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EFFECT OF AMMONIA ON THE FORMATION OF THMs IN DRINKING WATER CHLORINATION --- A CASE STUDY

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ABSTRACT

In a water supply system total Trihalomethanes (THMs) content in drinking water may vary considerably depending on water quality and treatment conditions. Most urban water treatment plants generally use chlorine as disinfectant. The effect of various parameters on the formation of THMs has been extensively studied for the last couple of decades throughout the world where it was found almost universally that an increase in the value of any of these parameters has positive effects on the formation of THMs except ammonia which exert negative effect in the process, but surprisingly this fact failed to attract due focus in THM study worldwide. In the backdrop of presence of high concentration of ammonia content in the drinking water source of Dhaka, especially in dry months of the year, the objective of this investigation was to evaluate the effect of ammonia on the formation of THMs in the water samples of the largest water treatment plant at Dhaka in Bangladesh. The water samples were tested for a wide range of parameters including pH, ammonia, UV_{254}, TOC, DOC and bromide following the standard methods of testing. THMs was measured by THM plus Method (Method:10132) using UV-VIS Spectrophotometer DR 6000(HACH, USA). A detailed quantitative study of the effect of ammonia on the trihalomethanes balance is obtained when water is chlorinated under varying conditions is discussed. Experiments at a residual free chlorine dose of 0.89 & residual total chlorine dose of 1.29 mg /L utilizing treated waters from the supply system having dissolved organic carbon (DOC) contents of 6.0 mg/L was applied with several doses of ammonia namely 0.0, 0.5, 1.0, 5.0, and 10.0 mg/L. The presence of ammonia at different concentrations was found to significantly reduce THMs formation at applied chlorine concentration but not elimination.

THMs formation decreased continuously with increasing ammonia concentration, and the decline is sharp during relatively low concentration of ammonia up to 3 mg N/L then remained near to flat slope after ammonia exceeded 3 mg N/ L. It is noticed that the formation of THMs significantly reduced with increasing ammonia concentration from 0 to 10 mg N/L in chlorinated drinking water. The suppression of THMs was prominent with increasing ammonia concentration from almost zero to 5 mg N/L. However, the formation of THMs remain low and constant after ammonia addition over 5 mg N /L. A general correlation for the prediction of THMs as a function of ammonia is presented. Its predictions are in good agreement with the observed results regardless of the fact that it is limited for this study. It is suggested to conduct similar study with diversified data. When the unwanted presence of significant amount of ammonia along with other organic pollutants in the river water as well as the threat of formation of perceived THMs are present in a water supply system like Dhaka extensive study should be taken on how to handle / utilize the source water ammonia judicially during water treatment like use of this ammonia to form chloramine as disinfectant of the treatment system.

Keywords: *Ammonia, chlorination, drinking water, Natural organic matter (NOM), THMs formation*

1. INTRODUCTION

Potable water is of utmost importance for all human beings Potable water by default satisfies .some defined criteria, like free from deterrent materials, colour, turbidity, odour, taste, pathogens, harmful metals, organic substances etcWater is synonim to life yet it can be detrimental if not treated properly . It may be resulted from . before human consumtionthe improper choice of treatment processes particularly disinfection)Bujar et al., 2013)

In water treatment, disinfection is one of the most significant milestones in the public health advances of the last century. It is a very important & indispensable water treatment process for drinking water safety, as it inactivates pathogens from drinking water. Since the beginning of the 20th century the disinfection process has been carried out routinely to exterminate and inactivate the pathogens from water used for drinking purposes. The disinfectants also serve as oxidants and are used for oxidizing iron and manganese; removing taste and colour; improving efficiency of coagulation and filtration; algal growth prevention in sedimentation basins and filters, and biological regrowth prevention in the water distribution system (U.S. Environmental Protection Agency (USEPA,1999).

