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Factors affecting the formation of DBPs by chlorine disinfection in water distribution system

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ABSTRACT

Chlorine is the most commonly used disinfectant in drinking water treatment plants in China. It reacts with natural and anthropogenic organic pollutants in the water distribution system, resulting in the formation of carcinogenic disinfection by-products (DBPs). In this study, the influences of residence time, pH, chlorine dosage, water temperature, bromide concentration, and pipe material on the formation of DBPs in the water distribution system were investigated. Studies and observations showed that the formation of trihalomethanes (THMs) and haloacetic acids (HAAs) was positively correlated with residence time, chlorine dosage, water temperature, and bromide concentration during the chlorine disinfection, and the main generated DBPs were determined to be dichloroacetic acid, trichloroacetic acid, and trichloromethane. With the increase of bromide concentration, the generation of Br-DBPs substantially increased. Elevating solution pH posed a promoting effect on THMs production, while exerted an inhibitory effect on HAAs formation. The generation of THMs and HAAs in the water supply network was also affected by pipe material, and the production level followed the order of plastic steel pipe > ductile iron pipe > PE pipe. Through exploring different influencing factors, it provides a reference for the control strategy of DBPs in the water supply network and achieves the purpose of realizing the residents' drinking water safety.

Keywords: Affecting factors; Chlorine; Disinfection by-products; Water distribution system

1. Introduction

In order to maintain the stability of microorganisms in the water distribution system, chlorine disinfection is often an indispensable procedure of drinking water treatment. However, the disinfectants (chlorine, chloramine, etc.) react with natural organic matter (NOM) and anthropogenic contaminants in water to generate unexpected disinfection by-products (DBPs) during killing the pathogenic microorganism [1–3]. Previous studies have demonstrated that these DBPs in drinking water have carcinogenicity, teratogenicity, mutagenicity, and long-term exposure may pose adverse effects on human health [4–7]. The disinfectant/DBP regulations issued by the US Environmental Protection Agency stipulate that the limits of haloacetic acid (HAAs) and trihalomethane (THMs) are 60 and 80 μg/L, respectively [8]. In China, the regulations of drinking water standard for dichloroacetic acid (DCAA), trichloroacetic acid (TCAA),

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trichloromethane (TCM), bromodichloromethane (BDCM), dibromochloromethane (DBCM), and tribromomethane (TBM) were 50, 100, 60, 60, 100, and 100 μg/L, respectively. For THMs, the summed ratio of the measured concentration of each DBP to their respective limit should not exceed 1 [9].

During the chlorine disinfection, hypochlorous acid (HOCl) is its main form for disinfection reaction. Being an electrophilic reagent, HOCl easily reacts with the electron-rich moieties of NOM. HOCl interacts with NOM through a series of reactions such as the electrophilic substitution, addition, and then generate DBPs via further hydrolysis [10]. According to the kinetic analysis, chlorine can selectively react with certain chemical functional groups, and the second-order rate constant of the reaction between chlorine and organic pollutants varies widely, from 0.1 to 10^9/M/s. The reactivity of different reaction sites follows the order of reduced sulfur groups > primary and secondary amines > phenols, tertiary amines >> double bonds >> other aromatic hydrocarbons, carbonyls, amides [11]. NOM contains various functional groups such as carboxylic, aromatic, amino, and hydroxyl group, which easily react with an oxidizing disinfectant. It is worth noting that chlorine also reacts with a bromine compound or an iodine compound in water to undergo the substitution or addition [12].

The formed concentration of DBPs in the water distribution system is related to the raw water quality and water treatment process. In general, the main influencing factors include the structures and properties of NOM, disinfectant dose, solution pH, water temperature, bromide concentration, and residence time [13–18]. Most of the studies on DBPs focus on the removal efficiency by the processes in water plants, the formation mechanism, the generation of predictive models, and analytical methods [19–21]. However, the influence of various factors on the generation of DBPs in the water supply network is still at the exploring stage, and the associated studies are relatively fewer [22]. Consequently, the main purpose of this study is to examine the factors affecting the formation of DBPs by chlorine disinfection in water supply networks. Understanding the roles of these factors and their influences on the fate of DBPs will benefit utilities to minimize DBPs in the water distribution system by adopting preventive strategies for their control.

2. Material and methods

2.1. Reagents and materials

The chemicals used in this study were analytical reagent grade or higher. HPLC grade methanol, HPLC grade methyl-tert-butyl ether (MTBE), 1,2-dibromopropane, THMs mixture standards, and HAAs mixture standards were all obtained from Anpel Laboratory Technologies Inc., (Shanghai, China). Anhydrous sodium sulfate (AR, 99%) was taken from Yonghua Chemical Co., Ltd., (Jiangsu, China). The other chemical reagents were obtained from Sinopharm Chemical Reagent Co., Ltd., (Shanghai, China). Ultrapure water produced by reverse osmosis equipment was used to prepare all experimental water.

The used pipes with different materials were cut from Jiaxing water supply network, namely PE pipe, ductile iron pipe, and plastic steel pipe and their pipe diameters were found to be 20, 15, and 10 cm, respectively. The pipe wall images were presented in Fig. 1.

2.2. Experimental procedures

Several important affecting factors such as chlorine dosage, residence time, solution pH, water temperature, coexisting bromine concentration, and pipe materials were considered to examine their effects on the formation of THMs and HAAs during chlorination of unchlorinated post-filtration water. Unless otherwise specified, chlorination was carried out in a ductile cast iron pipe reactor (Fig. 2). A temperature stick was used to maintain the required experimental temperature and a magnetic stirrer was used to simulate the flow of water in a pipe. Before the chlorination, phosphate buffer solution (200 mM) was used to maintain the pH stability in the reaction system. The chlorine stock solution (1,116 mg/L as Cl_2) was added to the bottle at the required dose, and the ductile cast iron pipe reactor were sealed separately until the required reaction time (0.5, 1, 2, 4, 8, 12, and 24 h) was reached. After the reaction, the residual chlorine was quenched with an ascorbic acid solution (0.5 M) and extracted with MTBE immediately for subsequent analysis of DBPs by GC-ECD. All experiments were replicated independently at least two times, and the error bars presented in the figures represent the standard deviation among the replicates.

The single factor static experiments were employed to examine the affecting factors including chlorine dosage, residence time, solution pH, water temperature, bromide concentration, and pipe materials through varying one parameter at a time from the baseline condition: chlorine dosage (0.5, 2, and 3 mg/L as Cl_2), residence time (0.5, 1, 2, 4, 8, 12, and 24 h), solution pH (6, 7, and 8), water temperature (10°C, 20°C, and 30°C), bromide concentration (0.2, 0.5, and 1 mg/L), pipe materials (PE, ductile cast iron and plastic steel).

