

Controlling Disinfection by-Products via Enhanced Coagulation: A Case Study, EL-Nobarya Drinking Water Treatment Plant, Egypt

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Abstract

The present study aims to reduce Trihalomethane (THMs) level, residual aluminum and control filter run time in EL-Nobarya drinking water treatment plant via the application of enhanced coagulation technique. The effectiveness of the treatment process was evaluated in terms of the removal of natural organic matter (NOM), and the decrease in THM levels in the period from January to August 2020. First, the optimal coagulant dose (60-70 mg/L) for NOM removal was determined on bench scale and then applied on the plant. The results showed that NOM and THM concentrations were reduced by 28-44% and 25-35%, respectively. The residual aluminum varied from 0.1-0.13 mg/L compared to 0.18-0.26 mg/L using conventional treatment. In addition, the filter run time increased from 12 hours to 24 hours saving about 250 m³ of washing water for each filter. Enhanced coagulation was adopted as a controlling strategy for THM in EL-Nobarya plant and EL-Beheira water&drainage company (BWADC). Further control of chlorine dose and point of injection was needed for more reduction in THM.

Keywords: Chlorination, Enhanced coagulation, Natural organic matter, Residual aluminum, Trihalomethanes.

Introduction

Chlorination is the most prevailing disinfection technique for ensuring the biosafety of drinking water and wastewater (Xu et al., 2023). However, the formation of harmful disinfection by-products (DBPs) such as trihalomethanes (THMs), halonitromethanes, haloacetonitriles, haloketones, haloaldehydes

and haloacetic acids (HAAs) through the reaction of chlorine with natural organic matter (NOM) is the main drawback of chlorination (Qian et al., 2021).

These compounds have adverse effects on aquatic environment and human health due to their high mutagenicity, cytotoxicity and carcinogenicity (Chen et al., 2019). Among DBPs, THMs is detected extensively in tap water with relatively high concentration. The

presence of THMs in drinking water have linked with heart abnormalities, premature births, colon, bladder and rectum cancers (**Li et al., 2017**). Due to the harmful effects of THMs, the US Environmental Protection Agency (EPA) set a maximum contaminant level (MCL) for THMs of 80 µg/L in groundwater and drinking water (**Mazhar et al., 2020**).

The formation of THMs in drinking water had been shown to be a function of various factors such as disinfectant concentration, NOM concentration, contact time, temperature and pH (**Albanakis et al., 2021**). The concentration and characteristics of NOM are considered to have the most significant influence on the generation of DBPs (**Golea et al., 2017; Chaves et al., 2019**). Thus, it is vital to efficiently eliminate NOM and other DBP precursors to protect water quality throughout water treatment. However, NOM cannot be directly measured, it is expressed through TOC, DOC or UV₂₅₄ (**Musteret et al., 2021**).

Coagulation process have traditionally been used for turbidity removal in water treatment plant and can also be optimized to be an effective means of removing NOM. Enhanced coagulation (EC) is one of the promising techniques used for improving the removal of disinfectant byproducts precursor through the addition of excess coagulant. D/DBP rule proposed by US Environmental Protection Agency (**USEPA, 1999**) requires the implementation of enhanced coagulation as a treatment technique for NOM removal as a means of limiting the formation of all DBPs. By applying EC technique, 50 - 75% of NOM can be removed by using aluminum and ferric salts or by cationic polymers, depending on the nature of the organic matter (**Sun et al. 2020; Sapingi et al., 2017**). The coagulant dose that yields optimal NOM or DBP precursor removal is the most important factor, but effective turbidity removal must still be provided (**Garcia, 2005**).

Enhanced coagulation is a valuable method for controlling DBP formation that does not require a significant capital investment and may even obviate the need for expensive and sophisticated treatment processes (**Ghernaout et al., 2021; Lopez-Maldonado et al., 2014**). Since, enhanced coagulation can be applied in developing countries where it is almost impossible to afford high technologies or costly upgrading current treatment processes.

