

REDUCTION OF TRIHALOMETHANE PRECURSORS USING ENHANCED COAGULATION

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ABSTRACT

Jar tests were conducted to simulate the conventional processes of water treatment. Enhanced coagulation used the traditional alum that is already applied in water treatment in Egypt, with a comparison with ferric chloride and mixture of alum and ferric chloride, the strategy of enhanced treatment adopted mainly organic matter and THMs beside turbidity, so the doses of coagulant were increased to achieve better removal of THMs which have an health issue. Conventional treatment processes including addition of aluminum sulphate or ferric chloride to the raw water followed by coagulation, sedimentation and rapid sand filtration are important step in water purification. They reduced the organic matter to about 42% and THMs to 37 using the baseline dose (25 mg/l). The enhanced coagulation process by increasing the dose of alum reduced the total organic carbon and THMs to 57 % and 54 % respectively. Using of ferric chloride at a baseline dose reduced TOC and THMs by 40 % and 36% respectively, while enhanced coagulation by increasing the dose of ferric chloride (at a dose of 35 mg/l) raised up the removal of TOC and THMs to 47 % and 44% respectively. The dual coagulant of alum and ferric chloride achieved 47% removal of TOC and 36% for THMs at a dosage of 25 mg/l, while the enhanced coagulation by increasing the dose of dual coagulant of alum and ferric chloride (35 mg/l) raising up the removal of TOC to 61% and THMs to 50%. The advantageous of the dual coagulant is higher reduction of residual aluminum and THMs, this attributed to the enhancement of flocculation which create activated adsorption sites of flocs surfaces.

Key words: Water, Water treatment, Coagulation, TOC, THMs.

INTRODUCTION

Natural organic matter (NOM) is defined as a complex matrix of organic materials present in all natural water. The hydrophilic fractions of NOM are composed mostly of aliphatic carbon and nitrogenous compounds, as carboxylic acids, carbohydrates and proteins. Hydrophobic NOM primarily consists of humic and fulvic acids (Matilainen *et al.* 2010). Particular increase of interest in the composition of humic substances in water is noted since the publication of research regarding the generation of trihalomethanes (THMs) during the chlorination of water. THMs precursors are mainly humic acids, metabolites of aquatic organisms, mono-, di- and tri-carboxylic acids, and aromatic carboxylic acids (Poleneni and Inniss, 2015 and Dojlido 2002).

Four derivatives of methane belong to THMs group: chloroform CHCl_3 , bromoform CHBr_3 , bromodichloromethane CHCl_2Br , dibromochloromethane CHClBr_2 . They also have a certain binding potential for multivalent metal ions and hydrophobic organic materials such as pesticides, therefore, the content of dissolved organic matter in drinking water ought to be kept low (Her *et al.*, 2003; Zhang *et al.*, 2020).

A number of regulations regarding the potential effects of DBPs have been developed by the United States Environmental Agency (US EPA, 1998). The disinfection/ disinfection by-product (D/DBP) Rule has set maximum permissible concentration of total THMs in drinking water to be 80 mg/l. The rule mandates utilities that using chlorine for disinfection, to remove total organic carbon as a measure for reducing DBP precursors.

Analyses of water collected in different water treatment plants and also in pool water confirm the possibility of THMs presence in water subjected to chlorination (Abdullah *et al.* 2003; Kim 2009; Chang *et al.* 2010; Pardakhti *et al.* 2011 and Lee *et al.* 2009).

The conventional chemical coagulation processes with aluminum sulphate and ferric chloride followed by sedimentation and rapid sand filtration are important step in water purification. Most of chemical coagulation processes were essentially designed for turbidity and particulate removal. However, these processes are not efficient for the removal of many occurring organic micro pollutants (Margot *et al.*, 2013; Stackelberg *et al.*, 2007; Ternes *et al.*, 2002; Troeger *et al.*, 2018).

Granular activated carbon (GAC) filtration is one of the advanced technologies for organic carbon that may increase the rate of removal (Margot *et al.*, 2013; Stackelberg *et al.*, 2007), but this is not always achieved (Troeger *et al.*, 2018 and complete extraction is often not done (McCleaf *et al.*, 2017; Troeger *et al.*, 2018).

Activated carbon removal efficiency for organic materials decreases with time (Kennedy *et al.*, 2015; McCleaf *et al.*, 2017) and this is adversely affected by the dissolved organic materials (Lavonen *et al.*, 2015 and McCleaf *et al.*, 2017).

The contents of some organic material in surface water are affected to great extent by upstream sources, such as wastewater treatment plant, and

other human activities (Gago-Ferrero *et al.*, 2017; Loos *et al.*, 2013; Soerengard *et al.*, 2019).

The development of awareness towards the presence of organic pollutants in surface waters, lead to review the water management titled “Drinking Water Directive (98/83/EC) and the organic substances of importance in the Water Framework Directive was included (Fuerhacker, 2009).

