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Effect of bromide and other factors on brominated trihalomethanes formation in treated water supply in Jordan

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ABSTRACT

Chlorine disinfection of drinking water containing natural organic matter leads to the formation of brominated trihalomethanes (THMs) in the presence of bromide ions. In the present study, the results of an experimental investigation into the factors forming THMs in the presence of bromide ions are presented. The experiments were conducted using samples collected from Zai water treatment plant in Jordan under different operating conditions such as reaction time, temperature, pH, TOC, and the ratio between bromide ions and initial chlorine dose. The experimental results showed that the formation of all brominated THMs, mainly $CHBr_3$), increased with increasing of any of these parameter values and decreased chloroform formation. $CHCl_2Br$ and $CHClBr_2$ increased with increasing the experimental parameters and thereafter remained constant or slightly decreased. Moreover, n(Br) values which represents bromine incorporation into THMs are almost in the range from 2.5 to 2.7. However, the values of n(Br) increased with increasing bromide concentration and remained constant or slightly decreased with increasing contact time under the studied range of bromide ion concentrations.

Keywords: Trihalomethanes; Brominated THMs; Brominated DBPs; Drinking water distribution system; Bromine; Jordan

1. Introduction

Some hazardous compounds can be formed as a result of chlorination process in water due to the reaction between chlorine and natural organic matter (NOM) [1]. Disinfectant by products (DBP's) have several potential health risks and it is necessary to keep them at the allowable limit [2,3] Trihalomethanes (THMs) are halogen-substituted single-carbon compounds with the general formula CHX_3, where X may be fluorine, chlorine, bromine, or iodine, or a combination

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thereof. The four types of THMs that have been widely reported in drinking water supply are bromoform ($CHBr_3$), dibromochloromethane (DBCM) ($CHBr_2Cl$), bromodichloromethane (BDCM) ($CHBrCl_2$), and chloroform ($CHCl_3$). The reaction pathways and factors influencing THMs formation are well established in the existing literature [4–11].

Bromine is more reactive agent than chlorine, and acts like a master species in the substitution process so when bromide is present in drinking water, hypochlorous acid (HOCl) oxidizes bromide ions (Br^-) to form hypobromous acid (HOBr), which is a stronger halogenating agent than HOCl [12,13]. Furthermore, the reaction shifts gradually from chlorinated THMs to brominated THMs as bromide acts like a substituting agent [14–16]. The brominated DBPs (Br-DBPs) are more toxic than their chlorinated analogs [17–19], however, to date; limited studies in the existing literature investigated the effect of bromide ions on the formation of the total THMs [20–24]. The occurrence of THMs in chlorinated waters in Jordan has been reported earlier [10,11,25]. Relatively less research has investigated the effect of bromide ions on the formation of the total THMs in Jordan under specific disinfection conditions, such as reaction time, pH, and temperature [10,11].

In 2006, the World Health Organization (WHO) has regulated the health-related guideline values for such compounds in drinking water, as shown in Table 1 [26].

The aim of the present study is to investigate the effect of bromine ion on the formation of THMs compounds in the samples collected from Zai water treatment plant in Jordan. Moreover, the effect of other parameters, such as total organic carbon (TOC), temperature, pH, the ratio between bromide ions and chlorine dose, and reaction time, on the formation of the four THMs species were also investigated.

2. Materials and methods

2.1. Water samples and conservation

The raw water is first pumped from the King Abdullah Canal in the Jordan Valley, situated 230 m below sea level, is pumped from the Deir Alla intake to Zai water treatment plant, 886 m above sea level near the city of Salt (32° 6′17.43′′N 35°43′2.13′′E). Treated water from the Zai treatment plant is then pumped to the Dabouq reservoir in West Amman, 1,032 m above sea level, as shown in Fig. 1. The capacity of the treatment plant is about 90 MCM per year [27]. The plant has a conventional method of treatment, comprising coagulation (by addition of $FeSO_4$ and cationic polymer to the rapid mixing tank), sedimentation, filtration, and chlorination.