THM concentration in El-Nobarya water treatment plant, EL-Nobarya city, Egypt, was high all over the year mostly exceeding the limit of 80 µg/L set by the USEPA. The plant uses a traditional low alum dose (30-35 mg/L) which led to only 9-20% removal of NOM, while 15-25% removal is needed according to the **USEPA D/DBP rule (Stage1 1998, 2006)**. In addition, residual aluminum usually exceeds the limit of 0.2 mg/L (**WHO 2017**). Therefore, the present study aims to improve the coagulation process in EL-Nobarya water treatment plant as a mean for reducing the THM and residual aluminium levels which resulting in better overall treatment efficiency. This work was applied on both bench scale (jar test) and large scale on the treatment plant constantly. This was the first applicable study of enhanced coagulation technique in a treatment plant in Egypt as a strategy to reduce THM level and improving the water quality produced by the treatment plant as a whole.

Materials and methods

Plant description

This study was performed from January to August 2020, in EL-Nobarya water treatment plant, El-Nobarya city, EL-Beheira, Egypt. The capacity of the plant is 2160 m³/h and it supplies drinking water to El-Nobarya city and the surrounding village. The treatment system of this conventional plant comprises of coagulation (using aluminium sulphate) followed by flocculation, sedimentation, five rapid sand filters, contact tank and rectangular clear well. Chlorine which used for disinfection is injected from two of five injection points and the Residual chlorine (RCl₂) is measured at the end of the clarifier and the clear water well (Figure 1). A low alum dose 30-35 mg/L is used in EL-Nobarya plant which led to only 9-20% removal of NOM. The total organic carbon (TOC) of El-Nobarya raw water ranges from 2-10 mg/L and varying seasonally, being high at summer and autumn months.

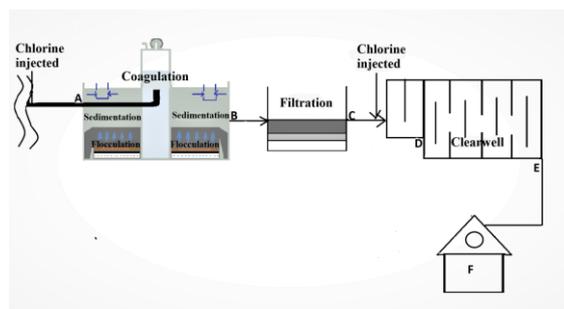


Figure 1: Schematic diagram for EL-Nobarya water treatment plant: A, B, C, D, E, and F are residual chlorine monitoring points.

Experimental setup

The present study was conducted to investigate the application of enhanced coagulation in El-Nobarya water treatment plant to control the high levels of THM, residual aluminum and short filter run time. The optimal coagulant dose for NOM removal was determined as a crucial step. The jar test was carried out within 30 min from collecting raw water at ambient temperature ($\sim 25^{\circ}\text{C}$) according to **Lytle (1995)**. The effect of alum dose on TOC, pH, turbidity, color, alkalinity, and residual aluminum concentrations was determined. Alum dose with optimum removal efficiency was then chosen to be applied in the treatment plant. Consequently, the effectiveness of the enhanced coagulation was evaluated in terms of the percentage removal of TOC (mg/L), and the decrease in THM level ($\mu\text{g/L}$).

Analytical methods

All chemicals were of analytical reagent grade, purchased from Sigma-Aldrich (USA), and the solutions were prepared with ultra-pure water produced from Millipore Super-Q plus water system (Millipore, USA). Analysis was conducted in the Central laboratory of Beheira water Company, Damanhur. The laboratory procedures were in accordance with protocols recommended in standard method (APHA, 2017). Water samples were collected once a week from El-Nobarya canal (branch from Nile River, 2Km far of the plant), clarifier and end of the clear well with high density polyethylene (HDPE) tanks with 20 L capacity. The jar tests were carried out using a Stuart scientific instrument flocculator (model SWI, UK). THMs were measured by GC-MS (ThermoQuest, USA) equipped with a $30\text{ m} \times 0.25\text{ mm} \times 0.25\text{ }\mu\text{m}$ DB-1701 column

(J&W Scientific, USA), with ECD detector and a quadrupole analyzer. Dissolved organic carbon (DOC) was measured in the pre-filtered samples (glass-fibre filters GFC of pore size $0.45\text{ }\mu\text{m}$) using a total organic carbon (TOC) analyzer (Tekmar-Dohrman Apollo 9000). pH was measured by a pH meter (Theromscientific, Orion star A111), while the turbidity was measured using a turbidimeter (Hach Instrument, model 2100N). Residual aluminum was analyzed using Eriochrom Cyanine R method. Total alkalinity (measured at pH 4) was determined by titration 0.05 N sulphuric acid using Bromocresol green and methyl red indicators. The color of pre-filtered samples ($0.45\text{ }\mu\text{m}$ membrane filter (Millex, Millipore) was measured in accordance with a chloroplatinate standard at 400 nm on a Shimadzu UV 2100 spectrophotometer (APHA, 2017).