Certain NOM fractions were preferentially removed like humic (hydrophobic) and higher molecular weight NOM are more effectively reduced (Owen *et al.*, 1993). It was found that a preferential reduction in ultraviolet absorbance (UVA) compared to dissolved organic carbon (DOC), indicated preferential removal of aromatic (Humic) NOM (Imai *et al.*, 2003; Reckhow and Singer, 1990).

Organic matter content in water is reduced by coagulation (Świdarska-Bróz *et al.* 2008; Matilainen *et al.* 2010; Alexander *et al.* 2012; Jeong *et al.* 2014). Removal depends on physico-chemical composition of water, pH value, type and dosage of coagulants. The most often used aluminium coagulants are aluminium sulphate $\text{Al}_2(\text{SO}_4)_3$ and aluminium chloride AlCl_3 .

Aluminum irons can remove NOM according to two mechanisms: (1) adsorption on aluminum hydroxide or iron hydroxide flocs and (2) formation of insoluble complexes such as aluminum or iron humates or fulvates (Jacangelo *et al.*, 1995; Randtke, 1993). The first mechanism is more dominant at higher coagulant dosages and higher pH conditions, whereas the

second mechanism is relatively more dominant at lower pH conditions (Krasner, and Amy, 1995).

Over 90 and 80% reduction is obtained by coagulation for turbidity and color respectively (Dąbrowska and Rosińska 2013). Research on treatment of water with different compositions confirms the effectiveness of the coagulation in the removal of organic matter (Hussain *et al.* 2013; Wang *et al.* 2013).

The concept of enhanced coagulation is used for controlling the precursors of DBPs, where effective coagulation dosages are used for reduction of DOC. Enhanced coagulation was selected as a measure because it was effective for the reduction of DOC and could be executed at water plants treating the surface waters using the existing processes (Owen *et al.*, 1993).

To meet the drinking water standards limits, in conventional water treatment, mixture of aluminum and iron salts was conducted to lower residual aluminum in finished water to an acceptable content (less than 0.2 mg/l) (Li and Mitch , 2018).

This study aims to investigate acceptable approach to enhance coagulation for TOC (DBD precursors), turbidity and aluminum minimization using aluminum and iron based coagulants in drinking water treatment, single and dual dosing with aluminum sulphate and ferric chloride coagulants. Once the condition for optimized coagulation has been determined by Jar tests, TOC was evaluated under baseline coagulation conditions and optimized

coagulation. In addition, the study aimed to compare the efficiency of aluminum sulfate, ferric chloride and mixture of the two coagulants for the removal of turbidity and organic substances from water samples collected from River Nile with reference to residual aluminum. Minimizing of operating cost is indirect objective should be achieved.

MATERIALS AND METHODS

Study area: Surface water samples were collected from the intake of Shubra Al-Khaymah water treatment plant on Ismailia canal (branch of the River Nile). This plant is located in north Cairo. The water was collected in November 2018, Different levels of TOC were recorded in each table (Table 1, Table 2 and Table 3) according to different doses of each coagulant.

Sampling Procedure: Water samples were collected in winter of 2018/2019 where high concentrations of organic matter were recorded. Water samples were collected in stopper polyethylene plastic bottles and amber bottles for TOC and THMs. All samples were stored in an ice box with preservation for each parameter and delivered immediately to the laboratory for analyses.

Materials: Coagulants tested were liquid aluminum sulfate $\text{Al}_2(\text{SO}_4)_3 \cdot n\text{H}_2\text{O}$ 8% Al_2O_3 (commercial grade), produced by Egyptian aluminum sulfate Co. of Egypt (ASCE), Ferric chloride, 29.6% Fe_2O_3 (commercial grade, India), and mixture of the two salts (50/50). For each raw

water sample, TOC was assessed before and after treatment under baseline coagulation conditions using the three different coagulants with different doses.

Procedures: Physical and chemical examinations were carried out according to Standard Methods for Examination of Water and Wastewater 23rd edition (APHA, 2017).

Table (1): The analyzed parameters, method of analysis and instruments

No.	Parameter	Unit Used	Used method.	Reference	Instrument
1	Turbidity	NTU	2130 B. Nephelometric Method	APHA, 2017	HACH2100N Turbidimeter
2	PH	-	4500.H,B Electrometric method	APHA, 2017	HACH sension3 PH Meter
3	Al, Fe	mg/l	EPA (200.7)	Maxfield and Mindak, 1985	ICP-OES Optima8300
4	Total organic carbon (TOC)	mg/l	5310 Total Organic Carbon (TOC)	APHA, 2017	Siever5310C
5	Trihalomethan (THM _s)	µg/L	EPA (551.1)	Hautman and Munch, 1997	GC-ECD Thermo-Trace1300
6	residual chlorine	mg/l	4500-CLG DPD Colorimetric Method	APHA, 2017	Lovibond Chlorine MD100