2.3. Analytical methods

Unchlorinated post-filtration water was collected from Shijiuyang Water Plant in Jiaxing, Zhejiang Province, and then the water sample was shipped to the laboratory through a polyethylene drum. The water quality parameters were determined using Standard Methods (2005) [23]. Total organic carbon (TOC) of the water sample was measured by TOC analyzer (multi N/C®3100, Analytikjena, Germany). The UV254 was measured by ultraviolet spectrophotometer (TU-1901, Persee, China). The pH of water sample was measured by a pH meter (PHS-3G, Leici, China). Anion concentration was determined by ion chromatography (ICS-900, Dionex, USA). The turbidity of the water sample was measured using a portable turbidimeter (2100AN, Hach, USA). The main water quality parameters were listed in Table 1. The DBPs detected in this study included four THMs (TCM, TBM, DBCM, and CDBM) and five HAAs (DCAA, TCAA, monochloroacetic acid (MCAA), monobromoacetic acid (MBAA), and dibromoacetic acid (DBAA)). According to U.S. EPA Method 551.1, THMs were analyzed by liquid/liquid extraction with methyl-tertiary-butyl-ether (MTBE) and analysis by GC/ECD (Shimadzu, GC-2014, Japan)

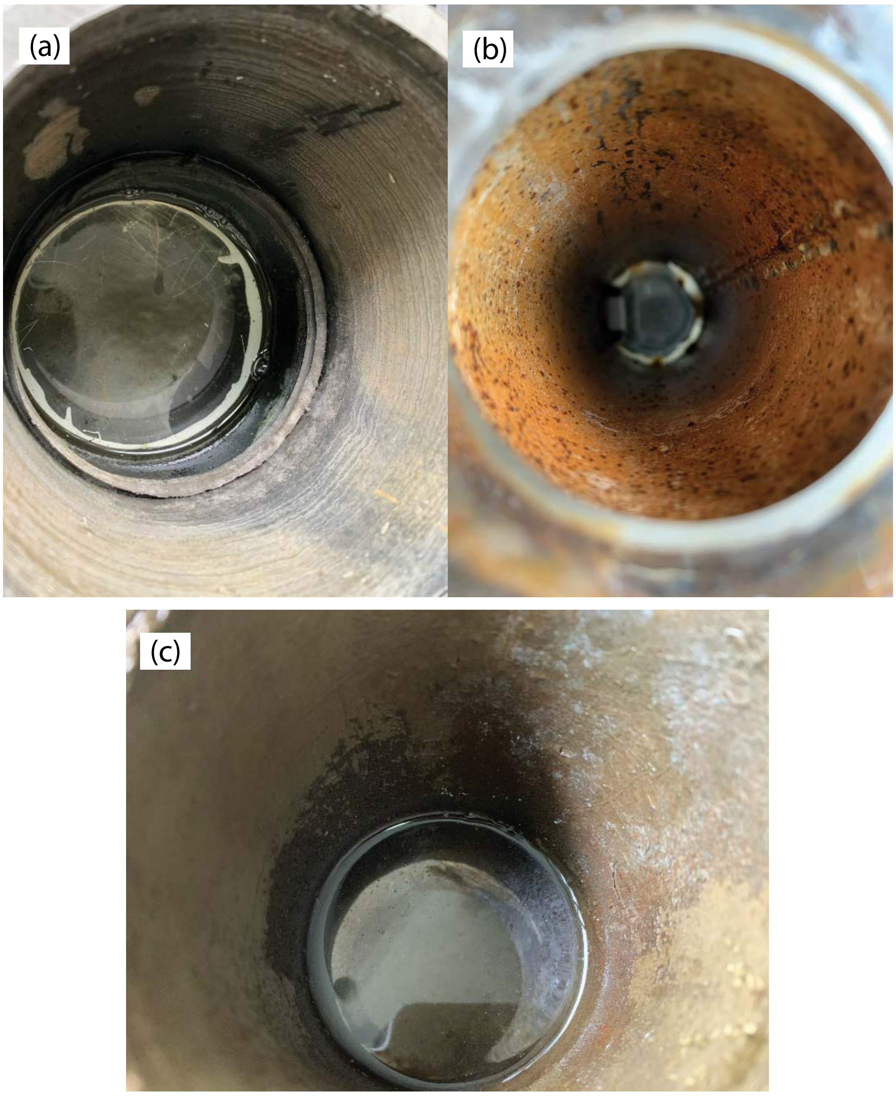

Fig. 1. Pipe wall images (a) PE pipe, (b) plastic steel pipe, and (c) ductile cast iron.

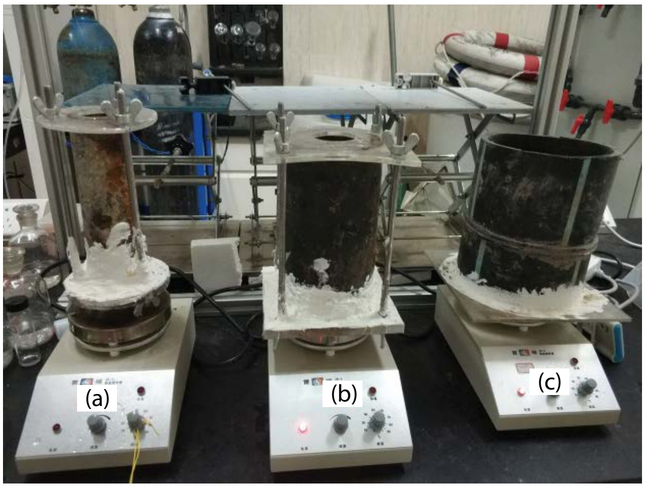

Fig. 2. Simulated pipeline reactor ((a) plastic steel, (b) ductile iron, and (c) PE).

Table 1
Main water quality parameters of unchlorinated post-filtration water (Shijiuyang Water Plant)

Parameter	Value
pH	7.31
TOC (mg/L)	3.15
Total hardness (as $CaCO_3$, mg/L)	137
Total dissolved solids (mg/L)	365
Oxygen consumption (COD_{Mn}, as O_2, mg/L)	1.32
Turbidity (NTU)	0.21
Bromide (μg/L)	84.42
Bromate (mg/L)	<0.005
Nitrate (mg/L)	1.48
Chloride (mg/L)	57.2
UV_{254} (cm^{-1})	0.03
Chroma	<5

[24]. HAAs were analyzed by liquid/liquid extraction with MTBE, then derivated by acidic methanol, and analyzed by GC/ECD according to USEPA method 552.3 [25].

3. Results and discussion

3.1. Effect of chlorine dosage on THMs and HAAs formation

Chlorine is the most commonly used disinfectant in drinking water treatment, and the relationship between the chlorine dose and the organic concentration in treated water is the decisive factor for the formation of DBPs. With the change of chlorine dosage, the concentration and species composition of DBPs constantly change. Fig. 3 showed the formation levels of THMs and HAAs within 24 h under different chlorine dosage. As expected, the production levels of THMs and HAAs increased with the increase of chlorine dosage [26,27]. The level of HAAs formation is significantly higher than that of THMs, which may be related to the precursor of DBPs in treated water. It has been reported that the relative distribution of THMs and HAAs is affected by the hydrophobic/hydrophilic distribution of NOM in chlorinated water [10,28]. Many studies have shown that hydrophobic organic precursors are important precursors of TCAA and THM, which can produce high yields of THM and TCAA during the chlorination process [29–31]. As for DCAA, several studies have reported that hydrophilic organic matter is as important or sometimes more important than hydrophobic organic matter, and that DCAA precursors are generally more hydrophilic than TCAA precursors [30,32,33].