The experiments in this study were conducted on the samples taken from Zai water treatment plant located near the city of Salt in Jordan. The procedure is based on collecting 2 L of samples from the influent (raw water) and the effluent streams (treated water following chlorination) of the plant. The samples were stored in a cooler at a given constant temperature (4°C). For THMs, measurement at each experiment after a chosen reaction time and under any operating conditions such as temperature, pH, TOC, and the ratio between bromide ions and initial chlorine dose, smaller samples were taken from the original sample in 40-ml septum vials containing few crystals of sodium thiosulfate (sufficient to eliminate any residual chlorine and thus stop the THMs formation reaction), filled to zero head space, and stored in the dark at the desired experimental conditions prior to analysis. The experimental conditions under which the effect of each parameter was studied were fixed as follows maintaining the conditions for other parameters fixed at some predetermined values.

Table 1
WHO guideline values for THMs (WHO, 2006)

Disinfectant byproducts	Guideline value (ppb)	Remarks
THMs	—	The sum of the ratio of the concentration of each to its respective guideline value should not exceed 1
TTHMs	150 μg/L until 2008 and 100 μg/L after 2008[a]	
$CHBr_3$	100	
$CHBr_2Cl$	100	
$CHBrCl_2$	60[b]	
$CHCl_3$	300	

[a] European Union guidelines.

[b] For substances that are considered to be carcinogenic.

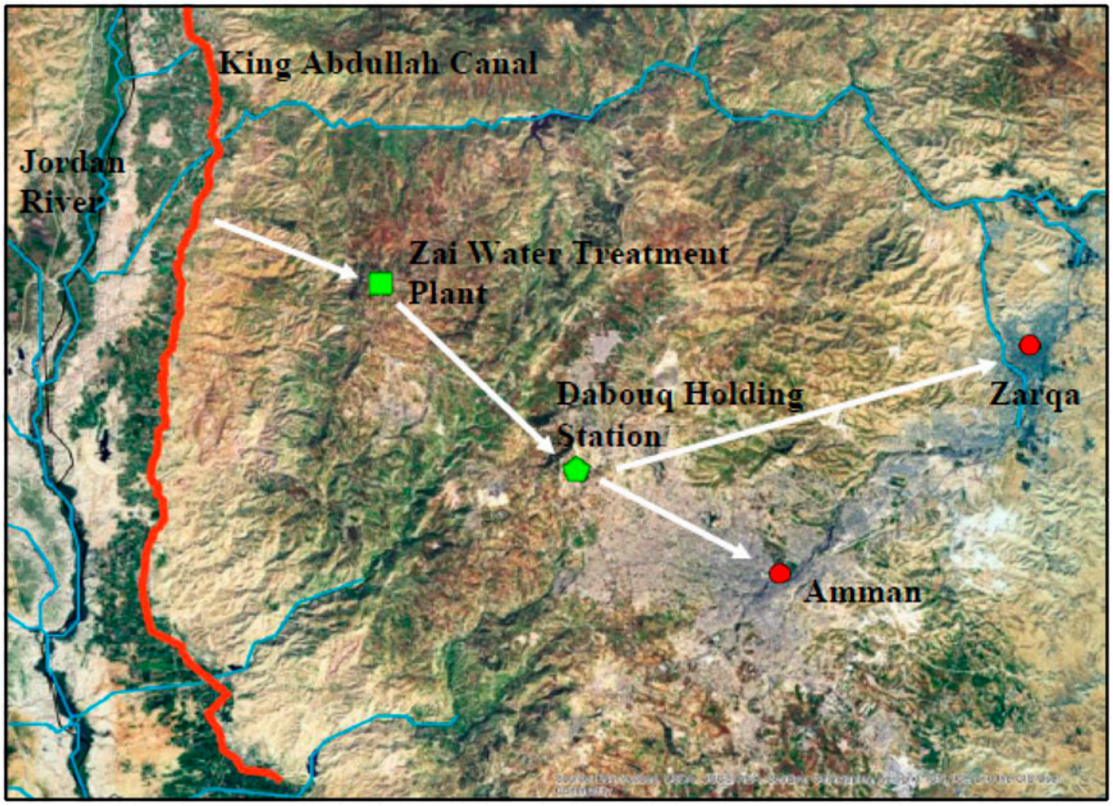

Fig. 1. The transferred water path from the KAC to the Zai water treatment plant, then to the Dabouq Holding Station, and then on to Amman and Zarqa (Image: Jordan's Water Resource Challenges and the Prospects for Sustainability [44]).

