Thai Environmental Engineering Journal Vol. 36 No. 2 (2022) : 69-77
www.eeat.or.th

DBPs Formation by Chlorination and Chloramination of Water and Treated Water at Short and Long Reaction Times

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Abstract

This work investigated disinfection by-products formation potential (DBPFP) by chlorination and chloramination. Raw water was collected from the U-Tapao canal, Songkhla, Thailand. This work used poly aluminum chloride (PACl) of 15 mg/L and aluminum chlorohydrate (ACH) of 1 percent w/v in coagulation experiments. DBPFPs were conducted on raw water and PACl-treated water and ACH-treated water at 1-day and 7-day reaction times. The raw water had a turbidity of 31.3 NTU and a DOC of 2.52 mg/L. The turbidity was reduced by PACl and ACH to levels below the water supply standard. DOC was reduced by 27 and 32 percent by PACl and ACH, respectively. The most prominent DBPFP found in chlorinated and chloraminated samples was trihalomethane formation potential (THMFP). THMFP levels of 171 and 425 µg/L were detected in the chlorinated raw water after 1-day and 7-day reaction times, respectively. In the chloraminated raw water, THMFP of 4.7 and 8.5 µg/L were detected, respectively. At 1-day reaction time, iodo-trihalomethane formation potential (I-THMFP), haloacetronitrile formation potential (HANFP), and trichloronitromethane formation potential (TCNMFP) of the chlorinated raw water were higher than that of the chloraminated raw water. At 7-day reaction time, I-THMFP of the chlorinated raw water was higher than that of the chloraminated raw water. HANFP and TCNMFP of the chlorinated raw water were lower than that of the chloraminated raw water. PACl and ACH are capable of lowering DBPFP, particularly THMFP and I-THMFP, in chlorinated water and HANFP in chloraminaed water. For treated water at 1-day and 7-day reaction times, chlorination had a greater impact on THMFP and I-THMFP than chloramination. The HANFP of chlorinated samples was higher than that of chloraminated samples after 1-day reaction time, whereas the HANFP of chloraminated samples was higher than that of chlorinated samples after 7-day reaction time.

Keywords : Aluminum chlorohydrate; Dissolved organic carbon; Haloacetronitriles; Halonitromethanes; Polyaluminium Chloride; Trihalomethanes

Introduction

Disinfection is an essential process at a drinking water treatment plant (WTP) that ensures the stability of microorganisms in the water supply as it passes though the water distribution system to the water consumer. Disinfectants such as chlorine and chloramine are commonly used in WTPs. The interaction between free chlorine residual and dissolved organic matter (DOM) in raw water and treated water is the most important factor for disinfection by-products (DBPs) formation during disinfection [1]. The components of DOM have a direct impact on the types of DBPs formation. Dissolved organic carbon (DOC) can form carbonaceous DBPs like trihalomethanes (THMs) when it reacts with disinfectants [2, 3]. In addition, the reaction of dissolved organic nitrogen (DON) with disinfectants can result in nitrogenous DBPs like haloacetonitriles (HANs) and halonitromethanes (HNMs) [2, 4, 5]. Chlorine is widely used for disinfection in Thailand and other countries.

The types of disinfectants can affect the formation of DBPs. The United States is now turning to chloramine as a tap water disinfectant instead of chlorine because it has a lesser risk of creating disinfection by-products (DBPs) than chlorine [6]. However, the formation of DBPs linked to chloramine, such as dihalo-acetonitriles, *N*-nitrosodimethylanine (NDMA), THMs, and dihaloacetic acid, can occur when chloramine reacts with DOM [7]. In the chloramination process, however, the inorganic chloramines monochloramine (NH_2Cl), di-chloramine ($NHCl_2$), and trichloramine (NCl_3) usually are detected. THMs, haloacetic acids (HAAs), haloaldehydes, haloacetamides, haloacetronitriles (HANs), and halonitromethanes can be formed when chloramines react with dry deposition particulate matter (HNMs) [8].

Coagulation is a common method for reducing turbidity in raw water. Coagulation has the potential to reduce DOC and color. A low dose of polyaluminium chloride (PACl) of 5-6 mg/L and aluminum chlorohydrate (ACH) of 1 percent w/v can reduce turbidity and color while also slightly lowering the potential for disinfection by-product formation (DBPFP) [9]. The type of coagulant used can affect pollutant removal performance. In this study, PACl and ACH were used as coagulants.

