

UTILIZING POLYALUMINUM CHLORIDE (PACl) AND ALUMINUM CHLOROHYDRATE (ACH) FOR TREATING RAW WATER AND DBPs FORMATION

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Abstract

This research is aimed at utilizing polyaluminum chloride (PACl) and aluminum chlorohydrate (ACH) for treating raw water from U-Tapao Canal, Songkhla, Thailand. Coagulation experiments were conducted to determine the effects of PACl and ACH dosages on turbidity, color, ultraviolet adsorption at a wavelength of 254 nm (UV-254), dissolved organic carbon (DOC), and disinfection by-product formation potential (DBPFP) reduction. Optimal dosages of PACl and ACH for turbidity reduction were 5 mg/L and 1% w/v for raw water from the first sampling and 6 mg/L and 1% w/v for the raw water from the second sampling, respectively. ACH coagulation offered a better efficiency on color, UV-254, and DOC reduction compared to the PACl coagulation. Low dosages of PACl and ACH could slightly reduce DBPFP. The ACH coagulation could be considered as an alternative process for the reduction of turbidity, color, and DBPs precursor for water treatment plants.

Keywords: Turbidity, Color, DBPFP, Trihalomethanes, Haloacetonitrile, Water treatment.

Introduction

Surface water is a significant raw water source for water supply. Sufficient quantity and satisfactory raw water quality are of concerns for water supply production. The expansion of communities, agriculture and industry could affect the surface water quality by increasing the level of suspended solids (SS), allochthonous organic matter, and other contaminants in water sources (Kawamura, 2000; Daneshvar, 2009). Turbidity and color are two essential parameters characterizing the quality of surface water and drinking water. In regards to

turbidity, bacteria, viruses, and parasites are pathogens in water that can attach themselves to suspended particles in turbid water (Dimijian, 2000). Therefore, turbidity promotes the growth of pathogens in water resources leading to waterborne disease outbreaks. Although it is not a direct indicator of health risks, many studies have shown a correlation between the removal of turbidity and removal of protozoa and pathogens (Herson *et al.*, 1984; McCoy and Olson, 1986; Gauthier *et al.*, 2003).

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In the case of color, several factors are responsible for different water colors such as environments, organic materials, and algal growth (WHO, 2003). Colorless water is considered as pure though it may be unsafe for human health. In general, colored water has an impact on human health and the aquatic environment (Bennett and Drikas, 1993). Water treatment plants must reduce turbidity and color in raw water to a suitable level. Water quality standards of the Metropolitan Waterworks Authority (MWA) and Provincial Waterworks Authority (PWA) in Thailand follow the Guidelines for Drinking-Water Quality of the WHO (WHO, 2011; Metropolitan Waterworks Authority, 2017; Provincial Waterworks Authority, 2019). The WHO's guidelines include the recommended limits on the following; turbidity less than 5 NTU, and apparent color less than 15 color units (WHO, 2011). Moreover, according to the United States Environmental Protection Agency (US EPA), the allowed quantity of turbidity in drinking water must be lower than 1 Nephelometric Turbidity Unit (NTU) (US EPA, 1999).

Coagulation-flocculation is a relatively simple physicochemical technique that is employed during the water treatment process (Duan and Gregory, 2003; Ghafari *et al.*, 2009). Coagulation is a process in which the charge particles in a colloidal suspension are neutralized by mutual collision with opposite ions. Then, the resulting flocs settle in a clarifier (Daneshvar, 2009; Li *et al.*, 2010). Aluminium sulphate (alum) is the most commonly used coagulant in water treatment plants, with low cost being its major attraction. The limitation of alum is that it is acidic, causing the pH and alkalinity of water in production to be low. However, the WHO's guidelines recommend that the pH of drinking water must be between 6.5-8.5, resulting in high turbidity when large quantities of alum are unable to be used in raw water. (Pornsak, 2010; WHO, 2011; Metropolitan Waterworks Authority, 2017). Moreover, Residual aluminum levels in treated water can often exceed acceptable limits at 0.2 mg/L (Metropolitan Waterworks Authority, 2017).

