

# CONTROL OF ORGANIC MATTER FROM SURFACE WATER IN THE COASTAL AREAS USING A HYBRID COAGULATION-MEMBRANE SYSTEM

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**ABSTRACT:** With the impact of climate change, it is impossible for the residents living in coastal areas to employ underground water due to the severe salinity intrusion, they switch to using surface water instead. Nevertheless, the surface water source often has high organic matter, turbidity, and occasionally gets salinity, which results in difficult and costly treatment. This paper evaluated the efficiency of using a hybrid coagulation and ultrafiltration (UF) process in treating surface water in coastal areas of Vietnam. A series of preliminary jar tests were conducted to optimize the pH condition, coagulant dose, and impact of salinity on turbidity and organic matter control. The tests with the hybrid coagulation-UF system were tried after that and fouling evidence was investigated. The jar test results rendered that pH of 6.5, Poly Aluminum Chloride (PAC) with a concentration of 10 mg/L, and pre-chlorination with 10mg/L NaOCl solution were the best conditions for UF pretreatment. Salinity influenced the coagulation process, not the membrane one. The removal efficiencies of turbidity and total organic carbon (TOC) removal were 88% and 66% after coagulation, and 95% and 67% after the hybrid processes, respectively. The presence of trihalomethanes (THMs), specifically, *Chloroform*, *Dibromochloromethane*, and *Bromodichloromethane* were detected in the disinfected water, but much lower than the standard. The THMs formation depended on the contacting time and chlorination doses. The proposed system initially showed some promising results in controlling the pollutants of this river source in the coastal area. Future studies would look at thoroughly the impact of salinity on THMs formation for this kind of water.

**Keywords:** Surface water, Hybrid coagulation-UF, THMs formation, Salinity impact

## 1. INTRODUCTION

Within the last few decades, because of climate change's impacts, salinity tends to penetrate deeper via the estuaries of rivers in the dry season, affecting the quality of water for domestic water supply and irrigation in the coastal areas. In addition, the use of plant protection chemicals in agricultural production, as well as the discharge of untreated industrial and domestic wastewater into the river are some main reasons causing high organic matter in these rivers. According to Chau et al. [1], 165 out of 1153 micropollutants examined were detected in surface waters, of which more than 100 contaminants occurred at  $\mu\text{g/L}$  level concentrations in Hanoi and Hochiminh city.

Meanwhile, water treatment systems from centralized water plants in the coastal areas currently apply conventional processes only which involve coagulation, flocculation, sedimentation, and rapid gravity filtration to treat sediment and part of organic matter. To increase the efficiency of organic matter treatment, there have been some applications of pre-chlorination, pre-ozonation, biological contact filters, or adsorption with activated carbon. Chlorine and chlorine-containing solutions are also used in Vietnam and other

developing countries to disinfect treated water at treatment plants. Nevertheless, in developed countries, to safely control organic matter in surface water without disinfection by-products (DBPs), some technologies such as membrane process [2,3], adsorption with activated carbon [4,5], pre-oxidation with ozone [3,6], disinfection with ultraviolet (UV) light [7] or a combination of them is often used in practice, at small scale or large scale [8-10]. The activated carbon had good efficiency in organic adsorption and trihalomethanes (THMs) control, however, it is still recommended to combine with membrane filtration [8] or with pre-oxidation and coagulation process as pretreatment [11].

In many previous studies, the hybrid coagulation-ultrafiltration (UF) membrane processes were often recommended since the main advantages of integrating coagulation with membrane filtration are the enhanced removal of natural organic matter and reduced membrane fouling [12-14]. Different coagulants were tried for organic matter control including  $\text{Al}_2(\text{SO}_4)_3$  [15,16],  $\text{FeCl}_3$  [17,18], PAC/PACl [19,20], bio coagulant [11,21] or even drinking water treatment sludge [22]. Among them, PAC or PACl has been used the most for water treatment as it creates less residual,

has high efficiency of flocs formation, and is more available in the market. However, for river waters with a high concentration of organic matter and seasonal salinity in coastal areas, it is necessary to find the best-operating conditions to ensure the water quality standard.