The U-Tapao canal in Songkhla, Thailand is used as the raw water source for three WTPs in Songkhla. The canal receives water from the Sadao reservoir and wastewater and treated wastewater from the community, agricultural, and industrial activities along the canal [3, 10]. The water qualities of the U-Tapao canal have presented with high turbidity and high color. This is caused by DOM, inorganic matter, plankton, and other microscopic organisms. The three WTPs use coagulation, sedimentation, filtration, and disinfection to remove suspended solids, colloids, color, and pathogenic microorganisms. The coagulant is PACl, and chlorine is used in the disinfection process. The remaining DOM in treated water can react with disinfectants to form DBPs. In addition, the most important considerations in the formation of DBPs are disinfection types and reaction times. The goal of this study is to compare the formation potential (FP) of THMs, iodo-trihalomethanes (I-THMs), HANs, and HNMs in raw water from the U-Tapao canal and treated water by PACl and ACH at 1-day and 7-day chlorination and chloramination.

Material and Methods

Sampling Sites and Sample Collection

Raw water from WTP was collected from the U-Tapao Canal, Songkhla, Thailand in January 2021 to determine its characteristics. The coagulation and disinfection experiments were then carried out using the raw water. All samples were stored at or below 4 ºC until analysis and experiments.

Experimental Procedure

The experimental procedure consisted of three parts. The first step was to determine the properties of the raw water, which included pH, alkalinity, turbidity, color, ultraviolet adsorption at 254 nm (UV-254), and DOC. The second part was the coagulation experiments. This work used PACl and ACH in the coagulation experiments using a jar-test apparatus. We used the optimal dosage of

PACl and ACH for turbidity reduction by coagulation from previous work. The optimal dosages of PACl [11] and ACH [9] were 15 mg/L and 1 percent w/v, respectively. The optimal dosage of turbidity was selected because the WTP operation mostly emphasizes turbidity reduction due to the perception of water consumers. As a results, the amount of coagulant used at WTPs must reduce turbidity to lower than the water supply standard. We set the jar-test apparatus for rapid mixing at 100 rpm for 1 minute and then on slow mixing at 60 rpm for 8 minutes, 40 rpm for 8 minutes, and 25 rpm for 5 minutes, respectively. pH was controlled at 6.8-7.2 by sodium hydroxide and sulfuric acid addition. After a 30-min rest period, the supernatant was measured for its turbidity and color. The treated water was measured for its pH, alkalinity, turbidity, color, UV-254, and DOC. The raw water and treated water were filtered by GF/F filter prior to the analyses of DOC and UV-254.

In the third experiment, the filtered raw water and treated water were used in the test. We used chlorine and chloramine as disinfectants. This work conducted the experiments in triplicate and used sodium hypochlorite solution for chlorination. We freshly prepared the chloramine (NH_2Cl) solution by adding NaOCl at the Cl_2:N mass ratio of 5:1 at pH 7-8.5 for 30 min and this was used in chloramination [12].

The experiments were conducted with the reaction times of one day and seven days. During the reaction times, the pH was kept constant at 7±0.2. At the end of the reaction times, free chlorine residuals of 3-5 mg/L for chlorinated samples [13] and monochloramine residuals of 1-3 mg/L for chloraminated samples [14] were measured.

We extracted the DBPs with methyl tert-butyl ether (MTBE); purity was 99.9% with 4-bromofluorobenzene as an internal standard [15]. The extraction conditions were modified from a previously reported procedure [16]. We mixed 17.5 µL of internal standard into 35 mL of water sample. After thoroughly shaking the sample, 7 mg sodium sulphate anhydrous, 1 mg copper (II), and 1 mg sulfate 5-hydrate were added. Then we added 2 mL of MTBE and shook for 1 minute. We collected MTBE in vials at a top layer of 2 mL and stored it below 4 °C before GC analysis. The GC conditions are described in more detail elsewhere [2]. This work analyzed four THMs species consisting of trichloromethane or chloroform (TCM), bromodichloromethane (BDCM), dibromochloromethane (DBCM), and tri-bromomethane (TBM), five I-THMs species consisting of dichloroiodomethane (DCIM), bromochloroiodomethane (BCIM), bromodi-iodomethane (BDIM), chlorodiiodomethane (CDIM), and triiodomethane (TIM), four HANs species consisting of trichloroacetonitrile (TCAN), dichloroacetonitrile (DCAN), bromo-chloroacetonitrile (BCAN), and dibromo-acetonitrile (DBAN), and one HNMs (trichloronitromethane, TCNM) of the disinfected samples at one day and seven days reaction times.