Recently, poly aluminum chloride (PACl) and aluminum chlorohydrate (ACH) have been developed for improving water qualities. Several research studies reported that PACl works effectively over a broader pH range than alum, which has less effect on the pH value of raw water (Duan and Gregory, 2003; Gregory and Dupont, 2001). Low levels of residual aluminium in treated water can be achieved, typically 0.01-0.05 mg (Yang *et al.*, 2011), with less sludge volume (Aguilar *et al.*, 2002) and more rapid flocculation

(Gregory and Dupont, 2001). Aluminium chlorohydrate (ACH) is a modern coagulant which has a high aluminium content ($\text{Al}_2(\text{OH})_5\text{Cl}$).

In practice, there is little difference between the performance of ACH and PACl in water treatment applications, even though ACH is more hydrated (Gebbie, 2001). So, considering the purity, dosage, decolorization efficiency, and quantity and quality of sludge production, ACH was found to be the best coagulant, giving 91% decolorization efficiency at a dose of just 100 mg/L. To achieve this high degree of decolorization efficiency, the dosage for PACl was found to be approximately 50% higher than that of ACH (Verma *et al.*, 2012). For DOC removal, some researchers confirmed the superiority of ACH compared to alum or PACl for DOC removal from wastewater especially at higher pH values (Sadrnourmohamadi and Gorczyca, 2015; Yonge *et al.*, 2016). It is possible that the removal of trihalomethanes (THMs) precursors may not be as complete as with alum (Shi *et al.*, 2007; Yan *et al.*, 2008; Sadrnourmohamadi and Gorczyca, 2015).

The U-Tapao canal, Songkhla, Thailand is used as raw water for three water treatment plants in Songkhla. The canal receives wastewater and treated wastewater from community, agricultural, and industrial activities (Musikavong and Wattanachira, 2013; Musikavong *et al.*, 2016; Phatthalung, *et al.*, 2016). The coagulation by PACl for the removal of disinfection by-products (DBPs) has been studied (Musikavong and Wattanachira, 2013; Musikavong *et al.*, 2016; Phatthalung *et al.*, 2016; Tongchang *et al.*, 2018). Raw water from the U-Tapao Canal, Thailand, during the months of January through to March present low turbidity with high color. This is caused by organic and inorganic matter, plankton, and other microscopic organisms. Therefore, the removal of suspended solids (SS), colloids, and color is of most importance. In addition, an alternative coagulant to PACl or alum requiring low dosage for water treatment processes in the removal of SS and color is significantly important. To the best of the authors' knowledge, no research has been conducted to study the effects of ACH on the removal of SS, color, surrogate for dissolved organic matter (DOM), and disinfection by-products (DBPs) formation potential. This research focuses on determining the optimal conditions for PACl and ACH coagulation to remove contaminants from the raw water of the U-Tapao canal and the highest DBPs in water. The obtained results could provide better understanding on the reduction of contaminants by ACH and could be applied to operations in water treatment plants.

Materials and Methods

Sampling sites and sample collection

Raw water was collected twice from the pumping station of the Hat Yai water treatment plant (WTP), Songkhla, Thailand, which is located adjacent to the U-Tapao Canal. The raw water samples were collected in November 2019 (the first sampling, RW#1) and January 2020 (the second sampling, RW#2) for monitoring the basic water characteristics, DBP formation potential (DBPFP) and conducting the coagulation experiments. All samples were stored below 4°C until analysis and experiments.

Experimental procedure and analytical methods

Basic water characteristics

The raw water samples were measured for their pH, temperature, turbidity, alkalinity, and color. The pH was directly measured using a Hach pH meter (accuracy of ± 0.01 pH unit). Alkalinity was determined following the standard method 2320 B (Titration method). Turbidity and color were measured by a turbidity meter (HACH, 2100 N) and a Genesys 10S UV/VIS spectrophotometer (Thermo Electron Corp. Madison, WI, USA).