The novel part of this study is to evaluate the impact of hybrid coagulation – UF membrane processes in the control of turbidity, organic matters, the impact of salinity, and the potential formation of disinfection by-products after disinfection for a typical type of surface water in coastal areas in Vietnam.

## 2. RESEARCH SIGNIFICANCE

Even though the research on organic removal from surface water by coagulation and membrane processes has been conducted in many studies, it has not been done thoroughly in Vietnam, in particular for the surface water in coastal areas, which are vulnerable to salinity intrusion with seasons. In addition, the formation of THMs from this kind of water after disinfection has not been studied comprehensively in Vietnam. This paper has shown the initial results that would help understand the proper treatment technology (role of coagulation process and membrane process) and potential responsive actions when using this kind of water as a supply source.

## 3. MATERIALS AND METHODS

### 3.1 Raw Water

Feed water was taken from the Hoat river in Thanh Hoa province, a coastal area of Vietnam from April to June 2022. The water has pH of  $8.5 \pm 0.4$ , total dissolved solids (TDS) =  $750 \pm 43$  mg/L, total organic carbon (TOC) =  $6.3 \pm 2.0$  mg/L, turbidity =  $43 \pm 15$  mg/L, color =  $150 \pm 2.0$  TCU,  $\text{NH}_4^+\text{-N}$  =  $0.23 \pm 0.1$  mg/L, salinity =  $785 \pm 52$  mg/L as NaCl, conductivity =  $1576 \pm 127$   $\mu\text{S}/\text{cm}$ . The river has quite a high pH as it flows through limestone

rocks. It is strongly influenced by tides so the salinity varies quite significantly. The organic matter is quite high, compared to the national standard COD ( $\text{KMnO}_4$ ) of 2 mg/L or equivalent to TOC of 2.3 mg/L for this particular water.

### 3.2 Experimental Diagram and Procedure

The lab-scale diagram is presented in Fig. 1. The feed water was coagulated with PAC powder (28-30%  $\text{Al}_2\text{O}_3$  purity, Viet Tri chemical co., Vietnam) with the optimum dose determined via jar test. Details of the jar test would be described after this. After coagulation (chemical mixing for 1 min, mixing speed of 200 rpm) and flocculation for 5 min, the water was settled for 15 min before being filtered by an ultrafiltration UF membrane unit (PVDF hollow fiber, pore size 0.04  $\mu\text{m}$ , surface area 0.046  $\text{m}^2/\text{unit}$ ). The conditions for coagulation and settling process were referred from previous studies [4,12,24]. The membrane was backwashed every 20 min for the duration of the 30s. As the limited volume of water source for lab scale testing, each run was lasting for 3 days. After each test, the membrane unit was soaked in NaOH 0.5 M (CAS 1310-73-2, Xilong Scientific, China) for 2 hours and HCl 0.5M (CAS 7647-01-0, Xilong Scientific, China) for 2 hours for cleaning before next tests.

Before the membrane testing, the Jar test was conducted to optimize the coagulant dose for the effective coagulation process. For this Jar test, stock PAC was prepared at 1000 mg/L. After that, the optimum coagulation process was conducted at different pH of 5.5, 6.0, 6.5, 7.0, 7.5, 8.0, different PAC concentrations (2mg/L - 20 mg/L), different salinity, and with and without pre-chlorination using NaOCl solution (CAS 7681-52-9, Xilong Scientific, China). The NaOCl solution was added before the PAC supplement, with a dose of 10 mg/L to assess the impact of the pre-oxidation process in organic control for this kind of water. The PAC dose for this test was selected concerning previous studies [19, 20]. The pH was adjusted using HCl acid and NaOH solution.

