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Factors influencing the formation of assimilable organic carbon during chlorination/chloramination of Cyclops

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ABSTRACT

Investigation of the formation of assimilable organic carbon (AOC) was performed during the chlorination/chloramination of Cyclops metabolite solutions under varied conditions, and the effects of metabolites generated by organisms on water security as well as the formation of assimilable organic carbon (AOC) was also researched. Furthermore, investigation of impacts of chlorine and chloramine doses, coagulant doses, reaction time and temperature on the formation of assimilable organic carbon (AOC) was also carried out during the disinfection of Cyclops metabolite solutions. It was revealed that the pattern of AOC concentration decreased following an increasing trend accompanied with increased temperature and reaction time. Disinfectant dosages exerted influences on AOC formation in different manners. With the increase in the dose of chlorination and chloramination, AOC concentration exhibited an apparent decrease, followed by an increase, while the chain of AOC concentration decreased a little bit. Nevertheless, AOC concentration experienced a consistent decrease as coagulant dosage was augmented. The formation of AOC can be suppressed through the proper scenarios. Moreover, the results revealed the fact that the minimum amount of AOC was produced by a combination of chlorination (4 mg/L) and coagulant (40 mg/L), and the interaction was conducted at 20°C of temperature together with encompassing reaction time of 6 h.

Keywords: Cyclops metabolite dissolution; Chlorination; Chloramination; Assimilable organic carbon (AOC)

1. Introduction

In recent years, there were increased incidences of overgrowth of Cyclops in eutrophic water, especially in drinking water sources, for instance reservoirs and fresh water lakes. Cyclops belong to Phyllum arthropod a, its appearance has a tough carapace. As a kind of zoo plankton , the body length of the adult Cyclops is one to several millimeters [1]. Cyclops give rise to issues in drinking water treatment, for instance clogging filters as well as penetrating sand filters. Human health is threatened by the Cyclops as they contaminate water supply chain, performing as the host of pathogenic parasites such as Schist o some and Eelworm that give birth to diseases in human body [2,3].

Chlorine and chloramine are utilized in water for promoting the formation of low molecular weight (LMW) oxygen-containing organic by-products. These compounds are products of the oxidative segregation of intricate natural organic matter (NOM), which has a normal presence in the water [4]. Carboxylic acids, benzoic compounds, aldehydes, ketones, and keto-acids are the common instances of this kind of low molecular weight (LMW) compounds [5].

Quantification of the organic substrates in drinking water can be carried out by using assimilable organic carbon (AOC). AOC, termed as an organic contamination, usually results into the decay of water quality because of the phenomenon of multiplication of heterotrophic bacteria in water supply and treatment systems. For the purpose of removing assimilable organic carbon (AOC), disinfection through the application of chlorine/chlora-

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mine technique is a typical measure in respect of water treatment. Moreover, it also eliminates the essential nutrients of heterotrophic bacteria that are mandatory for them to survive and multiply in the water stage. In addition, it obstructs them to expand their colonies in the drinking water supply system. Furthermore, these organic matter reacts with chlorine/chloramine (typically utilized as a disinfectant) that is likely to result into the growth of carcinogenic as well as mutagenic disinfection by-products. According to past reports, both chlorination and chloramination can cause the formation of halogenated disinfection by-products [6,7]. And the previous study [8] evaluated the formation of selected DBPs during chlorination of Cyclops metabolite solutions under various conditions, including reaction time, chlorine dosage, pH, temperature, and ammonia concentrations. Nevertheless, the organic carbon concentration is most probably one of the most imperative factors when it comes to curbing the associated biomass and heterotrophic bacteria functions in drinking water. In accordance with a report [9], approximately 50–70% of AOC fraction comprises biological molecules that have MW less than 1000 Da. It is apparently suggested that not all organic carbon compound is able to be utilized by bacteria in the water. Moreover, as a matter of fact, it merely is a low-molecular weight fraction, capable of being biodegraded by bacteria [10].

