



Pilot-scale drinking water treatment plant: effects of disinfection alternatives and filtration systems

Tarek A. Gad-Allah^a, Mohamed I. Badawy^a, Azza M. Abdel-Aty^a, Rizka K. Ali^a, Hazem Saleh^b, Yeomin Yoon^c, Mohamed E.M. Ali^{a,*}

^aWater Chemistry Lab, Department of Water Pollution Research, National Research Centre, 33 EL Bohouth Street (former El Tahrir Street), Dokki, Giza, P.O. 12622, Egypt, Tel./Fax: +2033371499; emails: alienv81@yahoo.com (M.E.M. Ali), tareqabdelshafy@yahoo.ca (T.A. Gad-Allah), badawy46@hotmail.com (M.I. Badawy), azzamy@hotmail.com (A.M. Abdel-Aty), rizka.kamel@yahoo.com (R.K. Ali)

^bDepartment of Civil Engineering, Faculty of Engineering, Menoufia University, Menoufia, Egypt, email: hazemsaleh1@hotmail.com

^cDepartment of Civil and Environmental Engineering, University of South Carolina, 300 Main Street, Columbia, SC 29208, USA, email: yoony@cec.sc.edu

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ABSTRACT

A pilot-scale drinking water treatment plant was designed and manufactured to control disinfection by-product (DBP) levels through better removal of natural organic matter (NOM) and the application of disinfectant alternatives that produce lower DBPs levels. The designed pilot plant consists of several stages including disinfection (by chlorine, chlorine dioxide, or ozone), coagulation–flocculation (using alum or alum + cationic polymer), and a sedimentation and gravity filtration system, such as sand or granular activated carbon (GAC). The efficiency of the designed plant was evaluated according to the water quality produced in terms of turbidity, pH, alkalinity, total organic carbon, and ultraviolet absorption at 254 nm (UV₂₅₄), phytoplankton, and DBP levels. Improved reductions of turbidity, algae, and NOM, and consequently lower DBP levels, could be obtained. The lowest DBPs levels were obtained through the treatment sequence: ClO₂/modified coagulation/GAC, due to the combination of the high oxidation powers of ClO₂ with the high adsorption capacity of GAC.

Keywords: Disinfection; NOM removal; Ozonation; Chlorine dioxide; Granular activated carbon

1. Introduction

Existing water treatment plants (WTPs) are under severe stress due to the pollution of natural water resources and increasing demand with population growth. In Egypt, most of the WTPs use conventional treatment methods, which consist of pre-chlorination, coagulation–flocculation, sedimentation, sand filtration, and post-chlorination [1]. This sequence ensures the production of safe, disinfected, potable water.

Fresh water usually contains 2–10 mgC/L as natural organic matter (NOM). NOM consists mainly of microorganisms,

complex organic matter (humic acid (HA) and fulvic acid), and naturally occurring degradation products (e.g., amino acids, fatty acids, phenols, sterols, sugars, hydrocarbons, urea, porphyrins, and polymers) [2]. Unfortunately, it was confirmed in the 1970s that chlorine reacts with NOM to produce a wide range of disinfection by-products (DBPs) [3–5]. Many of these DBPs have since been identified as carcinogens [4–6]. Trihalomethanes (THMs) and haloacetic acids (HAAs) are the two most abundant groups of DBPs found in chlorinated water. The United States Environmental Protection Agency (USEPA) regulates THMs and HAAs in drinking water at 80 and 60 µg/L, respectively [7]; while in Egypt, the permissible limits for these compounds are 100 and 80 µg/L, respectively.

* Corresponding author.

Thus, there is a dilemma in water treatment strategies between efficient disinfection and the formation of harmful DBPs. Two reasonable strategies have been proposed to control DBPs levels. They are (1) removal of NOM (the main precursor of DBPs) and (2) use of disinfectant alternatives that produce lower DBPs levels.

Various technologies (e.g., the magnetic ion exchange resin [MIEX] technique, activated carbon filtration [8,9], membrane filtration techniques, and advanced oxidation processes [9]) have been investigated for NOM removal with differing degrees of success, depending on the properties and amount of NOM. For example, the MIEX technique is the only technique that is able to remove even hydrophilic NOM, while the other techniques mentioned do not successfully remove all NOM fractions. However, from an economic point of view, coagulation and flocculation followed by sedimentation/flotation and filtration remains the most commonly used reliable technique [9].

Regarding the disinfectant alternatives, UV disinfection has been shown to be effective in removing most pathogens without contributing to the formation of toxic by-products. Nevertheless, the efficiency of UV disinfection fluctuates with water quality, such as turbidity, color, and suspended solids [10]. Also, DNA damaged by UV can be repaired by photoreactivation processes and/or dark repair, which can result in extensive bacterial re-growth [11]. Sonication of water and wastewater effluent treatment has also been reported. In this process, the mechanical, chemical, and heat effects of cavitation bubbles inactivate the microorganisms, with mechanical effects dominating the process. However, this type of disinfection consumes high specific energy [10]. Recently, radiation technologies using gamma rays or an electron beam as a powerful alternative to conventional disinfection methods have attracted attention. Studies have demonstrated the effectiveness of ionizing radiation for the destruction of organic contaminants and the inactivation of pathogenic microorganisms [11]. However, these disinfection alternatives suffer from high costs in application on a large scale. Only ozonation and chlorine dioxide, as disinfection alternatives, have shown comparable disinfection to chlorination with low application costs.

