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Assessment of disinfection of water containing high concentration of ammonia by chlorination

Subrata Paul, Md. Arif Istiaque Khan, Mashiat Hossain, Muhammad Ashraf Ali*

Bangladesh University of Engineering and Technology, Department of Civil Engineering, Dhaka-1000, Bangladesh, email: skpaul.2k10@gmail.com (S. Paul), khanarifistiaque@gmail.com (M.A.I. Khan), mashiath11@ce.buet.ac.bd (M. Hossain), Tel. +8801713-043325, email: ashraf@ce.buet.ac.bd (M.A. Ali)

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ABSTRACT

Surface waters in Bangladesh and many developing countries are characterized by high concentrations of organic matter and ammonia, particularly during the dry season. In the presence of high concentration of ammonia, break point chlorination often becomes impractical at treatment plant because of very high chlorine dose requirements; therefore, "chloramination" (i.e., chlorination at chlorine to ammonia ratio below "break point") is most often carried out. It has been found that "chloramination" at low chlorine to ammonia molar ratios (e.g., less than 0.6:1.0) could be effective for disinfection for water containing lower level of fecal contamination, even when initial ammonia concentration is relatively low. However, "chloramination" may not be effective for water with higher level of fecal contamination. Even before breakpoint is reached, a noticeable amount of free chlorine has been found to exist in water; and both mono-chloramine and this free chlorine act as disinfectant. For a known initial ammonia concentration, the optimum disinfection should be achieved at chlorine to ammonia weight ratio of around 5.0:1.0 to 6.25:1.0. But if ammonia concentration in water is high, chloramination at such chlorine to ammonia ratio would produce very high concentrations of chloramines, exceeding the WHO/USEPA guideline values. For water with such high initial ammonia concentration, a lower "chlorine to ammonia" ratio should be considered for chloramination.

Keywords: Chlorination; Chloramines; Disinfection; Free chlorine; Ammonia

1. Introduction

In Bangladesh, contribution of surface water in water supply is increasing. Surface water accounts for about 22% of Dhaka WASA water supply, which is expected to reach 70% by 2025, because Dhaka WASA is changing its focus to surface water instead of groundwater. Disinfection of surface water involves the oxidation of organic and inorganic substances and the elimination of bacteria and viruses in the raw water [1]. For disinfecting surface water, chlorine is the most widely used disinfectant because it is readily available, cost effective, easily applied and is more efficient than other disinfectants such as ozone, chlorine dioxide and potassium permanganate [2]. Surface waters in Bangladesh are characterized by high concentrations of organic matter and ammonia [3], and concentration of both these parameters varies significantly with season. Ammonia concentration exceeding 20 mg/L has been reported for Sitalakhya River water during dry season [4], which is the raw water source for the largest water treatment plant of Dhaka, the Saidabad Water Treatment Plant (SWTP). This gives rise to a lot of problem, including problem with disinfection. In order to address this issue, the SWTP installed a nitrification pre-treatment plant; the nitrification plant is designed to reduce ammonia concentration from a maximum of 15 mg/L to 4 mg/L at a flow rate of 450 MLD. Such high concentration of ammonia is unusual in source waters typically used as raw water

*Corresponding author.