Analysis of dissolved organic matter and DBPFP

For DOC, ultraviolet absorption at 254 nm (UV-254), specific ultraviolet absorption (SUVA), and DBPFP were analysed, in which the water samples were filtered using GF/F (Whatman GF/F, 0.7 μ m) before measurement. The filtered water samples were acidified with sulfuric acid (H_2SO_4) to pH=2 for preservation and stored at 4°C prior to analysis. DOC in water samples was determined by a combustion method (Standard Method 5310D) using a total organic carbon analyzer (TOC-V CSN, Shimadzu, Japan) (APHA, 1998). UV-254 was measured by the Standard Method 5910B using a Genesys 10S UV/VIS spectrophotometer (Thermo Electron Corp. Madison, WI, USA). UV-254 can be used as a quantitative indicator of the DOM with aromatic rings in the water (Findlay *et al.*, 2003). SUVA was calculated using the UV-254 absorbance normalized to the mg/L DOC concentration (Hong *et al.*, 2013).

The formation potential of DBPs, including trihalomethanes (THMs): chloroform; bromodichloromethane (BDCM); dibromochloromethane (DBCM); and bromoform, iodo-trihalomethanes (I-THMs); iodoform (TIM); dichloriodomethane (DCIM); bromochloriodomethane (BCIM); bromodiiodomethane (BDIM); and chlorodiiodomethane (CDIM), haloacetonitrile (HANs): trichloroacetonitrile (TCAN); dichloroacetonitrile (DCAN); bromochloroacetonitrile (BCAN); and

dibromoacetonitrile (DBAN), and trichloronitromethane (TCNM) were determined. The DBPFP were carried out according to the procedure in the standard method 5710B (APHA, 1998). A phosphate buffer was used to neutralize the water samples to pH 7.0 ± 0.2 . The free chlorine residual after seven-day reactions was between 3 and 5 mg/L. Methyl tert-butyl ether (MTBE); purity 99.9% with 4-bromofluorobenzene as an internal standard were used to extract DBPs from water samples according to US EPA Method 551.1. Gas chromatography with a micro-electron capture detector (Agilent 6890) and HP-5ms (5% diphenyl/95% dimethyl polysiloxane as the stationary phase, 30 m length, 0.32 mm inside diameter 0.25 mm film thickness) were used for analysis. The GC conditions were presented elsewhere (Phatthalung and Musikavong, 2019).

Aluminum residual in supernatant

After the jar test, the supernatant is filtered through the Whatman GF/C filters (Whatman GF/F, 0.7 μ m) and collected in 50 ml. Aluminum concentrations were analyzed with an Inductively Coupled Plasma-Optical Emission Spectrometer (ICP-OES, HP4500 and HP-7700, Agilent Technologies, Inc.). A set of calibrating solutions is prepared. Each solution contains accurately known concentrations of the analyte elements Al in this example. The next step in ICP-OES analysis is introducing the calibration solutions and sample solutions into the plasma and measuring the intensities of light at appropriate analytical lines. The concentrations of the aluminium in each sample solution are determined from the calibration graphs. The experiments were conducted in triplicate.

Coagulation experiments

Two coagulants: PACl and ACH were used in the coagulation experiments by using a jar-test apparatus (JLT leaching test jar-test-VELP scientific). Dosages of PACl and ACH were varied in the range of 4-8 mg/L and 1-3% w/v, respectively. Samples were placed in the jar-test apparatus and coagulants were injected into each of the samples. The jar-test apparatus was set on the rapid mixing speed of 100 rpm for 1 minute, and then on slow mixing speed of 60 rpm for 8 minutes, 40 rpm for 8 minutes, and 25 rpm for 5 minutes, respectively. The pH was controlled in the range of 6.8-7.2 by sodium hydroxide (NaOH) and sulfuric acid (H_2SO_4) addition. After a 30 min rest period, the supernatant was measured for its turbidity and color. The optimal conditions for turbidity and color removal were determined. The treated water at optimal conditions was filtered by GF/F filter prior to the analyses of DOC, UV-254, DBPFP, and

aluminium residual. The experiments were conducted in triplicate.