2.2. Effect of reaction time

The concentrations of THMs were determined after the following reaction times have elapsed: 0, 1.25, 5.25, 10.0, 18.0, 23.0, 29.25, and 49.25 h. The other parameters were fixed at 16˚C, pH 7.8, 1.14 mg/l TOC, 1.04 ppm free chlorine, and 0.42 ppb bromide concentration. THMs formation potential (THMFP) was evaluated for the sample taken from the chlorinated effluent stream after 168 h (7 d) according to US EPA method 551.1.

2.3. Effect of temperature

The study of the effect of temperature on the formation of THMs was carried out at temperatures of 5, 14, 28, and 40˚C. The experiment was conducted on sample of effluent stream (treated water following chlorination). The other parameters were fixed at TOC of 0.829 mg/l, pH of 7.71, initial free chlorine of 1.2 ppm, reaction time of 3 h, and bromide ion 0.5 ppb.

2.4. Effect of pH

The pH was checked and changed for each sample by the dropwise addition of 0.1 M HCl or 0.1 M NaOH. The pH values used in this part of the investigation were of 6.04, 7.15, 7.71, and 8.7 respectively. The other experimental conditions were fixed at temperature of 20˚C, TOC of 0.829 mg/l, free chlorine of 1.15 ppm, reaction time of 2.5 h, and bromide ion of 0.5 ppb.

2.5. Effect of chlorine dose

The residual chlorine checked and changed for each sample by the dropwise addition of standard chlorine solution prepared from 6% sodium hypochlorite solution following the Standard Method 5710B [28]. The concentrations of THMs were determined at the following conditions: 14˚C, 0.829 mg/l of TOC, pH 7.71, 1.5 h of reaction time, 0.5 ppb of bromide ion, and for the chlorine residuals values of 1.2, 1.75, and 2.88 ppm.

2.6. Effect of TOC

In order to study the effect of TOC on the THMs formation, drops of influent water (raw water) sample was added and mixed with a volume of chlorinated effluent stream sample to achieve and adjust the required TOC concentration for each experiment to investigate the effect of TOC. The following TOC values: 1.33, 1.82, and 2.04 mg/l, were considered in this study. With these TOC levels, the concentrations of THMs were determined for water samples under the conditions of 2 h reaction time, 15˚C temperature, and 0.55 ppb bromide ion concentration. In these experiments, there were very slight variations in pH and chlorine residuals for the three samples. These changes in pH are due to the presence of organic acids resulting in pH values of: 7.84, 8.01, and 7.88. Rapid chlorine decay reaction resulted in different levels of chlorine of 0.84, 0.7, and 0.6 ppm. For those reasons, an average pH value of 7.91 and an average chlorine residual of 0.74 mg/l were taken for those samples.

2.7. Analysis

The THMs concentrations were measured by liquid–liquid extraction followed by gas chromatography (GC) with an electron capture detector (ECD) (Model: Agilent 6890 N) based on US EPA method 551.1 [29].

Bromide concentration in water samples was measured by ion chromatogram (Model: DX-120, Dionex, USA) equipped with an IonPac AS14A analytical column and a conductivity detector (CD-20, Dionex).

The DPD-colorimetric method was employed for residual chlorine measurement following the Standard Methods 4500-Cl F [25].

The TOC analyzer employed was a DC-180 (3300) (Dorhman/Rosemount) with a non-dispersive infrared detector (NDIR).

The pH was measured using an ORION 960 Automatic System, with pH electrode (model No. 9172 BN).

3. Results and discussion

3.1. Effect of reaction time

THMs formation depends on several factors. One of them is the reaction time. Fig. 2 presents THMs' formation for each compound at different reaction time. In accordance with the literature findings [14–16], bromide ions acted like a substituting agent and, by increasing reaction time, chlorinated THMs shifted to more brominated THMs. It is also noticeable that reaction time is less effective on chlorinated THMs than that in brominated THMs. According to Fig. 1, the effects of reaction time on specific THMs showed variable results for $CHCl_3$, BDCM, and DBCM formation. It is shown in Fig. 2 that a slight decrease in $CHCl_3$ and initial increase in BDCM and DBCM, which was slightly decreased after 16 h then all of them ($CHCl_3$, BDCM, and DBCM) became constant with further increase in reaction time, while $CHBr_3$ was remarkably increased in most cases in the present study at controlled pH of 7.8, temperature of 16˚C, and at low concentration of bromide ion of 0.42 ppb. The same formation behavior was reported by Cowman and Singer [16].

