# Removal of natural organic matter from water by coagulation and flocculation to mitigate the formation of chlorine-disinfection by-products: a case study at Chinaimo water treatment plant, Vientiane capital, Laos

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# <u>Abstract:</u>

The reaction between natural organic matter (NOM) and chlorine during disinfection of water potentially forms trihalomethanes (THMs), which are classified as dangerous, carcinogenic disinfection by-products. Thus, this study aimed to investigate the removal of NOM by using coagulation and flocculation via jar tests of the raw water collected from the Chinaimo water treatment plant in Laos. Several different coagulants, such as  $Al_2(SO_4)_3$  (alum), polyaluminium chloride (PAC), and iron chloride (FeCl<sub>3</sub>), and the flocculant polyacrylamide (PAM) were examined to determine the optimal operational conditions (i.e. coagulant dosage, flocculant dosage, and initial pH). The removal efficiency was evaluated by turbidity, NOM measured as total and dissolved organic carbon (TOC and DOC), ultraviolet absorbance at 254 nm (UV-254), and trihalomethane formation potential (THMFP). Results showed that 60 mg/l of alum, 40 mg/l of PAC, and 80 mg/l of FeCl<sub>3</sub> were the optimal dosages for coagulation, while a 0.2-0.3 mg/l of PAM was effective for flocculation. Optimal initial pH values of 7.0, 6.0, and 8.0 were found for the alum, PAC, and FeCl<sub>3</sub> coagulants, respectively. At the optimal conditions, the removal efficiency of turbidity was over 90% in all cases, which was higher than that of NOM (i.e. DOC of 31-42%, TOC of 19-52%, UV-254 of 17-39%, THMFP of 44-48%).

One sentence summary: NOM can be removed from water by coagulation-flocculation with  $Al_2(SO_4)_3$  (alum) at a dosage of 60 mg/l, PAM dosage of 0.2 mg/l, and adjusted pH of 7.0.

<u>Keywords:</u> disinfection by-products, flocculation and coagulation, natural organic matter, trihalomethanes, trihalomethane formation potential.

Classification numbers: 2.3, 5.1

## Introduction

Chlorination is widely used for disinfection of water and wastewater since it is efficient, easily supplied and operated, and cost effective. Most municipal water treatment plants in Laos use chlorine  $(Cl_2)$  for disinfection [1, 2]. However, it has also been discovered that the use of chlorine poses potential health risks due to the formation of disinfection by-products (DBPs), such as trihalomethanes, which are recognised as carcinogenic halo-organic compounds.

THMs are formed from the chemical reaction of natural organic matter (NOM) and  $Cl_2$  during the disinfection process. NOM is widely described as a complex mixture of organic compounds that occur naturally in groundwater and

surface water. Two common types of NOM are humic acids and fulvic acids, which cause colour and odour in water bodies [3]. The presence of NOM in water sources does not cause serious effects on the human's health, but problems arise when water sources containing NOM are treated with Cl<sub>2</sub> and other chlorine related compounds during the disinfection stage. Chlorination of water containing NOM is believed to be the most important precursor to the formation of THMs and it enables the growth of microorganisms in the treatment unit or distribution system [4, 5]. Typically, four types of THMs are found in chlorinated water, including chloroform (CHCl<sub>3</sub>), dichlorobromomethane (CHCl<sub>2</sub>Br), dibromochloromethane (CHBr<sub>2</sub>Cl), and bromoform (CHBr<sub>3</sub>) [6]. THMs are also reported to be the dominant

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species of DBPs, followed by haloacetic acids (HAAs) in water systems [6]. The total concentration of THMs and the formation of individual THM species in chlorinated water strongly depend on the concentration and properties of NOM, type of disinfection chemical and dose, and operational conditions (e.g. reaction time, temperature, and pH). Legislation has been strictly regulated to control allowable DBP levels in drinking water. The maximum contaminant level for THMs is set at different levels in developed countries, such as 80  $\mu$ g/l in the US, 250  $\mu$ g/l in Australia, 100 µg/l in Canada, 10 µg/l in Germany, and 100 µg/l in the EU [7]. Moreover, according to the World Health Organization (WHO) and the United States Environmental Protection Agency (US EPA), the limits for a total of five HAAs and bromate in drinking water are 60 µg/l and 10 µg/l, respectively [8]. Some negative effects of THM exposure due to the usage of chlorinated public water supplies (e.g., drinking and bathing) are low birth weight, small gestational size, and cancer [9, 10].