Results and Discussion

Characteristics of raw water

The characteristics of raw water from the U-Tapao canal are presented in Table 1. The pH values of the raw water from the first (RW#1) and second sampling (RW#2) were 6.7 and 7.1, respectively. The raw water from the first and second sampling had different turbidity values: 45.4 NTU for RW#1 and 30.2 NTU for RW#2. The turbidity of raw water from the first sampling was higher than that of the second sampling. Color is a significant parameter for representing the quality of drinking water. Due to the perception of water consumers, clear water is required for drinking. Color values of 78.9 and 23.9 Pt-Co were determined for RW#1 and RW#2, respectively. The color of the raw water from the first sampling is higher than for the second sampling. By observing the color, the raw water from the first sampling had a brown color that was different from the dark moss color of the raw water from the second sampling. In general, the green color in water could have originated from algae.

Surrogate parameters for DOM were determined in terms of UV-254, DOC, SUVA, and DBPFP. The raw water had a similar UV-254 of 0.100 and 0.103 cm^{-1} for the first and second samplings, respectively. DOC is considered as the precursor of THMs' formation (Musikavong *et al.*, 2016). DOC in the raw water from the first and second sampling were 3.9 and 4.8 mg/L, respectively. DOC of raw water from the first sampling was lower than that of the second sampling. The SUVA values of 2.6 L/(mg.m) for the raw water from the first sampling and 2.1 L/(mg.m) for the raw water from the second sampling were determined. Some researchers indicated that the SUVA of DOM range from 1.0 to 6.0 L/(mg.m) in surface waters (Phatthalung and Musikavong, 2019) which is related to the aromatic carbon content in

DOM. SUVA of the raw water from the first and second sampling were similar.

Some researcher reported the UV-254, DOC, and SUVA of raw water of Hat Yai WTP which were 0.132 cm^{-1} , 3.9 mg/L and 3.4 L/(mg.m), respectively. However, the UV-254 and SUVA in this study were lower than that of the previous study, whereas DOC was higher than that of the previous study (Tongchang, 2018). This indicated that the characteristics of the raw water in the canal varied according to the time of sampling. In regards to the raw water characteristics, the water treatment plant must reduce the turbidity and color to be lower than the drinking water standards of 4 NTU and 15 Pt-Co, respectively (Provincial Waterworks Authority, 2019). In addition to the turbidity and color reduction, water treatment plants must reduce the UV-254 and DOC to be as low as possible, because high UV-254 and DOC reduction could result in the high possibility of DBPs reduction.

The optimum dose of coagulants for turbidity and color reduction

The purpose of these experiments was to determine the optimum dosage of PACl and ACH coagulation for the removal of turbidity and color in the raw water from the first and second samplings. The operation of the water treatment plant usually focuses on the reduction of turbidity and color to produce a clear water supply. Besides, the low turbid water can enhance the chlorine disinfection efficiency in the water treatment plant. Figure 1 presents residual turbidity and the percentage reduction of turbidity in the supernatant by PACl and ACH under a controlled pH of approximately 7.

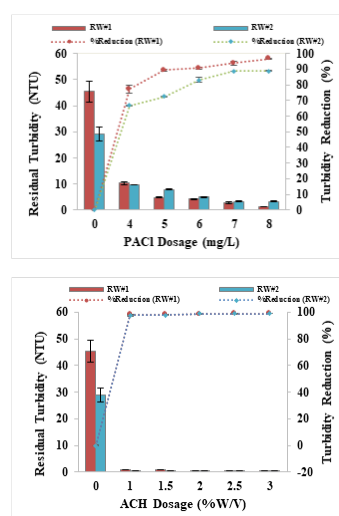


Figure 1. Residual turbidity and percentage reduction of turbidity in the supernatant by PACl (a) and ACH (b) at controlled pH 7.

Table 1. Characterization of raw water.