Analytical Methods

The pH was directly measured using a HACH pH meter. Alkalinity was determined following the standard method 2320 B (Titration method). Turbidity and color were measured by a turbidity meter and UV/VIS spectrophotometer. We carried out DBPFP for chlorinated and chloraminated samples, including THMs, I-THMs, HANs, and HNMs, which were determined according to the standard method 5710B [13], except for the level of monochloramine residuals for chloraminated samples, we followed the recommendation from the previous work [14].

Results and Discussion

Characteristics of Raw Water and Treated Water

Raw water characteristics are important in selecting the best approach to improve water quality. The pH and alkalinity of the raw water were 6.4 and 20 mg/L, respectively indicating that coagulation could be used for treating the raw water. Water consumers are commonly concerned about turbidity and color. The WTP must reduce turbidity and color to lower than the drinking water standard. The turbidity and color of the raw water were 31.3 NTU and 30.6 Pt-Co (Table 1), respectively. The turbidity value was higher than the standard of

4 NTU [17]. DOC and UV-254 in raw water were detected at 2.52 mg/L and 0.144 cm^{-1}, respectively. Raw water from the U-Tapao Canal had DOC and UV-254 values of 3.7-5.6 mg/L and 0.100-0.284 cm^{-1}, respectively [9, 18]. In this study, the DOC of raw water was lower than in previous studies. UV-254 was within the UV-254 range reported in previous work.

Using a 15 mg/L PACl and a 1 percent w/v ACH, the turbidity was reduced to below the Provincial Waterworks Authority standard of 4 NTU. Color was reduced from 30.6 Pt-Co to 3.67 Pt-Co (88 percent reduction) and 2.45 Pt-Co (92 percent reduction) using PACl and ACH coagulation, respectively. In terms of turbidity and color reduction, the PACl and ACH coagulations performed similarly. When it came to surrogates for DOM reduction, the ACH coagulation had a 32 percent reduction in DOC and a 67 percent reduction in UV-254, respectively. This was slightly higher than the 27 percent and 58 percent reductions in DOC and UV-254 by PACl coagulation, respectively. This finding was consistent with a previous study [9], which found that ACH coagulation at 1 percent w/v performed better on UV-254 and DOC reduction than PACl coagulation at 5-6 mg/L. Concerning the capacity of the coagulation process in five raw water sources in Japan, PACl coagulation-filtration reduced DOC and UV-254 by 29 to 65 percent and 63 to 84 percent, respectively [19]. Regarding the findings of this work and previous research, the coagulation process is more effective at reducing DOM with an aromatic character, as indicated by the UV-254 value.

DBPs Formation after Disinfection Process

The DBPFPs after 1 day of chlorination and chloramination is shown in Fig.1. For the raw water (untreated), PACl-treated water, and ACH-treated water after chlorination, THMFP of 171, 63.5, and 53.7 µg/L, were determined, respectively. The levels of I-THMFP, HANFP, and TCNMFP in the raw water (untreated), PACl-treated water, and ACH-treated water were less than 8 µg/L or not detected. In all the chlorinated water samples, TCM was the most prevalent THMs species. TCM distributions of 83, 71, and 56 percent were detected in the raw water, PACl-treated water, and ACH-treated water, respectively. DCIM, BDIM, and TIM were found in the raw water for I-THMs species, but only BDIM was found in the PACl-treated water and only DCIM in the ACH-treated water. For HANs species, TCAN, DCAN, and DBAN were found in the raw water. After PACl and ACH coagulation, only DCAN was detected in both water samples. The raw water and PACl-treated water both produced TCNM, but ACH-treated water did not.