Previous studies used mostly bench-scale setups to test the efficiency of the disinfection alternatives mentioned above. In this study, a complete pilot setup was designed and used in the investigation of water treatment using ozone and chlorine dioxide (as disinfection alternatives), accompanied with modified coagulation using an ionic polymer and filtration on sand or granular activated carbon (GAC). To gain insight into the role of NOM in water quality, NOM was measured as dissolved organic carbon (DOC); UV absorption at 254 nm provides an indication of NOM aromaticity. The DBPs formed were evaluated in each treatment scheme. In the present study, various ways will be applied to diminish DBP production, such as changing disinfection style (ClO₂-based, O₃-based disinfection or point (intermediate Cl₂ disinfection) or the elimination of the NOMs or DBP precursors before disinfection. Herein, different trends of pilot system for drinking water treatment (DWT) were implemented in pilot system for the delivery of high-quality drinking water. The efficiencies of different DWT system will be evaluated.

2. Experimental setup

2.1. Pilot-plant setup

Pilot conventional and modified drinking water treatment plants were set up at the National Research Center, Egypt. Raw water was collected from a site at the Nile River and stored in a feed tank with a storage capacity of 240 L; the feed tank was made of uPVC with stirring blades for complete mixing. Most parts of the pilot plant were constructed from 2-mm-thick plexiglass with steel supports (thickness: 10 mm), with the exception of the feeder tank. The feed tank was made of uPVC and 1-inch-diameter polypropylene pipes. Table 1 lists the pilot plant components. Fig. 1 shows the plant layout.

The designed pilot plant includes primarily the stages of disinfection, coagulation–flocculation, and a sedimentation and gravity filtration system, such as sand or GAC. The design of this plant allowed the examination of different treatments, as depicted in Fig. 2. The examined treatment schemes are given below:

- Scheme 1: Cl₂/conventional coagulation
- Scheme 2: Cl₂/conventional coagulation/sand filtration/Cl₂
- Scheme 3: Cl₂/modified coagulation/sand filtration/Cl₂
- Scheme 4: Cl₂/modified coagulation/GAC filtration/Cl₂
- Scheme 5: Cl₂/modified coagulation/Cl₂/sand filtration/Cl₂
- Scheme 6: ClO₂/modified coagulation/sand filtration/Cl₂
- Scheme 7: ClO₂/modified coagulation/GAC filtration/Cl₂
- Scheme 8: O₃/modified coagulation/sand filtration/Cl₂
- Scheme 9: O₃/modified coagulation/GAC filtration/Cl₂

Table 1
Components of the pilot plant (Q = 240 L/d)

| Item | Item name | Description |
|------|-----------------|--|
| 1 | Feed tank | 50 × 50 × 50 cm |
| 2 | Serpentine pipe | Diameter = 1 inch Total length = 7.00 m Length for chlorination = 6.6 m (detention time = 20 min) Length for alum treatment = 0.4 m (detention time = 60 s) |
| 3 | Flocculation | Dimensions 30 × 20 × 10 cm Detention time = 36 min |
| 4 | Sedimentation | Dimensions 40 × 15 × 40 cm Detention time = 144 min |
| 5 | Ozonation | Inner diameter = 6 cm Length = 120 cm Detention time = 20 min |
| 6 | GAC | Inner diameter = 5 cm Length = 1.3 m Average loading rate = 5.1 m ³ /m ² h EBCT = 15 min |
| 7 | RSF | Inner diameter = 5 cm Length = 1.3 m Rate of filtration = 5.1 m ³ /m ² h EBCT = 15 min |

RSF, rapid sand filter; EBCT, empty bed contact time.