Fig. 4 illustrated the relationship between the morphology of THMs or HAAs and chlorine dosage. During the chlorination process, the proportion of Cl-THMs increased with the increase of chlorine dosage, especially the production of TCM, accounting for up to 50% of total THMs. At the same time, DCAA and TCAA were found to be the main HAAs produced through chlorination of water samples. The production levels of DCAA and TCAA greatly increased with the prolongation of reaction time, while MCAA slightly increased with the increase of chlorine dosage. The enhancement of DCAA and TCAA yields might be due to the presence of more hydrophobic NOM in raw water. According to the report by Sun et al. [34], the hydrophobic NOM was the dominant precursor for DCAA and TCAA, the hydrophobicity of these precursors might follow the order of TCAA > DCAA.

It is interesting to note that the yields of Br-HAAs and Br-THMs (DBAA, BDCM, and CDBM) increased with increasing chlorine dosage. During the chlorination, as the chlorine dosage increased from 0.5 to 3 mg/L, and the maximum yields of DBAA, CDBM, and BDCM increased from 1.8, 1.0, and 1.0 µg/L to 7.4, 4.3, and 4.3 µg/L, respectively. It is clear that increasing the chlorine dosage can substantially improve the yields of Br-HAAs and Br-THMs. This can be explained by the chlorination of bromide to HOBr, which exhibits stronger halogenation activity than HOCl, causing the increase in the yield of Br-DBPs [17,35,36].

3.2. Effect of retention time on THMs and HAAs formation

Fig. 5 showed the level of THMs and HAAs as a function of residence time after chlorination of post-filtration water. It can be seen that THMs and HAAs increased with the extension of chlorination time. The generation of DBPs can be regarded as the continuous chemical reaction between disinfectant and organic matter, thus the reaction time also affects the final yield of DBPs. A similar conclusion can be found everywhere [15,37–39]. In addition, the generated HAAs were mainly composed of DCAA and TCAA, and the former was significantly increased with the extension of reaction time, from 5.3 to 22 µg/L after 24 h chlorination. The formed THMs were mainly composed of TCM and BDCM. After 24 h chlorination, TCM and BDCM increased from the initial 7.5 and 1.6 µg/L to 12.4 and 8.9 µg/L, respectively. Furthermore, the generation level of THMs is significantly lower than that of HAAs, which may be related to the

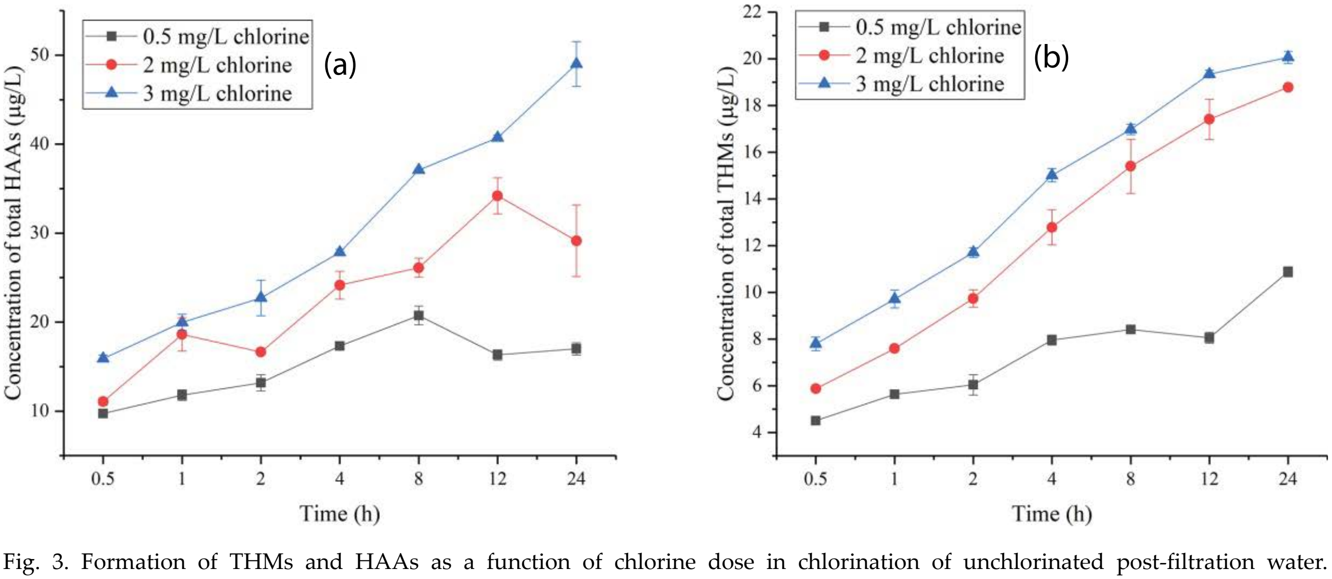

Fig. 3. Formation of THMs and HAAs as a function of chlorine dose in chlorination of unchlorinated post-filtration water. Experiment condition: $[Cl_2]$ = 0.5–3 mg/L, temperature = 20°C ± 1°C, and pH = 7 (a) total HAAs and (b) total THMs.