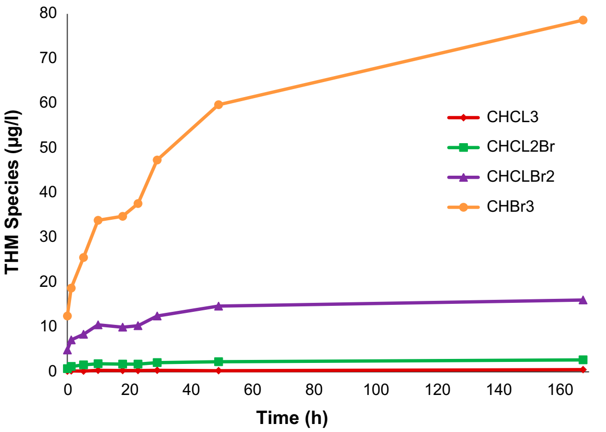

Fig. 2. The effect of reaction time on the formation of THMs species (16˚C, pH 7.8, TOC = 1.14 mg/l, free chlorine = 1.04 ppm, and bromide ion = 0.42 ppb).

It can be seen in Fig. 2 that, at longer reaction times, $CHBr_3$ has become the dominant THMs species. It is seen that $CHBr_3$ has become 79 times higher than $CHCl_3$ after 7 d of reaction time. This figure also shows that brominated THMs species increased predominantly up to 10 h and formation of $CHBr_3$ increased linearly after 50 h of the reaction time. It can be concluded that at all reaction time the formation of THMs species was in the order of $CHBr_3 > CHClBr_2 > CHCl_2Br > CHCl_3$. The increase in THMs may be attributed to the reactions between the NOM and HOBr as well as shifting in chlorinated THMs to brominated THMs as stated earlier in this study and hypothesized in previous literature, and this consequently leads to higher ratios of brominated THMs to total THMs [10–12,14,30]. The mechanism affecting the formation of the different THM species, in presence of bromide ion during water chlorination, is influenced by the fact that the HOCl rapidly oxidizes Br^- to HOBr forming a mixture of HOCl/HOBr, and these two active species react with the fast reactive sites within NOM to form brominated THMs. It is reported that HOBr is approximately 20 times more reactive with NOM than HOCl [12];

henceforth, the brominated THMs are formed first with bromine consuming the available sites on NOM.

3.2. Effect of temperature

It is generally acknowledged that chemical reactions increase with temperature, with the Arrhenius equation being widely accepted to describe such a relationship. In the present study, increasing temperature increases the rate of reaction leading to increase in THMs formation as illustrated in Fig. 3.

The highest formation rate of THMs occurs in the summer due to the high microbial activity which shifts to increase in TOC to promote THMs formation while the lowest formation rate of THMs occurs in winter due to the low microbial activity. The characteristics and composition of organic precursors have been found to be significantly influenced by microbial effect and seasonal variations [31,32]. For instance, Uyak et al. found that the reactivity of organic matter varied annually, achieving maximum concentrations in autumn when the proportion of hydrophobic material was at its peak, before falling to a minimum in winter [32].

It is seen in Fig. 3 that when temperature was increased from 5 to 40˚C $CHCl_3$ and $CHCl_2Br$ concentrations did not vary significantly with increasing temperatures. The formation of $CHClBr_2$ showed a slight increase from 8.56 to 17.33 ppb under the same conditions, while $CHBr_3$ was the dominant compound at all temperature values. At 40˚C, concentration of $CHBr_3$ was around 50 times higher than $CHCl_3$. Similar observations were made by other researchers [33]).

3.3. Effect of TOC

The presence of NOM in water is the major reason for the formation of THMs compounds in water during the reactions among chlorine, halogens, and NOM [3]. The effect of TOC on THMs was conducted for initial TOC concentrations of 1.33, 1.82, and 2.04 mg/L in this paper. As displayed in Fig. 4, the results showed that increasing TOC concentrations up to 1.82 mg/l shifted formation of THMs. All THMs species show similar trends, however, it is obvious that the most effect was observed to be on the formation of $CHBr_3$ species which is the dominant compound for all initial TOC concentrations. It is also noticed that the formation of the THMs species is in the order of $CHBr_3 > CHClBr_2 > CHCl_2Br > CHCl_3$. These findings indicate that bromide is more active than chlorine in the substitution process. However, the formation of $CHCl_2Br$ and $CHClBr_2$ showed a slight decrease in concentrations at TOC equals 2.04 mg/l, while $CHBr_3$ was the dominant compound at all TOC values.