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at water treatment plants. While chloramines (prepared through controlled mixing of ammonia and chlorine) have been successfully used for disinfection of water [5], there is limited literature on the formation and effectiveness of chloramines during chlorination of water containing naturally occurring ammonia. In the presence of high concentration of ammonia, break point chlorination often becomes impractical because of very high chlorine dose requirements. Chlorination of high-ammonia water results in formation different quantities of mono-, di- and tri-chloramines, depending on chlorine to ammonia ratio [6], and these species have different disinfection potential. If chlorine to ammonia molar ratio is maintained below 1:1, it is called "combined residual chlorination" or chloramination. On the other hand, if it is allowed to be over 1.6:1, it is called "breakpoint chlorination" [7]. In the presence of variable concentration of ammonia in water, it becomes difficult to adjust chlorine dose to achieve desired disinfection. Ammonia may lessen the formation of THMs by reacting with free chlorine [8–13]. Mono-chloramine offers several advantages over chlorine; most essentially lower THMs and HAAs by-product concentrations [6,14–15]. Chloramines provide the advantage of increased residual activities in the distribution system and control of bacterial biofilm re-growth in the distribution system [16]. On the other hand, di- and tri-chloramines give rise to unpleasant odor in treated water. The types of chloramines formed vary with chlorine dose, and often the outcome is uncertain disinfection and generation of odor problem. Previous research conducted on chloramination indicate that though chloramines have less disinfection capability than chlorine dioxide and free chlorine [17], they have significant potential in disinfecting bacterial population in raw water on a plant scale [18].

According to WHO, the guideline value of maximum acceptable concentration (MAC) for chloramines in drinking water is 3 mg/L [19,20]. This MAC is based on a risk evaluation for monochloramine only, as monochloramine is usually the predominant chloramine species and information on dichloramine and trichloramine toxicity is insufficient to establish guidelines. Besides, United States Environmental Protection Agency (USEPA) in 1998 and Water Quality Association (WQA) in 2013 provided a standard of 4.0 mg/L for total chloramines concentration in drinking water [21,22] and this level can be exceeded under emergency conditions such as a major line break or a natural disaster. Previous studies suggest that monochloramine species are in predominant form in Cl_2:NH_3 molar ratio 1.2:1 [23] but di- and tri-chloramines take the lead after gradual increase of Cl_2:NH_3 weight ratio till the breakpoint. In the presence of variable ammonia concentration, formation of chloramines species varies; higher ammonia concentration indicates greater chloramines concentration. Therefore, it is necessary to understand the disinfection capability of chloramines at concentrations within acceptable limits for these species. The primary objective of this study was to determine the dominating chloramines species and their disinfection capability at different chlorine to ammonia ratios and also to provide guideline for chlorine dose for effective disinfection in the presence of high concentration of ammonia in water.

2. Methods and apparatus

In order to understand the disinfection capability of chlorine in the presence of ammonia, disinfection experiments were carried out at different chlorine to ammonia ratios using raw water samples containing different levels of FC (fecal coliform). For preparing raw water samples, groundwater (free from chlorine, ammonia and FC) collected from a deep tube well was amended with different but small quantities of domestic wastewater (as a source of fecal coliform). Low/negligible concentration of nitrogenous and organic compounds in groundwater is expected to limit possible formation of nitrogenous DBPs and organic chloramines upon addition of chlorine [24]. Domestic wastewater was used as the source of fecal coliform because untreated domestic sewage is also the main source of pollution of river water surrounding urban centers in Bangladesh. Table 1 shows the characteristics of groundwater used for preparation of raw water samples.

2.1. "Combined residual" chlorination experiments

To understand disinfection ability of different chlorine species which form at low chlorine to ammonia ratios, batch experiments were conducted. Raw water samples prepared as described above were taken in a series of beakers. Required quantities of ammonia stock solution, prepared with reagent grade ammonium chloride, were then added to the beakers to achieve desired level of initial ammonia concentration. Then, chlorine stock solution, freshly prepared with bleaching power each day, were added to the beakers to achieve the desired chlorine dose and chlorine to ammonia ratio. The contents of the beakers were thoroughly mixed with a glass rod for two minutes, and then they were allowed to stand for 30 minutes. Samples were then collected from the beakers for analysis of FC by Membrane Filter Method. All experiments were carried out at room temperature (22–25°C).