Natural organic matter (NOM), termed as the intricate matrix of naturally found organic contents available in natural waters, is commonly taken into consideration as an originator of AOC. Most researches on AOC have been performed based on the NOM-containing raw water. Liu [11] used drinking water in Cheng-Ching Lake and water in a northern suburb of China, as empirical water for the purpose of researching AOC, respectively. Increasing numbers of research works have revealed that natural organic matter (NOM) is not the one and only source of AOC precursors. In reservoirs, in addition to NOM, Cyclops, algae, humus and other contents are also existent. They also pose to be the originators of assimilable organic carbon that may generate a large amount of AOC [12]. Ozonation of phytoplankton is capable of making substantial contribution to AOC as the surface water is utilized as a source in respect of drinking water treatment [4]. Phytoplankton cells are quite famous for being affluent with biological nitrogen in the forms of proteins, amino acids, and amines, together with being established to the formation of organic carbon load from the ozonation [4]. A past research work by Muller [13] revealed that increase in the heterotrophic plate count (HPC) had direct association with the augmented AOC concentrations arising out of the ozonation of the phytoplankton biomass. As a traditional method, HPC can reap AOC concentration according to bacteria quantity. However, it is complicated and time-consuming. This study applies a fresh technique: the bases of flow cytometry (FCM).

In comparison with algae, bacteria and other micro organisms, Cyclops has larger size. This is why it puts forth suggestion that the retained biomass of amino acids, protein, fat and other organic contents possesses a larger capacity for the formation of AOC. In this way, it appears to be a matter of interest to discover the way the metabolites generated by these organisms exert influence on the water security, in addition to contributing to the generation of AOC.

There have been made consistent attempts in order to find out the identities as well as the detriments of AOC, especially for the purpose of modelling their development and controlling their formation, but information regarding their association with chlorination of Cyclops metabolite solutions is not known. This study aims at performing the evaluation of the concentration of AOC all through the chlorination of Cyclops metabolite solutions under several situations, involving reaction time, disinfection dose, and temperature as well as coagulation dosage.

2. Materials and methods

2.1. Chemicals and materials

The use of analytical reagent grade chemicals was made for the preparation of all of the solutions. Preparation of stock solutions of aluminium sulphate ($Al_2(SO_4)_3{\cdot}18H_2O$) was done using the deionised water. Fresh preparation of the mono chloramine solution (500 mg/L) was also done through a mix of a free chlorine solution with an ammonium chloride (NH_4Cl) solution at an initial Cl/N mass ratio of 4/1. Preparation of a free chlorine (HOCl) stock solution (2500 mg/L as Cl_2) was carried out from a 4% sodium hypochlorite (NaOCl) solution, followed by periodical standardization performed by DPD/FAS titration (Standard Methods 4500-Cl, 1998). Preservation of the disinfectant solutions was carried out at 4°C of temperature, followed by bringing to room temperature well before use.

2.2. Preparation of the cyclops metabolite solution

Primarily, the Cyclops was received from the localities of Mopanshan Reservoir in Harbin that is an essential source of drinking water for the residents of Harbin city. Culturing of Cyclops was carried out in aerated 25 L glass aquaria that were filled using the crude water from the reservoir. Furthermore, aquaria were retained at a steady temperature (15°C), leading to exposure to a steady photo period (12 h light/12 h dark). Cyclops culture is carried out for a period of 10 d, under this situation. Addition of huge numbers of Cyclops was made in a 1 L beaker using the deionised water. Subsequent to 1 d, filtration of the Cyclops suspensions was performed with the help of a 0.45 μm membrane for the purpose of eliminating poised solids, in addition to preserving in dark at a temperature of 4°C and analyzing within one week of time so that the variations in the components could be cut down.