For drinking water treatment, the universal disinfectants are chlorine and its compounds. Higher oxidizing potential of chlorine ensures a minimum level of residual chlorine throughout the distribution system and protects against recontamination of microbes. On the other hand it cost the lowest. These properties have made it popular in the treatment industry. During the conventional treatment process, chlorine may be added to drinking water as chlorine gas, dry calcium hypochlorite or sodium hypochlorite solution. Each of these methods of disinfection forms free chlorine when disinfectants are applied to water, destroys pathogenic organisms (American Chemistry Council [ACC], 2018).

Chlorine remains the most effective and inexpensive disinfectant albeit there are several disinfectants and disinfection strategies like chlorination, chlorine dioxide, chloramination, ozonation, granular activated carbon with post chlorination, ultraviolet ray radiationare available .etc)Reiff, 1995; Clark et al., 1998; Chowdhury et al., 2007.(

1.1 The Challenge of Disinfection Byproducts

Disinfection byproducts (DBPs) are chemical compounds formed unintentionally when oxidants like chlorine and other disinfectants react with naturally occurring organic matter (NOM) in source water. Although, acute microbial contamination protection is the top priority, drinking water systems must also control DBPs. Even though, chlorination worked wonderfully as disinfection process, the use of chlorine as disinfectant allows the formation of carcinogenic halo-organic compounds as disinfection by-products (DBPs) which poses potential health risks (Rook, 1974; Beller et al., 1974). Among the identified DBPs, the trihalomethanes (THMs) are widely reported in drinking water supply (Richardson 1998, 2005). The species distribution of TMHs includes chloroform ($CHCl_3$), bromodichloromethane ($CHBrCl_2$) (BDCM), dibromochloromethane ($CHBr_2Cl$) (DBCM)and bromoform ($CHBr_3$) (Singer & Reckhow, 1999).

The sum of chloroform, BDCM, DBCM, and bromoform concentrations is referred to as total trihalomethanes or TTHM (Bujar et al., 2013). With the concern of THMs carcinogenic affect to human health, albeit having limited data, EPA set the first regulatory limits for TTHM in 1979 with its Total Trihalomethane (EPA, 1979).

Research on the formation and control of the THMs is required although the concentration of THMs in drinking water is generally relatively low. It is due to the fact that a perceived suspected carcinogen can be easily distributed through the public water supply system.

Looking at potential health risks of these THMs, many countries, after the USA, around the world, i.e., the U.K., Japan, France, Australia, etc. and World Health Organization (WHO), have regulated these

compounds in their drinking water quality standard. The US Environmental Protection Agency has set a maximum contaminant level (MCL) of 80 µg/L for THM (USEPA, 1999). Canada recently set drinking water guidelines stating a TTHM maximum acceptable level of 100 µg/L (Federal-Provincial Subcommittee on Drinking Water (Canada), 1996). Moreover, recently most of the European countries regulated THMs in their water at the MCL of 100 µg/L (European Economic Community Directive [EECD],1997). In Bangladesh, there is no standard level of total THM or its species as such for drinking water but for chloroform it is set at 90 ppb (Department of Environment [DoE], 1997).

The formation of THMs and the factors influencing its formation including the raw water quality parameters before chlorination have been a research focus during the past three/four decades. Around 22 parameters have been identified in a number of widely discussed modeling approaches for THM, and the major factors which drew attention of the researchers are chlorine dose, chlorine demand, total chlorine, TOC, DOC, UV_{254}, pH, temperature, reaction time and bromide ion concentration. (Stevens & Symons, 1977; Engerholm & Amy, 1983; Amy et al., 1987; Singer, 1994; Black et al., 1996; Cowman & Singer, 1995; Rathbun, 1996; Carlson & Hardy, 1998; King et al., 2000; Westerhoff et al., 2000, Rodriguez et al., 2000; 2002; Clark et al., 2001; Gang et al., 2002; Sadiq & Rodriguez, 2004; Sohn et al., 2004; Chen et al., 2008, Motasem et al., 2013).