A conventional water treatment system normally comprises of coagulation-flocculation, sedimentation, rapid sand filtration, and disinfection. Coagulation-flocculation process is used to remove common physical parameters of surface water, such as suspended solids, turbidity, and colour. For NOM removal, it was reported that treatment efficiency was strongly affected by many factors, including the characteristics of raw water (e.g., nature and properties of NOM particles) and operational conditions (e.g., type and dose of coagulants/flocculants, pH, ionic strength, temperature, and turbidity) [11]. Other advanced treatments, such as adsorption with activated carbon, ion exchange, electro-coagulation, bio-filtration, membrane filtration, and advanced oxidation, have been investigated for NOM removal [5]. However, in terms of cost, coagulation and flocculation is generally considered to be an effective and economical option for NOM removal compared to other advanced alternatives, especially in the case of largecapacity water treatment plants [11]. Thus, the removal of NOM from surface water by using coagulation-flocculation technique should be investigated in detail, and performed at a real water treatment plant to demonstrate the practical applicability.

The Chinaimo Water Treatment Plant (CWTP) is a main water supply source of Vientiane, the capital of Laos. CWTP was established in 1980 with an initial capacity of 40,000 m<sup>3</sup>/day. Currently, the plant is operated with a capacity of 120,000 m<sup>3</sup>/day to supply tap water for 156,335 households over the 7 districts of Vientiane. A conventional water treatment process is designed and operated at CWTP, in which raw water is collected from the Mekong River at the water intake and pumping station located on the boundary of Xaysathan (upstream side) and Phonsavang village (downstream side). The current water treatment process

at CWTP focuses on removal of common pollutants, such as turbidity, colour, and microorganisms. Although the water quality currently produced by CWTP satisfies the national standard (i.e., Ministry of Natural Resources and Environment, Decree No. 81/MONRE issued in 2017 [12]) and is safe for people's health, the removal of NOM has been not considered during the treatment.

Therefore, the objective of this study is to investigate the optimal operational conditions for NOM removal by chemical coagulation and flocculation, which was carried out through a case study at CWTP. The optimal initial pH of water, types and optimal dosages of coagulants (i.e., alum as  $Al_2(SO_4)_3$ , PAC and FeCl<sub>3</sub>), and flocculant dosages (i.e., PAM) for NOM removal via jar tests were examined. The treatment efficiency is evaluated by considering the removal percentage of turbidity and NOM, in which NOM is measured by total and dissolved organic carbon, ultraviolet absorbance at a wavelength of 254 nm, and trihalomethane formation potential.

#### Materials and methods

# *Raw water samples collection, preservation, and characterisation*

Raw water samples are collected from the water intake of CWTP by using the grab sampling method with 10 high density polyethylene (HDPE) tanks with 20 1 capacity. The grab sampling procedure includes 2 sampling times, where the interval between samples is 16 hours, thus the experimental water samples were obtained from mixed samples. The sampling procedure was taken at a specific time when the pumping station is operating at the average daily flow rate. Since the pH and dissolved oxygen (DO) of the water sample can change rapidly once the sample is removed from the flow, these parameters were measured on-site during the grab sampling.

Afterwards, all samples were preserved by sodium thiosulfate  $(Na_2S_2O_3)$  to eliminate biological reaction, hydrolysis of organic compounds and complexes, and water evaporation. It was reported that  $Na_2S_2O_3$  is a satisfactory dechlorinating agent that neutralizes any residual halogen and prevents the continuation of bactericidal actions during the transfer and chilling of samples at 4°C.

The properties of raw water were characterized by physical-chemical parameters including pH, temperature, turbidity, TOC, DOC, UV-254, THM content, and THMFP. The above factors are important to assess the occurrence of NOM in water. Due to the heterogeneous and undefined character of NOM, surrogate parameters (i.e., TOC, DOC, and UV-254) are normally used for measurement [6]. Also, UV-254 provides an indication of NOM concentration and the DBPs formation potential when  $Cl_2$  is added for disinfection [13].