Parameters	First sampling (RW#1)	Second sampling (RW#2)
pH	6.7	7.1
Temperature (°C)	27.9	30.5
Turbidity (NTU)	45.4	30.2
Alkalinity (mg/l as CaCO ₃)	29	38
Color (Pt-Co)	78.9	23.9
DOC (mg C/L)	3.9	4.8
UV-254 (cm ⁻¹)	0.100	0.103
SUVA (L/(mg.m))	2.6	2.1

In the case of PACl coagulation, the turbidities of raw water was 45.4 and 30.2 NTU for the first and second samplings, respectively, reduced to the value of 10 NTU when a 4 mg/L PACl dosage was used. The turbidity of raw water was gradually reduced when increased PACl dosages from 5 to 8 mg/L for both raw waters from the first and second sampling were applied. The turbidity reduction efficiency was in the range of 67-97%. When ACH was used as a coagulant, it was remarkable that the turbidity of raw water from the first and second sampling rapidly decreased to values lower than 1 NTU with a dosage of 1% w/v. The increase in ACH dosages marginally improved turbidity reduction. ACH dosages from 1.0 to 3.0% w/v could reduce turbidity by more than 95%.

The color of raw water from the first and second samplings decreased to 6.3 and 8.2 Pt-Co when PACl of 4 mg/L was employed (Figure 2). Increasing PACl from 5 to 8 mg/L gradually reduced the color of raw water. The removal efficiency of color by PACl dosages from 4 to 8 mg/L ranged from 66 to 97%. Concerning the reduction of both turbidity and color together, PACl of 5 and 6 mg/L were the optimal dosages for coagulation of the raw water from the first and second samplings, respectively. By using these optimal dosages, the obtained percentage reduction of turbidity and color were slightly different from the highest percentage reduction of turbidity and color. When ACH was used as a coagulant, the color in raw water from the first and second samplings could rapidly be removed by ACH of 1% w/v. The increase in ACH

dosage from 1.5 to 3% w/v slightly increased color removal. ACH dosage from 1 to 3% w/v could remove color by 87 to 97 %. The optimal dosage for turbidity and color reduction by ACH was determined at 1% w/v.

Aluminum residual in the supernatant

After the coagulation process, the residual aluminum in the supernatant was determined. In the case of PACl coagulation, the residual aluminum in the supernatant of the raw water from the first and second sampling experiments were 0.12 and 0.29 mg/L, respectively. When ACH was used, the residual aluminum in the supernatant of the raw water from the first and second sampling experiments were 0.03 and 0.01 mg/L, respectively. It can be stated that the supernatant of PACl coagulation contained a higher residual aluminum concentration than that of ACH coagulation. This obtained result is in accordance with another researcher (Sawitri, 2009). The residual aluminum in water after ACH coagulation was lower than that of PACl coagulation.

The aluminum in drinking water should be seriously monitored. WHO and MWA recommends that the concentration of aluminum in drinking water from large water treatment facilities and small treatment facilities (with the number of water consumers lower than 10,000 people) must be equal or lower than 0.1 mg/L and 0.2 mg/L, respectively (WHO, 2006; Metropolitan Waterworks Authority, 2017). The Hat Yai WTP produces a water supply to serve approximately 420,000 people (Tongchang *et al.*, 2018) so it is considered as a large water treatment facility. According to obtained results in this study, the residual aluminum concentration in the supernatant from the PACl coagulation was higher than that of the recommended value from WHO, whereas the residual aluminum in the supernatant from ACH coagulation was within the recommended concentration (WHO, 2006).