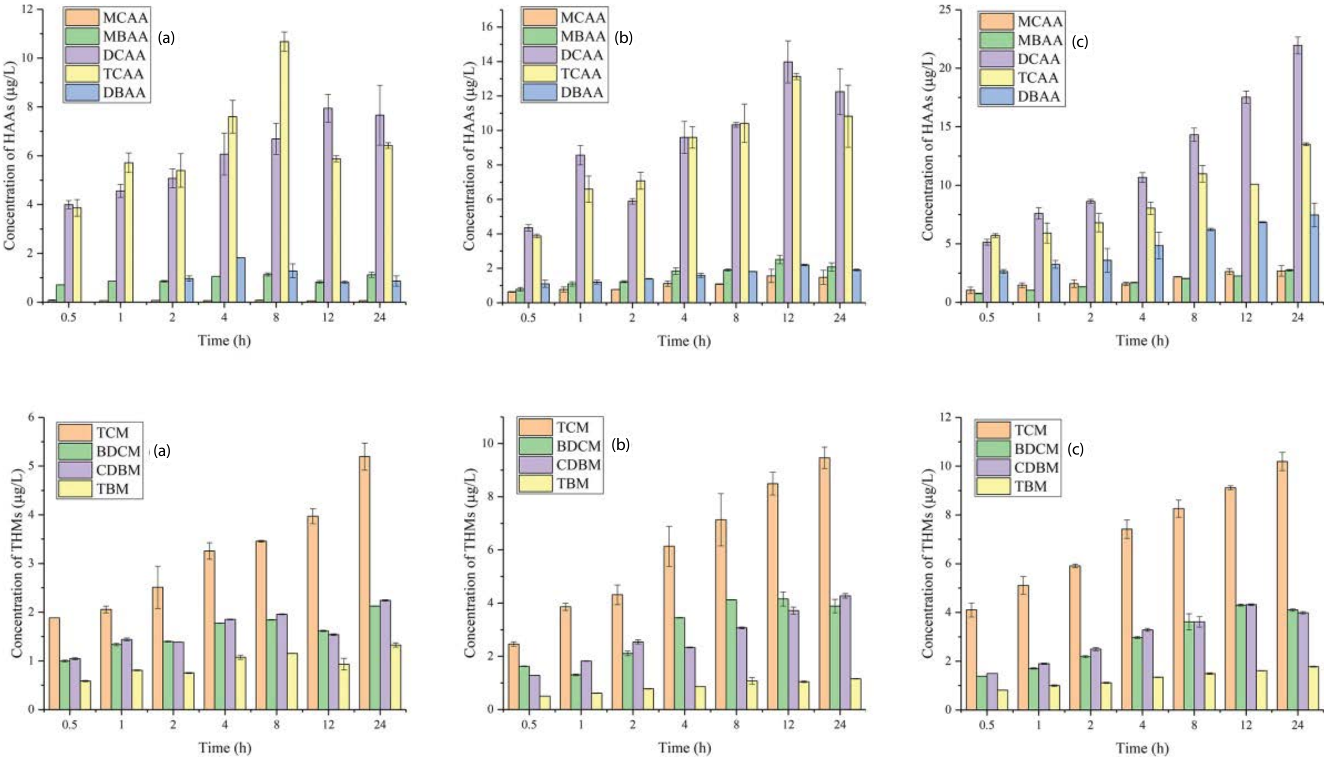

Fig. 4. Morphology of THMs and HAAs as a function of chlorine dose in chlorination of unchlorinated post-filtration water. Experiment condition: $[Cl_2]$ = 0.5–3 mg/L, temperature = 20°C ± 1°C, and pH = 7 (a) 0.5 mg/L, (b) 2 mg/L, and (c) 3 mg/L.

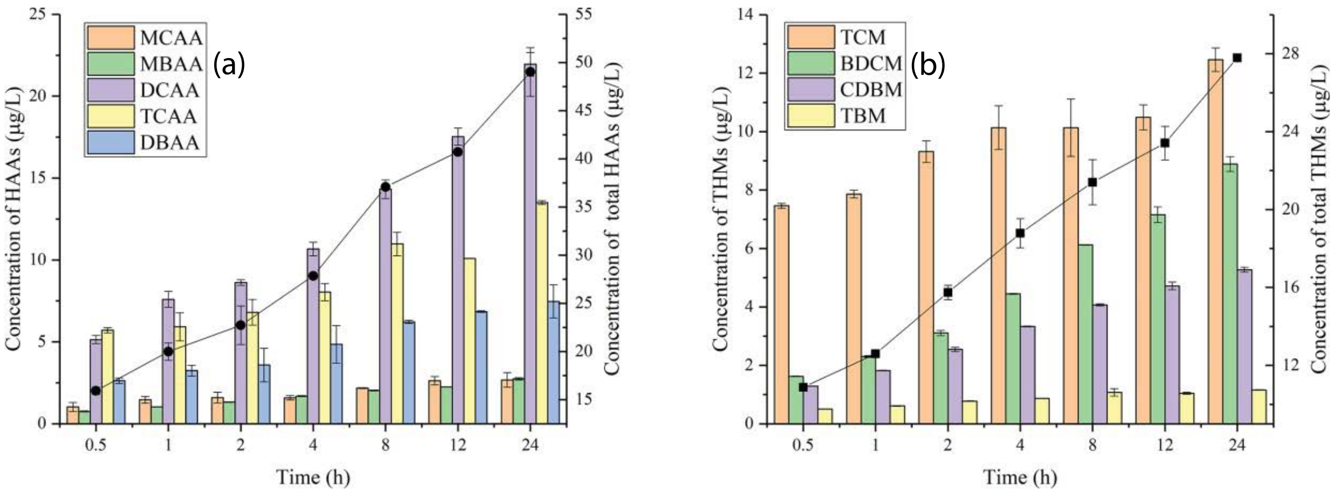

Fig. 5. Formation of THMs and HAAs as a function of retention time in chlorination of unchlorinated post-filtration water. Experiment condition: $[Cl_2]$ = 3 mg/L, temperature = 20°C ± 1°C, and pH = 7 (a) HAAs and (b) THMs.

properties of precursor, especially the hydrophilicity and hydrophobicity in treated water.

3.3. Effect of pH on THMs and HAAs formation

Fig. 6 showed the effect of pH on the production levels of THMs and HAAs during chlorination. Previous studies have shown that the stability of disinfectants was highly dependent on pH, and pH also affected the decay rate of disinfectants and the morphology of precursors [40–42]. As can be seen from Fig. 6, the effect of pH on the formation of HAAs was different from that of THMs during chlorination. As consistent with earlier studies, THMs concentration increased with increasing pH and reaction time [43]. After 24 h chlorination, the production levels of THMs at pH 6, 7, and 8 were determined to be 14.5, 28.2, and 30.4 μg/L, respectively. When pH rose from 6 to 8, the concentration of THMs increased more than twice after 24 h chlorination.

Base catalyzed reaction is generally considered to play an important role in the formation of THMs [44,45]. Contrary to THMs, the generation of HAAs was inhibited with elevating pH from 6 to 8, which might be caused by the difference between the precursors of THMs and HAAs. Early studies also found that some significant differences existed between the precursors of THMs and HAAs [18,46].

Fig. 7 presented the effect of pH on THMs and HAAs species during the chlorination of unchlorinated post-filtration water. The HAAs species formed during the chlorination were found to be mainly DCAA, followed by TCAA and DBAA. The main species of THMs was identified to be TCM, followed by BDCM and CDBM. The experimental results showed that the production level of DCAA at pH 8 was significantly lower than other pH values, while the production levels of MCAA and MBAA were almost unaffected by pH. For THMs, the production levels of TCM and BDCM under alkaline conditions were significantly