All samples were brought to room temperature (25 °C) prior to jar testing.

one liter samples were performed using standard jar test procedure. Two

kinds of coagulants dosing approaches, including single dosing aluminum sulphate and ferric chloride and dual dosing. Initial rapid mixing was conducted at 200 rpm for 1 min. followed by slow mixing at 40 rpm for 20 min. Then the flocks were allowed to settle for 20 min., the turbidity of the supernatant was measured by turbidity meter, PH was measured by PH meter, Total organic carbon (TOC) was measured by TOC analyzer Trihalomethan (THMs) was measured by gas chromatography with electron capture detector, Iron and residual aluminum was measured by ICP.

This research was conducted using jar testing to simulate drinking water treatment processes in laboratory. This instrument is a widely accepted and used method for understanding the effects of treatment processes changes at lab scale (AWWA, 1992).

Jar tests that simulated the plants conditions (pH of coagulation, coagulant type and dose) is termed “baseline coagulation” were done using alum as coagulant. The aim of “baseline” jar tests was to compare the jar test results with the full-scale plant data, and serve as a level for comparison with the optimized coagulation conditions.

Enhancing coagulation was performed using a series of bench-scale jar tests. With different coagulant dosages (the baseline dosage used by the plant). TOC concentrations were measured with UV- Persulfate TOC analyzer, model Phoenix 8000, USA after filtration of the samples through 0.45 μm membrane.

RESULTS AND DISCUSSIONS

Physical and chemical examinations of water samples collected from the intake of Shubra Al-Khaymah WTP during winter 2018/2019. The pre-chlorine dose was 5.5 mg/l according to break point procedure.

Table (2) shows that the raw water contains a relatively high turbidity, pH and organic matter, and upon using chlorine, slight reduction of pH was reported. Figure (1) shows the influence of alum doses on TOC and THMs removal. Using alum reduced the turbidity, pH and organic matter (TOC). The percentage of removal of organic matter (TOC) and Trihalomethane (THMs) are increasing by increasing alum doses as shown in Figure 1. The percentage of removal of organic matter (TOC) and by-products (THMs) is more than 55% complying with that reported by (Ndabigengesere, and Narasiah, 1998).by increasing alum doses pH was reduced this increases the precipitation of aluminum in water and we see decreasing residual aluminum in water as shown in Figure2. Trihalomethane (THMs) is reduced with increasing alum doses as shown in Figure 3.

Table (2): Effect of alum dose on reduction of turbidity, TOC, THMs and residual aluminum

Dose of liquid alum. Sulphate (mg/l)	Turbidity (NTU)	pH	TOC (mg/l)	TOC removal (%)	THMs ($\mu\text{g/L}$)	THM removal (%)	R. Al (mg/l)	R. Cl (mg/l)
Raw Water	8.5	8.31	6.4	0%	N/A	N/A	0.05	N/A
Raw Chlorinated	7.9	8.09	5.91	8%	88.3	0%	0.04	2.85
10	1.4	7.5	3.93	35%	66.18	25%	0.25	2.4
15	1.3	7.44	3.88	39%	63.65	28%	0.21	2.4
20	1.20	7.4	3.76	41%	59.02	33%	0.18	2.4
25 Baseline	1	7.36	3.53	45%	55.69	37%	0.16	2.4
30	0.95	7.31	3.22	50%	50.3	43%	0.14	2.4
35	0.84	7.23	2.75	57%	40.83	54%	0.12	2.4