Table 1
Characteristics of groundwater used for preparation of raw water samples

Sl. No.	Parameter	Concentration
1	pH	7.1
2	Carbon-di-oxide, mg/L	71.0
3	Total alkalinity, mg/L as $CaCO_3$	216.0
4	Total hardness, mg/L as $CaCO_3$	256.0
5	Chloride, mg/L	55.0
6	Total Ammonia as NH_3-N, mg/L	< 0.017
7	Nitrate (NO_3-N), mg/L	0.30
8	Nitrite (NO_2-N), mg/L	< 0.008
9	Orthophosphate, mg/L	0.325
10	Sulfate, mg/L	14.2
11	Iron (Fe), mg/L	0.01
12	Manganese (Mn), mg/L	0.022
13	Arsenic (As), mg/L	< 0.001

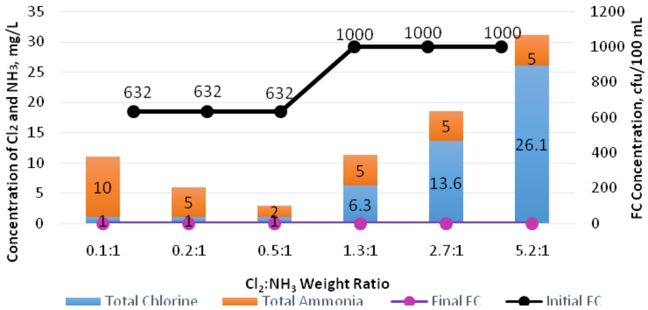

Fig. 1. Disinfection of water with low initial FC concentration (632 to 1,000 cfu/100 mL) at low chlorine to ammonia ratio.

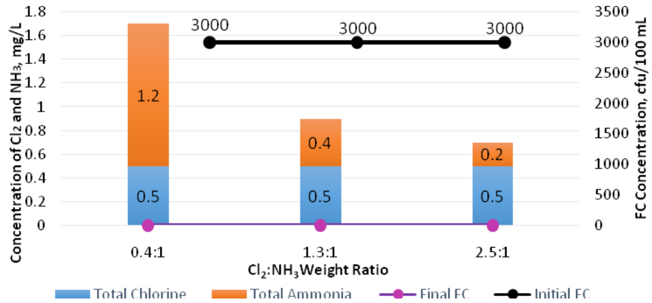

Fig.2. Disinfection of water with low initial FC concentration (3,000 cfu/100 mL) at low chlorine to ammonia ratio.

2.2. Combined residual and breakpoint chlorination experiments

These chlorination experiments were carried out in Jar Test apparatus. Groundwater samples containing a fixed concentration of ammonia (achieved through addition of ammonia stock solution) were taken in six beakers. Chlorine stock solutions were then added to the beakers such that Cl_2:NH_3 molar ratio in the beakers varied from 0.60:1.0 to 2.4:1.0. The contents of the beakers were slowly mixed in the jar test apparatus for about 20 minutes. Subsequently, samples were taken out from each beaker for determination of total residual chlorine, free chlorine and chloramines. Residual chlorine was measured by DPD colorimetric method; free chlorine, mono-, di-, tri-chloramines were measured by DPD titrimetric method [20]. After that, a fixed quantity (a few milliliters) of domestic wastewater was added to each beaker (as well as to a "blank" beaker) as a source of FC. This allowed exposure of water containing approximately a fixed initial concentration of FC to different levels of chlorine and chloramines species in different experimental beakers. The contents were then mixed in the Jar Test apparatus, and allowed to stand for 30 min.

Finally, samples were taken from each beaker for determination of FC by Membrane Filtration Method. The residual FC concentration in each beaker was then matched with the chlorine/chloramines species concentration in the respective beakers to assess the disinfection capacity of different chlorine/chloramines species. No effort was made to adjust pH of water, and pH of water samples varied from 6.6 to 7.5. All experiments were carried out at room temperature (22–25°C).