### Jar test experimental design

Jar test apparatus: in this study, a common jar test apparatus containing six paddles corresponding to six 1.0-1 beakers (i.e., B1-B6) was used. An rpm gauge at the topcentre of the system allowed the control of mixing speed in all beakers (i.e. 20 rpm in 3 mins for initial rapid mixing or 200 rpm in 17 mins for slow mixing flocculation). The jar test system simulates the coagulation and flocculation process at CWTP to investigate the practicability of removal of suspended colloids and organic matter from water. Thus, the jar test control procedure was based on the real operational parameters at CWTP.

*Coagulants and flocculants preparation*: during the jar test experiments, alum, PAC, and FeCl<sub>3</sub> were used as coagulants, while PAM was used as the flocculant. The preparation of the above reagents is described below.

- Coagulants, including alum, PAC, and  $\text{FeCl}_3$ , at different dosages (i.e., 10, 20, 40, 60, 80, and 100 mg/l) were prepared from their corresponding 1% stock solutions and distilled water.

- The flocculant PAM, at different dosages (i.e., 0.05, 0.10, 0.15, 0.20, 0.25 and 0.30 mg/l), was prepared from a 0.01% (100 mg/l) stock solution. PAM is an anionic organic polymer used widely in water treatment as a coagulant aid with inorganic coagulants to enhance performance due to its high molecular weight and long polymer chains.

Samples preparation: raw water samples were used to test with the three coagulants (i.e.,  $Al_2(SO_4)_3$ , PAC, and FeCl<sub>3</sub>) in duplicate experiments. A total of 576 samples were used during the jar test experiments. The alkalinity and pH of all samples were measured first. Then, a pH adjustment was carried out by using 1 N NaOH and 1 N HNO<sub>3</sub> solutions.

Jar test experimental design: a summary of the jar test experiments is presented in Table 1. During the experiments, 3 types of coagulants, including alum, PAC, and FeCl<sub>3</sub>, and flocculant PAM were used. Each substance was divided into 4 experiments (Table 1).

The experiments were conducted by varying the dosage of the 3 coagulants in a range of 10-100 mg/l and flocculant in a range of 0.05-0.30 mg/l at an initial pH range of 4.0-9.0. During experiments 1, 2 and 3, the turbidity, DOC concentration, and UV-254 value were measured and considered to determine the optimal dosage of coagulant, pH, and flocculant. In experiment 4, all parameters, including turbidity, DOC, TOC, UV-254, and THMFP, were simultaneously evaluated to compare the treatment efficiency between different coagulants.

The jar test operation was conducted at room temperature conditions (20°C) in a duplicate-mode experimental design. All chemicals were analytical grade supplied by Water Specialist Supply Co., Ltd (www.wssthailand.com). The solutions and reagents were prepared by using distilled water.

#### Analytical methods and calculation

All samples before analysis were preserved according to the standard methods of APHA, AWWA, and WEF (2005) [14]. The physical and chemical parameters were then analysed and measured under laboratory conditions in accordance with the standard of APHA, AWWA, and WEF [14]. Specifically, the pH was determined by using a pH meter (Model: Eutech, cyber scan 510 PC) and turbidity was measured by using a turbidity meter (Model: HACH 2100 P). The UV-254 absorbance measurements were carried out by a UV/vis Spectrometer (Model: Jasco V-530 at a wavelength of 254 nm with a 1 cm quartz cell). Before UV-254 analysis, the samples were filtered through a prewashed membrane filter with pore size of 0.45 µm to remove turbidity. For NOM parameters, the TOC measurement was performed with a TOC analyser (Model: Tekmar-Dohrman Phoenix 8000), whereas for DOC, samples were firstly filtered through glass-fibre filters (GFC) of pore size 0.45 µm before TOC analysis. THM concentration was determined by the liquid-liquid extraction gas chromatographic method, in which the total concentration of the four THMs (chloroform, bromodichloromethane, dibromochloromethane, and bromoform) was reported as TTHM in units of µg/l. The