However, in the presence of ammonia, higher concentrations of chlorinated THMs compared to brominated THMs were documented in scientific literature [34–37]. The rate constant for NH_2Cl formation between HOCl and NH_3 is three orders of magnitude larger than that of HOBr formation. Consequently, ammonia remarkably inhibits the formation of HOBr and the formation of Br-DBPs [34–37].

3.4. Effect of Br/Cl_2 ratio

Chlorine dose is one of the major precursors of the formation of THMs. In addition, the presence of bromide ions also can form THM compounds, but due to bromide

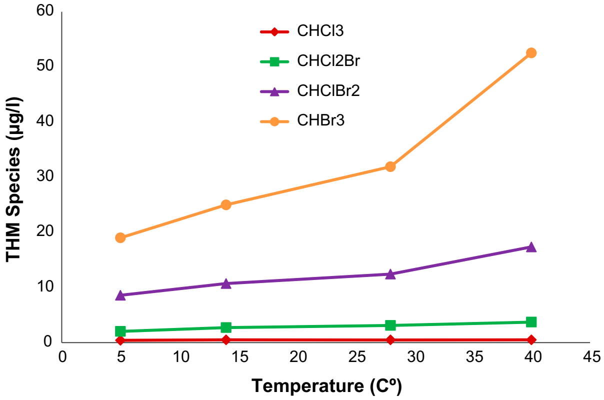

Fig. 3. The effect of temperature on the formation of THMs species (TOC = 0.829 mg/l, pH 7.71, free chlorine = 1.2 ppm, reaction time = 3 h, and bromide ion = 0.5 ppb).

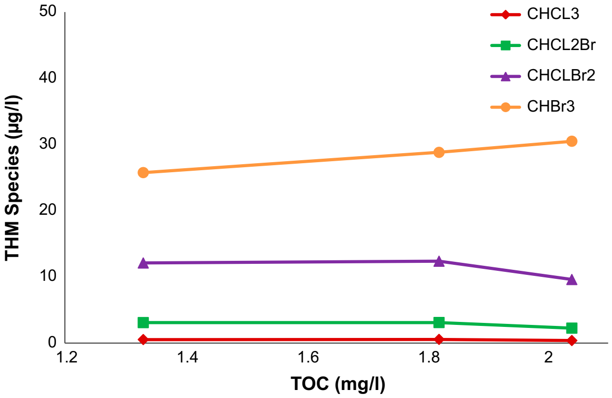

Fig. 4. The effect of TOC on the formation of THMs species (Reaction time = 2 h, 15˚C, and bromide ion = 0.55 ppb, pH).

substitution process and for a deep understanding, it is necessary to observe and identify the relationship and the effect of Br/Cl_2 ratio on the formation of THM compounds. The effect of Br/Cl_2 ratio on the formation of each THM compounds in this study is plotted in Fig. 5. Generally, increasing the initial chlorine dose will increase the formation rate of each THMs species because chlorine is more available to the oxidation process which later leads to more bromide substation to form brominated THMs. Increasing bromide to chlorine ratio leads to decrease in $CHCl_3$, $CHCl_2Br$, $CHClBr_2$, and $CHBr_3$ concentrations, where increasing bromide to chlorine ratio from 0.173 to 0.285 has remarkably decreased $CHCl_3$ concentration from 0.634 to 0.453 ppb. Also, increasing bromide to chlorine ratio from 0.173 to 0.285 has slightly decreased $CHBr_3$ concentration from 23.898 to 23.213 ppb. Overall, it seems that similar trends were observed for $CHCl_2Br$ and $CHClBr_2$.

In general, it appears that all findings related to the formation of THM compounds were shifted from chlorinated THMs compounds to brominated THMs compounds in accordance with the literature findings [12,38].