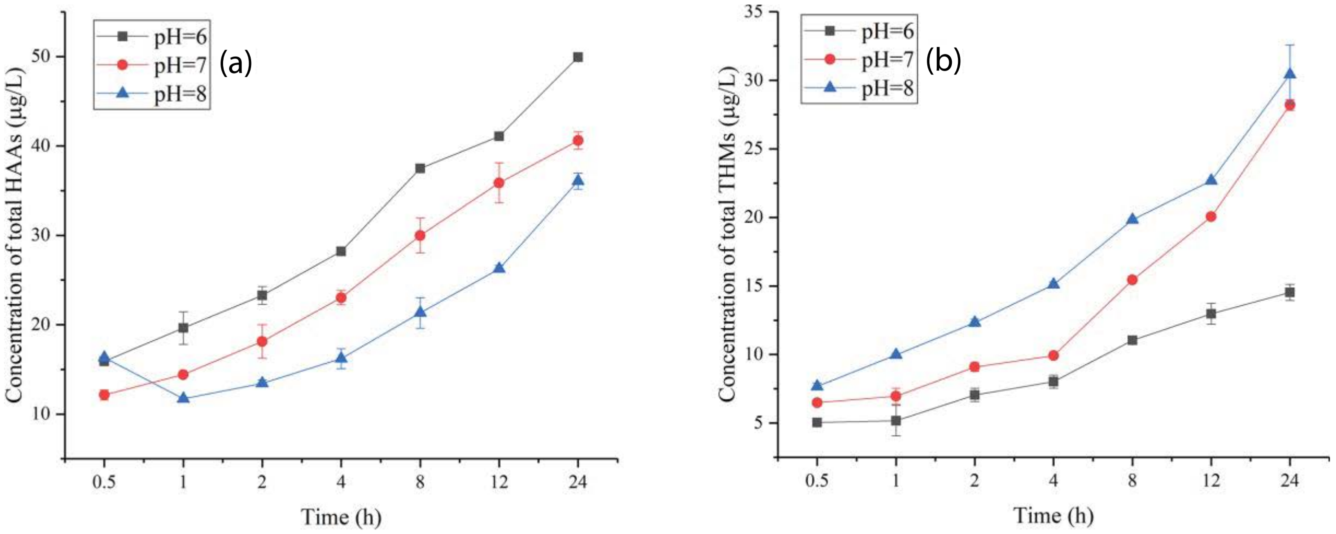

Fig. 6. Formation of THMs and HAAs as a function of pH in chlorination of unchlorinated post-filtration water. Experiment condition: $[Cl_2]$ = 3 mg/L, temperature = 20°C ± 1°C, and pH = 6–8 (a) total HAAs and (b) total THMs.

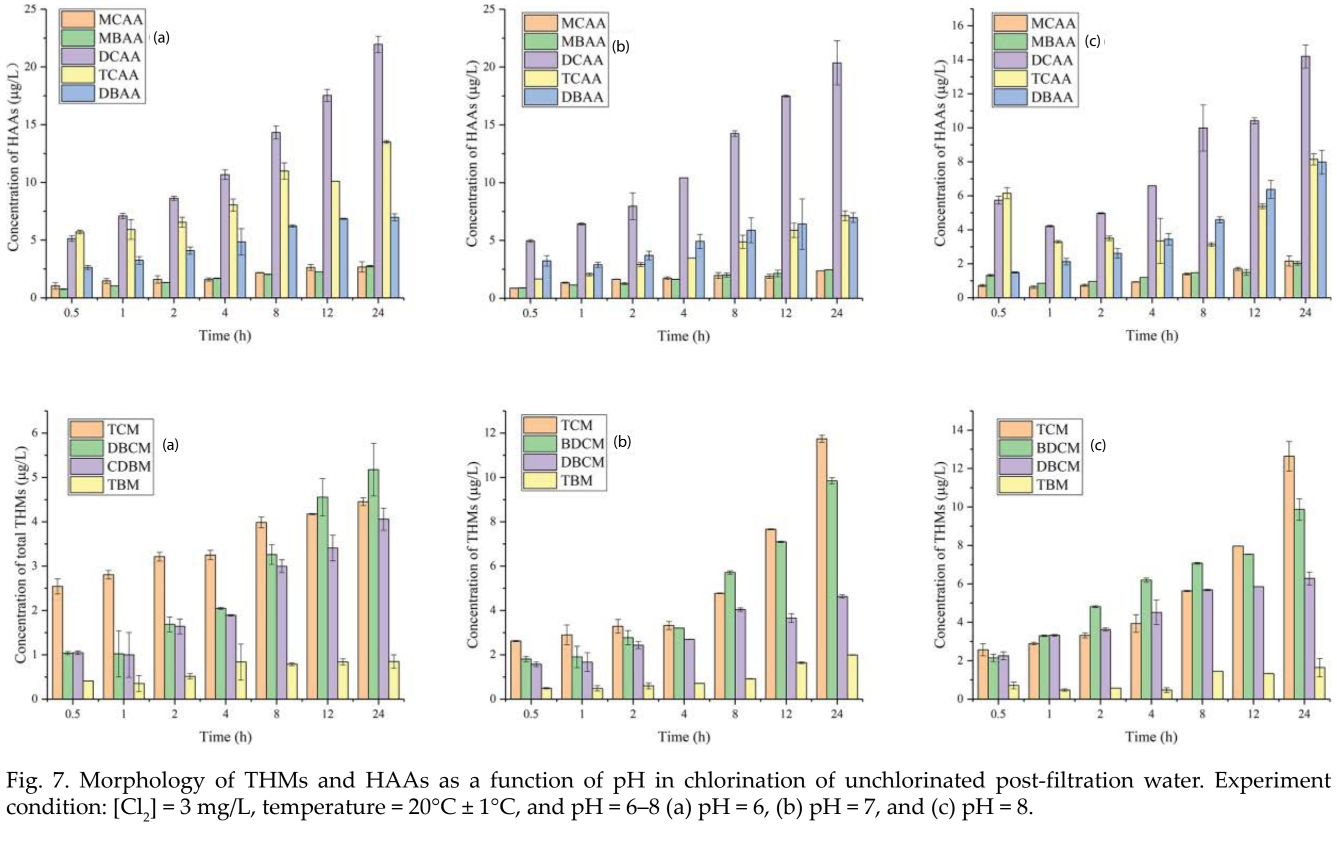

Fig. 7. Morphology of THMs and HAAs as a function of pH in chlorination of unchlorinated post-filtration water. Experiment condition: $[Cl_2]$ = 3 mg/L, temperature = 20°C ± 1°C, and pH = 6–8 (a) pH = 6, (b) pH = 7, and (c) pH = 8.

higher than under acidic or neutral conditions. Uyak et al. [47] believed that the speciation and reactivity of chlorine was highly pH dependent. Therefore, the demand for chlorine in alkaline solution is lower than that in acidic solution, while the formation of TCM increased in alkaline solution [48]. Since the formation and hydrolysis rate of each individual HAAs species are different, the effect of pH on the formation of HAAs is more complicated. As described by Hua et al. [44], the oxidation rate of different types of organics varies with increasing pH. Therefore, pH control may be an effective method to reduce the formation of DBPs in water distribution system.