The general behaviors of these parameters in THMs formation are briefly summarized as follows:

- With increase in pH, NOM and reaction time the THMs formation increases
- With temperature increase, reaction rates generally increase, yielding higher rates of formation of THMs.
- The chlorine demand increases with increasing NOM content, which is measured as Ultraviolate absorbance at 254 nm (UV_{254}), TOC or DOC, as well as slow reactions in the pipes and pipe materials itself. The TOC, DOC and UV_{254} have strong correlations with THMs formation, while TOC, DOC and UV_{254} are strongly correlated with each other.
- In areas where bromide ion concentrations are high (e.g., coastal areas), it can be an important factor in the modeling of THMs formation. Increased formation of brominated THMs is resulted due to the presence of bromide ions, resulting in a corresponding decrease in the formation of chlorinated THMs.

It is worth mentioning that all these parameters have a positive effect that is with the increasing quantity of these parameters in the water the formation of THM increases; only one parameter whenever present in the water is found to decelerate or suppress the formation of THM which is ammonia. Furthermore, from the perspective of DBP control the reaction between ammonia and chlorine is a very important aspect (Amy et al., 1984). The effect of ammonia concentration on the formation of DBPs remained out of focus in most of the cases during the chlorination of drinking/natural waters (Tieu et al., 1982; Sohn et al., 2004, Sun et al.,2009) or has not been studied as extensively as the other factors related with THM formation mechanism.

Indeed, a very limited number of references are found in the literature regarding the role of ammonia in the THM formation mechanism when compared with the references of other associated parameters. The reason for such ignorance may be developed due to the fact that the presence of ammonia in the river water or in the source raw water for drinking are very negligible especially in the developed countries. Furthermore, a health-based guideline has not been derived by WHO since ammonia is not of direct importance for health in the concentrations to be expected in drinking-water (World Health Organization [WHO] ,1996). Unfortunately, for couple of decades the excessive presence of ammonia in river water and / or drinking water sources has become a serious problem along with other water quality problems in the drinking water industry of the developing countries (Bittner A., 2000; Fu Qing et al., 2012; Yuk F H., Mekkonnen M T., 2018). This is also especially true for the capital city of Bangladesh (Motallib et al.,2016; Serajuddin et al., 2018) As such due attention to this parameter is also needed in the study of THM formation mechanism. In general, the studies on drinking/natural water have demonstrated that with increasing ammonia concentration the concentration of total THMs is decreased (Tieu et al., 1982; Duong et al., 2003; Sohn et al., 2004; Yang & Shang, 2004).

Though a large portion of the city population consumes chlorinated public drinking water yet almost no previous attempts to assess, monitor, and predict THM concentrations or parameters influencing thereof in public drinking water have been reported in this country, lest the study on the influence of ammonia in the formation of THM. Hence our research may be regarded as almost the first one of this kind.

1.2 Objective of the Study

The objective of this study is to investigate the role / influence of ammonia at different concentrations on the formation of THMs during the chlorination process of Sitalakhya river water being used for producing drinking water in the largest drinking water treatment plant in Dhaka.

2. MATERIALS AND METHODS

It is found that the main variables which affect formation of THMs are contact time, temperature, pH, total organic carbon, and chlorine dosage (Mishra et al., 2016). However, the effect of ammonia in drinking water in formation of THMs rarely drew attention of most of the researchers. The aim of this research was to assess the importance and the effect of ammonia, the largely ignored variable in the formation of THMs in the drinking water of Dhaka city. In order to observe the importance of ammonia in THMs formation the study has been performed in experimental plant scale. Measurements were performed during 2019-20. UV-Vis spectrophotometry was used while determination of THMs.