No.	Experiment 1			Experiment 2			Experiment 3			Experiment 4			
	Coagulant dose (mg/l)	pH initial	PAM dose (mg/l)	Coagulant dose (mg/l)	pH initial	PAM dose (mg/l)	Coagulant dose (mg/l)	pH initial	PAM dose (mg/l)	Coagulant dose (mg/l)	pH initial	PAM dose (mg/l)	
1	10	7.0	0.10	a	4.0	0.10	a	b	0.05				
2	20	7.0	0.10	a	5.0	0.10	a	b	0.10	a	b	c	
3	40	7.0	0.10	a	6.0	0.10	a	b	0.15	a			
4	60	7.0	0.10	a	7.0	0.10	a	b	0.20				
5	80	7.0	0.10	a	8.0	0.10	a	b	0.25	Be corresponding to each coagulant			
6	100	7.0	0.10	a	9.0	0.10	a	b	0.30	-			

Table 1. Summary of Jar test experiments.

Coagulant:  $Al_2(SO_4)_3$ , PAC, and FeCl<sub>3</sub>; flocculant: anion polymer (PAM); a: optimal coagulant dose obtained from experiment 1; b: optimal pH obtained from experiment 2; c: optimal flocculants dose obtained from experiment 3.

TTHM measurement was carried out by a head-space gas chromatograph ECD detector (Model: Perkin Elmer, Autosystem XL) and column Supelco 241 35-U PTE<sup>tm</sup>-5 under the following conditions: carrier gas N<sub>2</sub> and He flow rates of 2 ml/min, injection temperature of 220°C, oven temperature of 55°C for 15 min, and detector temperature of 300°C. The THMFP was determined by measuring THMs formed after adding Cl<sub>2</sub> (~20 mg/l) to all samples within a reaction time of 4 h at 35°C. THMFP was calculated from the difference in concentration between the instantaneous THMs (inst THMs) and terminal THMs (term THMs). Inst THMs is the THM concentration in water measured while sampling. In contrast, the total THMs (TTHM) or term THMs is the THM concentration measured at the end of the reaction between Cl<sub>2</sub> and precursor in the water supply system.

#### **Results and discussion**

#### Characterisation of raw water

The characterisation results of raw water collected from the Mekong river at the CWTP water intake is presented in Table 2. Specifically, the pH was in a range of 7.48-8.20 and turbidity was 13.00-15.60 NTU. Organic substances measured in the form of DOC, TOC, UV-254, and THMFP, were detected in large quantities (i.e., 1.82-3.98 mg/l, 2.61-4.72, 0.054-0.514 cm<sup>-1</sup>, and 87.53 µg/l, respectively). However, the concentrations of total THMs were not detected ( $\leq 1$ ) in the raw water since disinfection with Cl<sub>2</sub> had not yet occurred at this stage and, thus, the reaction between organic substances and Cl, has not taken place.

 Table 2. Characteristics of raw water collected at CWTP water intake.

Parameter	Unit	Raw water	National Standard (Lao PDR)
pH		7.4-8.2	5-9
Turbidity	NTU	13.0-15.6	<20
Dissolve organic carbon (DOC)	mg/l	1.82-3.98	2 <sup>(3)</sup>
Total organic carbon (TOC)	mg/l	2.61-4.72	4(4)
Ultraviolet at 254 nm (UV-254)	cm <sup>-1</sup>	0.054-0.514	-
Total THMs <sup>(1)</sup>	μg/l	ND <sup>(2)</sup>	-
THMFP	μg/l	87.53	_

<sup>(1)</sup>Total THMs is measured and calculated by the concentration of CHCl<sub>3</sub>, CHBrCl<sub>2</sub>, CHBr<sub>2</sub>Cl, and CHBr<sub>3</sub>; <sup>(2)</sup>ND= Not detected due to the detection limit of the analysis method; <sup>(3)</sup>According to US-EPA standard; <sup>(4)</sup>According to WHO standard.

As compared to the Laos National Standard of Raw Water Sources issued by the Ministry of Natural Resources and Environment, Decree No. 81/MONRE (2017), the water quality at the CWTP water intake satisfied the regulations. For NOM parameters (i.e., TOC, DOC, UV-254) and DBPs

(i.e., TTHMs and THMFP), there is still no standard to apply to raw water in Laos at the moment. The results of raw water characteristics were then used as a background data to evaluate the removal efficiency during the coagulation flocculation simulated by the jar test experiments conducted in this study.