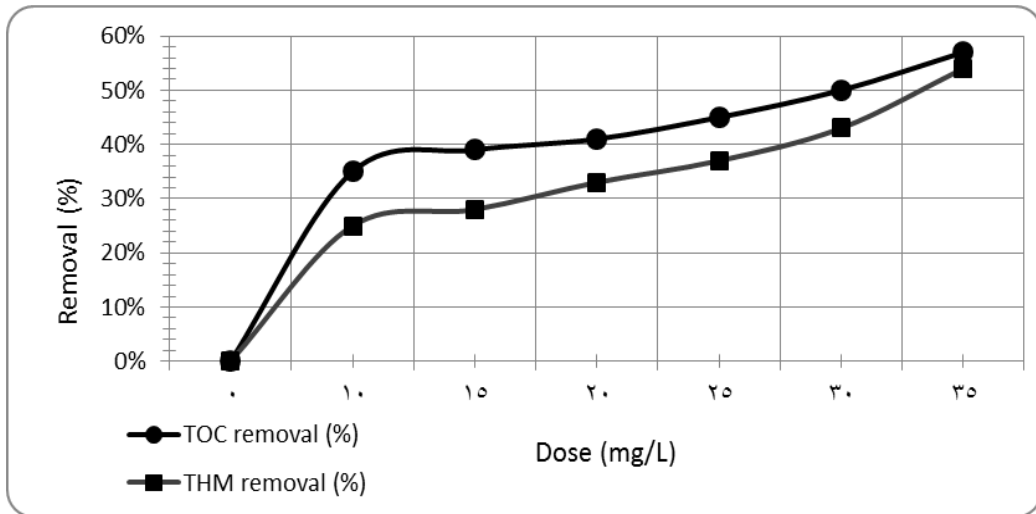


Figure (1): Removal percentage of TOC and THMs using aluminum sulphate

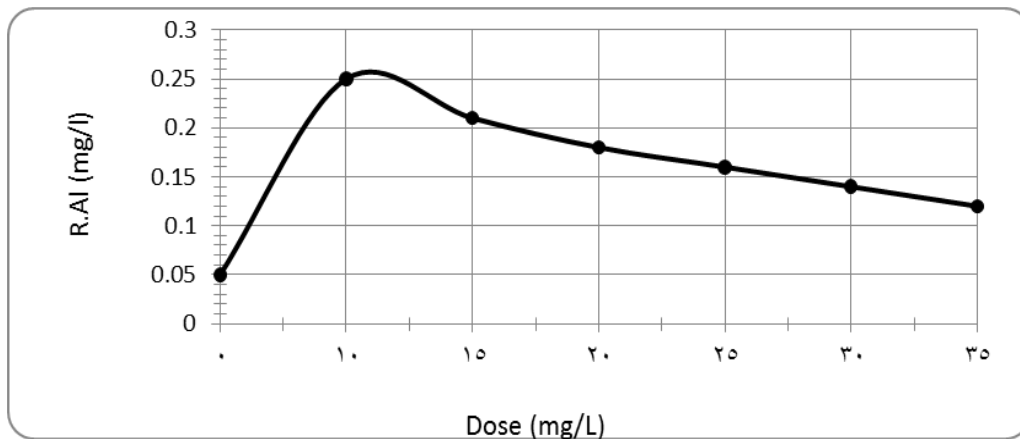


Figure (2): Relation between doses of aluminum sulphate and residual aluminum

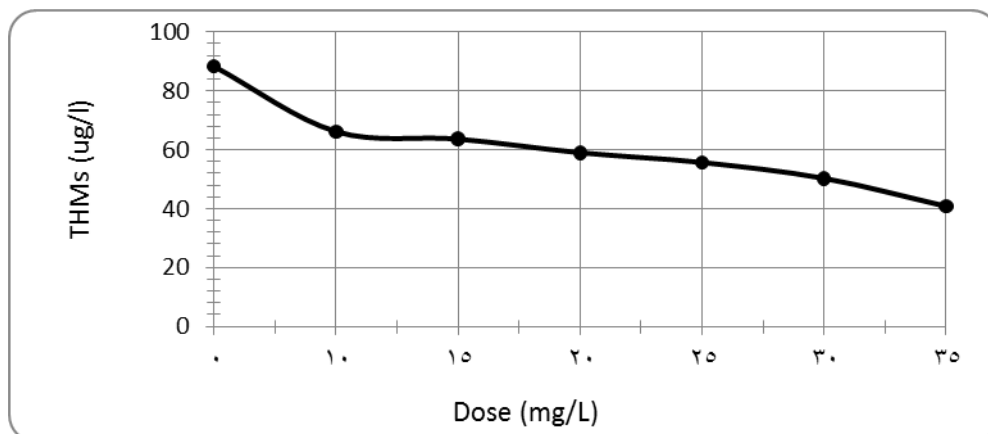


Figure (3): Relation between doses of Aluminum Sulphate and Trihalomethanes

Upon using of ferric chloride, turbidity, pH, organic matter (TOC) and byproducts (THMs) are reduced with increasing the dose until 35 mg / l, the percentage of removal of organic matter (TOC) is more than 47% and removal of byproducts (THMs) is about 44% agreeing with that reported by. (Pikkarainen, *et al.*, 2004). There is no baseline for FeCl_3 because it is not currently used, and we depend in our experiment on the already used coagulant (alum), it was supposed to be as alum (25 mg/l). increasing the dosage of ferric chloride, resulted in corresponding residual iron in water in small dosages, but it decrease as the dosage increase as a result of enhancement of settleability due to growing up of flocs and become more compact and heavy causing low residual iron in water (Fig. 5). THMs were reduced with the higher dosages (Fig. 6).