3. Results and discussion

3.1. Disinfection with combined residual chlorine

Figs. 1 and 2 show the results of chlorination experiments carried out with relatively low initial FC concentration (up to 3,000 cfu/100 mL) at relatively low chlorine to ammonia ratios (maximum weight ratio 5.2:1) that would promote formation of chloramines. Total chlorine concentration and total ammonia concentration at each weight ratio are shown by stacked columns. Initial ammonia concentration in these experiments varied from 0.2 to 10.0 mg/L. Fig. 1 shows the results of two sets of experiments where initial FC concentrations were 632 and 1,000 cfu/100 mL; while Fig. 2 shows the results of another set of experiments carried out with initial FC concentration of 3,000 cfu/100 ml. The figures show that in all cases zero residual FC concentration could be achieved. Thus, it appears that for samples containing low initial FC concentrations, effective disinfection could be achieved even at very low chlorine to ammonia ratios that result in formation of chloramines. For these experiments, however, chloramines concentrations were not measured.

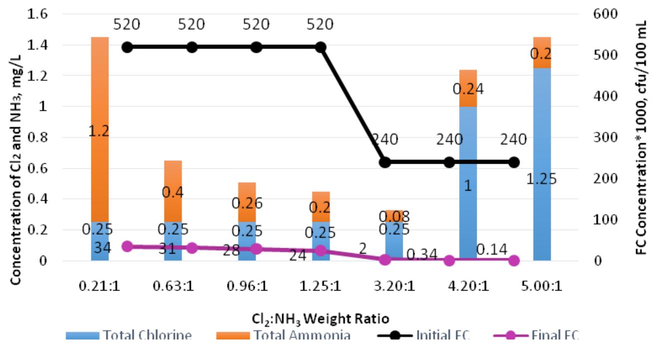

Fig. 3. Disinfection of water with relatively high initial FC concentration at low chlorine to ammonia ratio.

Fig. 3 shows the results of chlorination experiments carried out with relatively high FC concentration (up to 5,20,000 cfu/100 mL) at relatively low chlorine to ammonia ratios (maximum weight ratio 5.0:1) that would promote formation of chloramines. Fig. 3 shows the results of two sets of experiments where initial FC concentrations were 520,000 and 240,000 cfu/100 mL. Fig. 3 shows that in all cases, significant residual FC concentrations were detected. This means that if initial FC concentration is very high (which is the case for water of many rivers in Bangladesh during dry season), chlorination at lower chlorine to ammonia ratios would not be effective for achieving complete disinfection (i.e., zero residual chlorine). In other words, chloramines may not be effective for disinfection of water with high initial FC concentrations. For these experiments, however, chloramines concentrations were not measured. Additional experiments were then carried out to assess effectiveness of disinfection of chloramines species.

3.2. Formation of chloramines species and their disinfection capacity

Figs. 4–7 show the results of experiments carried out to assess effectiveness of chlorine and chloramines species in removing FC from water. Fig. 4 shows the results of a set of experiment, where water samples containing different levels of chlorine and chloramines (achieved through addition of chlorine at different doses to water containing 0.75 mg/L of ammonia, as described in Section 2.2) were inoculated with very high concentration of FC (6,40,000 cfu/100 mL). The Fig. 4 shows concentration of chlorine and chloramines species as well as residual FC concentration in each of the 6 beakers. In this set of experiment, highest chloramines (including mono-chloramine) concentration was reached at a chlorine-to-ammonia weigh ratio of 5.0:1.0; monochloramine and dichloramine concentrations at this ratio were 1.60 and 0.4 mg/L, respectively; free chlorine concentration of 0.4 mg/L was also detected.

Although complete disinfection was not possible before "breakpoint", the FC concentration was significantly reduced from a very high value of 6,40,000 cfu/100 mL to 40 cfu/100 mL at chlorine to ammonia weight ratio of 5.0 to 1.0; at this ratio, monochloramine reached its peak at 1.6 mg/L. These results confirm significant disinfection capability of chloramines, particularly monochloramine. Complete disinfection (i.e., zero residual FC) was achieved after "breakpoint" in the presence of higher concentration of free chlorine, confirming stronger disinfection capability of free chlorine under similar conditions. Before the breakpoint is reached, chloramines species should be dominant forms of chlorine species in water. However, Fig. 4 shows that appreciable amount of free chlorine remains in solution before "breakpoint"; this free chlorine must have contributed to disinfection, together with monochloramine, before "breakpoint".