# *Optimal conditions for coagulation - flocculation and water treatment efficiency*

*Optimal dosage of coagulants:* a jar test experiment was carried out to determine the optimal dosage of coagulants. The  $Al_2(SO_4)_3$  PAC, and FeCl<sub>3</sub> concentrations were varied in a range of 10.0-100.0 mg/l, corresponding to beakers 1-6, whereas an initial pH of 7.0 and an PAM polymer dose of 0.10 mg/l were kept constant. The results were evaluated based on turbidity, DOC, and UV-254 of the water samples pipetted from beaker after static settling.

Specifically, the turbidity decreased linearly along with the increase of  $Al_2(SO_4)_3$  dosage (Fig. 1). The highest turbidity removal efficiency, 97.31%, corresponding to a turbidity of 0.35 NTU, was obtained at an  $Al_2(SO_4)_3$  dosage of 100 mg/l.

In contrast, when PAC and FeCl<sub>3</sub> were used, the turbidity removal efficiency fluctuated with the increase of PAC and FeCl<sub>3</sub> dosage. The highest turbidity removal efficiency was 95.04% (i.e., turbidity of 0.65 NTU) at a PAC dosage of 40 mg/l, and 94.80% (i.e., turbidity of 0.78 NTU) at a FeCl<sub>3</sub> dosage of 60 mg/l.

In terms of DOC, the concentration measured from settled water fluctuated with an increase of coagulant dosage. Accordingly, the highest DOC removal efficiencies were 34.88, 51.76, and 55.35% (i.e. corresponding to DOC concentration of 1.68, 1.92, and 1.21 mg/l), which were found at alum, PAC, and FeCl<sub>3</sub> dosages of 40.0, 40.0, and 60.0 mg/l, respectively.

In the case of alum, the UV-254 sharply fluctuated when alum dosage was increased. In contrast, a slight change was found in the PAC and FeCl<sub>3</sub> case. The lowest UV-254 intensity of settled water was 0.0554 cm<sup>-1</sup>, 0.0528 cm<sup>-1</sup>, and 0.0842 cm<sup>-1</sup> obtained for alum, PAC, and FeCl<sub>3</sub> dosages of 60, 40, and 100 mg/l, respectively.

Previous studies also investigated the removal of NOM and DBPs in the Tigris river (Baghdad) by using alum and FeCl<sub>3</sub> via jar tests [15]. However, the results showed a different trend, in which the increase of alum and FeCl<sub>3</sub> dosage resulted in the decrease of turbidity and NOM. Similarly, another study also showed that when the FeCl<sub>3</sub> amount was increased from 10 to 80 mg/l, the removal efficiency of NOM also increased [16].

When taking all results (i.e. turbidity, DOC, and UV-254) and the cost aspect into consideration, the final optimal



Fig. 1. Change of turbidity (A), DOC (B), and UV-254 (C) at different coagulants dosage.



Fig. 2. Change of turbidity (A), DOC (B), and UV-254 (C) at different initial pH.

 $Al_2(SO_4)_3$ , PAC, and FeCl<sub>3</sub> dosage was chosen as 60.0, 40.0 and 80.0 mg/l, respectively. At the chosen  $Al_2(SO_4)_3$  dosage of 60 mg/l, the turbidity and DOC removal efficiency was not much different at dosages of 100 and 40 mg/l (Figs. 1A and 1B). Similarly, at the chosen FeCl<sub>3</sub> dosage of 80 mg/l, the removal efficiencies of all parameters did not change much (Fig. 1C). These chosen optimal values were then used for further experiments.

*Optimal initial pH of raw water:* the adjustment of pH is an important factor strongly affecting coagulation and flocculation. In this experiment, the initial pH of the raw water samples was varied in a range of 4.0-9.0, corresponding to beakers 1-6. The optimal dosage of  $Al_2(SO_4)_3$  (60 mg/l), PAC (40 mg/l), and FeCl<sub>3</sub> (80 mg/l) obtained from Experiment 1, and PAM polymer dose of 0.10 mg/l, were added to all beakers.