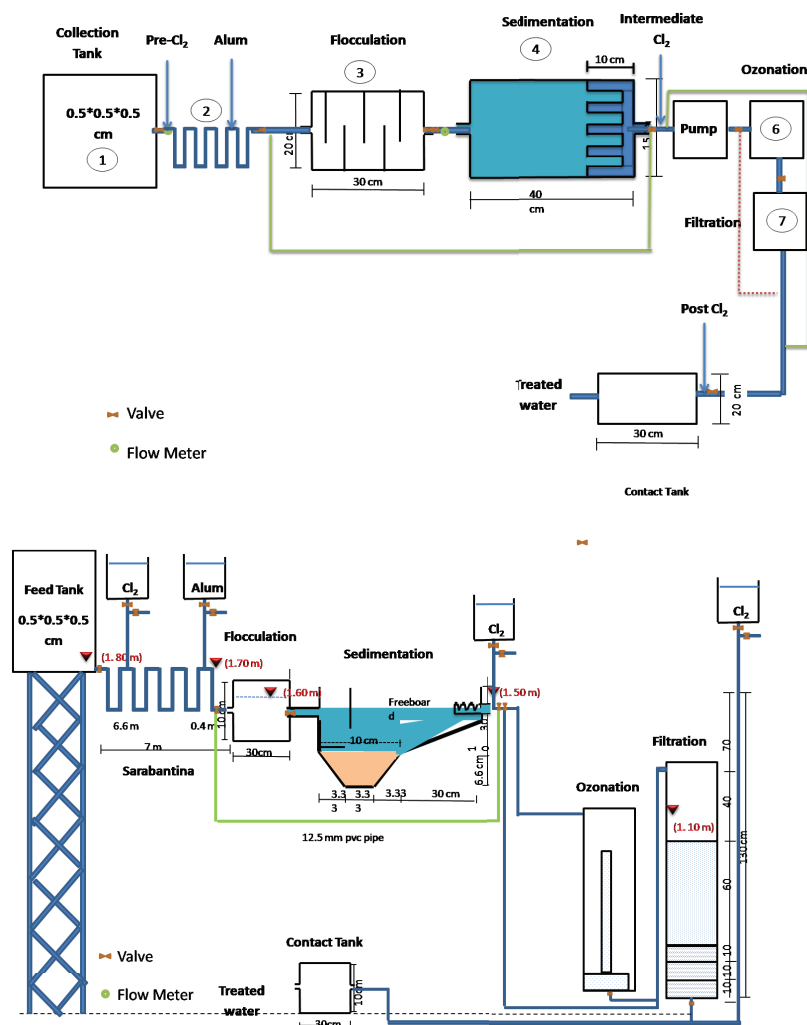


Fig. 1. Pilot plant layout (a) projection view and (b) side view.

Modified coagulation with aluminum sulfate (alum) and cationic polymer (Sigma–Aldrich, Germany) for removing DBP precursors, inorganic compounds, and particulate matter took place in a rapid mixing tank by injecting the chemicals in a serpentine pipe. Filtration via sand or activated carbon was examined to enhance the adsorption of organic matter as well as DBPs formed in the pre-chlorination step. Backwashing of the filters was carried out when needed. Chlorination points at different positions were examined for the reduction of DBPs formation: thus, pre-, intermediate, and post-chlorination locations were tested. Chlorine alternatives, such as chlorine dioxide and ozone, were also examined. Chlorine dioxide stock solutions were prepared by collecting gaseous ClO_2 generated through the reaction between H_2SO_4 and NaClO_2 according to standard method 4500- ClO_2 B [12]. The stock solution was standardized using a UV spectrophotometer at 360 nm [13]. Ozone was generated from oxygen gas using an ozone generator (Ozononia, CFS 1A, Switzerland). The ozone generator was turned on for a minimum of 15 min prior to the start of a run to allow the output to stabilize.

Samples were collected from each scheme every 6 h for a duration of 72 h of continuous operation of the treatment

system. The water quality was monitored during the treatment process by the determination of physico-chemical parameters, such as turbidity, pH, and alkalinity. Chlorine residuals were analyzed at the time of sampling using the *N,N*-diethyl-*p*-phenylenediamine ferrous titrimetric method [12]. Factors related to DBP formation, such as total organic carbon (TOC), which gives an indication of the amount of NOM, ultraviolet absorption at 254 nm (UV_{254}) for the determination of aromatic organic compounds, and specific ultraviolet absorption at 254 nm (SUVA_{254}) were calculated, obtained by dividing a sample's ultraviolet absorption at a wavelength of 254 nm (UV_{254}) (where $m = 1$) by its concentration of DOC (in mg/L).

The DBPs were analyzed for all samples including four chlorine-containing and bromine-containing THM (THM4) compounds and five chlorine-containing and bromine-containing HAA (HAA5) compounds. All of the samples were refrigerated at 4°C and analyzed within 2 weeks. THMs and HAAs were quantified by liquid/liquid extraction followed by gas chromatography and electron capture detection (GC/ECD) according to standard methods No. 6232 and 6251, respectively [12].