Fig. 5 shows the results of a set of experiments where water samples containing different levels of chlorine and chloramines (achieved through addition of chlorine at different doses to water containing 1.0 mg/L of ammonia, as described in Section 2.2) were inoculated with FC at a concentration of 75,000 cfu/100 mL. It shows that before "breakpoint", monochloramine concentration increased as chlorine to ammonia ratio increased, reaching its peak value of 3.1 mg/L at a chlorine-to-ammonia weight ratio of 6.75 to 1.0. It also shows that increasing reduction of FC with increasing monochloramine concentration. At the peak monochloramine concentration of 3.1 mg/L, the FC concentration was significantly reduced from a high value of 75,000 cfu/100 mL to 4 cfu/100 mL, confirming significant disinfection capability of monochloramine. However, as noted above, some free chlorine was also detected in water samples prior to reaching "breakpoint", which must have contributed to disinfection.

It is evident that as ammonia concentration in raw water increases, peak concentration of chloramines (particularly monochloramine), prior to reaching break point, increases upon chlorination. For example, for initial ammonia concentration of 0.75 and 1.0 mg/L, highest concentration of monochloramine recorded was 1.6 and 3.1 mg/L, respectively. As noted earlier, ammonia concentration in surface water in Bangladesh is often very high; at Saidabad Water Treatment Plant, a nitrification plant has been installed to reduce ammonia concentration in raw river water from 15 mg/l to 4 mg/l. Therefore, similar additional experiments were carried out with higher initial raw water ammonia concentration of 3.0 mg/l and 5.0 mg/L, in order to assess formation of chloramines and their disinfection capabilities.

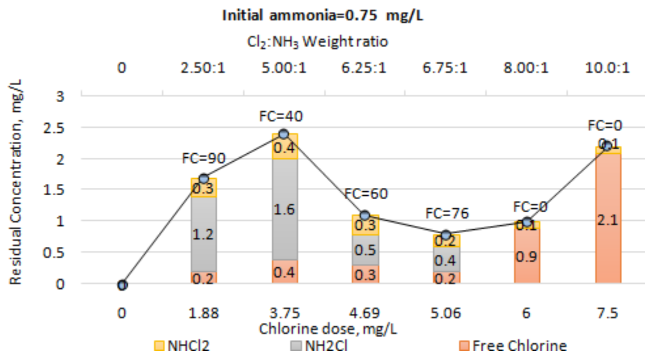

Fig. 4. Results of disinfection experiment, where water samples containing different levels of chlorine and chloramines (achieved through addition of chlorine at different doses to water containing 0.75 mg/L of ammonia, as described in Section 2.2) were inoculated with very high concentration of FC (6,40,000 cfu/100 mL).

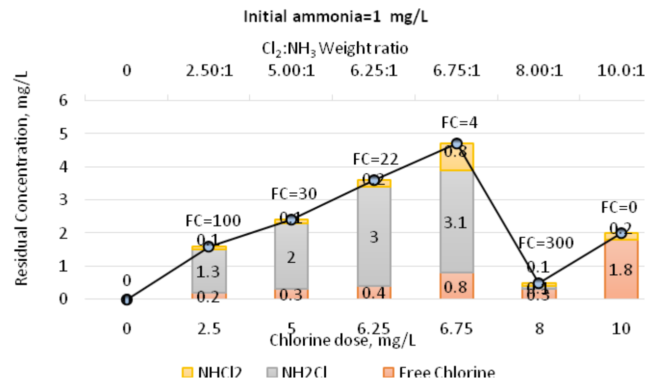

Fig. 5. Results of disinfection experiment, where water samples containing different levels of chlorine and chloramines (achieved through addition of chlorine at different doses to water containing 1.0 mg/L of ammonia, as described in Section 2.2) were inoculated with FC at a concentration of 75,000 cfu/100 mL.