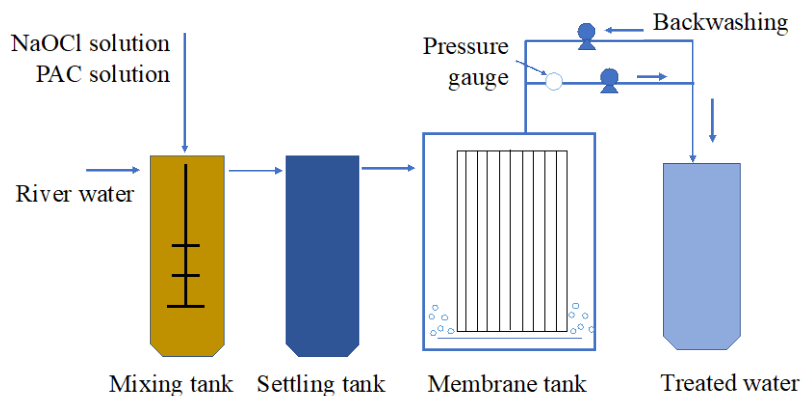


Fig. 1 Experimental diagram

### 3.3 Sampling and Analysis

The organic fractions from the raw and treated water were isolated using nonionic macroporous sorbents XAD7 and XAD4 (Merck, US). The principle of this method is that as the water is passed through two columns containing Amberlite XAD-7 and Amberlite XAD-4 respectively, the hydrophobic fraction of organic matter will be adsorbed by Amberlite XAD-7 and the transphilic fraction will be sorbed on Amberlite XAD-4. The hydrophilic fraction will be in the effluent of the XAD-4 column. The fractionation method was described in detail elsewhere [24].

To understand the organic matter in the raw water, it was subjected to TOC, UV254, and florescent analysis. The Total organic carbon (TOC) of the sample was measured using a TOC analyzer (VCPH, Shimadzu, Japan), and UV254nm was determined by Spectrophotometer (UH5300, Hitachi High Technologies, Japan). The organic content was also further analyzed for Fluorescence spectra using Spectrofluorometer FluoroMax-4 (HORIBA, Japan) at 254nm wavelength. Turbidity was measured by Portable Turbidity Meter (HI-83749-02, Hanna, USA). Other parameters including pH, conductivity, total dissolved solids TDS, and salt percentage were determined by the Smart sensor device (AR8011, China). The analysis of the above water quality parameters was performed according to the procedures in "Standard methods for the examination of water and wastewater" [25].

Moreover, the clean and fouled membrane samples were sent for Scanning Electron Microscope (SEM) using the Tabletop Microscope (TM4000plus, Hitachi, Japan).

To verify the formation potential of DBPs such as trihalomethanes (THMs), the treated samples were disinfected with NaOCl solution with a dose of 10 mg/L and were sent to a qualified lab at the Institute of Occupational Health for analysis of THMs compounds (*Chloroform*, *Dibromochloromethane*, *Bromodichloromethane*, and *Bromoform*).

## 4. RESULTS AND DISCUSSION

### 4.1 Raw Water Fractionation

According to the method, it is possible to separate the hydrophobic, hydrophilic, and transphilic fractions. The hydrophobic fraction usually contains large molecular weight particles, the hydrophilic fraction-operationally defined as a non-humic fraction composes of lower MW compounds (e.g., polysaccharides, amino acids, protein, etc.) and the transphilic fraction comprises particles with molecular weight in between [24].

For this particular river, it was found that the hydrophobic, hydrophilic, and transphilic fractions were 84%, 9%, and 7% respectively. It agreed with the previous comment that the hydrophobic fraction of organic matter in most natural waters was from 60-90% [24]. Even though the hydrophobic fraction had the largest molecular weight, it was not the main contribution to membrane fouling, but the hydrophilic fraction with low molecular weight compounds [14] as it could go inside the membrane pores. In addition, as the humic substances have higher molecular weight, it is easy for them to bind together when coagulants are added, and they are the major fraction to be removed during coagulation [13].

### 4.2 Optimization of pH and PAC dose via Jar Test

As the organic matter was high and the UF membrane, in general, can not control completely this source of contaminant, the effort was made to optimize the coagulation process before the membrane system. Besides the utilization of PAC, an attempt was tried to enhance the organic removal using strong oxidants such as NaOCl solution.

Fig. 2 depicts the jar test results at different pH and different PAC doses with and without pre-chlorination with 10 mg/L NaOCl solutions. While Table 1 presents the effect of salinity on TOC reduction.