2.3. Analytical methods

Measurement of the chlorine disinfectant concentration was taken with the help of N,N-diethyl-p-phenylenediamine/ferrous ammonium sulphate (DPD/FAS) titration [14]. A fresh technique was followed by this experiment in order to determine AOC, on the bases of flow cytometry (FCM). The quantity of bacteria in a test specimen suggests an indirect reflection of the concentration of AOC in water (1.0×10^7 bacteria = 1 μg/L AOC). This technique possesses huge sensitivity that employs quite a harsh cleaning mechanism in respect of the entire tools utilized to determine the AOC. In every situation, a 20 mL specimen was passed across a germ-free 0.22 μm Millipore filter, followed by cooling to normal room temperature and heating at a temperature of 70°C for a period of 1 h. For the purpose of ensuring the development of micro organisms, addition of 200 μL of mineral elements was done into all of the 20 mL water specimens. Table 1 shows the ingredients in the mineral solution. Quite a little quantity of Evian water (having natural bacteria), was utilized for inoculating each water specimen in such a way that the concentration of bacteria amounted to be approximately 5×10^3 cells/mL. Culture of the specimens was carried out at a temperature of 30°C in an incubator for the purpose of reaching the steady stage. Furthermore, recording of bacteria was taken with the use of flow cytometry [15]. Thereafter, the determination of the concentration of AOC in all of the samples was carried out.

2.4. Experimental procedures

The stock solution was concentrated with Cyclops metabolite solutions using the deionised water for the purpose of making testing solutions of 4 mg/L as TOC. Applications of the chlorine dosage as well as mono chloramine dosage of 20 mg/L as Cl_2 were made to Cyclops metabolite solutions (4 mg/L as TOC) that underwent bugging at a pH of 7.0 with deionised water in the 250 mL glass bottles, in addition to their incubation at 20 ± 1°C, subsequent to 48 h as the baseline scenario. In respect of this research work, reaction time, disinfections dosages, temperature and coagulant dose pose to be the impacting factors of disinfection whereby, all of the factors were changed, one parameter at a time, from the baseline situation: reaction time (6, 12, 24, 48, 72 h), chlorine and chloramine dosages (2, 4, 6, 8, 10, 20 mg/L as Cl_2), temperature (5,10, 20, 30°C), coagulant dosages (20, 30, 40, 50 mg/L as $Al_2(SO_4)_3$). Subsequent to the response of disinfection, solutions were satiated using sodium sulphite followed by extraction for later disinfection analyses. The concentration of AOC identified in the current research work was initiated from the Cyclops metabolite solutions.

Table 1
The ingredients in the mineral solution

Solution number	Ingredients	Concentration (g/L)	1 L mineral solution required the amount of reserve liquid
1	$Na_2HPO_4 \cdot 2H_2O$	1.28	998 mL
	KH_2PO_4	0.3	
	$(NH_4)_2SO_4$	1.77	
	$CaCO_3$	8.0	
	$MnCl_2 \cdot 4H_2O$	1.15	
	$CuSO_4 \cdot 5H_2O$	0.146	
2	$CoCl_2 \cdot 6H_2O$	0.13	1 mL
	ZnO	0.4	
	H_3BO_3	0.124	
	$MgCl_2 \cdot 6H_2O$	13.42	
	$Na_2MoO_4 \cdot 2H_2O$	1.04	
3	EDTA	3.0	1 mL
	$FeCl_3$	24.0	

3. Results and discussion

3.1. Effect of disinfection dosage

Fig. 1 Concentration of AOC as functions of disinfectant dosage after 6 h chlorination and chloramination of Cyclops metabolite dissolution at pH 7.0, temperature: 20 ± 2°C. The error bars represent the standard deviation of replicate measurements (*n* = 3).