3.4. Effect of water temperature on THMs and HAAs formation

Fig. 8 showed the effect of water temperature on the production levels of HAAs and THMs during chlorination. As can be seen, with the increase in water temperature, the generation of THMs and HAAs presented an obvious upward trend. When water temperature was 10°C, 20°C, and 30°C, after 24 h chlorination reaction, the HAAs concentrations were found to be 31.1, 48.5, and 61.1 μg/L, respectively, and the THMs concentrations were determined to be 17.9, 25.9, and 36.2 μg/L, respectively. Since the formation of DBPs was endothermic reaction, raising water temperature can accelerate the reaction rate between the disinfectant and organic matter, thus leading to an increase in the formation of THMs and HAAs [27]. Furthermore, water temperature exerted more significant impact on the formation of HAAs than THMs, which might be related to the formation process of two kinds of DBPs. Taking the *m*-hydroxyphenol with a substituent on the benzene ring as a precursor, the carbon between the two hydroxyl groups is the main attack site of chlorine and HAAs are prone to be generated after ring opening. Since the more open bonds are needed, the chemical energy required to generate HAAs is higher than THMs, causing water temperature exerts a more significant effect on HAAs formation [49].

Fig. 9 presented the composition of HAAs and THMs produced under different water temperature. It was obvious that the level of Cl-DBPs was much higher than that of Br-DBPs, which might be attributed to the low concentration of bromide in water samples. Under different water temperatures, HAAs were dominated by DCAA and TCAA. Due to the presence of chlorine in solution, Cl-HAAs tended to increase in the whole process. Interestingly, it can be found that water temperature exerted relatively obvious effect on the generation of DBCM and BDCM. For the composition of THMs, the Br-THMs dramatically increased as water temperature increased from 10°C to 30°C, which can be attributed to the enhancement of Br substitution reaction rate. From a kinetic point of view, it can be seen as a manifestation of the higher reactivity of HOBr species that reacted with NOM immediately after the chlorination initiated [36].

3.5. Effect of bromide concentration on THMs and HAAs formation

Bromide widely exists in natural water source and can be oxidized to HOBr by chlorination, which exhibits greater halogenation activity than HOCl [36]. Fig. 10 exhibited the formation level of THMs and HAAs during chlorination at different bromide concentrations. When bromide concentration was set at 0.2, 0.5, and 1 mg/L, after 24 h chlorination reaction, the formation level of HAAs was found to be 28.5, 50.6, and 67.5 μg/L, respectively, and the production level of THMs was determined to be 38.3, 57.7, and 73.3 μg/L, respectively. A similar trend was also found in earlier studies [50]. It is speculated that the bromide in water is involved in multiple reaction cycles with NOM and plays a catalytic role in the interaction of NOM with halogen [51]. Chloride or bromide ions would be simultaneously released as DBP precursors are attacked to release the halogenated DBPs. While excessive free chlorine will immediately oxidize bromide ions to HOBr species that react easily with NOM and initiate the circular reaction [52–54].

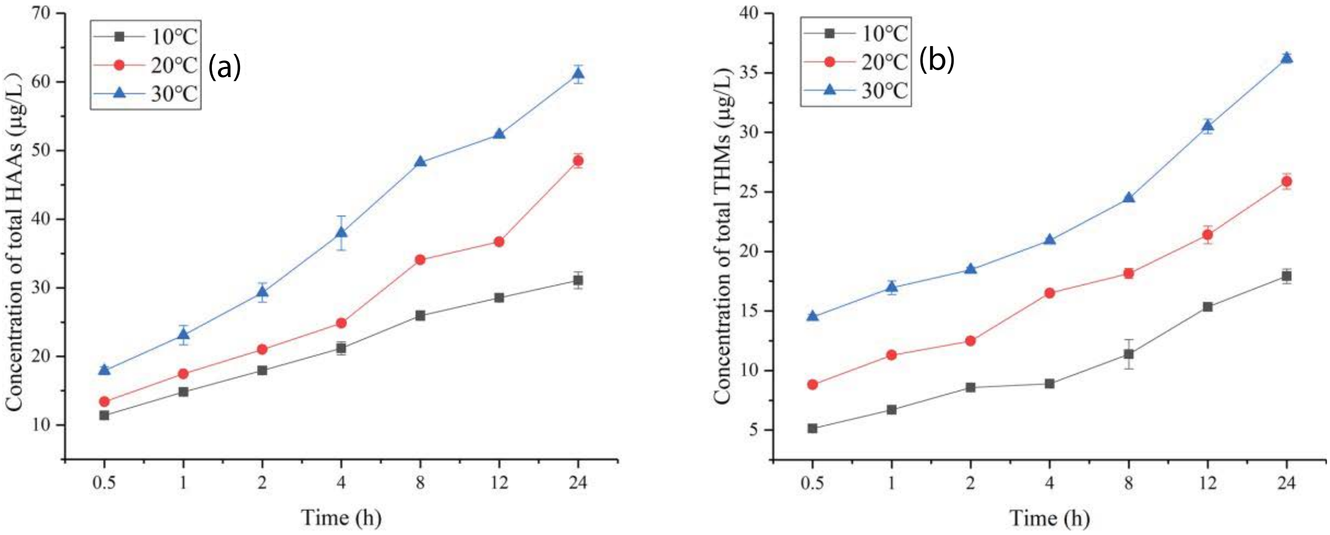

Fig. 8. Formation of THMs and HAAs as a function of water temperature during the chlorination of unchlorinated post-filtration water. Experiment condition: [Cl_2] = 3 mg/L, temperature = 10°C–30°C, and pH = 7 (a) total HAAs and (b) total THMs.