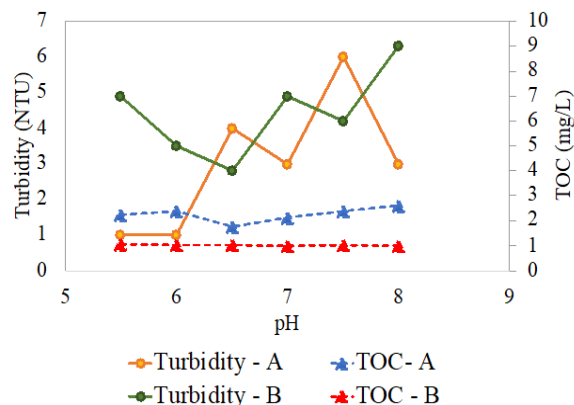


Fig. 2 Impact of pH on turbidity and TOC control (PAC = 10 mg/L, Case A: without pre-chlorination, Case B: with pre-chlorination).

It can be seen from Fig. 2 that pH had a strong impact on turbidity, which was confirmed in some previous studies [4,20]. The pH of 6.5 seems to be the best condition for both cases of with or without pre-oxidation with NaOCl solution. This finding is in line with Zhang et al. [23], in which the pH of 6.5 was found also the best pH condition in their study. The result again confirmed that the pre-oxidation helped improve the organic matter removal as the organic matter was degraded substantially by strong oxidants (the red line was lower than the blue line).

Overall, a pH of 6.5 was chosen for the jar test of different PAC doses.

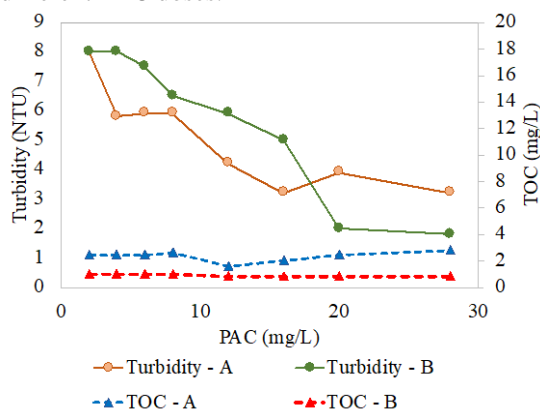


Fig. 3 Impact of PAC doses on turbidity and TOC control (pH = 6.5, Case A: without pre-chlorination, Case B: with pre-chlorination)

The impact of Pthe AC dose was illustrated in Fig. 3. Again, the turbidity removal was strongly influenced by Pthe AC dose, and organic matter was more affected by pre-chlorination. Previous studies also found that organic removal by the coagulation process was normally low, e.g., 30-70% only [19,23,26]. In this study, the TOC removal was from 50-60%. As the increasing dose of PAC helped reduce the turbidity, but not the organic matter, the PAC concentration of about 10-12 mg/L would compromise both technical and economical aspects. For this particular study, PAC concentration of 10 mg/L would be selected for later filtration tests for this kind of raw water. The impact of PAC on turbidity control was also found in many previous studies [10, 12, 14]. Additionally, as this raw water was affected by salt intrusion occasionally in the dry season, an effort was made to evaluate the impact of salinity on turbidity and TOC control in the preliminary jar test.

The salinity was observed to influence the pre-oxidation and coagulation with PAC as can be observed in Table 1. Compared with the blank one, a higher dose of NaCl solution would lead to higher organic concentration, but lower turbidity. This was due to the effect of salt on the aggregation. Specifically, salt ions collect suspended particles and bind them together, increasing their weights and thus their likelihood of settling to the bottom [27, 28]. In addition, it is quite interesting to observe the slight increase of organic matter with the concentration of NaCl solution.

Previous studies suggested that increasing salt might be conducive to leaching organic matter from watershed soils, thus increasing its presence and concentration in the water [29], while Steele and Aitkenhead-Peterson [30] believed that salt increased the leaching of dissolved organic carbon

from abscessed and cut vegetation, which led to the higher organic matter in the water. Nevertheless, in this study, there were no watershed soils or vegetation, hence, the possible explanation may be that the addition of salt possibly created more breakdowns of complex organic matter to detectable carbon levels by the TOC analyzer. As a result, the TOC concentration was recorded higher with higher salinity. One thing should be noted the residual  $\text{Cl}^-$  was very low (from 10 mg/L to 0.14 mg/L), meaning that significant amount of  $\text{Cl}^-$  was consumed to oxidize the organic matter in the water.