Results and Discussion

General characteristics of raw and treated water in El-Nobarya plant

The characterization of water samples from El-Nobarya plant intake is presented in Table 1. Coagulation-flocculation process in the plant is used to remove common physical parameters, such as suspended solids, turbidity, and color. The monthly average values of the water quality parameters of treated water through year 2019 are displayed in Table 2.

Table 1. Raw water quality parameters in El-Nobarya plant in year 2019.

Parameter	Mean	Minimum	Maximum
pH	8	7.8	8.2
Turbidity (NTU)	13.5	11	16
TOC (mg/L)	6	2	10
Color	95	80	110
Alkalinity (mg/L)	150	140	160
Temperature ($^{\circ}\text{C}$)	20.5	11	30

As clear from Table 2, THMs concentration ranged from 55-120 $\mu\text{g/L}$ with an annual average of 85 $\mu\text{g/L}$. More than 60 % of the values exceeds the Stage I limit of 80 $\mu\text{g/L}$ set by USEPA (1998). It was noticed that THMs values of water samples are corresponding to TOC values. The obtained results were basically consistent with that reported by **Durmishi et al. (2016)** and **Albanakis et al. (2021)**. Moreover, TOC level was high during summer and autumn months and this may be attributed to the fluctuation of natural water

characteristics in different seasons (Liu *et al.*, 2012; Dong *et al.*, 2021). These results agreed with that previously recorded (Garcia *et al.*, 2019; Saidan *et al.*, 2013). In addition, residual

aluminum in treated water ranges between 0.18-0.26 mg/L (Table 2), mostly exceeding the limit set by the WHO (Oram, 2017) of 0.2 mg/L.

Table 2. Water quality of clarified and treated water samples of El-Nobarya plant in year 2019.

Month	TOC (mg/L)			Turbidity (NTU)		^a R.Al (mg/L)	^b R.Cl ₂ (mg/L)		Temp, °C	THM (µg/L) after treatment
	Raw	Treated	%R	Clarified	Treated		Clarifier	Clearwell		
January	5.1	4.6	9	3.1	0.5	0.17	0.8	2.5	15	70
February	5.1	4.6	9	3.4	0.6	0.18	0.6	2.3	15	75
March	5.5	4.8	13	3.2	0.55	0.19	0.8	2.3	18	87
April	4	3.3	18	3.2	0.5	0.21	0.8	2.5	25	65
May	3.5	2.8	20	3.5	0.6	0.23	0.8	2.8	25	60
June	3	2.6	14	3.7	0.48	0.23	0.6	2.8	28	55
July	7	6.2	13	2.9	0.5	0.26	0.8	2.5	30	115
August	8.5	6.9	20	2.8	0.55	0.24	0.6	2.3	30	120
September	7	6.1	13	3.2	0.6	0.23	0.6	2.3	22	103
October	8.5	6.8	20	3.3	0.5	0.22	0.8	2.5	20	109
November	5.7	4.8	17	3.1	0.53	0.2	0.5	2.3	13	82
December	5.5	4.6	17	3.6	0.5	0.18	0.8	2.5	14	80
MEAN	5.7	4.8	15	3.25	0.53	0.21	0.7	2.4	21.3	85
±SD	1.78	1.45	4	0.267	0.045	0.027	0.116	0.182	6.3	21.9

^aR.Al is the residual aluminium after treatment.

^bR.Cl₂ is the residual chlorine.