The bromine incorporation factor (BIF) provides a useful measure to explain the shifting in THMs formation at varied ratios of Br/Cl_2. For instance, if chlorine is in excess and the amount of available bromide is low, $CHCl_3$ will be the dominant species. Subsequently, the chlorinated THMs decrease as the ration of Br/Cl_2 increases [39]. However, in the present study, Br/Cl_2 ratio was investigated, while in the previous studies, the inverse ratio (Cl_2/Br) was investigated. Uyak and Toroz observed that molar TTHMs concentration increased with bromide ion concentration for a given chlorine dose under low Cl_2/Br ratio conditions (less than approximately 60 mol/mol). However, a slight decrease in molar TTHMs appeared at higher Cl_2/Br ratios (approximately 60–270 mol/mol) with increasing bromide concentration, for given chlorine dose, which is similar to the general trend of the data discussed in the present study. The relationship between Cl_2/Br ratio at higher values and molar TTHM reported here supports and expands on the work previously published by Uyak and Toroz.

3.5. *Effect of pH*

The effect of pH on the formation of $CHCl_3$, $CHCl_2Br$, $CHClBr_2$, and $CHBr_3$ was also investigated in the present paper at pH values of 6.04, 7.15, 7.71, and 8.7 while other reaction conditions were fixed at 20˚C, 0.829 mg/l of TOC, 1.15 ppm of free chlorine, 2.5 h of reaction time, and 0.5 ppb of bromide concentration. The results are shown in Fig. 6 which illustrate that increase in pH from 6.04 to 8.7 yielded the dominant species of $CHBr_3$ while $CHCl_3$ formation was the least one among other THMs species. Formation of $CHBr_3$ increased sharply after pH 7.5 while the formation of the THMs species was almost constant. Concentration of $CHBr_3$ was 30 times higher than $CHCl_3$ at pH 6.04 while this difference increased to more than 55 times at pH 8.7. Controlling chlorination pH values has been used by water utilities to reduce the formation of THMs [40]. Therefore, Fig. 6 indicates that pH control may be effective to inhibit the rate of brominated THMs formation to pH up to 8.

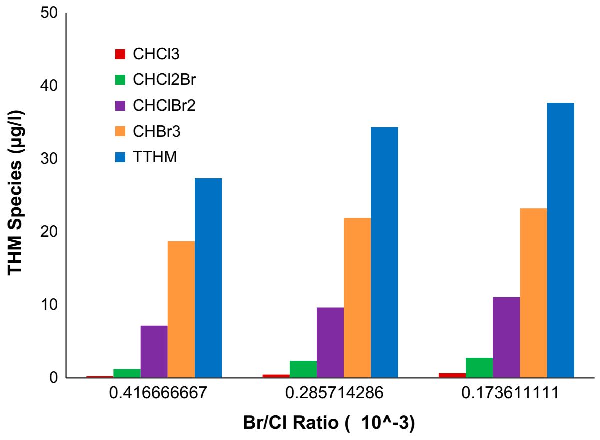

Fig. 5. The effect of Br/Cl_2 ratio on the formation of THMs species (14˚C, TOC = 0.829 mg/l, pH 7.71, reaction time = 1.5 h, and bromide ion = 0.5 ppb).

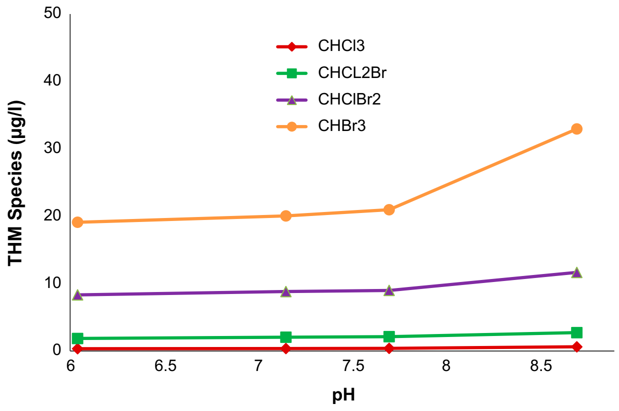

Fig. 6. The effect of pH on the formation of THMS species (20˚C, TOC = 0.829 mg/l, free chlorine = 1.15 ppm, reaction time = 2.5 h, and bromide ion = 0.5 ppb).