Reduction of DOC and UV-254

In this section, reductions of DOC and UV-254 by PACl and ACH coagulation were carried out. DOC and UV-254 of the supernatant at the optimal condition of turbidity and color for the raw water from the first and second sampling were determined. PACl could reduce DOC and UV-254 from the raw water from the first and second samplings by 26.1 and 7.1%, and 63.7 and 78.3%, respectively. For ACH, DOC and UV-254 of raw water from the first and second sampling could be reduced by 53.2 and 30.7%, and 75.6 and 76.4%, respectively (Figure 3 and Figure 4). The percentage reduction of UV-254 was higher than DOC. The ACH coagulation could remove higher levels of DOC compared to PACl

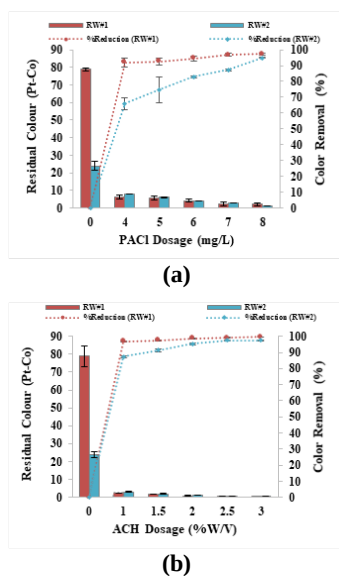


Figure 2. Residual Color and percentage removal of color in the by PACl (a) and ACH (b) at controlled pH 7.

coagulation. PACl of 15 mg/L was used to treat the raw water of Hat Yai WTP. This condition was able to reduce the UV-254 and DOC by 69 and 27%, respectively. The reduction of the surrogate parameter for DOM by PACl coagulation corresponds well with previous work (Jaichuedee *et al.*, 2019).

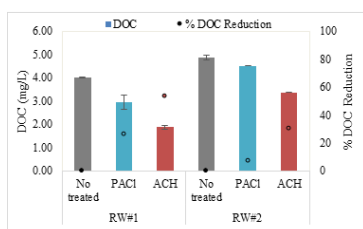


Figure 3. DOC and DOC Reduction (%).

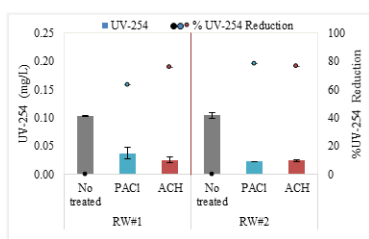


Figure 4. UV-254 and % UV-254 Reduction.

DBPFP and DBPFP reductions

DBPFP represents the possibility for dissolved organic matter to form the highest DBPs in water. This is because the conditions for testing DBPFP were set at a high amount of free chlorine residual (3-5 mg/L), optimal pH for DBP formation (pH at 7 ± 0.2), prolonged incubation time (7 days), and optimal temperature ($25 \pm 2^\circ\text{C}$). Therefore, it could not be used to represent the DBP in the water supply. For the carbonaceous DBPs, THMFP of 562 and 673 $\mu\text{g/L}$ were detected in raw water from first and second sampling, respectively. Chloroform was the dominant THMFP species detected, followed by BDCM and bromoform. DBCM was detected in the raw water at the second sampling. I-THMFP in the raw water from the first and second sampling was 5.1 $\mu\text{g/L}$. BCIM was the dominant of I-THMFP species followed by BDIM and CDIM. Other I-THMFP species could not be detected. In the case of nitrogenous DBPs, HANs of 1.26 and 1.31 $\mu\text{g/L}$ was detected in raw water from the first and second sampling. TCAN was the dominant HAN species. DBAN was found as a minority in second sampling (RW#2). Other HAN species could not be detected in RW#1 of first sampling. TCNM of 0.2 $\mu\text{g/L}$ was detected only from the raw water from the second sampling (RW#2) (Figure 5).

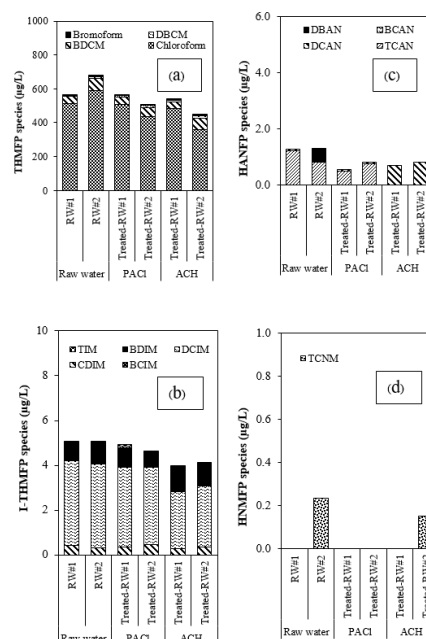


Figure 5. THMFP (a), I-THMFP (b), HANFP (c) and HNMF (d) for raw water.