Table 1. Impact of salinity on turbidity and TOC control

Samples	Blank river water	River water + 1 mg/L NaCl	River water + 1.5 mg/L NaCl	River water + 5 mg/L NaCl
pH	8.13	8.19	8.17	8.09
EC ( $\mu\text{S}/\text{cm}$ )	1336.00	2900.00	3820.00	9410.00
TDS (mg/L)	670.00	1450.00	1910.00	4710.00
Salinity (%)	0.06	0.15	0.19	0.48
Initial turbidity (NTU)	36.00	20.30	19.60	17.40
Initial TOC (mg/L)	5.86	6.59	6.59	6.79
After adding 10mg/L NaOCl				
Turbidity (NTU)	32.00	17.80	16.50	15.90
TOC (mg/L)	4.52	6.45	5.83	6.72
After adding 10mg/L NaOCl + 10mg/L PAC				
Turbidity (NTU)	4.20	2.70	2.50	2.10
TOC (mg/L)	1.95	2.49	2.32	2.57
Residual $\text{Cl}^-$ (mg/L)	0.14	0.15	0.18	0.23

### 4.3 Evaluation of Removal Efficiency of Testing the System

#### 4.3.1 Organic and turbidity removal efficiency

Under the optimization condition, which employed pre-oxidation with 10mg/L NaOCl solution and 10mg/L of PAC coagulant at pH=6.5, the efficiency of removing turbidity and organic matter was observed. The membrane unit was controlled at constant pressure of 0.4 bar, thus the permeate flux varied with time. It was observed that

the permeate flux reduced from 35 L/m<sup>2</sup>/h to 25 L/m<sup>2</sup>/h within each cycle.

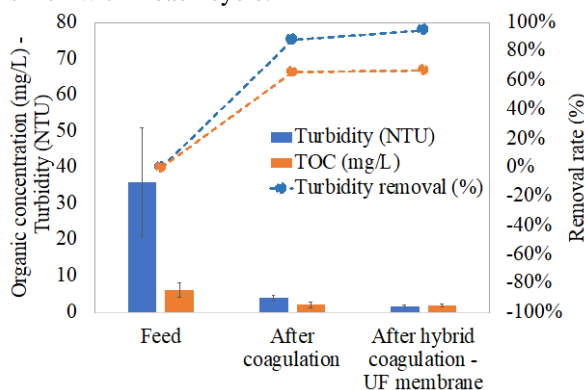


Fig. 4 Organic and turbidity removal by the hybrid coagulation-membrane system

Fig. 4 illustrates the removal efficiency of organic matter and turbidity after the test with a hybrid coagulation-membrane system, in which turbidity was reduced up to 88% after coagulation and 95% after the hybrid processes, while the organic matter was controlled up to 66%. The UF membrane contributed very little in the TOC removal. Normally, the organic retaining by UF membranes was proved limited as in some previous research [9, 10]. The coagulation plays an important role in contaminant control. It was found from the study that the TDS and salinity did not change after the hybrid processes. For this particular water, using the pre-oxidation with 10mg/L NaOCl solution and 10mg/L of PAC coagulant at pH=6.5 itself successfully controls the organic matter to meet the national standard.

Nevertheless, the UF membrane helped reduce the turbidity to the required value for turbidity ( $\leq 2$  NTU) for the water supply quality standard. Compared to several similar studies, the TOC removal rate was a bit higher in this study which could be explained by the high hydrophobic fractions (e.g., high humic-like substances) of 84%. This fraction would be more easily formed agglomeration, bigger flocs which were easily retained by the UF membrane [12]. The fouling evidence and the organic removal can be seen via fluorescence and SEM results as below.

#### 4.3.2 SEM results

The precipitation of contaminants on the membrane surface was illustrated obviously via the SEM images at a magnification of 2500X. (Fig. 5). For the pristine membrane image, it was possible to see the membrane holes. Nevertheless, there were lots of precipitates and aggregates on the membrane surface after the 3-day runs. Those could be salt precipitate and organic compounds. That was the reason leading to flux decrease over time and the

disappearance of humic-like substances in the treated water in 3D fluorescence EEM images.