2.1 Study Area

Saidabad water treatment plant is the largest treatment plant of Bangladesh located in t capital city Dhaka. The raw water is drawn from the Sitalakhya River in eastern periphery of the city. Dhaka is one of the most densely populated cities in the world with more than fifteen million population. The city's climate has seasonal variation. Annual average rainfall of the city is 2,123 millimeters and 87% of the annual rainfall occurs during monsoon season (between May and October). The mean temperature of the city is 26^0C with monthly average temperature ranging from 19^0C in January to 29^0C in May. With such seasonal variations, the raw water quality of the rivers also varies significantly. Water samples from the Saidabad water treatment plant was collected and tested for this study (Figure. 1) (Serajuddin et al., 2018)

2.2 Sample Collection and Analysis

Presently Dhaka city with a population of fifteen million people, receives about 22% of drinking water from Sitalakhya river as the raw water source. Due to very nature of the raw water even accidental inadequate and improper treatment technique may causes occurrence of THMs in the water supplied. The largest drinking Water Treatment Plant of Dhaka draws raw water from the Sitalakhya river and was established and put into operation in 2002. Dhaka Water Treatment Plant uses the conventional water treatment processes, except with a unique biological pretreatment unit.

To conduct this study adequate quantity of supplied treated water sample was collected from residential area and five samples were prepared with five different doses of ammonia, namely, 0.001,0.5,1.0,5.0,10.0 mg/L. The primary characteristics of the sample water was as follows: pH=7.08, H_3 –N = 0, Free chlorine=0.89 mg/L, Total chlorine= 1.29 mg/L, DOC=6.0 mg/L, UV254 = 0.0616, DO=9.06, Temperature= 23.0 0 C. The laboratory analysis was done on December 12, 2019.

The water samples were tested for a wide range of parameters including pH, ammonia, UV254, TOC, DOC and bromide. Sample containers were dark amber glass sterile bottles with PTEF lined screw caps. The samples were cooled immediately in an ice cooler and then brought back to SWTP laboratory. Whatman GF/F filter (Whatman Japan Ltd., Tokyo, Japan, nominal pore size 0.7 mm) was used for filtering the water. Measurement of pH & Temperature was done using multimeter sensION+ MM150 (HACH, USA). Ammonia concentration in this study was measured by USEPA approved Nessler method (HACH method 8038) using a DR-6000 UV-VIS spectrophotometer (HACH, USA). UV_{254}

samples, after filtration, were analyzed using a DR-6000 UV-VIS spectrophotometer (HACH, USA). DIUF water was used as blank.