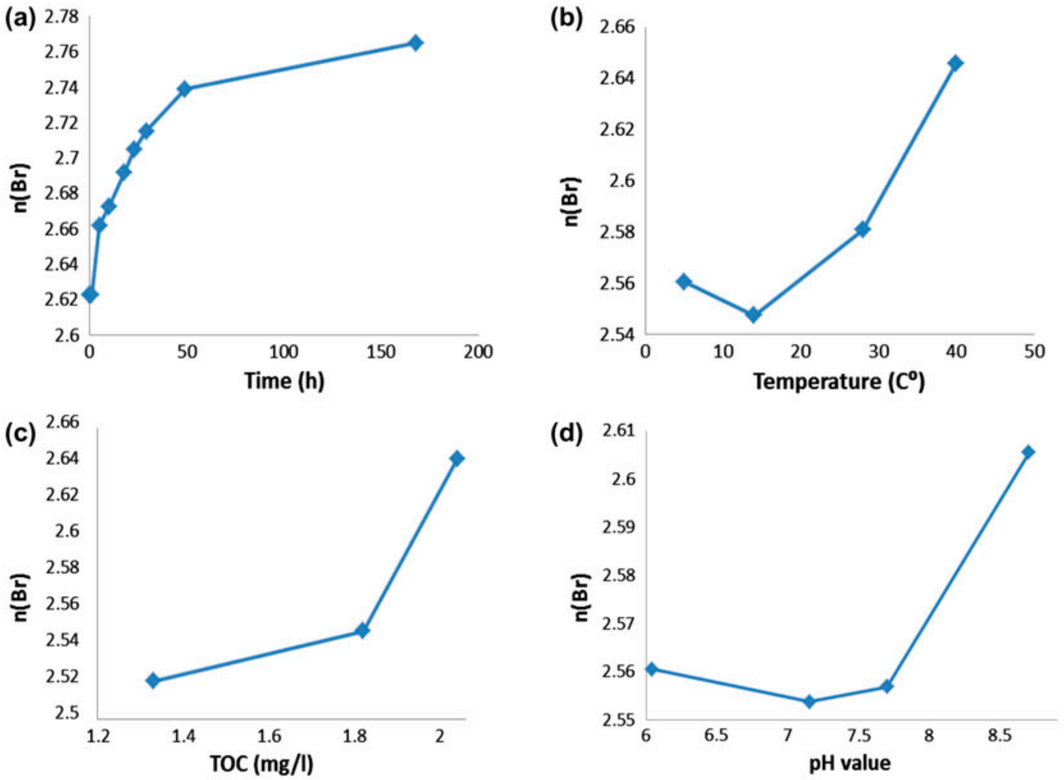

Fig. 7. Bromine incorporation into THMs vs. Reaction time (a) (16°C, pH 7.8, TOC = 1.14 mg/l, free chlorine = 1.04 ppm, and bromide ion = 0.42 ppb), (b) Temperature (°C) (TOC = 0.829 mg/l, pH 7.71, free chlorine = 1.2 ppm, reaction time = 3 h, and bromide ion = 0.5 ppb), (c) TOC (mg/l) (reaction time = 2 h, 15°C, and bromide ion = 0.55 ppb), (d) pH (20°C, TOC = 0.829 mg/l, free chlorine = 1.15 ppm, reaction time = 2.5 h, and bromide ion = 0.5 ppb).

3.6. Incorporation of bromide into THMs

Bromine is easily incorporated into organic compounds, and most of the bromine is bound to organic carbon in natural systems [41]. Bromide incorporation into THMs, which is denoted as n(Br) herein, was also studied in the present paper. The n(Br) is used to determine the formation of brominated THMs. In addition, n(Br) gives an indication of how THMs species change with certain parameter in presence of bromide ions. The values of the n(Br) can be calculated by the following Eq. (1) [42]:

$$n(\mathrm{Br}) = \frac{[\mathrm{CHCl_3}] \times 0 + [\mathrm{CHCl_2Br}] \times 1 + [\mathrm{CHClBr_2}] \times 2 + [\mathrm{CHBr_3}] \times 3}{[\mathrm{TTHM}]} \qquad (1)$$

where TTHMs (μmol/L) is the sum of the concentrations of the four species: [$\mathrm{CHCl_3}$], [$\mathrm{CHCl_2Br}$], [$\mathrm{CHClBr_2}$], and [$\mathrm{CHBr_3}$].