Table (3): Effect of ferric chloride dose on reduction of turbidity, TOC, THMs

Dose of ferric chloride (mg/l)	Turbidity (NTU)	pH	TOC (mg/l)	TOC removal (%)	THMs ($\mu\text{g/L}$)	THM removal (%)	Fe (mg/l)	R. Cl (mg/l)
Raw Water	8.5	8.31	6.4	0%	N/A	N/A	0.2	N/A
Raw Chlorinated	7.9	8.09	5.91	8%	88.3	0%	0.18	2.85
10	1.5	7.2	4.60	28%	76.82	18%	0.5	2.5
15	1.4	7.1	4.13	35%	65.69	26%	0.42	2.5
20	1.30	7	3.98	38%	61	31%	0.36	2.5
25 Baseline	1.19	6.95	3.80	41%	58.3	34%	0.30	2.5
30	1	6.88	3.60	44%	53.36	39%	0.27	2.5
35	0.9	6.75	3.45	47%	49.62	44%	0.23	2.5

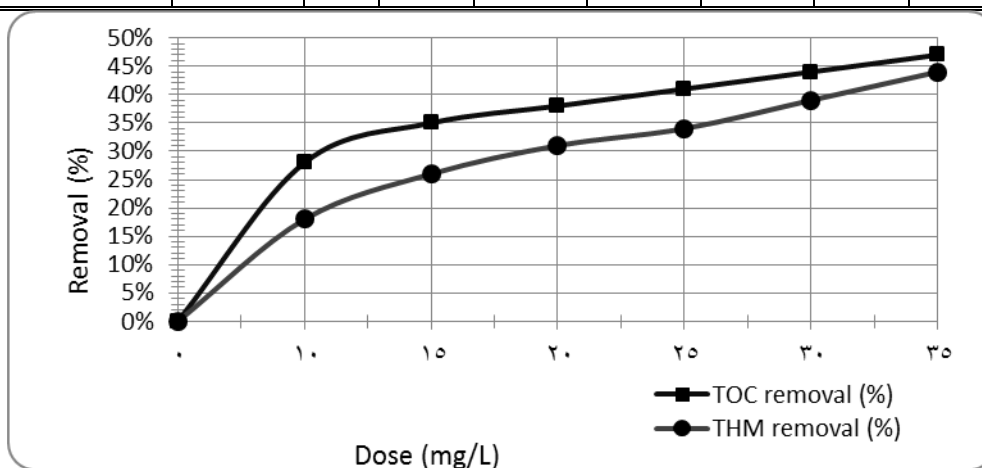


Figure (4): Removal percentage of TOC and THMs using Ferric Chloride

Table (4) Effect of mixed solution of Aluminum Sulphate and Ferric Chloride on reduction of turbidity, TOC, THMs and residual aluminum

Dose	Turbidity (NTU)	pH	TOC (mg/l)	TOC removal (%)	THMs ($\mu\text{g/L}$)	THM removal (%)	R. Al (mg/l)	Fe (mg/l)	R. Cl (mg/l)
Raw Water	8.5	8.31	6.4	0%	N/A	N/A	0.05	0.2	N/A
Raw chlorinated	7.9	8.09	5.91	8%	88.3	0%	0.04	0.18	2.85
10 ppm	1.2	7.4	3.88	39%	62.55	29%	0.18	0.25	2.6
15 ppm	1.15	7.35	3.78	41%	60.31	32%	0.16	0.23	2.6
20 ppm	1.10	7.3	3.67	43%	56.2	36%	0.13	0.18	2.6
25 ppm	0.92	7.2	3.42	47%	52.43	41%	0.11	0.15	2.6
30 ppm	0.85	7.15	2.9	55%	45.89	48%	0.09	0.12	2.6
35 ppm	0.79	7.12	2.2	65%	35.13	60%	0.07	0.10	2.6