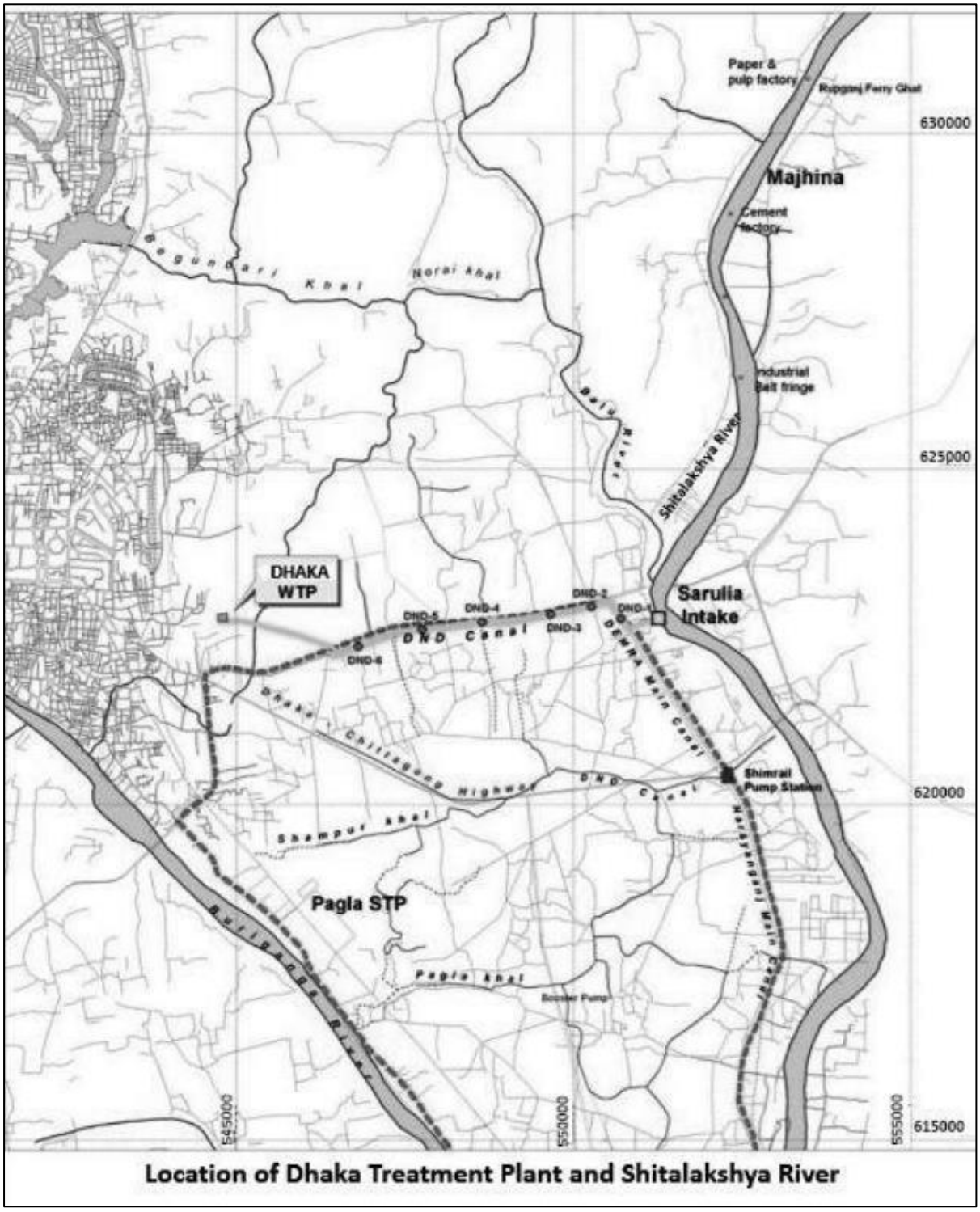

Figure 1: Raw water source and its transmission network from Sitalakhya River to WTP

TOC concentration was measured by using HACH method no.-10129 LR, Test 'N Tube™ Vials (0.3 to 20.0 mg/L) DR-6000 UV-VIS spectrophotometer (HACH, USA). After TOC measuring, Dissolved Organic carbon (DOC) was determined by acidifying the filtered samples, in which Inorganic Carbon (IC) converted into Carbonic acid (H_2CO_3), also known as dissolved inorganic carbon (DIC). DOC concentration was measured by using HACH method no.-10129 LR, Test 'N Tube™ Vials (0.3 to 20.0

mg/L) DR-6000 UV-VIS spectrophotometer (HACH, USA). Bromide was measured by HACH method no. 8016 DPD Method (0.05 - 4.50 mg/L Br_2) using DR-6000 UV-VIS spectrophotometer (HACH, USA). Trihalomethane concentration was measured by HACH method no.-10132 THM Plus™ Method (10 - 600 µg/L CHCl3) using DR-6000 UV-VIS spectrophotometer (HACH, USA).

Among the THMs species chloroform is always found with the highest concentration in drinking water, and very often more than 90% of the total concentration of the THMs are represented by this species (Bhujar, 2013). In non-coastal zones, bromoform generally is not found where study showed that amongst various THMs species, the contribution of chloroform was highest (97.99 to 98.71%) (Mishra, 2016). In our study in absence of the facility to test the other three forms of THM only chloroform was measured, and an extra five percent was added to ascertain the total THM and Chloroform & THM are used interchangeably in this paper. In the water samples under this study the bromide concentration level were beyond the detectable limit.

3. RESULTS AND DISCUSSIONS

The effect of ammonia on THM formation in chlorinated drinking water sample collected from the largest water treatment plant at Dhaka is shown in Figure 2. It can be seen from the graph that with increasing ammonia concentration the trends for the THMs formation are decreasing continuously, and the decline is sharp during relatively low concentration of ammonia up to 3 mg/L and then maintains a gentle slope after ammonia exceeded 3 mg- N/ L. This is a similar phenomenon to that of chlorination with ammonia or mono chlorination in drinking water (Carlson and Hardy, 1998; Nissinen et al., 2002) and also in secondary effluent chlorination of wastewater (Sun et al., 2009).