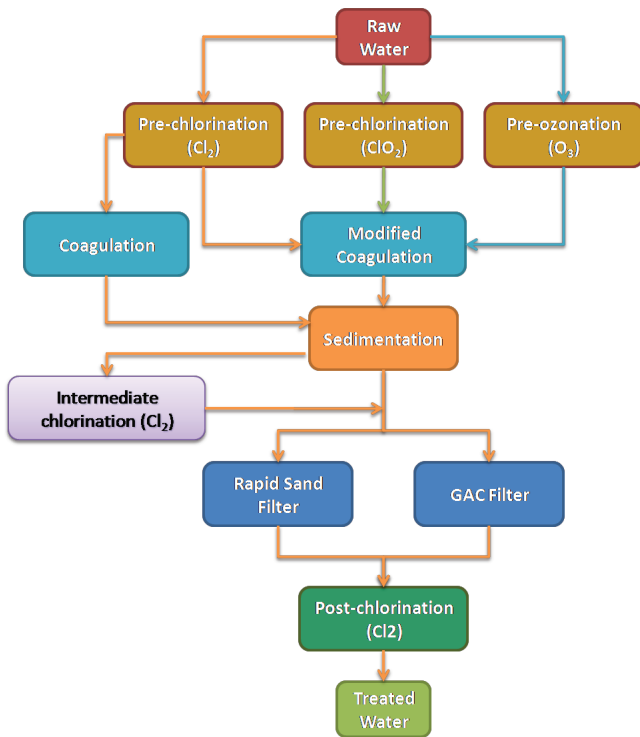


Fig. 2. Treatment schemes used in the pilot plant.

The concentration of phytoplankton present in water samples was detected using a Sedgwick-Rafter funnel and preserved in Lugol's iodine solution [12] to assess algal removal. Identification of the algal community structure was examined using identification keys according to [12,14]. A Sedgwick-Rafter counting cell was used for phytoplankton counting.

3. Results and discussion

3.1. Efficiency of the pilot plant on water quality

To evaluate the water quality after the application of the different treatment processes, 12 water samples were collected from the outlet of the pilot plant for each treatment system. Physiochemical characteristics, such as alkalinity, pH, turbidity, and electric conductivity (EC), were analyzed in the treated water (Figs. 3(a)–(d)). The results (Fig. 3(b)) showed that the modified coagulation improved turbidity reduction vs. the conventional scheme. The efficiency of turbidity removal ranged from 78% to 88.3%. Also, average alkalinity levels in the treated waters were 82–120 mg/L, with a corresponding removal efficiency of 14%–39% (Fig. 3(d)). However, the changes in the pH and EC levels (Figs. 3(a) and (c)) of the treated water over time were not significant, as observed during continuous operation of the pilot-scale plant.

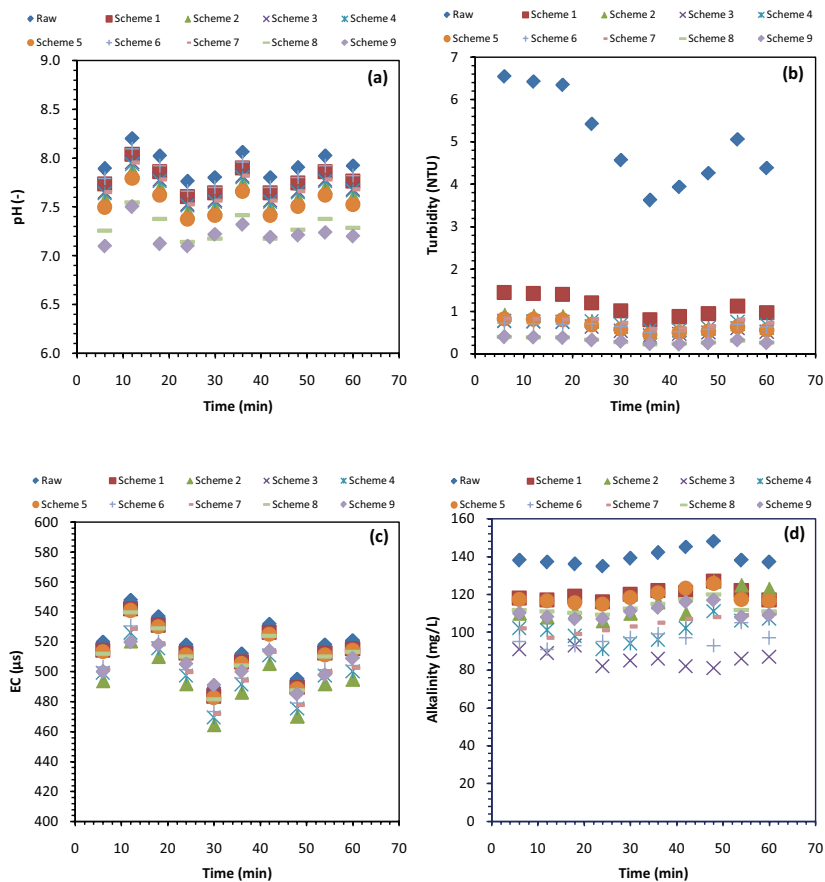


Fig. 3. Time variation of (a) pH, (b) turbidity, (c) EC and (d) alkalinity using different treatment schemes.

3.2. Efficiency of the designed pilot plant for the removal of parameters related to DBP formation

The levels of TOC and DOC have a significant impact on DBP formation upon reaction with chlorine. Other indicators for the level of NOM are ultraviolet absorption at 254 nm (UV_{254}) and specific ultraviolet absorption at 254 nm ($SUVA_{254}$), which is the ratio of UVA_{254}/DOC . Particularly, $SUVA_{254}$ is an indicator of NOM aromaticity in the water. The efficacy of the different treatment schemes in the removal of TOC, DOC, UV_{254} and $SUVA_{254}$ are shown in Figs. 4(a)–(d).