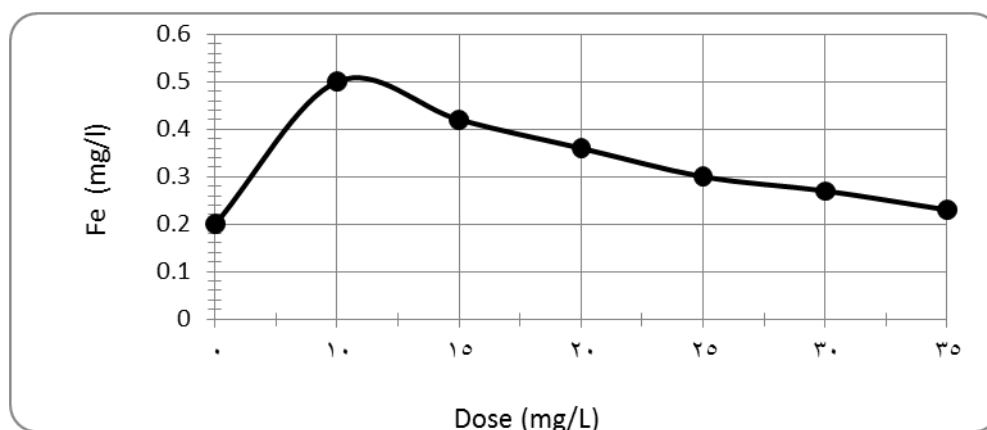


Figure (5): Relation between doses of Ferric Chloride and Fe

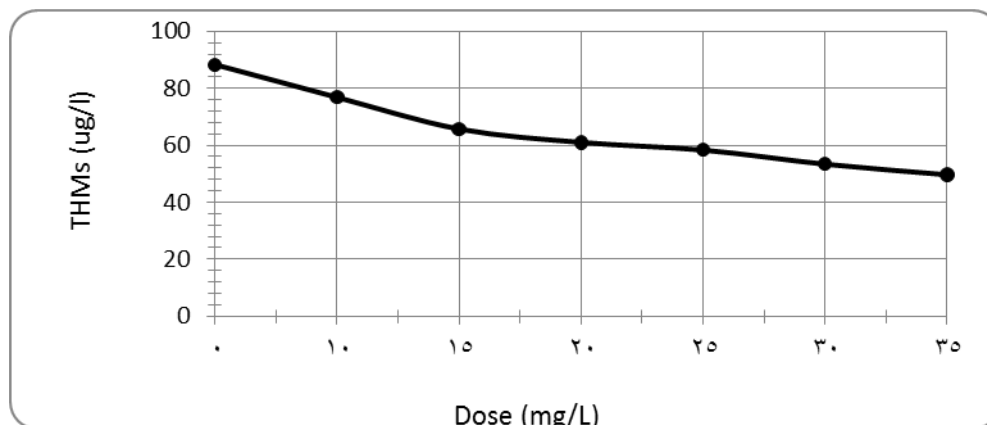


Figure (6): Relation between doses of Ferric Chloride and Trihalomethane

Using mixture of Aluminum Sulphate and Ferric Chloride at a dose of 35 mg/l, the percentage of removing organic matter (TOC) was about 65% and removal of THMs was about 60% as shown in(Figure7). Agreeing with that reported by (Crozes, *et al.*, 1995). The TOC and THMs removal in the effluents with all three coagulants was higher than the baseline control 25 mg/l of aluminum sulphate which is the common dose applied in the water treatment plant of shoubra alkHEMA. Increasing the dose of coagulants resulted in decreasing of pH, which enhance precipitation of aluminum and iron hydroxide, consequently, low residual and iron were detected (Fig. 8). Precipitation of Iron increase in water by increasing of increasing dual mixture doses due to presence of Aluminum Sulphate in dual mixture which increase of activity of Precipitation of Iron in water by forming very large and heavy flocs which is increasing of Precipitation of Iron in water as shown in

(Figure9). Trihalomethane (THMs) is reduced with increasing dual mixture doses as shown in (Figure 10).

The organic matter as TOC and the THMs reduction in the plant effluent with the three coagulants were higher than the baseline dosage. At normal dosages in the plants which depends essentially on turbidity removal, the removal of TOC and THMs were they reduce the organic matter to about 45% and THMs to 37 using the baseline dose (25 mg/l). The enhanced coagulation process using alum reduced the total organic carbon and THMs to 57 % and 54 % respectively by aluminum sulfate. Using of ferric chloride at a baseline dose reduced TOC and THMs by 41 % and 34% respectively, while enhanced coagulation (at a dose of 35 mg/l) raised up the removal of TOC and THMs to 47 % and 44% respectively. The dual coagulant of alum and ferric chloride achieved 61% removal of TOC and 50%. At baseline dose, residual aluminum was 0.16 mg/l using alum and 0.11 mg/l using dual mixture (Fig. 2, 8), but at enhanced doses residual aluminum dropped to 0.06 mg/l for dual coagulant and 0.12 mg/l for alum. Residual iron was reduced to 0.1 mg/l at 35 mg/l as FeCl_3 , while it was 0.15 mg/l at baseline dose (Fig. 9).