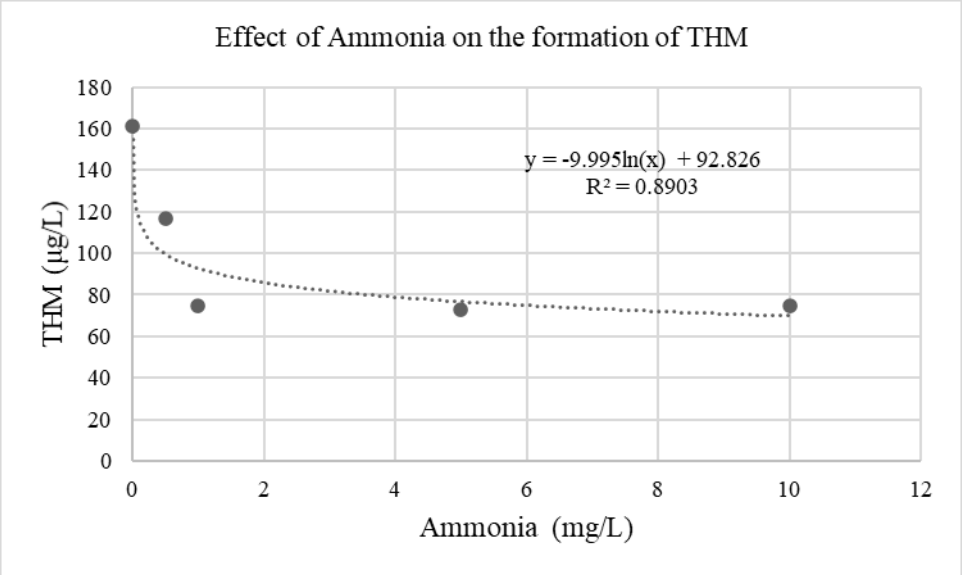

Figure 2: Effect of Ammonia on the formation of THM

Regression analysis suggest there exists an excellent logarithmic negative correlation between the concentration of ammonia and the concentration of the resulted THMs, with the increase in the ammonia concentration the resulting THMs concentration decreases. The R^2, coefficient of determination value of the correlation equation is 0.89 (r=0.94) which signifies excellent correlation between the two parameters.

During chlorination, ammonia when present in the water can react with free chlorine quickly before other pollutants can react and change to combined chlorines (chloramines) in water (Scully & Hartman, 1996; Fayyad & Al-Sheikh, 2001). Ammonia's effects on the formation of THMs are dependent on their reactivity to chlorine in competing with the NOM. Additionally, there activity of combined