Fig. 1 shows in this investigation, AOC concentration encountered an evident decrease to 2.4 mg/L as the chlorine dosage amounted to be 4 mg/L. The reaction curve was spiked with chlorine of 10 mg/L, while the chain of AOC concentration displayed a slight decrease. With the chlorine dose of 20 mg/L, the AOC concentration decreased to 7.07 mg/L. The initially decreasing tendency suggests that both chlorine and chloramine were, at times, found producing moderate amounts of AOC. AOC is primarily composed of more low molecular weight (LMW) as well as fewer high molecular weight (HMW) compounds, while several of the lower MW underwent oxidization by chlorine disinfectant into non biodegradable organic matter. This finding validated the fact that AOC concentration achieves minimum value with 4 mg/L chlorine. The achievement could be because of the oxidation of organic matters either by chlorine or chloramines, which led to a rise of AOC concentration, together with the use of AOC by bacteria, which gave rise to a decrease of AOC [16]. The concentration of chlorine over 4 mg/L, AOC concentration increase was observed, which suggested that the oxidants such as chlorine could decompose high molecular compounds into low molecular weight (LMW) organic matters that are expected to simply raise AOC concentration. At least, AOC concentrations were inclined to be a bit lower in the specimens that were treated at 20 mg/L disinfectant due to the fact that

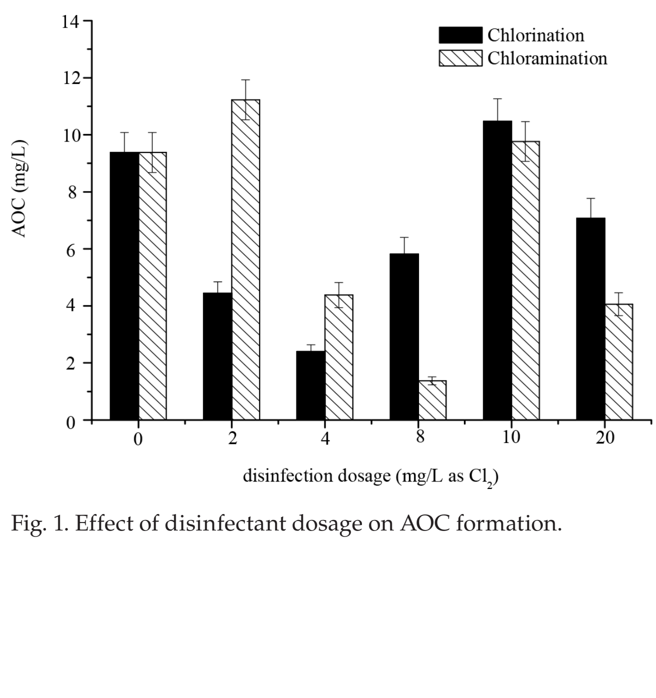

Fig. 1. Effect of disinfectant dosage on AOC formation.

free chlorine/chloramine residual effectively inactivated low molecular weight (LMW) organic matter.

The differences and similarities in the patterns between chlorination and chloramination are quite imperative to observe. In this bench-scale experiment, AOC concentration achieves the minimum value 4 mg/L chlorine and 8 mg/L chloramine. Moreover, the pattern exhibits an apparent spike at 2 mg/L chloramination. AOC concentration initially decreased and then increased while chlorine concentration was from 0 to 8 mg/L. However, AOC concentration initially increased and then decreased while chloramine concentration was from 0 to 8 mg/L. While disinfectant dosages range from 8 to 20 mg/L, suggesting that the patterns were very similar. It is generally acknowledged that chloramine is a smaller efficient disinfectant and sterilization ability in comparison with chlorine [17].

3.2. Effect of coagulant

Fig. 2 Concentration of AOC as functions of temperature after 6 h chlorination and chloramination of Cyclops metabolite dissolution at pH 7.0, chlorine dosage: 4 mg/L as Cl_2, chloramination dosage: 4 mg/L as Cl_2. The error bars represent the standard deviation of replicate measurements (n = 3).