Results showed that the turbidity of the settled water in the three cases of coagulants investigated decreased sharply when the initial pH increased from 4 to 5-6 (Fig. 2A). When pH increased to 8-9, the turbidity did not change as much as with the alum, but the turbidity continued to decrease with FeCl<sub>3</sub>. For PAC, an opposite trend was found, as turbidity increased with high pH. Accordingly, the highest turbidity removal efficiencies were 90.00%, 96.75%, and 95.64% obtained at pH of 5.0, 6.0, and 8.0 for alum, PAC, and  $\text{FeCl}_3$ , respectively.

When DOC was examined, the optimal effective pH value was easily found as the peaks of curves. DOC values of 2.31, 1.41, and 1.56 mg/l were found for alum, PAC, and FeCl<sub>3</sub> coagulant, respectively, as seen in Fig. 2B. Accordingly, the highest DOC removal efficiency was 29.57, 43.37, and 34.18% at initial pH values of 7.0, 6.0, and 8.0, respectively. In terms of UV-254, a pH of 7.0, 6.0, and 8.0 was also effective for alum, PAC, and FeCl<sub>3</sub>, respectively, during the coagulation (Fig. 2C).

Previous studies on the removal of NOM by using coagulation with alum, NaAlO<sub>2</sub>, and PAC at a pH range of 5.0-10.0 have been conducted [17]. These findings showed that a pH of 6.0-8.0 is optimal for removing NOM. This can be explained by the fact that alum has a low solubility in a pH range of 5.7-6.2. When the alum dosage is added in excess amounts, alum will form Al(OH)<sub>3</sub>, which enhances the removal of turbidity. In contrast, when the pH is below 5.7, alum will dissolve in water in the form of a cations such as Al<sup>3+</sup>, Al(OH)<sub>2</sub><sup>+</sup>, and Al(OH)<sup>2+</sup>. These cationic forms are able to give a neutralization charge at the surface of colloidal particles. On the other hand, if pH is in a basic range, the cationic states will change to Al(OH)<sub>4</sub><sup>-</sup>.



Fig. 3. Change of turbidity (A), DOC (B), and UV-254 (C) at different PAM dosages.

Another study on turbidity and NOM removal used coagulation with PAC and alum in water from the Yellow river of China [18]. The results showed that an initial pH of 6.0 is more efficient to remove turbidity, DOC, and UV-254 with the removal efficiencies of 86%, 45%, and 55%, respectively. With a pH lower than 6.0, PAC will dissolve well in water and change form to a monomer and the cationic polymers  $Al_{13}O_4(OH)_{24}^{7+}$ ,  $Al^{3+}$ , and  $AlOH^{2+}$ . Similarly, when the pH is lower than 8.0, the FeCl<sub>3</sub> coagulant will also change to a cationic monomers like Fe<sup>3+</sup>, FeOH<sup>2+</sup>, and Fe(OH)<sub>2</sub><sup>+</sup> [19, 20]. Under this condition, NOM has a high density of negative ions (anion) and coagulants in cation form, which enhance the neutralization and precipitation as well.

In this study, when the three parameters turbidity, DOC, and UV-254 were considered, the optimal initial pH was chosen as 7.0, 6.0, and 8.0 for  $Al_2(SO_4)_3$ , PAC, and FeCl<sub>3</sub>, respectively. The above optimal pH values were then used to in further experiments.

*Optimal PAM polymer dosage:* in this experiment, the PAM polymer dosage was varied in a range of 0.05-0.30 mg/l, corresponding to beakers 1-6. The optimal  $Al_2(SO_4)_3$  dosage of 60 mg/l, PAC dosage of 40 mg/l, and FeCl<sub>3</sub> dosage of 80 mg/l, obtained from Experiment 1, and corresponding optimal pH of 7.0, 6.0, and 8.0, obtained from Experiment 2, were constant across all beakers.

Figure 3A shows that the increase of polymer dosage promoted the flocculation in the case of alum and FeCl<sub>3</sub> coagulants. Specifically, the turbidity of settled water in these cases decreased sharply along with the increase of PAM dosage. However, when the coagulant PAC was used, an increase of PAM polymer dosage over 0.2 mg/l caused a lower performance as the turbidity of water increased. Therefore, in terms of turbidity removal, a PAM dosage of 0.3 mg/l was effective when using coagulant as alum and FeCl<sub>3</sub>, whereas a PAM dosage of 0.2 mg/l should be used with PAC.