chlorines is much weaker than that of free chlorine, and the combined chlorines are slower to form THMs during the reaction with dissolved organic matter (DOM) in water. From the present study it is found that with increasing ammonia from 0.001 to 5.0 mg N/ L in chlorinated sample of drinking water the concentration of THMs is decreased sharply. This can be explained by the Cl_2/N mass ratio determining the change from chlorination to monochloramination. When the Cl_2/N mass ratio is under 5:1 and the pH value is between 6.5 and 8.5 monochloramine is the dominating form in the chlorinated system (Qiang & Adams, 2004). In this study the initial pH of the samples was 7.08. In sample 1 the background concentration of ammonia was almost zero (0.001 mg N/ L) and its initial Cl_2/N mass ratio was 6.82:1, while there was 42% residual chlorine in the form of free chlorine. With the further addition of ammonia up to 5.0 mg N/L, the THM/$CHCl_3$ formation was significantly restrained by the monochloramine formed. Moreover, it is interesting to find that, with increasing ammonia concentration from 2 to 10 mg N/ L in the chlorinated drinking water the formation of chloroform is significantly reduced. We can infer, as above, that chloramines in the reaction system and the monochloramine dominated in the equilibrium system of chlorine. It is reported, furthermore, that chloramines are much weaker oxidants than HOCl and the reaction between the ammonia and chlorine is faster than that between chlorine and THMs precursors (Karen & Keith, 1998; Adams, 2005). Therefore, with increasing ammonia concentration from almost zero to 5 mg N/L in chlorinated drinking water the restrained of chloroform was prominent. However, after ammonia addition over 5 mg N/L the formation of chloroform was low and almost constant. From Figure 2 it can be seen that the formation of THMs in sample 2 (where Ammonia concentration 0.5 mg/L) is 27% less than the formation of THMs in sample 1 (where Ammonia concentration is almost zero, 0.001mg/L) & in sample 3 (where Ammonia concentration 1.0 mg/L) formation of THMs is 27% less than sample 2 & 54% less than sample 1.In sample 4 (where Ammonia concentration is 5 mg/L) the THMs formation is only 1% less than that of sample 3.This is because that with increasing ammonia concentration the free chlorine was consumed to the greatest extent which is evident from observing the free Cl_2 concentration which is zero after reaction time. It is reported in a study on chlorination of waters containing humic substances that theoretically, after free chlorine is converted to chloramines the production of THMs should stop (Amy et al., 1984), but with chloramine formation the THMs is found to form in parallel. The hydrolysis of monochloramine to form free chlorine will be reduced with increasing ammonia concentration though monochloramine is thought to hydrolyze before reacting with DOM to form HOCl (Yang et al., 2007). As such, with ammonia addition over 5 mg N/ L, the low and constant level of THMs formed in chlorinated drinking water. This similar phenomenon is seen in chlorination with ammonia or monochloramine in secondary effluent chlorination of wastewater (Sun et al., 2009).

The water treatment industry may now renew their investigation to check Chloramination as an alternative chlorination like as in 1930s. Chloramines are less expensive among the alternative drinking water disinfectants in addition to the well-known advantages of using chloramines for disinfection like stability of residuals in the system, chemical inertness towards THMs.

4. CONCLUSIONS AND RECOMMENDATIONS

It is observed in this study that Ammonia whenever present during drinking water treatment with chlorine containing organic/ humic substances reduces THMs production substantially as was seen in some other studies elsewhere. However, cent percent elimination of THMs was not observed. During the present study when the ammonia concentration was gradually increased during the chemical reaction of drinking water with chlorine the rate of formation and concentration of THMs are found to be restrained. It is also noted that ratio of chlorine to ammonia was a factor in suppressing THMs. However, increase pattern of ammonia do not follow linear relation in suppressing formation of THMs as was also seen in previous studies elsewhere. Due to the high ammonium levels in water, chlorine is mainly transformed to chloramine.

This behavior of ammonia in the chlorination of organic matters in drinking water has vital consequences for **urban** water companies that use a drinking water source enriched with low to high substantial levels of ammonia depending on seasons for example the river water such as the drinking

water source of Dhaka. This ammonia converted into chloramine during chlorination may judiciously be considered for chloramine disinfection as a THMs control strategy.
The initial formation rate of THMs competes with the relatively rapid reaction of chloramine formation despite that the THM formation reaction is comparatively slower. After conversion of chlorine to chloramine THMS production supposed to stop but THMs forms in parallel with chloramine formation.

While many aspects of earlier THMs research worldwide are relevant to THMs formation and control in Bangladesh, some characteristics of the Bangladesh water industry visibly differ to other parts of the world, and therefore presents unique challenges, such as source waters that are rich in ammonia as well as THMs precursors, long distribution systems, and Bangladesh's climate. When the unwanted presence of significant amount of ammonia along with other organic pollutants in the river water as well as the threat of formation of perceived THMs are present in a water supply system like Dhaka extensive study should be taken on how to handle / utilize the source water ammonia judicially during water treatment like use of this ammonia to form chloramine as disinfectant of the treatment system.

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