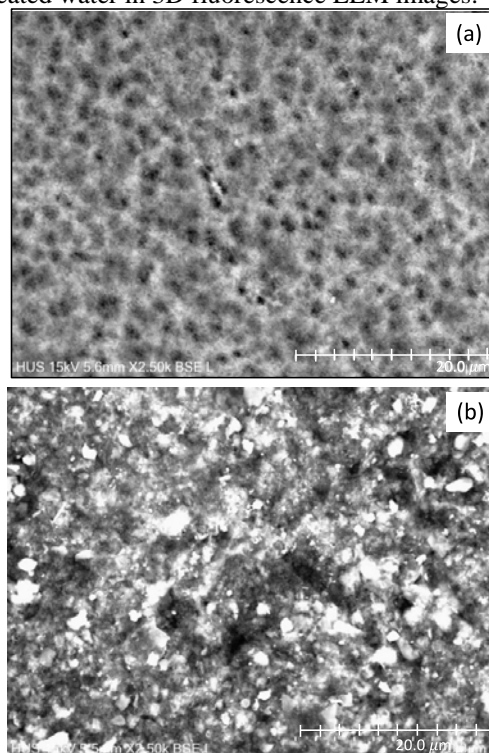


Fig. 5 SEM images at a magnification of 2500 X

#### 4.3.3 Fluorescence result

Fig. 6 presents the synchronized fluorescence spectra and 3D fluorescence excitation-emission matrix of the feed water and treated water. It can be seen clearly that the feed water had much higher fluorescence intensity, which means it had much more contaminated organic matter than that of the treated one (Fig. 6).

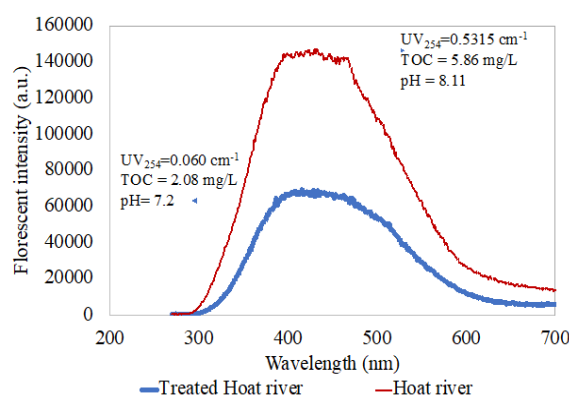


Fig. 6 Synchronized fluorescence spectra

There were multi peaks between the emission wavelength of 400nm and 500nm, which indicated the presence of humic-like substances [31]. The spectra were quite consistent with the organic contents in which the fluorescence intensity's peaks decreased when the TOC values decreased from

5.86 mg/L to 2.08 mg/L, and the UV absorbances at

254nm reduced from 0.5315 cm<sup>-1</sup> to 0.060 cm<sup>-1</sup>.