However, the use of PAM polymer had little effect on NOM removal evidenced by DOC. As shown in Fig. 3B, the DOC concentration in all cases changed slightly when PAM dosage was varied. Accordingly, the NOM removal efficiency was around 30-40% in all cases. A PAM dosage of 0.2 mg/l was seemly effective for the alum and PAC cases, whereas a PAM dosage of 0.3 mg/l resulted in high NOM removal for the FeCl<sub>3</sub> coagulant.

In terms of NOM removal determined by UV-254, trends different from those measured by DOC were found (Fig. 3C). However, a PAM dosage of 0.2 mg/l still showed as an effective dosage for alum and PAC case. Also, a PAM dosage of 0.3 mg/l caused a high performance in the FeCl<sub>3</sub> case.

At the end of this experiment, when all results were considered simultaneously, the optimal dosage of the PAM polymer was chosen as 0.20, 0.20, and 0.30 mg/l for  $Al_2(SO_4)_3$ , PAC, and FeCl<sub>3</sub>, respectively. These optimal values were used in the final experiment to compare the performance of different coagulants.

*Comparison of different coagulants:* experiment 4 was conducted based on the results obtained from experiments 1, 2, and 3. Specifically, the optimal coagulant dosage (i.e.,  $Al_2(SO_4)_3$  dosage of 60.0 mg/l, PAC dosage of 40 mg/l, and FeCl<sub>3</sub> dosage of 80.0 mg/l), optimal initial pH of water sample (i.e. 7.0 with  $Al_2(SO_4)_3$ , 6.0 with PAC, and 8.0 with FeCl<sub>3</sub>), and optimal PAM dosage (i.e., 0.20 g/l with  $Al_2(SO_4)_3$ , 0.20 mg/l with PAC, and 0.30 mg/l with FeCl<sub>3</sub>), were used. The jar test in experiment 4 compared the removal efficiency of the different coagulants. In this experiment, 5 parameters were considered to evaluate the treatment efficiency, including Turbidity, DOC, UV-254, TOC, and THMFP (Table 3 and Fig. 4).

	Optimal conditions				Influent $(C_{in})^{(*)}$ and effluent $(C_{eff})^{(*)}$ concentration								
Type of coagulant	Coagulant (mg/l)	Initial pH	Polymer (mg/l)	Turbidity (NTU)		DOC (mg/l)		TOC (mg/l)		UV-254 (cm <sup>-1</sup> )		THMFP (µg/l)	
				C <sub>in</sub>	C <sub>eff</sub>	C <sub>in</sub>	$C_{e\!f\!f}$	C <sub>in</sub>	C <sub>eff</sub>	C <sub>in</sub>	$C_{e\!f\!f}$	C <sub>in</sub>	C <sub>eff</sub>
$Al_2(SO_4)_3$	60.0	7.0	0.20	14.30	1.29	1.98	1.18	2.61	1.31	0.054	0.035	87.53	45.40
PAC	40.0	6.0	0.20	14.48	1.31	1.82	1.25	2.65	1.26	0.075	0.061	87.53	48.91
FeCl <sub>3</sub>	80.0	8.0	0.30	14.36	0.73	2.11	1.22	4.72	3.81	0.078	0.047	87.53	45.41

Table 3. Result of jar test operation for removal of turbidity and NOM from raw water with different coagulants at optimal conditions.

(\*) $C_{in}$  values were extracted from the results of raw water characteristic shown in Table 2; (\*\*) $C_{eff}$  values were the results obtained in experiment 4.



Fig. 4. Removal efficiency of turbidity (A), TOC (B), DOC (C), UV-254 (D), and THMFP (E) at the optimal operational conditions for coagulation and flocculation.

When all results were compared (Fig. 4 and Table 3), the FeCl<sub>3</sub> coagulant at an optimal dosage of 80 mg/l showed the highest removal efficiency of turbidity and NOM. Other optimal conditions were pH of 8.0 and PAM polymer dosages of 0.30 mg/l. However, it is practically unsuitable to use FeCl<sub>3</sub> as a coagulant in water treatment plant due to its yellow colour, which affects the aesthetics of drinking water.