THMFP levels in raw (untreated) water, PACl-treated water, and ACH-treated water were 4.7, 4.8, and 4.4 µg/L for chloramination, respectively. The levels of I-THMFP, HANFP, and HNMFP in the aforementioned water samples were less than 1 µg/L or not detected. (Fig 1). For THM species, only TCM was found in the raw water, PACl-treated water, and ACH-treated water. In the case of I-THM species, DCIM was detected in the water samples after PACl and ACH coagulation but not in the raw water. Only DCAN was found in the raw water for HAN species, but not in the PACl-treated or ACH-treated water. TCNM was found in all the water sample tests.

Table 1 Characterization of raw water and treated water

Parameters	Units	Raw water	Treated water	
			PACl (15 mg/L)	ACH (1 percent W/V)
Turbidity	NTU	31.3±0.5	2.5±0.2	0.97±0.4
Color	Pt-Co	30.6±0.1	3.67±0.07	2.45±0.20
DOC	mg/L	2.52±0.01	1.84±0.06	1.71±0.14
UV-254	cm^{-1}	0.144±0.002	0.061±0.005	0.047±0.011

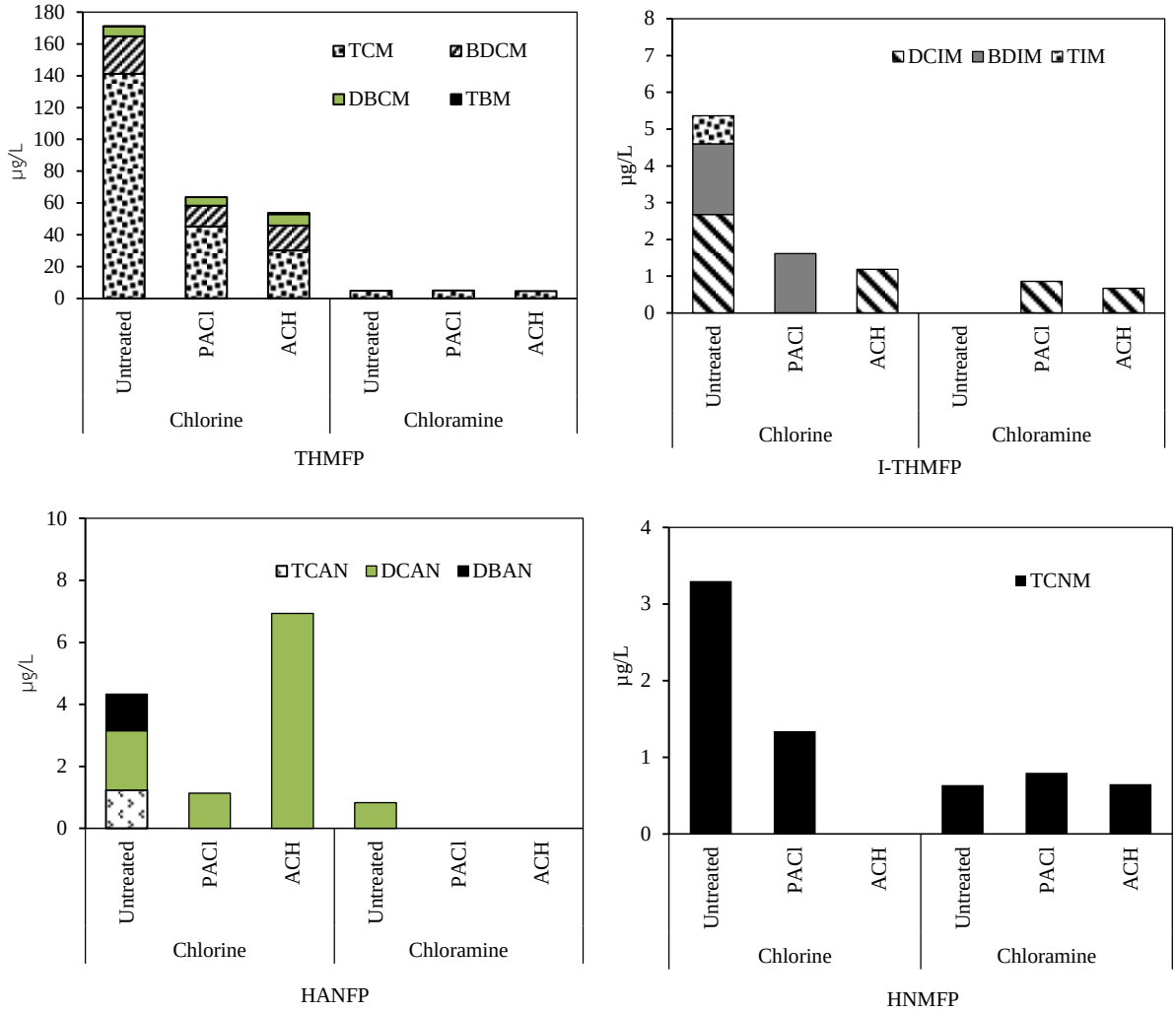

Fig. 1 DBPs formation during chlorination and chloramination after 1-day reaction time