Fig. 2 emphasizes the findings of AOC formation with coagulant, subsequent to 6 h of chlorination and chloramination of Cyclops metabolite solutions, under the baseline situations, at various coagulant doses. The amounts of AOC experienced a decrease with the increase in the dose of aluminium sulphate from 10 mg/L to 40 mg/L. When utilized in quantities of coagulation dosages more than 30 mg/L, the AOC concentration amounted to be merely 0.15 mg/L with chlorine and 0.43 mg/L with chloramines, correspondingly. The key result suggested that addition of aluminium sulphate during coagulation is capable of augmenting AOC removal. The formation of AOC experienced more reduction by the aluminium sulphate at low coagulant dose as compared with that at higher ones. With the aluminum sulfate dose is increased, more aluminum oxide is formed during coagulation, AOC is removed through three main mechanisms including adsorption, coprecipitation, and precipitation. All three of these mechanisms can independently contribute towards AOC removal [18]. Coagulation removed the components of sample selectively, the higher the MW, the higher the removal, leading to a dramatic decrease of the average MW of the sample [19].

In this experiment, AOC removal increased up to 99% with the addition of aluminium sulphate on the doses of coagulant. In accordance with Hem and Efraimsen [9], almost 50–70% of AOC fraction comprises biological molecules that have MW less than 1000 Da. AOC encompasses a broad range of compounds, for instance sugars, organic acids, nucleic acids, amino acids, aldehydes, ketones and alcohols [15]. The reason behind the reduction of the AOC concentration is that a large amount of Cyclops metabolic organic matter possesses a larger molecular weight (MW) distribution, indicating the distribution peak for the majority of natural organic matter (NOM) sources surpassing 1.5 kDa, which, thereby, suggests the potential for AOC elimination effectiveness of coagulant (with MWs of <700 g/mol in respect of even the most hydrated alum sulphates) [20]. Nevertheless, it is required to be taken into notice that the lower MW organic compounds are still expected to be able to be left over, following the coagulation. Low molecular weight (LMW) oxygen-containing organic left in water samples are quite easily treated by chlorine and chloramine in the subsequent disinfectant mechanism. Coagulation eliminates extremely large molecules. The rest of components remained after coagulation and can react with the disinfectant chlorine to form chlorinated DBPs. The aluminium sulphate and disinfectant deliver a synergistic improvement in AOC removal, in addition regulating a trace of AOC concentration.

3.3. Effect of reaction time

Fig. 3 Time-dependent concentration of AOC from chlorination and chloramination of Cyclops metabolite dissolution at pH 7.0, chlorine dosage: 4 mg/L as Cl_2, chloramine

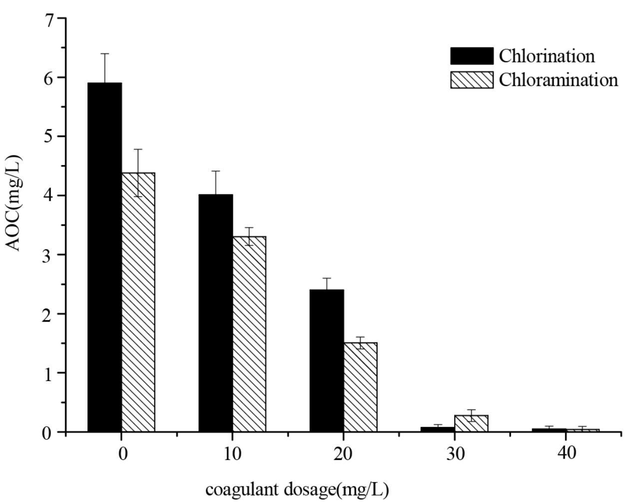

Fig. 2. Effect of coagulant dosage on AOC formation.

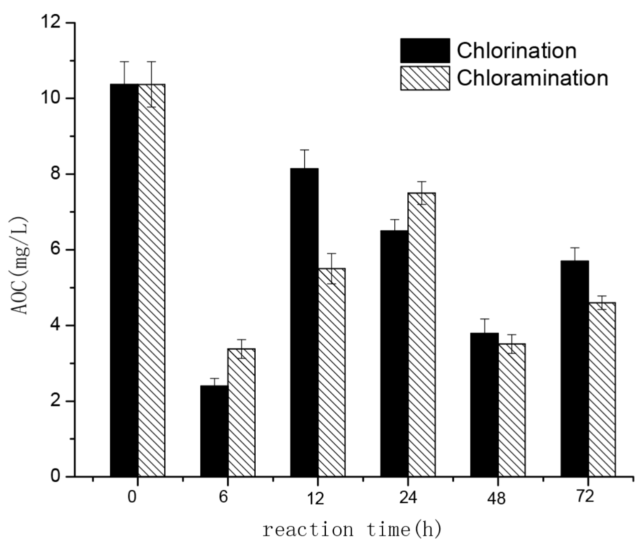

Fig. 3. Effect of reaction time on AOC formation.