In Fig. 4(a), 46% reduction in the UV_{254} value was obtained by modified coagulation using alum/cationic polymer, compared with 30% removal via conventional coagulation. When using treatment schemes with modified coagulation and Cl_2 as the pre-disinfectant, followed by sand or GAC filters, the UV_{254} values were reduced by 46% and 53.5%, respectively. UV absorption was reduced by 72% for enhanced coagulation (alum/cationic polymer) coupled with intermediate chlorination and a sand filter compared with 30% removal after conventional coagulation. The removal percentages for the treatment schemes with modified coagulation and ClO_2 as the pre-disinfectant followed by a sand or GAC filter were 74.6% and 77.6%, respectively. In the case of using ozone in the pre-disinfection, the removal percentage of UV absorption was 80% after sand filter use. This suggests that ozone is able to destroy the aromatic rings of organic compounds in water more effectively than the other disinfectants tested.

Changes in the DOC of treated water over time, using the different treatment schemes, are illustrated in Fig. 4(c). The experimental results from the analysis of treated water showed that the average DOC levels ranged from 1.84 to 3.2 mgC/L, equivalent to 26.7% to 69% removal, respectively. It was observed that application of ClO_2 or ozone as the pre-disinfectant reduced the DOC level in water produced from the pilot plant (Fig. 4(c)). Also, water produced from the pilot plant equipped with a GAC filter showed a lower level of DOC and higher removal percentages due to the high adsorption capacity of GAC relative to sand. For example, the water produced from the pilot plant showed that 26% and 36% DOC removal could be achieved in cases of intermediate chlorination and ozone as a pre-disinfectant, respectively, followed by a GAC filter. Removal of DOC by ozone was not very high. This may be attributable to ozone reacting with large, aromatic NOM species, transforming them to smaller, less reactive compounds. Thus, an ozone treatment system should not be expected to decrease significantly the total amount of DOC in the water; however, a considerable reduction in THMs and HAAs was observed when chlorine was added for post-disinfection. The slight reduction in DOC, apparent after commissioning the treatment plant, may be related to adsorption of ozonated NOM species on the media of the filters that followed the ozone contact chamber.

The time variation in $SUVA_{254}$ level in the treated water is shown in Fig. 4(d). The use of modified coagulation reduced the values of $SUVA_{254}$ by 15%, compared with conventional

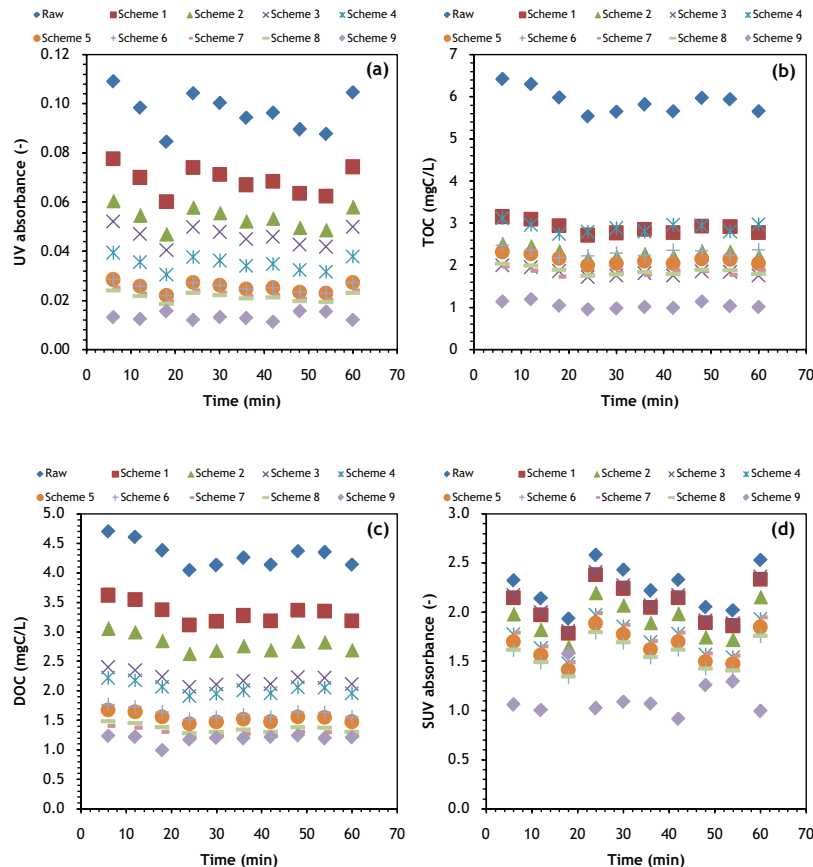


Fig. 4. Changes in parameters related to DBP formation during continuous operation of the pilot plant.

coagulation (only 8%). Moreover, 20% and 26% reductions in $SUVA_{254}$ values after sand and GAC filters, respectively, were observed when Cl_2 was used as the disinfectant. The use of ClO_2 in the pre-chlorination phase reduced the $SUVA_{254}$ by 33% and 36% after sand and GAC filters, respectively. Upon using ozone and intermediate chlorination in the pre-chlorination resulted in a decline in the UV, by 78% and 74%, respectively, as shown in Fig. 5. As well decrease in TOC and DOC is achieved using ozone and intermediate chlorination in the pre-disinfection by 64% and 68%, respectively. This attributed to oxidation of HA substance to less reactive species.