The chlorination affected the formation of DBPs more than chloramination. THM concentrations were 36, 13, and 12 times higher in raw water, PACl-treated water, and ACH-treated water, respectively, than in chloramination with a one-day reaction time. THMFP by chlorination was detected in treated water in Japan using PACl coagulation-filtration with scales ranging from 30 to 150 µg/L, whereas less than 1 to around 5 µg/L of THMFP was produced by chloramination. This result was in accordance with a prior investigation; chloramination significantly decreased the formation potential of THMs [19]. With a one-day reaction time, chlorination produced higher concentrations than chloramination for I-THMs and HANs.

Taking into account the reduction of DBPFP by PACl and ACH coagulation at the day reaction time, after 1 day chlorination, THM and I-THM formation in PACl-treated water was slightly higher than in ACH-treated water. THMs and I-THMs formation were reduced by 63 and 70 percent, and 69 and 78 percent, respectively, when PACl and ACH coagulation were used. The PACl coagulation and filtration process reduced overall total THMFP of five raw water samples in Japan by 46 percent [19]. When using the PACl coagulation, the formation of HANs in PACl-treated water was reduced by 74 percent, while it was detected in higher concentrations than in the raw water when using ACH with chlorination. The formation of TCNM was

reduced by 59 and 100 percent in the PACl-treated and ACH-treated waters, respectively. Coagulation can decrease the formation of THMs, I-THMs, HANs, and TCNM. After 1 day of chloramination, a few THMs, HANs, and TCNM were formed. This is similar to the one-day chlorination process, in which the formation of THMs, I-THMs, and TCNM in ACH-treated water was were slightly lower than in the PACl-treated water, whereas HAN formation was not detected in either the PACl-treated water or ACH-treated water.

The DBPFPs of chlorination and chloramination after 7 days of reaction time are shown in Fig.2.Chlorination had a greater effect on the formation of THMs and I-THMs than chloramination, which was similar to the formation of these DBP species after one day. In contrast to the formation of HANs after one day, chloramination had a greater effect on HAN formation than chlorination. At 7-day chlorination, THMFP of 425, 417, and 166 µg/L, I-THMFP of 24.3, 4.4, and 0.66 µg/L, HANFP of 5.1, ND, and 1.3 µg/L, HNMFP of 0.85, 3.7, and 3.8 µg/L were measured in the raw water (untreated), PACl-treated water and ACH-treated water, respectively.