Optimization process

Optimal dosage of coagulant

The current water treatment process at El-Nobarya focuses on removal of common pollutants, such as turbidity and microorganisms. Therefore, investigation the optimal coagulant dose for NOM removal was carried out using alum doses ranged from 20-100 mg/L in an increment of 10 mg/L (Table 3). pH is controlled by the addition of the coagulant itself due to its acidic effect. The results showed that increasing the alum dose led to a decrease in the values of turbidity, organic matter, color, pH, alkalinity and residual aluminum as

previously mentioned by Kalavathy *et al.* (2017), Malik *et al.* (2017) and Phanutda and Ho. (2019). In general, residual Al and TOC are pH dependent and lowest solubility values of different Al species were observed at pH 6.2-6.8 (Ruyuan., 2015; Yang *et al.*, 2010). Accordingly, increasing coagulant dose lower residual Al as the pH decreased from ~ 8 to 7.24 (Rafiee., 2014). The point of diminishing returns (PODR); the point at which a 10 mg/L increase coagulant dose no longer reduces the TOC concentration by at least 0.3 mg/L; was determined to identify the optimum alum dose should be adopted. Doses of 60-70 mg/L was employed instead of 80 mg/L (point of diminishing return) due to health and economical view point (USEPA., 1998).

Table 3. Effect of increasing alum dose on water quality.

Parameters	Raw water	Dose (mg/L)								
		20	30	40	50	60	70	80	90	100
Turbidity (NTU)	14	4.4	3.4	2.6	2.1	1.75	1.4	1.1	0.98	0.9
pH	8.02	7.83	7.76	7.64	7.58	7.5	7.42	7.34	7.3	7.2
TOC (mg/L)	6.5	5.9	5.5	5.1	4.7	4.3	4	3.7	3.5	3.3
RAI (mg/L)	-	0.26	0.21	0.18	0.15	0.13	0.12	0.11	0.1	0.09
Color (Unit)	80	36	27	19	18	15	15	13	13	11
Alkalinity (mg/L as CaCO ₃)	160	154	152	150	146	144	140	138	134	130

Applying the enhanced coagulation process

The USEPA (2006) D/DBP rule stage 1 and 2 stated that there is no need for applying strategy for THM controlling if the source water or the

treated water TOC is less than 2 mg/L. However, the previous results showed that the lowest value of TOC was 3.3 mg/L, therefore a new strategy should be applied to reduce the TOC and consequently THMs concentrations from drinking water. Enhanced coagulation was

adopted on the treatment plant with a minimum dose of 60 mg/L and shifted to 60-70 mg/L due to the variation in the plant flow during the day to follow water requirements and storage tank level. The results revealed that the application of enhanced coagulation process led to a TOC percentage removal varied between 28-44%. The obtained results agreed with that reported by Sillanpaa *et al.* (2018) and Narayan *et al.*

(2011) who recorded that more than 60% of TOC removed by enhanced coagulation process against only 13% with conventional coagulation (Matilainen *et al.*, 2010). Moreover, a noticeable reduction in THM concentrations (Table 4) was observed compared with that previously displayed in Table 2 and nearly all the samples didn't exceed the maximum level of 80 µg/L.

Table 4: Effect of Enhanced coagulation (January- April 2020).

Month	TOC (mg/L)			Turbidity NTU		RAI (mg/L)	RCl ₂ (mg/L)		Temp. °C	THM (µg/L)
	Raw	Treated	TOC % removal	Clarified	Treated	Treated	Clarifier	Clearwell	(Raw)	Treated
January	5.05	3.2	32.5	1.7	0.37	0.1	0.7	2.5	14	57
February	3.9	2.67	39	1.95	0.44	0.107	0.75	2.47	12	45
March	5.37	3.85	28	1.5	0.35	0.1	0.65	2.45	15	63
April	5.45	3.45	37	1.8	0.4	0.12	0.7	2.3	16	57
May	3.4	2.2	34	1.6	0.4	0.11	0.7	1.8	26	47
June	4.2	2.8	33	1.5	0.4	0.12	0.6	2.5	27	64
July	4.4	2.6	39	1.5	0.3	0.13	0.8	2.2	26	58
August	7	4.2	44	1.4	0.4	0.11	0.65	2	27	85

^aRAI is the residual aluminium after treatment.

^bRCl₂ is the residual chlorine.