3.3. Efficiency of the pilot plant in reducing DBP formation

THMs and HAAs are the most important groups of DBPs. The results obtained for THMs and HAAs variation with time in a continuous-flow mode of the pilot-scale plant using different treatment schemes are shown in Fig. 6. Both THMs and HAAs formation were reduced by

more than 25% after water treatment schemes of Cl_2 /modified coagulation/sand filter compared with the water treatment scheme of Cl_2 /conventional coagulation/sand filter. This was attributed to better removal of TOC and DOC (the main precursors of DBPs) via modified coagulation (Fig. 5). The use of intermediate chlorination and ozone as a pre-disinfectant had a positive effect on the reduction of DBPs. The system of ozone/enhanced coagulation/GAC filter showed the lowest average values of THMs and HAAs, 21.83 and 18.95 $\mu\text{g/L}$, respectively, due to the combination of the high oxidation powers of O_3 with the high adsorption capacity of GAC. This combination reduced the DBP precursors in the water. Thus, the most effective treatment sequence was O_3 /modified coagulation/GAC filter. Fig. 7 showed the efficiency of DBPs reduction in the pilot plant using different treatment system. Notably, the formation of DBPs was reduced in pilot treatment using O_3 /modified coagulation, intermediate ClO_2 /modified coagulation and intermediate Cl_2 /modified coagulation in comparison to conventional drinking water treatment. This attributed to oxidation of HA substance to less reactive species to be reacted with Cl_2 or less abundance of hypochlorous acid in water.

3.4. Efficiency of the pilot plant regarding algae removal

Table 2 presents the effects of optimal conditions for algal coagulation removal during the pilot study. During the period of the study, the dominant algal species were diatoms, green algae, and blue-green algae. Diatoms represented the major percentage of the algal community, 84.6%; green algae amounted to 9.8%; and blue-green algae represented 5.6% of the total algal count. These results are consistent with those from previous studies [15,16].

Generally, the cationic polymer enhanced the percentage of algal removal (92.3%) compared with the control (alum treatment, 86.3%). With the cationic polymer, diatoms showed higher removal, followed by green algae (80.8%). These results are consistent with those of Huang and Yeh [17], who showed that algae species exhibit noticeable

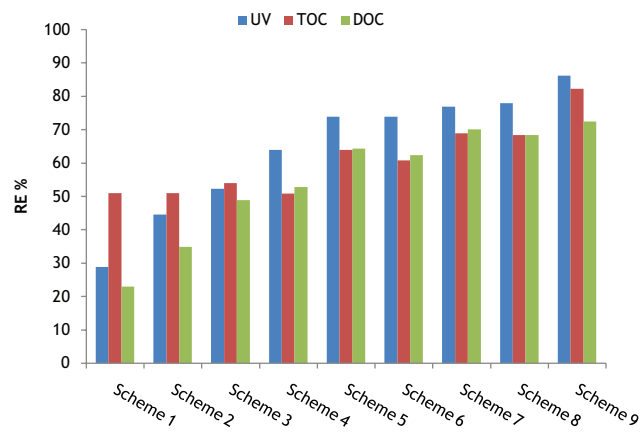


Fig. 5. Efficiency of the pilot plant with different operation for removal of DBPs precursors.

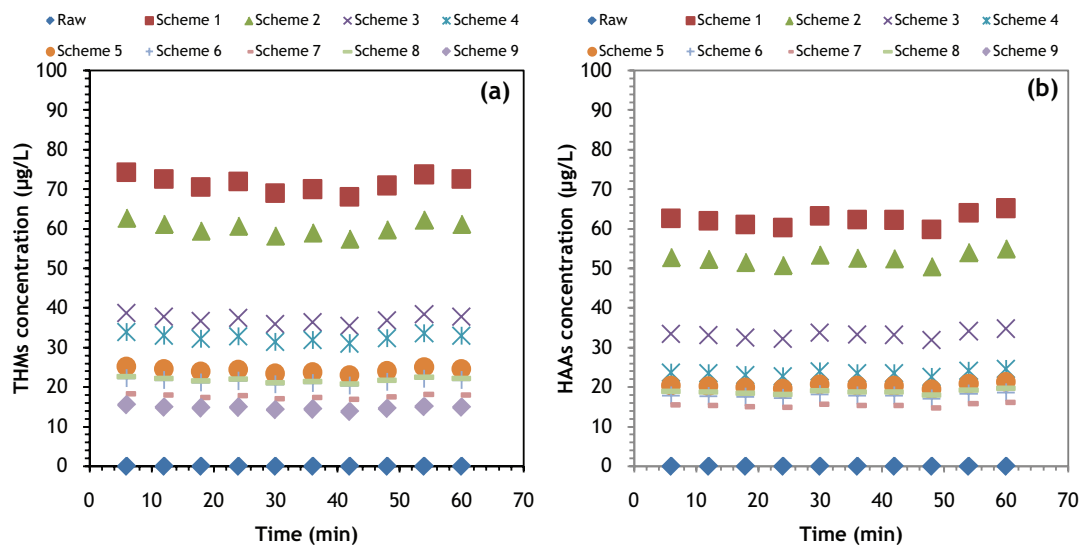


Fig. 6. (a) THMs and (b) HAAs changes with time in the continuous-flow mode of the pilot plant.