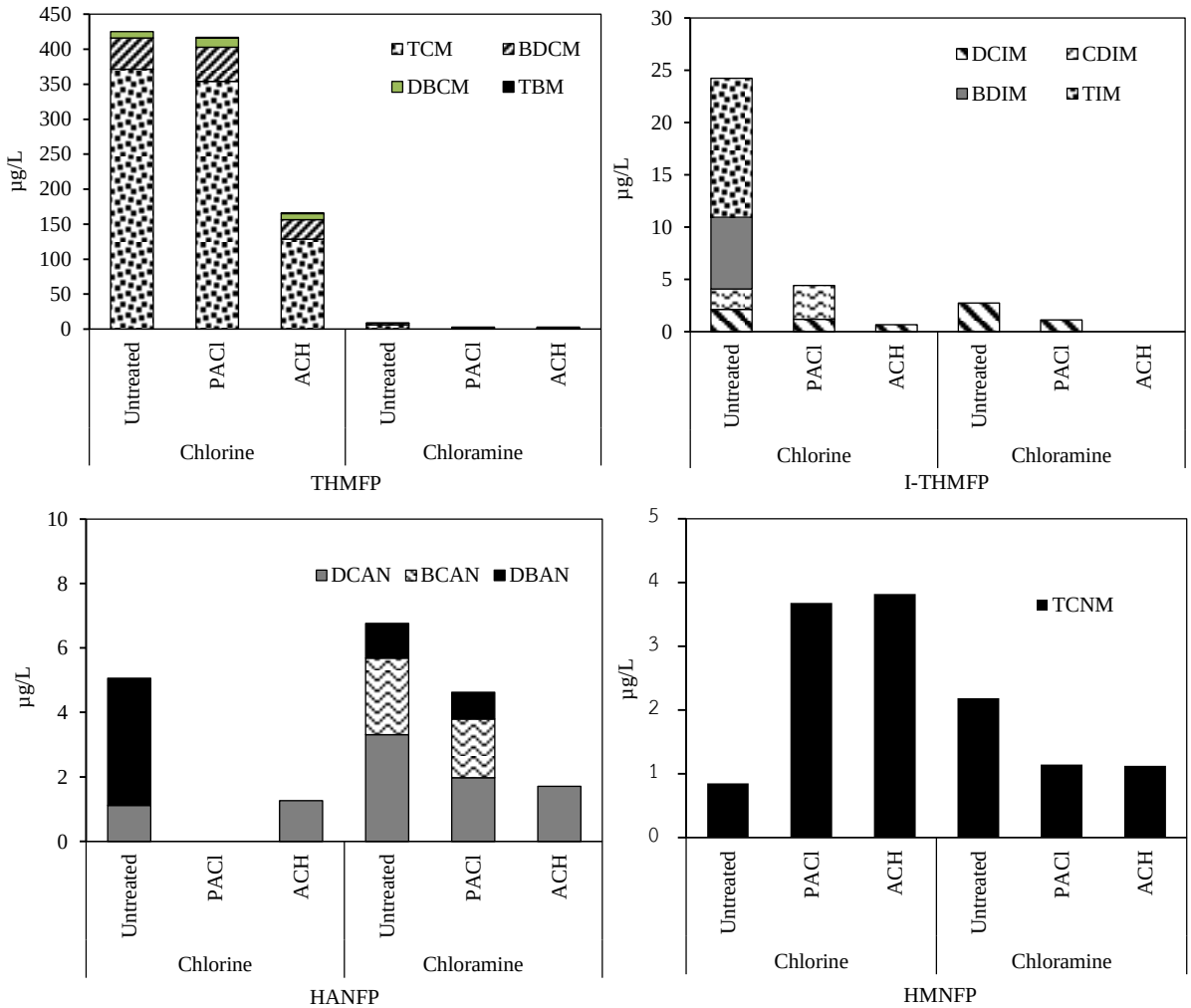

Fig. 2 DBPs formation during chlorination and chloramination after 7-day reaction time

TCM was the major THM species found in all the chlorinated water samples, like these samples after one-day reaction time. TCM distributions of 88, 85, and 78 percent were determined in the raw water, PACl-treated water, and ACH-treated water, respectively. From one day to seven days after chlorination, THMFP levels in the raw water, PACl-treated water, and ACH-treated water increased 2.5, 6.6, and 3.1 times, respectively. DCIM, CDIM, BDIM, and TIM were found in the raw water, CDIM and BDIM were found in the PACl-treated water, and only DCIM was found in the ACH-treated water for I-THMs species. At 7 days of chlorination, CDIM was the only I-THM species found in the raw water and PACl-treated water, but not at one day. TCAN is a HAN species that was detected after one day of chlorination but not after seven days, whereas DCAN was detected at a level that was incomparable to one day and DBAN was detected at a level that was higher than one day of chlorination. DCAN was detected in the ACH-treated water at levels that decreased over time. HAN species were not found in the PACl-treated water. In the PACl-treated water and ACH-treated water, TCNM was detected at a higher level than in one day of chlorination.