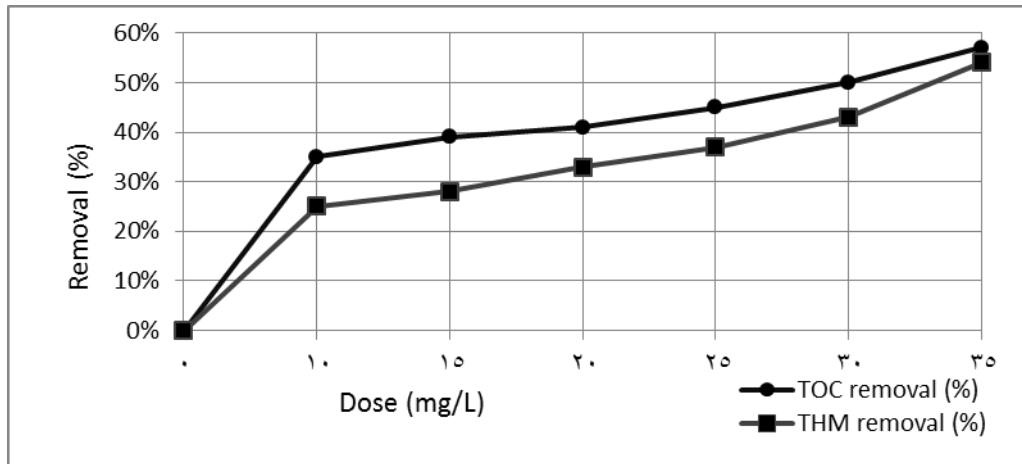


Figure (7): Removal percentage of TOC and THMs by coagulant mix

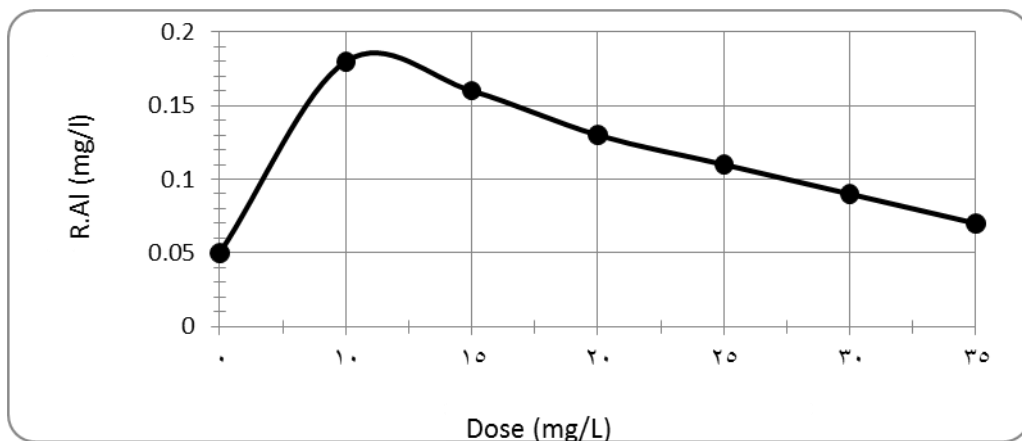


Figure (8): Relation between doses of Mix of Aluminum Sulphate Ferric Chloride and residual Aluminum

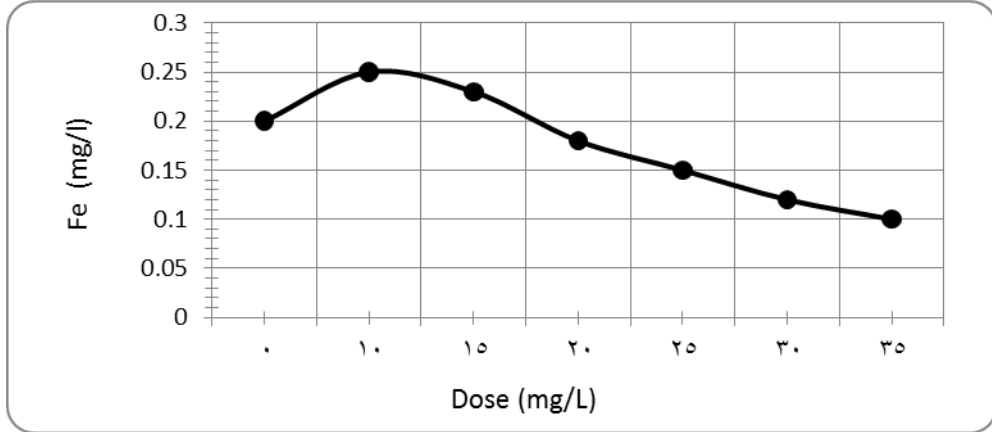


Figure (9): Relation between doses of Mix of Aluminum Sulphate Ferric Chloride and Fe

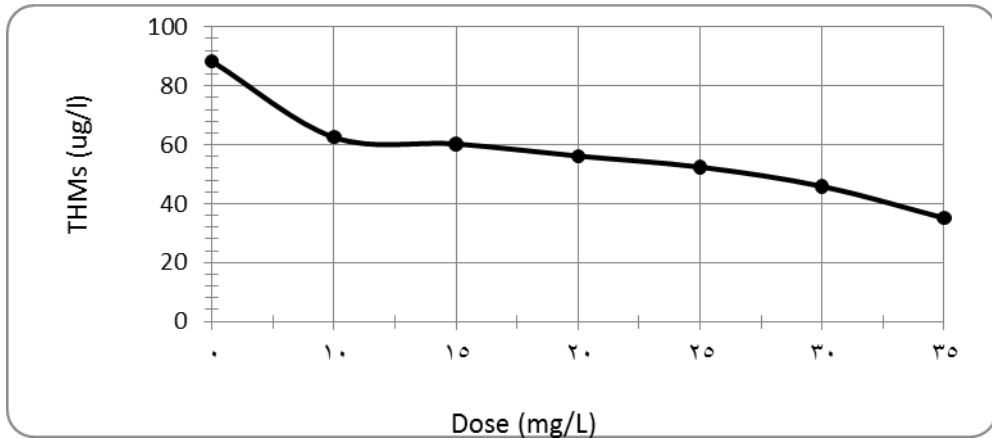


Figure (10): Relation between doses of Mix of Aluminum Sulphate Ferric Chloride and Trihalomethane