responses to various combinations of pre-oxidation and coagulation processes. When polymer was used as a coagulant aid with alum, the cell removal of both green algae and diatoms improved. Small doses of polymer could significantly reduce the required alum dose. Conventional water treatment facilities can remove the algal cells but do not remove potentially taste- and odor-forming compounds. Water treatment technology, especially the use of activated carbon, should be adopted to ensure that these are kept at or below guideline levels. Results with GAC as a filter medium are given in Table 2, which showed that GAC + modified coagulation were more effective for different algal group removal compared with a sand filter. Responses of the

dominant algal species to GAC treatment showed that most algal species were removed, with the exception of centric diatoms (*Cyclotella comta* and *Melosira granulata*) and pennate forms (*Diatoma elongatum* and *Synedra ulna*). By adding GAC as a polishing filter after a sand filter, algal removal increased to 96.6%. Variation in the response of algae to coagulation–flocculation treatment may be attributable to changes in their morphological characteristics and physiological activity.

The results in Table 2 give also information about using the alternative oxidants, chlorine dioxide and ozone, for algal removal instead of pre-chlorination. The combination of optimal chlorine dioxide and alum doses (2 and 25 mg/L) and using sand as a filter, showed high decreases in algal counts, especially diatoms, where the percentage removal was 95.6%. In the case of using GAC treatment as a filter aid in combination with chlorine dioxide, the percentage removals of diatoms, green algae, and blue-green algae increased compared with sand. The results showed that algal species were affected by 2 mg/L chlorine dioxide, with the exception of *Diatoma elongatum*, *Cyclotella comta*, *Melosira granulata* (diatoms), and *Coelastrum microporum*, *Pediastrum clathratum*, and *Actinastrum hantzschii* (green algae). The combination of optimal ozone and alum doses (3 and 25 mg/L, respectively) with sand and GAC showed that the responses of the three algal groups in the two cases were nearly equivalent. The results of the present study are consistent with previous investigations by many researchers who demonstrated that other pre-oxidants, such as ozone, chlorine dioxide, and permanganate, can improve algae removal by coagulation and filtration processes in drinking WTPs [18–20]. These pre-oxidants improve algae coagulation by deactivating algal cells, reducing cell stability, and liberating extracellular organic matter (EOM). The effects of the oxidant on algae removal during water treatment are dependent on oxidant type, oxidant dosage, and the algal species. For example, ozone improved the coagulation of green algae (*Scenedesmus quadricauda*) but not of diatoms (*Cyclotella* sp.) [21]. This is most likely due to differences in the surface characteristics of the cells. Bernhardt and Clasen [22] reported that EOM from

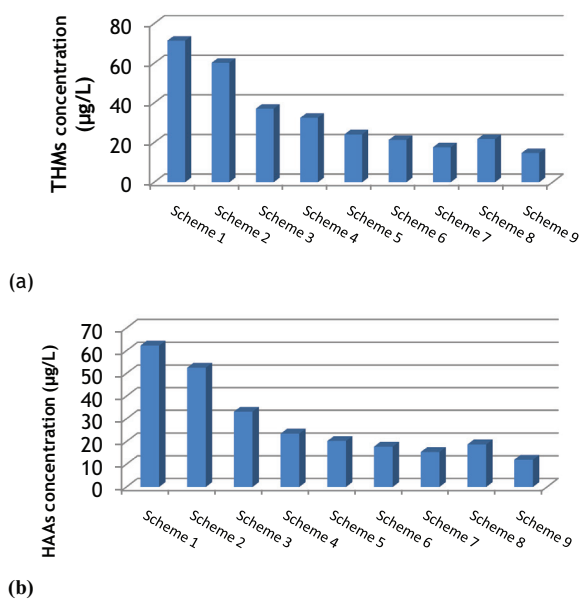


Fig. 7. Efficiency of different system of DBPs reduction (a) THMs and (b) HAAs changes with time in the continuous-flow mode of the pilot plant.