The relationship between n(Br) and reaction time is presented in Fig. 7(a). The results show that initially the n(Br) values increased by reaction times till 60 h, after that n(Br) values did not change significantly. The n(Br) values were reported to be between 1 and 3 [43]. However, in the present study, the n(Br) values varied from 2.62 to 2.76. This higher range of n(Br) values is attributed to the substitution of bromide. However, the prolonged contact time decreased the proportion of mono-bromine THMs, whereas it increased the proportions of di-bromine and tri-bromine THMs accordingly, as clearly seen in Fig. 2.

The relationship between n(Br) and temperature is almost similar to the relationship between reaction time and n(Br). The n(Br) values slightly increased

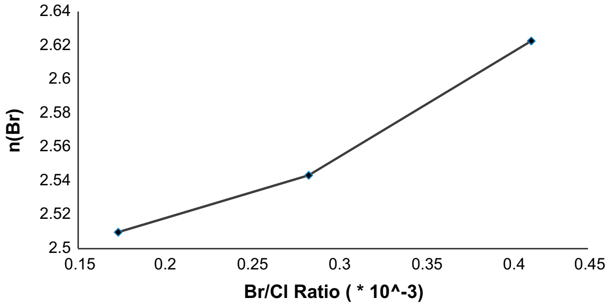

Fig. 8. Relationship between bromide incorporation into THMs and bromide to chlorine ratio (14°C, TOC = 0.829 mg/l, pH 7.71, reaction time = 1.5 h, and bromide ion = 0.5 ppb).

with increasing temperature values (Fig. 7(b)). In this case, n(Br) values ranged from 2.56 to 2.64, which are so close to the values stated in Fig. 7(a).

The results shown in Fig. 7(c) indicate above-mentioned similar trend between TOC and n(Br) values, where n(Br) values vary from 2.52 to 2.64. These results emphasize that the value of n(Br) depends mainly on the level of bromide substitution and for this reason the values of n(Br) vary at different TOC concentrations as seen in Fig. 7(c).

The relationship between pH values and n(Br) values, shown in Fig. 7(d), presented some fluctuations. The n(Br) values ranged between 2.553 and 2.605. As discussed above, all these results confirm that bromide plays a key role in the formation of THM compounds which shifted from chlorinated THMs to brominated species in the conditions studied in this study.

As might be expected, bromide incorporation into THMs yielded the n(Br) values which were to be almost constant for varying experimental conditions. Fig. 8 illustrates the relationship between ratio of bromide to chlorine and the n(Br) factor. It is noticeable that all n(Br) values for lower ratios of bromide to chlorine are so close with small differences, however, increasing bromide to chlorine ratio lead to an increase in n(Br) values. These results also indicate that brominated THMs formation is more favorable to occur in the presence of bromine.

4. Conclusion

The present study focused primarily on the influence of reaction time, temperature, TOC, pH, n(Br), and Br/Cl ratio on the formation of the major four THMs species for the samples collected form Zai water treatment plant in Jordan. Based on the experimental results and analysis the following conclusions can be drawn as the following:

(1) Bromide ions play a key role more than chlorine in the formation of THMs species and in the substitution process. Accordingly, THMs species gradually shifted from chlorinated THMs to brominated THMs (from $CHCL_3$ to $CHCl_2Br$ to $CHClBr_2$ to $CHBr_3$). $CHBr_3$ is the dominant THM compound at all values of all parameters.
(2) Increased pH, TOC, reaction time, temperature, and Cl dose values lead to increasing the rate of the formation of THMs species.
(3) The values of n(Br) are almost constant and slightly increasing for all parameters and these values are depending on the rate of bromine substitution. However, all of pH, TOC, reaction time, temperature, and Br/Cl ratio have less effective influence on the formation of chlorinated THMs than brominated THMs.
(4) The work undertaken regarding the effect of bromide ions and other factors on the formation of THMs has concentrated on laboratory-based investigations looking at conditions typically found in present day water supply systems, to understand how THMs are formed *in situ* under ambient conditions through the distribution system, to allow for an accurate portrayal of system response under typical operational conditions. With these findings, informed decisions could be made during water treatment under specific "high risk" water quality conditions.

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