dosage: 4 mg/L as Cl_2, temperature: 20 ± 2°C. The error bars represent the standard deviation of replicate measurements (*n* = 3).

Fig. 3 highlights time-dependent formation of AOC concentration from the chlorination and chloramination of Cyclops metabolite dissolution. The AOC concentration's initial concentration amounts to be 10.37 mg/L. After 6 h duration, it decreased from 10.37 mg/L to 2.4 mg/L and 3.38 mg/L, respectively with the measured disinfectants. Prolonging reaction time resulted into an increase in the AOC concentration, and AOC were achieved at 8.14 mg/L with chlorination and 5.5 mg/L with chloramination at 12 h, correspondingly. Thereafter, the levels experienced decrease with the increase in response time. Furthermore, AOC concentration variation tended to be 3.79 mg/L with chlorination and 3.51 mg/L with chloramination. At 72 h, the AOC concentration experienced a slight increase yet again. It was taken into observation that both the two disinfectants had achieved the lowest AOC concentration of reaction time after 6 h. The decreasing tendency reveals that both chlorine and chloramines, at times, were discovered generating normal quantities of AOC [10]. The explanation of this tendency can be made for more LMW organic matter, together with the fewer high MW compounds in the Extracellular organic matter (EOM) from Cyclops, based on the reference values in respect of MW distribution. Approximately 60% of lower MW compounds underwent oxidization by chlorine and chloramine within a time period of 6 h. Conversely, the explanation of the mounted productions of AOC at 12 h of incubation can be made with the help of the oxidation of microbial by-products having higher MW into lower MW organic matter (less than 1000 Da) [21]. Subsequent to oxidation, the high amount of low MW organic carbon compounds makes contribution to the formation of AOC in the EOM. The following increase could be explained in two ways that are: the oxidation of organic matters by chlorine/chloramines, leading to a rise in AOC concentration on the first; and disinfectants efficiency of disinfectants decreased gradually on the second. Chloride and Cyclops metabolite solutions form compound by means of addition reaction, substitution reaction and redox reaction. The oxidation of biological contents composed to more biodegradable and assailable compounds, for instance oxalic acid [12]. In accordance with chlorination generates low-molecular-weight compounds, in addition to enhancing the consumption of high-molecular-weight matter by bacteria.

3.4. Effect of temperature

Fig. 4 AOC Concentration as functions of temperature after 6 h chlorination and chloramination of Cyclops metabolite dissolution (4 mg/L as TOC) at pH 7.0, chlorine dosage: 4 mg/L as Cl_2, chloramine dosage: 4 mg/L as Cl_2. The error bars represent the standard deviation of replicate measurements (*n* = 3).