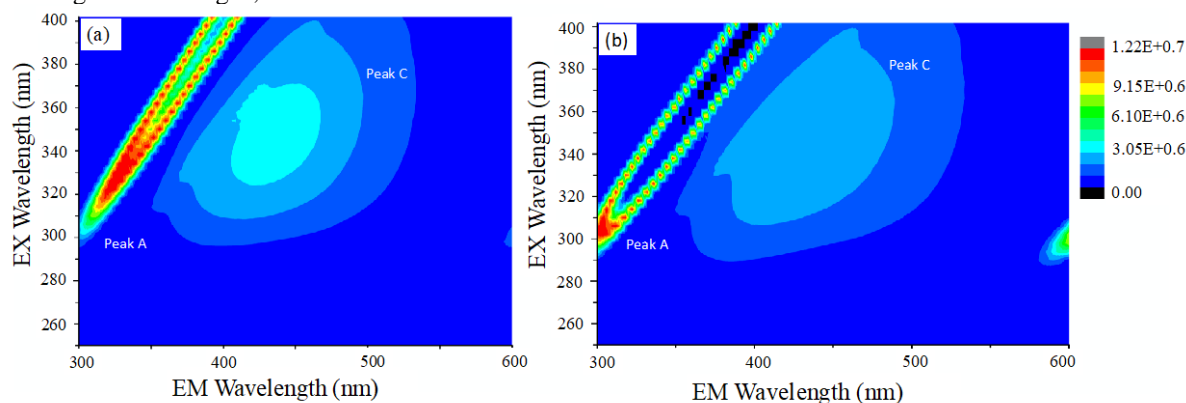


Fig. 7 The 3D fluorescence excitation-emission matrix of raw water (a) and treated water (b)

ND: Not detected, Testing method: EPA 551.1

Additionally, the presence of humic-like organic compounds was quite different between the raw water and the treated one in 3D fluorescence EEM. The high fluorescence intensity (depicts in red and orange color) in peak A was much less in treated water (Fig. 7b) compared to the feed water (Fig. 7a), indicating the efficiency of the testing treatment system in organic removal, in particular the humic-like substances. There was no evidence of protein-like substances based on the EEM images when compared with the standard classification [32]. From Fig. 7b, there was still a small amount of humic-like substance (in peak A) remaining in the treated water sample. A previous study revealed that humic acid-like compositions predominantly contributed to regulated DBP yields [33], thus an attempt was made to assess the THMs formation after the disinfection of the treated water.

#### 4.4 Evaluation of THMs formation

To understand more about the potential formation of THMs for this kind of water, different disinfectant doses (residual chlorine were 1.5 mg/L and 0.75 mg/L), and different retention times after disinfection (2 hrs and 24 hrs) were applied with the permeate samples. It should be noted that the accepted range of residual chlorine in the water supply in Vietnam is from 0.2 – 1.0 mg/L, thus, the disinfectant doses were chosen for the test as above. The results are presented in Table 2.

Table 2. THMs formation

THMs	1.5mg/L residual Cl <sup>-</sup>		0.75mg/L residual Cl <sup>-</sup>		Max. values
	2hrs	24hrs	2hrs	24hrs	
Chloroform	27.55	8.03	20.42	5.26	300
Dibromochloromethane	19.65	3.01	8.07	2.06	100
Bromodichloromethane	22.87	5.72	14.89	3.78	60
Bromoform	ND	ND	ND	ND	100

Note: Unit: µg/L.

It can be seen from Table 2 that it was possible to detect some trihalomethanes (THMs) in the treated water such as *Chloroform*, *Dibromochloromethane*, *Bromodichloromethane*. The presence of *Dibromochloromethane*, *Bromodichloromethane* could be due to the industrial discharge to the river somewhere upstream. The same phenomenon was reported in previous research [34, 35]. Table 2 showed that higher disinfectant doses would lead to higher values of THMs. It was consistent with the findings of Liu et al. [36] in which higher concentrations of total DBPs were found under higher chlorine dosages, higher temperatures, and lower pH. Nevertheless, they found that longer contact times led to higher DBPs formation. It contradicted this study in which the THMs were high at the beginning of the chlorination process, and reduced after 24 hours. The possible explanation is that there was some evaporation during the sample transport to the lab after 24 hrs.

The evaporation occurred when water was exposed to air [34] via the samples' bottle caps. In addition, the contact time in Liu et al. [35] study was short (within three hours) which is still probably the effective reaction time for THMs formation. Overall, all the detected THMs values were lower than the national standard, which would not be of concern to the residents using this water source for domestic use upon treatment and disinfection with chlorine dioxide.

## 5. CONCLUSIONS

For the surface water source in coastal areas, salinity would vary with seasons and impact the coagulation doses in the hybrid coagulation-UF membrane processes. It helped improve the precipitation of suspended solids, but not the removal of the organic matter. The combination of pre-oxidation with NaOCl solution, coagulation with PAC and ultrafiltration helped remove the

turbidity up to 95% and organic matter up to 66%.

There was the presence of THMs in the treated water being disinfected with NaOCl, but still lower than standard values. Future research would evaluate the impact of salinity and contact times on THMs formation in the surface water near coastal areas.

## 6. ACKNOWLEDGMENTS

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