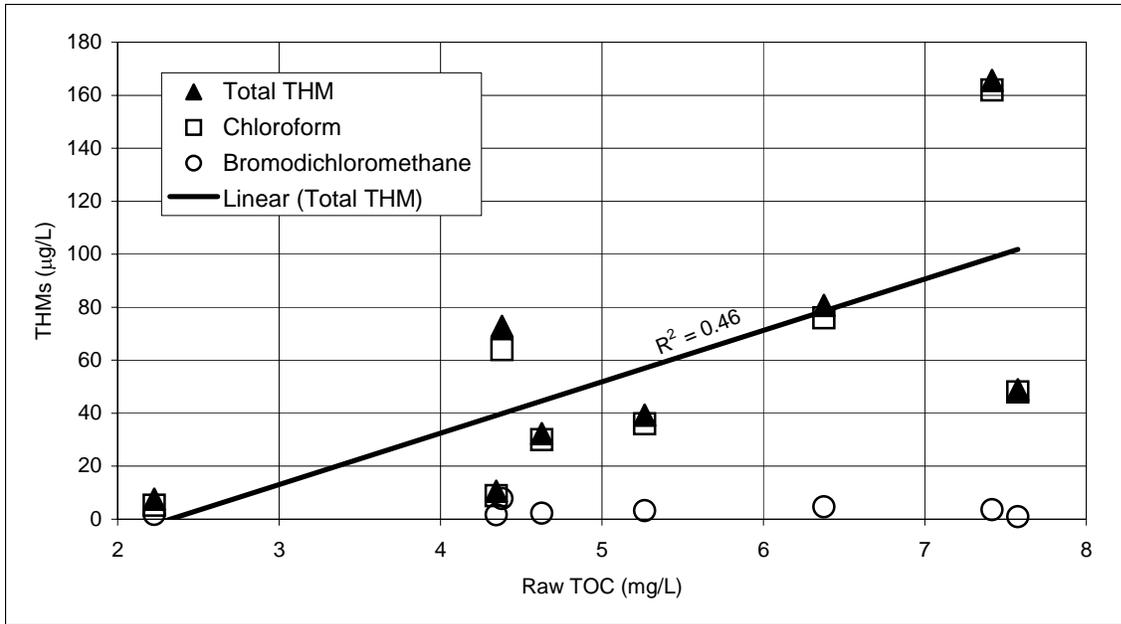


Figure 10. THM components and total THM concentrations of the finished water versus TOC of the raw water, second sample round.

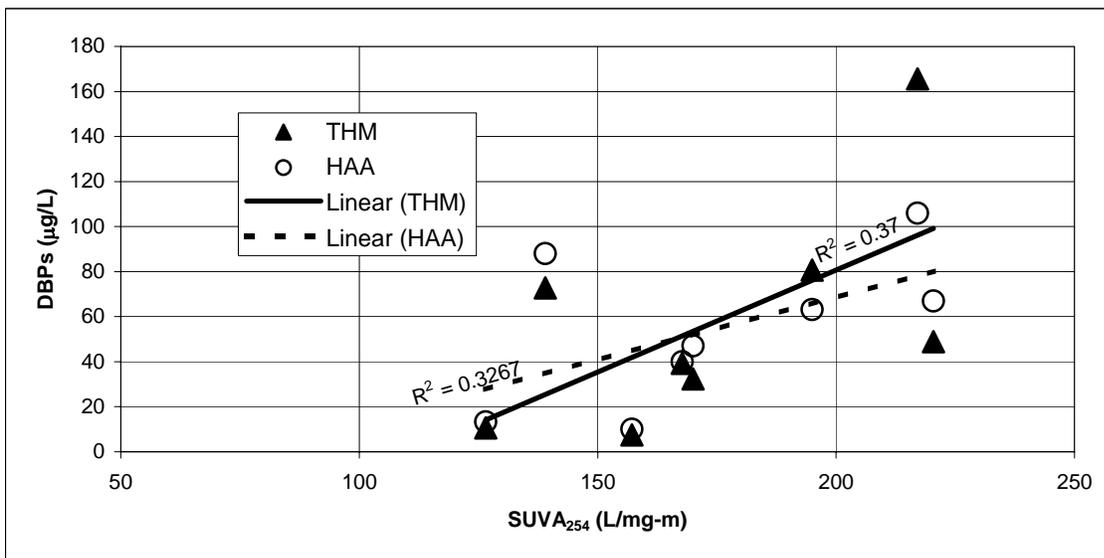
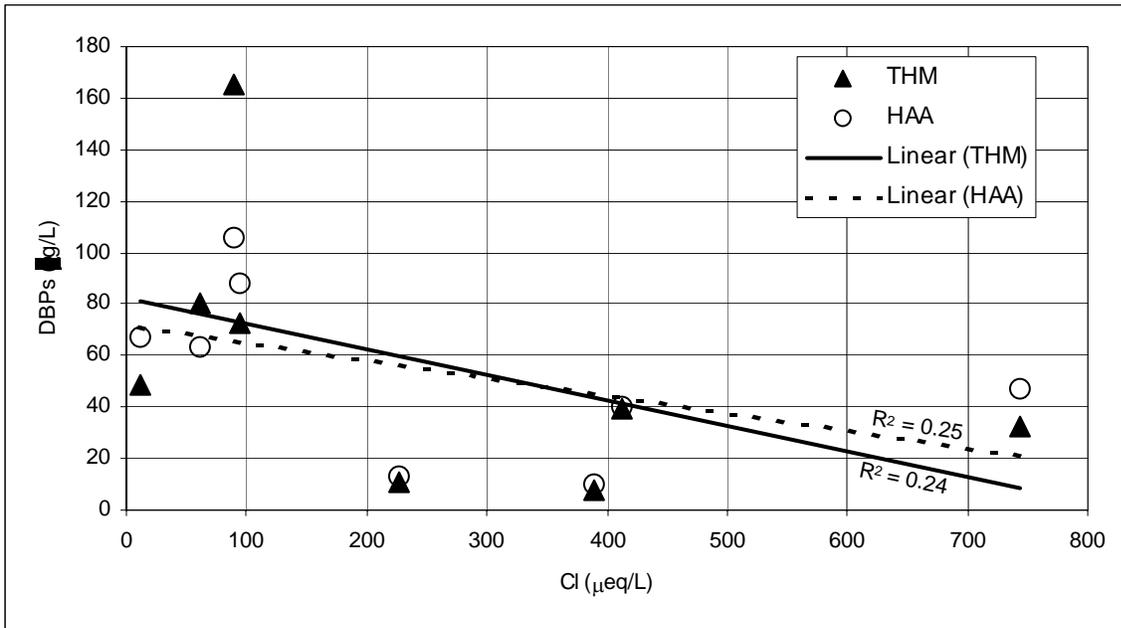


Figure 11. Total THM and HAA concentrations in finished water versus SUVA<sub>254</sub> in raw water, and trend lines, second sample round.



Figure

Figure 12. Total THM and HAA concentrations in the finished water versus Cl concentrations and trend lines, second sampling round.