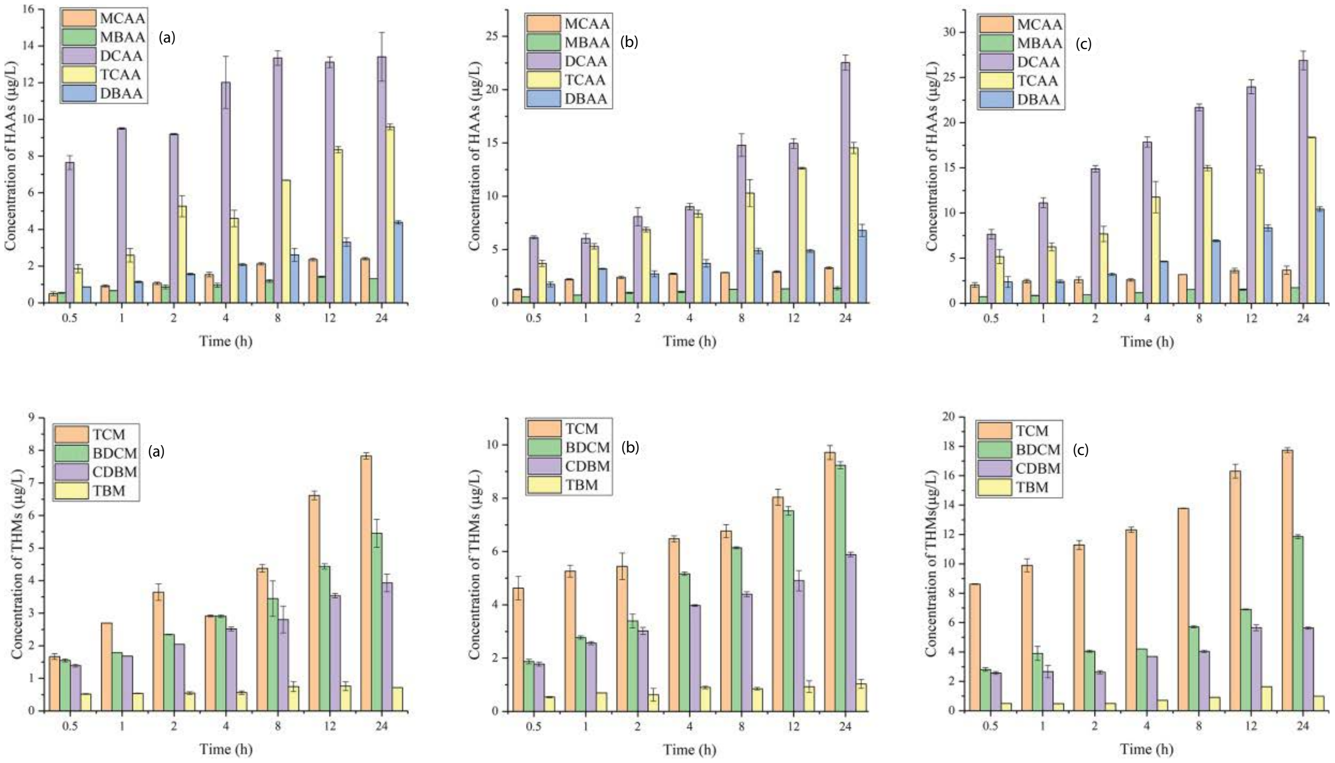

Fig. 9. Composition of THMs and HAAs as a function of water temperature during the chlorination of unchlorinated post-filtration water. Experiment condition: $[Cl_2]$ = 3 mg/L, temperature = 10°C–30°C, and pH = 7 (a) 10°C, (b) 20°C, and (c) 30°C.

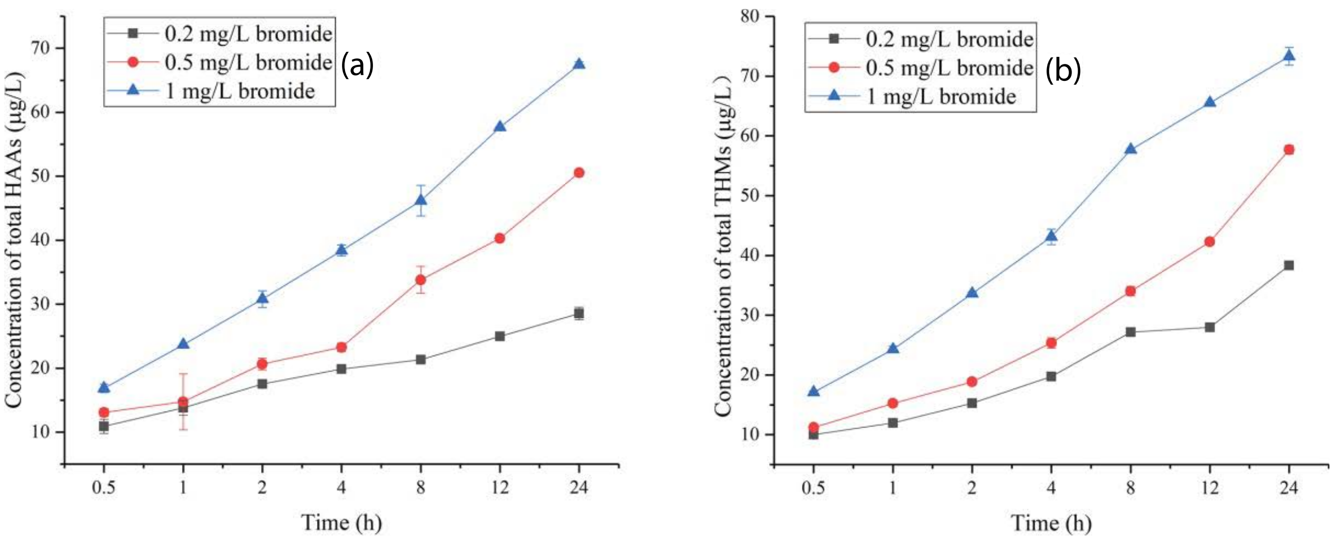

Fig. 10. Formation of THMs and HAAs as a function of bromide concentration during the chlorination of unchlorinated post-filtration water. Experiment condition: $[Cl_2]$ = 3 mg/L, $[Br^-]$ = 0.2–1 mg/L, temperature = 20°C, and pH = 7.0 (a) total HAAs and (b) total THMs.

Fig. 11 presented the effect of bromide concentration on the composition of THMs and HAAs after chlorination. The results showed that with the increase of bromide concentration, the formation forms of THMs and HAAs gradually were changed to bromide species. When bromide concentration was set as 0.2, 0.5, and 1 mg/L, after 24 h chlorination reaction, DBAA was generated at concentration of 15.3, 18.5, and 22.8 μg/L, respectively, and MBAA was produced at concentration of 3.73, 8.85, and 13.52 μg/L, respectively. THMs also increased with the increase of bromide concentration, and the level of Br-THMs production gradually increased. When bromide concentration increased from 0.2 to 1 mg/L, the TBM increased from 9.27 to 33.65 μg/L after 24 h of chlorination. Early studies on the effects of bromide on the morphology and yield of THMs and HAAs also found that the concentration of Br-DBPs increased with increasing bromide concentration [55,56]. That is, high bromide levels can be transformed into a variety of Br-DBPs species [26,57].

3.6. Effect of pipe material on THMs and HAAs formation

Earlier studies reported that the decay of disinfectants occurred not only in water but also on the surface of pipelines [58]. The reaction of residual chlorine with pipe materials and attached microorganisms will also result in attenuation of residual chlorine, which varies with the change of pipe materials [58,59]. The pipeline material also poses a significant effect on the level of disinfectant by-products [60]. Fig. 12 shows the effect of pipe material on the production levels of THMs and HAAs. As can be seen, the formation of THMs and HAAs is similar, with the highest generation of plastic steel pipe, followed by a ductile iron pipe, and the lowest generation of PE pipe. Since the pipe material is taken from the actual water supply pipe system, and the ductile iron pipe and plastic steel pipe have been seriously corroded, the scale deposits on the pipe wall and humus caused by microbial humification may be the reasons for the high content of THMs and HAAs in the ductile iron pipe and plastic steel pipe. However, the THMs and HAAs in PE pipe are significantly lower than the other two pipes, which can be explained by the fact that the inner wall of the PE pipe is relatively smooth, and it is not easy to scale deposits and attach microorganisms.