Fig. 4 presents the AOC concentration subsequent to 6 h of chlorination and chloramination of Cyclops metabolite dissolution, under the baseline situations, at four varied temperatures. Furthermore, it is also taken into observation from Fig. 4 that a decrease in the AOC concentration with chlorine was experienced from 10.24 mg/L at 5°C to 2.4 mg/L at 20°C and further increased to 6.7 mg/L at 30°C. The AOC variation patterns related to chloramine possessed identical attributes as the matching tendencies related to chlorine. Among the different temperature treatment plants, AOC achieves the minimum value at 20°C and the oxidizability of chloramine is quite feeble as compared with that of chlorine. It was generally acknowledged that chloramine is renowned for being a less efficient disinfectant in comparison with chlorine [17]. Our results revealed the fact that slightly heating treatment at disinfectant condition effectively hydrolyzed some of the AOC. Chlorination/chloramination of organic compounds usually turns them less bio-available, owed to the fact that halogenation of organic compounds is mostly involved as a reason for microbial persistence at low temperature [22]. Subsequent to 6 hours of time, metabolic organic matters were partially consumed and high molecular compounds were broken down into low molecular weight (LMW) organic matters that contributed to the AOC increment. With the increase in the temperature, the AOC concentration decreased. The appropriate temperature of 20°C is expected to deliver a healthier atmosphere for disinfectant reaction. Furthermore, there would be considerable improvement in the rate of chemical reaction between chlorine/chloramine and organic matter because of the impact of temperature. Hydrolysis is an endothermic process. In comparison with temperature of 20°C, high molecular compounds could be oxidized or hydrolyzed by chlorine/chloramine at temperature of 30°C. Temperature has a intimate relationship with reaction rate. Reactive activated molecule relative content increase with a increase of temperature. Rate of chlorination expedite, in favor of hydrolysis. Consequently, the response rate of disinfectants with biological contents in the water specimens is likely to boost because of the impact of temperature [16].

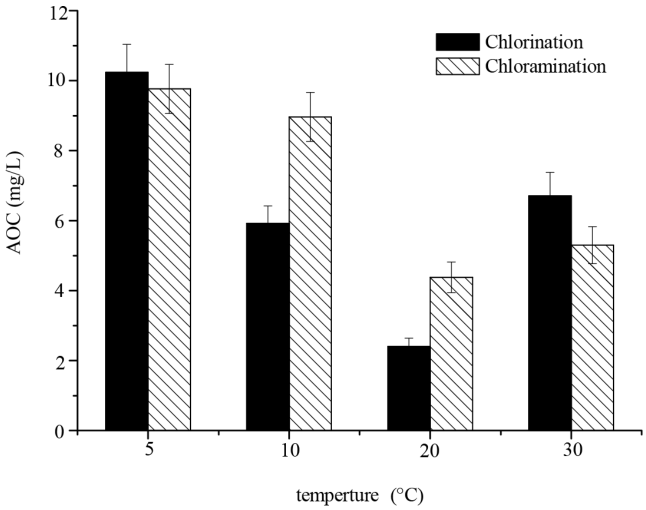

Fig. 4. Effect of temperature dosage on AOC formation.

4. Conclusions

The bench-scale experiments were carried out for the purpose of evaluating the AOC variations in disinfectant procedure of several control variable plants. This paper carried out the investigation of the relationship among AOC, disinfectant dosage, coagulant dosage, response time, and temperature effect when the raw water houses Cyclops.

On the bases of the findings received, the following conclusions can be derived:

(1) The variation of AOC concentrations by disinfectant dosages was because of organic matters oxidation resulted by chlorine and chloramine, that led to a fluctuant curve of AOC concentration. That is why choosing a reasonable disinfection dosage is quite crucial for the safety of water. The result validates the fact that the raw water with Cyclops could ensure the safety with 4 mg/L chlorine and 8 mg/L with chloramine.

(2) The variation attributes have exhibited the limitations on the efficiency of coagulant for AOC in respect to potable treatment. Normally, decrease in the AOC concentration was experienced through the addition of coagulant that led to further making AOC formation less than 0.5 mg/L.

(3) The results indicated that AOC concentration varied with reaction time extension. AOC concentration followed a decreasing trend at 6 h of time, in addition to attaining the minimum. As a result, a considerable AOC yield is not avoidable as lasting reaction time is applied for disinfection. Furthermore, an appropriate approach for controlling this parameter, subsequent to disinfection, is vital.

(4) Temperature affects reaction rate of disinfectant and oxidation/hydrolysis of organic matter. AOC achieves the minimum value at 20°C and the oxidizability of chloramine is quite feeble as compared with that of chlorine. The appropriate temperature of 20°C generated a healthier atmosphere for disinfectant reaction.

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