CONCLUSION

This study was performed as enhanced coagulation that will provide additional removal of organic matter (TOC) and the corresponding THMs that have a health issue. The protocol for enhancement adopted an effective dosage for removal of organic matter as a precursor for THMs, and utilization of the active adsorption sites on surface of flocs generated during coagulation and flocculation. The baseline for comparison taken 25 mg/l which is the average dose applied in the plant of Shubra Al-Khaymah WTP. The percentage removal of TOC for alum, ferric chloride and dual coagulant were improved by 12%, 6% and 14% respectively. The corresponding improvements of THMs for the coagulants were 17%, 10% and 14% respectively. The percentage additional reduction of residual aluminum for alum and the dual coagulant were 25% and 54.5% respectively. Residual iron improved by 33.3%. These results were carried out on the settled water so after filtration they will be better. THMs concentration for baseline 52.43 μ g and after enhancement it became 35.13 μ g lower than the max permissible limit which is 100 μ g. residual aluminum after improvement become 0.12 mg/l for alum and 0.07 mg/l for dual coagulant, while the max limit is 0.2 mg/l. Residual iron was 0.1 mg/l, and the max limit is 0.3 mg/l.

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خفض مسببات تكوين ثلاثي هاليد الميثان باستخدام الترويب المحسن

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المستخلص

في هذه الدراسة تم استخدام تجربة الكؤوس لمحاكاة المعالجة التقليدية التي تتم في محطات مياه الشرب وفي هذه الدراسة تم استخدام الشبة المعتاد استخدامها في محطات مياه الشرب بمصر ومقارنتها مع مروبات مختلفة اخرى مثل كلوريدات الحديد الثلاثية وخليط من الشبة مع كلوريدات الحديد الثلاثية كمحسنات لعملية الترويب. تهدف استراتيجية الترويب المحسن بالأساس الي محاولة تقليل تركيزات كلا من المواد العضوية ومتبقيات عملية التطهير جنباً الي جنب والتحكم في العكارات المرتفعة. تم رفع الجرعات الخاصة بالمراد المروبة للحصول علي افضل ازالات للمواد ذات التأثير علي الصحة (متبقيات التطهير). ينبغي الإشارة الي أن خطوات المعالجة التقليدية تبدأ بعملية إضافة المروبات المختلفة مثل الشبة وكلوريدات الحديد الثلاثية ثم عمليات الترويب والتنديف يليها عملية الترسيب وفي النهاية الترشيح. بمقارنة المعالجة التقليدية وما حققته من ازالات وهي ٤٥% للمواد العضوية و ٣٧% لمتبقيات التطهير عند جرعة الشبة = ٢٥ مجم/لتر. بزيادة تركيز الشبة لتحقيق الترويب المحسن تم تقليل المواد العضوية الكلية وكذلك متبقيات التطهير الي ٥٧% و ٥٤% علي التوالي. عند استخدام كلوريدات الحديد بجرعات أساسية = ٢٥ مجم/لتر تحققت ازالات للمواد العضوية الكلية وكذلك متبقيات التطهير الي ٤١% و ٣٤% علي التوالي. وبزيادة تركيز كلوريدات الحديد الثلاثية الي ٣٥ مجم /لتر لتحقيق الترويب المحسن تحققت ازالات للمواد العضوية الكلية وكذلك متبقيات التطهير الي ٤٧% و ٤٤% علي التوالي. باستخدام محلول من الشبة وكلوريدات الحديد الثلاثية كمروب بجرعة = ٢٥ مجم/ لتر تحققت ازالات للمواد العضوية الكلية وكذلك متبقيات التطهير الي ٤٧% و ٤١% علي التوالي. وحقق الترويب المحسن باستخدام نفس الخليط بجرعات اعلي = ٣٥ مجم/لتر ازالات للمواد العضوية الكلية وكذلك متبقيات التطهير الي ٦٥% و ٦٠% علي التوالي يتضح من النتائج ان خليط من الشبة وكلوريدات الحديد الثلاثية حقق اعلي نسب للإزالة والذي يعزي الي تحسين عملية التنديف بشكل عام.