Table 2
Efficiency of treatment scheme in algal removal

| Algal groups | Counts of raw water (Org./mL) | Counts (% R) | | | | | | |
|--------------------|-------------------------------|--------------|----------|----------|----------|----------|----------|----------|
| | | Scheme 1 | Scheme 3 | Scheme 4 | Scheme 6 | Scheme 7 | Scheme 8 | Scheme 9 |
| Diatoms | 4,500 | 510 | 240 | 250 | 200 | 150 | 160 | 120 |
| | | (88.7%) | (94.7%) | (94.4%) | (95.6%) | (96.7%) | (96.4%) | (97.3%) |
| Green algae | 520 | 130 | 100 | 70 | 70 | 50 | 50 | 40 |
| | | (75%) | (80.8%) | (86.5%) | (86.5%) | (90%) | (90.4%) | (92.3%) |
| Blue-green algae | 300 | 90 | 70 | 60 | 40 | 30 | 30 | 30 |
| | | (70%) | (76.7%) | (80%) | (86.7%) | (90%) | (90%) | (90%) |
| Total algal counts | 5320 | 730 | 410 | 380 | 310 | 230 | 240 | 190 |
| | | (86.3%) | (92.3%) | (92.8%) | (94%) | (95.7%) | (95.5%) | (96.4%) |

Alum ($Al_2(SO_4)_3 \cdot 16 H_2O$) dose = 25 mg/L.

Pre- and post-chlorination with chlorine dose 5 and 2 mg/L, respectively.

cultures of green and blue-green algae and diatoms behaved like ionic and non-ionic polyelectrolytes. The amount and properties of EOM produced varied with algae species, oxidant type, and oxidant dosages. EOM, depending on the concentration and molecular weight, can enhance or hinder flocculation. Low dosages of ozone can cause release of EOM, which aids coagulation, but at higher doses, ozone affects EOM structure (e.g., by lowering molecular size), which may hinder subsequent coagulation [23] (Figs. 8 and 9).

3.5. Mechanism of DBPs formation in ozone-based and intermediate chlorination-based scheme for DWT

In ozone-based scheme, O_3 interacts with great fraction of NOMs and degrade them into less reactive compounds. Consequently, the ozone treatment system should not be anticipated to decrease high amount of DOC in the water but relatively should reduce the amount of THMs and HAAs formed when chlorine is added for post-disinfection. This recommended that the ozone treatment was undeniably affecting the reactivity of the NOM, and produced low DBPs level-treated water (Scheme 1). In case of intermediate chlorination system/enhanced coagulation, the application of enhanced coagulation reduced the values of DOC as well as division Cl_2 dose. The lower dose of Cl_2 and availability of NOMs are the minor amount of DBPs formed (Scheme 2). Consequently, the intermediate chlorination with enhanced coagulation is more feasible trends rather than that of O_3 -based trend as cost-effective routes for production of high-quality water with low DBPs level.

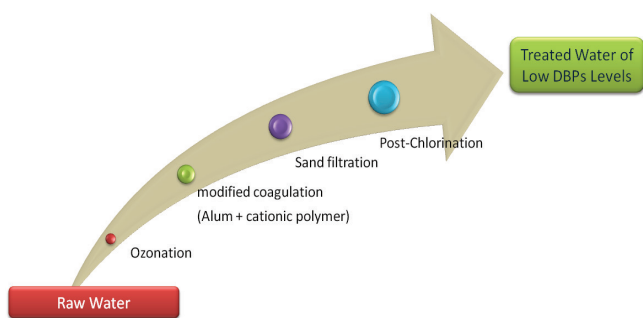


Fig. 8. Proposed Scheme 1: ozone-based DWT system.

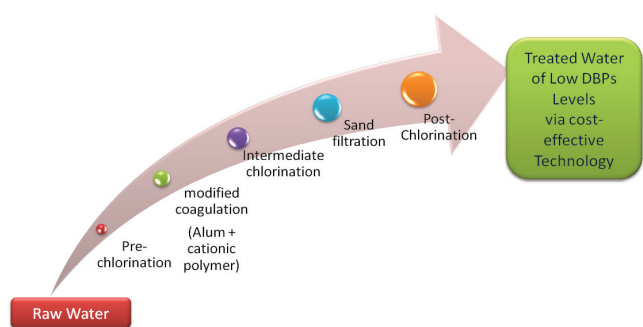


Fig. 9. Proposed Scheme 2: intermediate chlorination-based DWT system.

4. Conclusions

Different water treatment strategies were investigated using a custom-designed, pilot-scale treatment plant. Relative to conventional water treatment, better water quality was obtained using the treatment sequence of O_3 (3 mg/L)/modified coagulation/sand filtration and application of intermediate chlorination)/modified coagulation/sand filtration. This may be due to several factors:

- Modified coagulation enhances the removal of algal species, DOC, and suspended particulates. Thus, the possibility of DBPs formation was reduced.
- Replacing chlorine with ozone resulted in stronger oxidation of NOM (the main precursor of DBPs).
- Division amount of chlorine leads less available oxidant to NOMs.
- Filtration with sand contributed to the removal of NOM and DBPs from the water produced.

Conclusively, the intermediate chlorination with enhanced coagulation is more feasible trends rather than that of O_3 -based trend as cost-effective routes for production of high-quality water with low DBPs level.

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