The PACl of 5 mg/L could reduce THMFP, I-THMFP, and HANs from the raw water from the first sampling to 560, 4.9, and 0.5 $\mu\text{g/L}$, respectively. For the raw water from the second sampling, THMFP of 502, I-THMFP of 4.6, and HANs of 0.8 $\mu\text{g/L}$ could be detected from treated water by a PACl of 6 mg/L. The DBPFP slightly reduced when using a low dosage of PACl in the coagulation of raw water. The obtained results in this study were similar to previous work (Jaichuedee *et al.*, 2019). For DBPFP reduction by ACH of the raw water from the first sampling, a 1% w/v dosage could reduce THMFP, I-THMFP, and HANs to 443, 3.9, and 0.8 $\mu\text{g/L}$, respectively. For the case of the raw water from the second sampling, THMFP of 443, I-THMFP of 4.12 HANs of 0.80 and TCNM of 0.15 $\mu\text{g/L}$ could be determined when a ACH of 1% w/v was employed. The low dosage of PACl and ACH for coagulation could slightly reduce DOM. There was the possibility of carbonaceous and nitrogenous DBPs forming in the treated water.

The purpose of this work was to determine the optimum dosage of PACl and ACH coagulation for the reduction of turbidity and color in the raw water and their DBPFP. Low dosages of PACl and ACH coagulation had a good efficiency for the reduction of turbidity and color. PACl and ACH are used to remove color and colloidal matter present in untreated raw water. Both of them have a similar mechanism to improve water quality. Color and colloidal matter are removed by adsorption onto the metal hydroxide hydrolysis products formed; and

are sometimes referred to as sweep-floc coagulation (Gebbie, 2006; Yonge, 2012). However, ACH has several properties that set it apart from PACl. It is the most concentrated soluble aluminum-based coagulant available and has high basicity. It contains more metal, its dosage of ten is half that needed for PACl and also required less alkalinity in the water than PACl, because of its high basicity. Besides, the aluminum metal in ACH is more cationic than that in PACl (Gebbie, 2006; Yonge, 2012). The increased charge lets ACH work at lower metal dosages, further lowering chemical solids generation and residual aluminum levels in finished water (Gebbie, 2006). Regarding this results, ACH coagulation could be considered as an alternative process for water treatment plants. This is because it can help reduce chemical solids and alkali usage while helping water supply plant operations.

Conclusions

This is the first study reporting a comparison between PACl and ACH for the reduction of turbidity, color, and surrogate parameters for DOM, and DBPFP. The effects of different dosages of PACl and ACH on turbidity and color removal were determined. The optimal dosages were 5-6 mg/L and 1% v/w for PACl and ACH respectively. Low dosages of PACl and ACH coagulation had a high efficiency on turbidity reduction. ACH coagulation had a high efficiency on color removal in comparison to PACl coagulation. A low dosage for PACl coagulation could reduce DOC at 26.1 and 7.1% for first and second sampling and UV-254 at 63.7 and 78.3, respectively. THMFP, I-THMFP, and HANFP was slightly reduced and TCNMFP could not be reduced. When a low dosage of ACH was used for coagulation, the average percentage reductions of DOC and UV-254 were 53 and 30 and 75 and 76% for first and second sampling, respectively. THMFP, I-THMFP, HANFP, and TCNMFP were slightly reduced similar with PACl coagulant. However, ACH can provide significant advantages over PACl, including lower residual aluminum levels in the treated water and better performance in color reduction. ACH coagulation could be considered as an alternative process for water treatment plants.

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