Fig. 6 shows the results of a set of experiments where water samples containing different levels of chlorine and chloramines, achieved through addition of chlorine at different doses to water containing 3.0 mg/L of ammonia, were inoculated with a high FC at a concentration of 4,40,000 cfu/100 mL. It shows a peak monochloramine concentration of 5.8 mg/L at a chlorine-to-ammonia weight ratio of 6.25 to 1.0; as before monochloramine concentration increased with increasing chlorine to ammonia ratio. The most effective disinfection (i.e., zero residual FC) was achieved at the peak monochloramine concentration, even at very high initial FC concentration. It should be noted however that the peak monochloramine concentra-

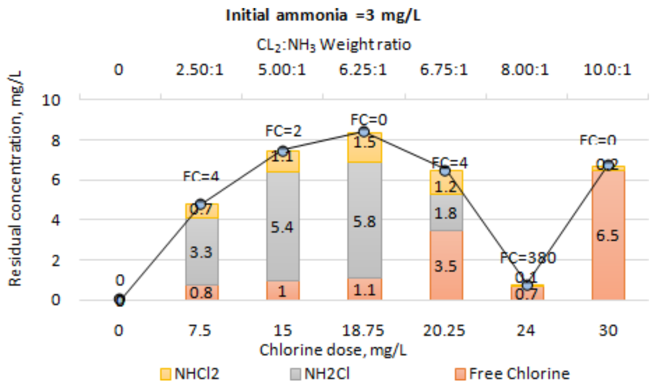

Fig. 6. Results of a set of experiment where water samples containing different levels of chlorine and chloramines, achieved through addition of chlorine at different doses to water containing 3.0 mg/L of ammonia, were inoculated with a high FC at a concentration of 4,40,000 cfu/100 mL.

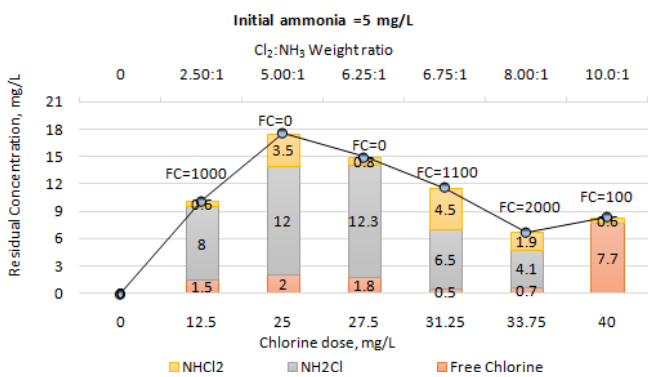

Fig. 7. Results of disinfection experiment, where water samples containing different levels of chlorine and chloramines, achieved through addition of chlorine at different doses to water containing 5.0 mg/L of ammonia, were inoculated with a very high FC concentration of 8,40,000 cfu/100 mL.

tion (i.e., 5.8 mg/L) reached exceeded the WHO/USEPA drinking water guideline/standard for monochloramine/chloramines by a large margin. Fig. 6 also shows effective disinfection after "breakpoint", but at a very high chlorine dose of 30 mg/L.

The results of another set of experiments are shown in Fig. 7 where water samples containing different levels of chlorine and chloramines, achieved through addition of chlorine at different doses to water containing 5.0 mg/L of ammonia, were inoculated with a very high FC concentration of 8,40,000 cfu/100 mL. It also shows effective disinfection (i.e., zero residual FC) during "chloramination" even in the presence of very high initial FC, but in the presence of very high monochloramine concentration (≥ 12 mg/L) exceeding drinking water guideline/standard. Thus, while monochloramine serves as an effective disinfectant, particularly at high concentrations, operators at treatment plants must consider the allowable limits of chloramines during fixing the dose of chlorine for water containing high concentration of ammonia.