Effect of enhanced coagulation on residual aluminum and filter run time

It is proved that residual aluminum concentrations from overdosing will results in significant deterioration of water quality as this metal are suspected to be harmful to human and may lead to diseases such as Alzheimer's syndrome, osteoporosis, anemia, and anorexia (Bachir, 2016; Krupińska., 2020). By applying enhanced coagulation, residual aluminum never exceeds the limit of 0.2 mg/L as it varied from 0.1-0.13 mg/L compared to 0.18-0.26 mg/L using conventional treatment depending on the pH of the water. The optimum hydrolyzing pH for aluminum salts ranged between 5.5 - 7.7 under which positively charged Al species are produced (Chaukura *et al.*, 2020). Moreover, the average value of clarified water turbidity decreased from 3.2 to 1.66 throughout the year. Consequently, the filter run time increased from 12 hours to 24 hours saving about 250 m³ of washing water for each filter and helped in maintaining the storage tank level to be consistent specially at mid-day where water withdraw is maximum.

Conclusions

Water sources have been contaminated by natural and man-made pollutants. The situation

is worse in developing countries due to economic restrictions, lack of education, and law application problems. Therefore, applying effective well-operated treatment processes are essential for the production of safe drinking water. Enhanced coagulation is one of the promising techniques used for water treatment. The present study was performed to apply the enhanced coagulation technology in El-Nobarya water treatment plant to reduce the high levels of THM, residual chlorine and control the filter run time. The results obtained confirmed that the TOC removal by EC process was 28-44% with a significant decrease in THM (25-35%). Employing enhanced coagulation process also lowered the residual aluminum levels (never exceeds limits of 0.2 mg), clarifier turbidity and consequently, increased filter run time (12-24 hr). These results were representative on both laboratory and large scale applications which solve the problems of aluminum residual failure and the short filter run time in the plant.

Future Perspectives

The present study reduced THM concentrations comparable to 2019 by more than 25-35 %, but still exceed the standard limit specially with high TOC and temperature. According to D/DBP RULE Stage 1 and 2 (USEPA, 2006) system with existing THM concentrations

approaching or exceeding the maximum limit are more likely to modify disinfection practice to reduce THM value through:

- Disinfection optimization (disinfectant, dose, and reaction time) should be applied as they play significant role in *controlling DBPs formation* (Dong et al., 2021).

- Chlorine dose adjustment: developing a disinfection profile to determine disinfectant dose via **EPA CT** tables should be carried out for the plant as there is a linear dependency between the chlorination dosage and THM formation (Zhou, 2019).

- Changing chlorine injection point: Two of five points are used constantly in the plant as changing injection points means changing in contact time with direct effect on THM formation (Plewa et al., 2017).

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الملخص العربي

عنوان البحث: التحكم في المنتجات الثانوية للتطهير عن طريق التخثر المحسن: دراسة حالة، محطة معالجة مياه الشرب النوبارية ، مصر

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^٢ شركة مياه وصرف البحيرة (BWADC) ، البحيرة ، مصر.
^٣ الشركة القابضة لمياه الشرب والصرف الصحي، القاهرة، مصر.

تهدف الدراسة الحالية إلى تقليل THMs والألومنيوم المتبقي والتحكم في وقت التشغيل القصير للمرشح في محطة معالجة مياه شرب النوبارية من خلال تطبيق تقنية التخثر المحسن. تم تقييم فعالية عملية المعالجة من خلال حساب النسبة المئوية لإزالة المواد العضوية الطبيعية (NOM) وانخفاض تركيزات THMs خلال الفترة من يناير حتى أغسطس ٢٠٢٠. أولاً، تم تحديد جرعة التخثر المثلى (٧٠-٦٠ مجم / لتر (علي النطاق العملي، ثم تطبيقها على محطة المعالجة. وقد خلصت الدراسة إلى انخفاض تركيزات NOM وTHM بنسبة ٤٤-٢٨٪ و ٣٥-٢٥٪ على التوالي. وتراوح تركيز الألومنيوم المتبقي بين ٠,١-٠,١٣ ملجم / لتر مقارنة بـ ٠,١٨-٠,٢٦ ملجم / لتر باستخدام المعالجة التقليدية. بالإضافة إلى ذلك، زاد وقت تشغيل الفلتر من ١٢ ساعة إلى ٢٤ ساعة مما يوفر حوالي ٢٥٠ متر مكعب من مياه الغسيل لكل مرشح. ومن ثم تم تبني التخثر المعزز كإستراتيجية تحكم في THMs بمحطة النوبارية وشركة مياه البحيرة والصرف الصحي. (BWADC) ولكن لا يزال هناك حاجة إلى مزيد من التحكم في جرعة الكلور ونقطة الحقن لمزيد من التخفيض في مستوى THMs.