4. Conclusions

In this study, the influence of several key factors including chlorine dosage, residence time, pH, water temperature, bromide concentration, and pipe material on the generation level and composition of THMs and HAAs in the water distribution system was studied. The main conclusions were listed as follows:

- After chlorination, the generation levels of DBPs in the water distribution system increased with the increase of chlorine dosage. It was found that the generation level of HAAs was substantially higher than that of THMs and the main DBPs were identified to be DCAA, TCAA, and TCM.
- The formed THMs and HAAs in the pipe network both showed an increasing trend with the extension of residence time, and the produced HAAs were identified to be TCAA and DCAA, while the generated THMs were identified to be TCM and BDCM.
- As pH increased from 6.0 to 8.0, the generation of THMs increased, whereas the production of HAAs decreased. Increasing pH posed the most marked effect on the formation of TCM and BDCM, while
- With the increase of water temperature, the generation of THMs and HAAs presented an obvious upward trend, and its influence on HAA formation was more remarkable than that of THMs.
- The presence of bromide in the water distribution system promoted the formation of DBPs, especially Br-DBPs.

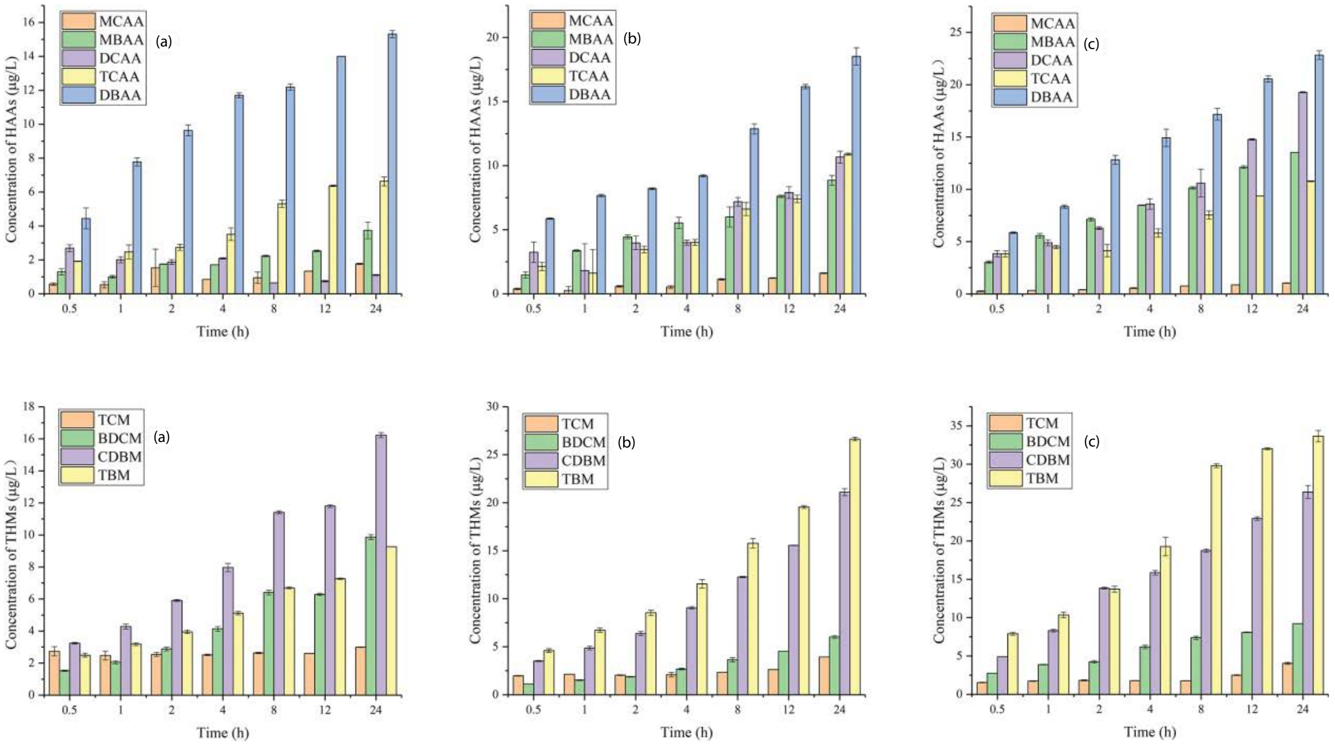

Fig. 11. Composition of THMs and HAAs as a function of bromide concentration during chlorination of unchlorinated post-filtration water. Experiment condition: [Cl_2] = 3 mg/L, [Br^-] = 0.2–1 mg/L, temperature = 20°C, and pH = 7.0 (a) 0.2 mg/L, (b) 0.5 mg/L, and (c) 1.0 mg/L.

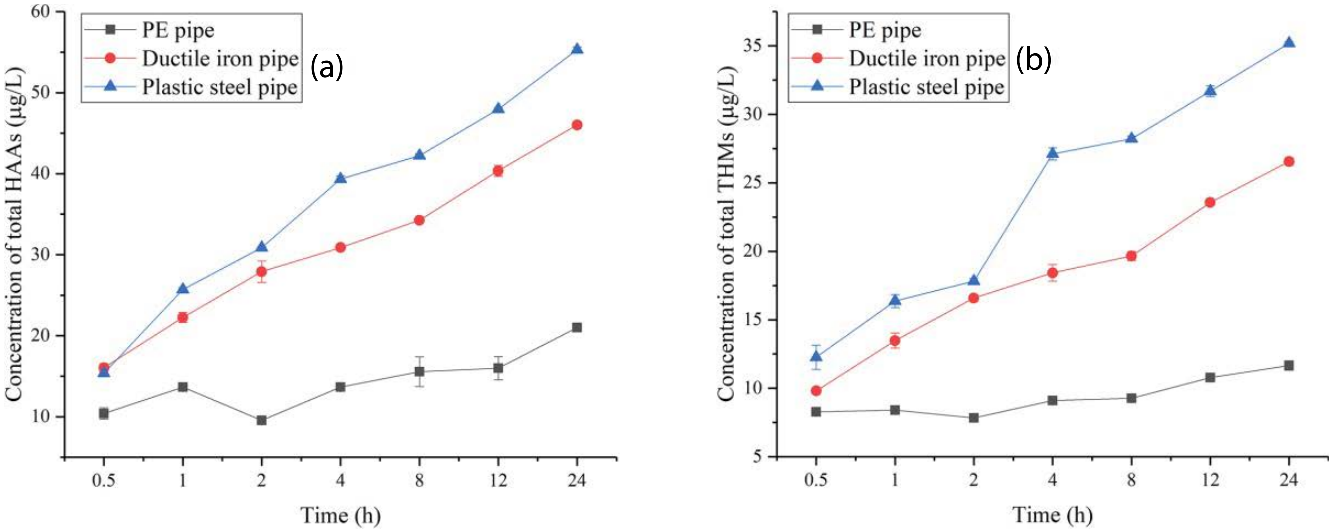

Fig. 12. Effect of pipe material on the formation of THMs and HAAs during chlorination of unchlorinated post-filtration water. Experiment condition: [Cl_2] = 3 mg/L, temperature = 20°C, pH = 7, and pipe material = PE pipe, ductile iron pipe, and plastic steel pipe (a) total HAAs and (b) total THMs.

- The effect of pipe material on DBPs formation followed the order of plastic steel pipe > ductile iron pipe > PE pipe, which might be related to the pipe properties and the corrosion degree.

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