The results of batch experiments presented in Figs. 4–7 show the presence of some free chlorine along with chloramines at low chlorine to ammonia ratios. Analysis of two treated water samples collected from the Saidabad Water Treatment Plant (SWTP) during dry season (when raw water contains high concentration of ammonia) confirmed simultaneous presence of free chlorine and chloramines during chloramination. Ammonia, free chlorine and total chloramines concentration in the two water samples were 4.80 mg/L, 0.5 mg/L, and 3.6 mg/L; and 4.04 mg/L, 0.4 mg/L, and 3.3 mg/L, respectively.

3.3. Chloramines and free chlorine as a function of ammonia concentration in water

For a fixed chlorine to ammonia ratio, peak monochloramine concentration increases as initial ammonia concentration increases. On the other hand, for a fixed initial ammonia concentration, peak monochloramine concentration is reached at chlorine to ammonia weight ratio of about 5.0:1.0 to 6.25:1.0. Therefore, for a known initial ammonia concentration, the most effective disinfection through "chloramination" should be achieved at chlorine to ammonia weight ratio of around 5.0:1.0 to 6.25:1.0, when monochloramine concentration reaches its peak. But for higher initial ammonia concentration, chloramination at such chlorine to ammonia ratio would produce very high concentrations of chloramines, exceeding the WHO/USEPA guideline/standard values. For such high initial ammonia concentration, lower chlorine to ammonia ratio may be considered for chloramination. Additional experiments were carried out to determine chloramines concentration as a function of initial ammonia concentration for two different "chlorine to ammonia" ratios.

Figs. 8 and 9 show chloramine concentrations as a function of the initial ammonia concentration for two "chlorine to ammonia" ratios. At a low ammonia concentration of 0.25 mg/L, monochloramine concentrations are quite low, 0.06 mg/L and 0.2 mg/L, respectively at chlorine to ammonia weight ratios of 2.5:1 and 5.0:1; At these concentrations, the corresponding total chloramines concentration are 0.11 mg/L and 0.4 mg/L, respectively. As initial ammonia concentration increases, concentration of monochloramine also increases. At ammonia concentration of 5.0 mg/L, concentration of monochloramine reached 8.0 and 12.0 mg/L, respectively at chlorine to ammonia ratio of 2.5:1 and 5.0:1.

From Figs. 8 and 9 it is easy to identify residual chloramines levels that are expected during chloramination of water containing different concentrations of ammonia. For example, if ammonia concentration in water is 2.0 mg/L, chloramination could be carried out at chlorine to ammonia weight ratio of 5.0:1, without significantly exceeding the WHO standard for chloramines. However, for ammonia concentration of 3 mg/L, the chlorine to ammonia ratio may have to be brought down in order to limit chloramines concentration close to WHO guideline value. It should be kept in mind though that monochloramine would be more effective in disinfecting water containing relatively lower level of FC, as was observed from batch experiments carried out in this study.