On the other hand,  $Al_2(SO_4)_3$  is more effective than PAC as the turbidity and NOM removal efficiency obtained with  $Al_2(SO_4)_3$  was higher than with PAC. The optimal conditions, however, were different. Specifically, a high removal efficiency was achieved at an  $Al_2(SO_4)_3$  dosage of 60 mg/l, pH 7.0, and PAM dosage of 0.20 mg/l. Meanwhile, PAC showed maximum effectiveness at an optimal dosage of 40 mg/l, pH 6.0, and PAM dosage of 0.20 mg/l.

Table 4. The cost of coagulants and flocculants.

Type of coagulant	Coagulant dosage (mg/l)	Flocculant dosage (mg/l)	Price of Coagulant dollar/m <sup>3</sup> raw water	Price of Flocculant dollar/m <sup>3</sup> raw water	Total price dollar/m <sup>3</sup> raw water
Al <sub>2</sub> (SO <sub>4</sub> ) <sub>3</sub>	60.00	0.20	0.042	0.0014	0.0434
PAC	40.00	0.20	0.140	0.0014	0.1414
FeCl <sub>3</sub>	80.00	0.30	0.112	0.0021	0.1141

Note:  $Al_2(SO_4)_3=0.7$  dollar/kilogram<sup>(\*)</sup>; PAC=3.5 dollar/kilogram<sup>(\*)</sup>; FeCl\_3=1.4 dollar/kilogram<sup>(\*)</sup>; <sup>(\*)</sup>The market price was obtained at the time of purchase, which was issued by Water Specialist Supply Co., Ltd.

When considering the cost of the chemicals in Table 4,  $(Al_2(SO_4)_3=0.7 \text{ dollar/kilogram}, PAC=3.5 \text{ dollar/kilogram})$  and FeCl<sub>3</sub>=1.4 dollar/kilogram) and the practical situation in the CWTP water supply system, it is recommended to

use  $Al_2(SO_4)_3$ , instead of PAC and  $FeCl_3$ . For PAM polymer, the usage is similar. The total cost of chemicals of all three conditions was estimated as 0.0434, 0.1414 and 0.1141 dollar/m<sup>3</sup> raw water, for alum, PAC, and FeCl<sub>3</sub> respectively. Therefore, it is concluded that  $Al_2(SO_4)_3$  is the most suitable coagulant to use in an actual water production system by adjusting the optimal dosage, initial pH, and flocculant dosage.

#### Conclusions

This research investigated the optimal operational conditions of the chemical coagulation and flocculation process with different coagulants (i.e. alum, PAC, and FeCl<sub>2</sub>) to remove turbidity and NOM from surface water collected at the CWTP water intake. The results showed that the optimal dose of coagulants for alum is 60 mg/l, PAC is 40 mg/l, and FeCl, is 80 mg/l. The increase of coagulant dosage affected the removal efficiency of turbidity and NOM, in which the removal efficiency of turbidity was higher than that of NOM. The optimal initial pH for the coagulation and flocculation process with alum, PAC, and FeCl, was 7.0, 6.0, and 8.0 respectively. Therefore, in the treatment process for the removal of turbidity and NOM, it is suggested to adjust the initial pH of raw water to the above optimal values to improve the performance. The addition of the anion polymer PAM significantly affected the removal efficiency of turbidity, whereas little effects were found in the case of organic substances. The optimal dose of the anion polymer for flocculation when using different coagulants such as alum, PAC, and FeCl, were 0.20, 0.20, and 0.30 mg/l respectively. In terms of THMFP, this study showed that the coagulation and flocculation treatment under experimental conditions resulted in high removal efficiencies. Specifically, the THMFP concentration as measured in settled water satisfied the standards of WHO and US-EPA after coagulation-flocculation. Thus, it is concluded that the optimal conditions found during the jar test experiments in this study were able to maximize the treatment efficiency for reducing the formation of THMs. In addition, a comparison of the suitability and cost of the coagulants indicated that the effective coagulant to be applied in CWTP is alum,  $Al_2(SO_4)_2$ . Actually, this cost effective coagulant is currently being used at CWTP.

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