At 7 days after starting the reaction, the formation of DBPFPs via the chloramination process was considered. THMFP concentrations of 8.5, 2.8, and 2.6 µg/L, I-THMFP concentrations of 2.8, 1.1, and ND. µg/L, HANFP concentrations of 6.8, 4.6, and 1.7 µg/L, and HNMFP concentrations of 2.2, 1.2, and 1.1 µg/L were determined in the raw water, PACl-treated water, and ACH-treated water, respectively. TCM and BDCM were found in all the samples, while DBCM was found in both the raw and PACl-treated water. TCM in the raw water increased slightly compared to one day, whereas TCM in the PACl-treated water and ACH-treated water decreased compared to one day, while BDCM and DBCM were formed for the first time after 7 days of chloramination. Only DCIM was found in the water samples from I-THM species, as well as in one day of chloramination. More species that could not be detected in one day were formed after 7 days of chloramination in the case of HAN species, including BCAN, and DBAN. TCNM formation was detected at a higher level in all the water samples than in the samples taken one day after chloramination.

After 7 days of reaction time, THMs, and I-THMs could be produced at a higher concentration by chlorination than chloramination. Chlorination of raw water, PACl-treated water, and ACH-treated water produced 50, 149, and 63 times higher THM concentrations than chloramination, respectively. Chlorination formed higher concentrations than chloramination for I-THMs formation with a seven-day reaction time. Interestingly, the amount of HANs produced by chloramination was greater than that produced by chlorination.

After chlorination, THMs and I-THMs were formed at lower levels in the ACH-treated water than in the PACl-treated water. THMFP levels in the PACl-treated and ACH-treated water were reduced by 2 and 61 percent, respectively. I-THMFP levels in the raw water were reduced by 82 percent using PACl coagulation, and I-THMFP could be removed by 97 percent using ACH coagulation. PACl coagulation could completely remove HANs, whereas ACH coagulation was found to reduce 75 percent of HANFPs, respectively.

Considering the reduction of DBPFP by PACl and ACH coagulation after 7 days of chloramination, THMs and I-THMs formation were reduced by 67 and 69 percent, when PACl and ACH coagulation were used, respectively. I-THMFP levels in the raw water were reduced by 59 percent using PACl coagulation. ACH coagulation could completely remove I-THMFP. HANFP levels in the PACl-treated and ACH-treated water were reduced by 46 and 75 percent, respectively. TCNMFP could be reduced by 48 and 49 percent using PACl and ACH coagulation, respectively. In reduced levels of THMs and I-THMFP, ACH coagulation performs better than PACl coagulation.

Conclusion

The turbidity of raw water from U-Tapao, Songkhla, Thailand was found to be 31.3 NTU and the DOC was 2.52 mg/L. Turbidity was reduced to levels below the

water supply standard of 4 NTU using PACl of 15 mg/L and ACH of 1 percent w/v. With PACl and ACH, DOC was reduced by 27 and 32 percent, respectively. The most prominent DBPFP found in the chlorinated and chloraminated samples was THMFP. THMFP levels of 171 and 425 µg/L were detected in the chlorinated raw water after 1-day and 7-day reaction times, respectively. In the chloraminated raw water, THMFP of 4.7 and 8.5 µg/L were detected. At 1-day reaction time, the I-THMFP, HANFP, and TCNMFP of the chlorinated raw water were greater than that of the chloraminated raw water. At 7-day response time, I-THMFP, HANFP, and TCNMFP of the chlorinated raw water were 24.3, 5.1, and 0.85 µg/L, respectively, whereas those of the chloraminated raw water were 2.8, 6.8, and 2.2 µg/L. PACl and ACH are capable of lowering DBPFP, particularly THMFP and I-THMFP, in chlorinated raw water and HANFP in chloramined water. ACH coagulation reduced DBPFPs more effectively than PACl coagulation. For treated water at 1-day and 7-day reaction times, chlorination had a greater impact on THMFP and I-THMFP than chloramination. At 7-day reaction time, the TCNMFP of treated water of the chlorinated sample was higher than that of chloraminated sample. The HANFP of the chlorinated samples was higher than that of chloraminated samples after 1-day reaction time, whereas the HANFP of the chloraminated samples was higher than that of the chlorinated samples after 7-day reaction time. Due to the lower level of DBPFP than chlorination, chloramination might be used as the disinfection process of a water treatment plant. The formation of HANs in treated water may be prevented by a one-day reaction period after chloramination.

Acknowledgement

This research was funded by the Center of Excellence on Hazardous Substance Management (HSM), Bangkok, Thailand, under contract number HSM-PJ-CT-18-20. The authors would also like to express their gratitude to the Faculty of Engineering at Prince of Songkla University, Hat Yai Campus for their assistance with the equipment.

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