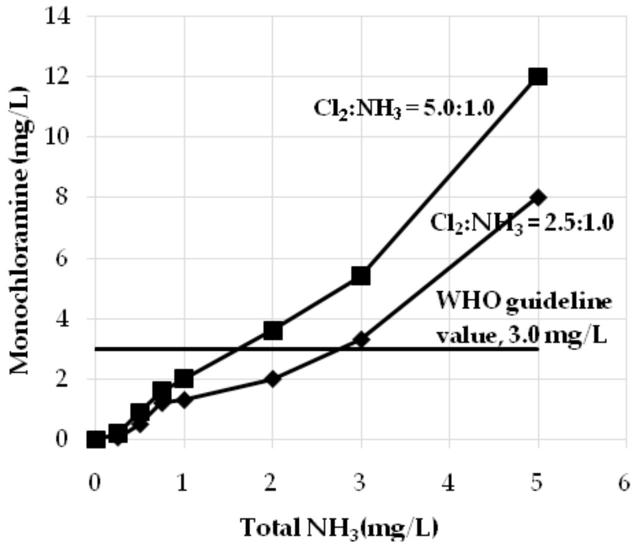

Fig. 8. Concentration of monochloramine produced during chlorination of water samples, with various initial ammonia concentrations, at two different Cl_2:NH_3 weight ratios (i.e., 2.5:1.0 and 5.0:1.0).

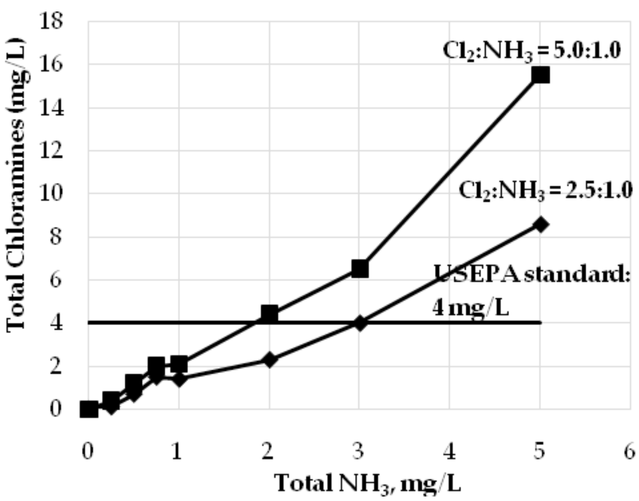

Fig. 9. Concentration of total chloramines produced during chlorination of water samples, with various initial ammonia concentrations, at two different Cl_2:NH_3 weight ratios (i.e., 2.5:1.0 and 5.0:1.0).

4. Conclusion

In the presence of naturally occurring high concentration of ammonia in water, "breakpoint chlorination" often becomes impractical due to very high chlorine dose requirements. In such cases, combined residual chlorination or "chloramination" becomes the only option. Batch disinfection experiments carried out in this study suggest that chloramination with very low chlorine to ammonia weight ratios (e.g., less than 2.5:1.0) may not be effective for disinfection (i.e., for achieving zero residual FC) of highly contaminated water with high concentration of FC, irrespective of initial ammonia concentration. Disinfection becomes more effective as chlorine to ammonia weight ratio increases up to about 6.25:1.0 due to the presence of the dominant chloramine species, monochloramine. It has also been found that for Cl_2:NH_3weight ratio below 6.25:1 (i.e., before "breakpoint" is reached), appreciable amount of free chlorine remains in solution (together with chloramines) and contribute to disinfection. Presence of free chlorine along with chloramines in two water samples collected from the Saidabad Water Treatment Plant (SWTP) during dry season (when raw water contains high ammonia) confirms simultaneous presence of free chlorine and chloramines in water during "chloramination" at water treatment plant. Therefore, for water containing high concentration of ammonia, chloramination with chlorine to ammonia weight ratio between weight ratio 2.5:1–6.25:1 (molar ratio 0.6:1–1.5:1) could be effective for disinfection. But for higher initial ammonia concentration, chloramination at higher chlorine to ammonia ratio would produce very high concentrations of chloramines, exceeding the WHO/USEPA guideline/standard values for monochloramine/chloramines. For water containing high concentration of ammonia and for which "breakpoint chlorination" is not feasible, the chlorine dose should be set such that it does not result in chloramines concentration exceeding the corresponding drinking water standards/guidelines. It should be noted however that presence of organic matter, especially nitrogenous organic matter, would complicate the disinfection scenario with possible formation of nitrogenous DBPs and organic chloramines [24,25]. This issue needs to be studied in greater details.

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