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The Effects of Pre-Ozonation on Drinking Water Quality Parameters

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Abstract
The study investigated the effects of pre-ozonation on the parameters such as turbidity, color, chlorophyll a and TOC on a pilot scale. The investigation results indicate that the amount of required ozone to remove TOC, color and turbidity depends on the quality of inlet water, and the efficiency of water ozonation depends on the process factors such as ozone dose, temperature, pH and ozone-water contact time. The study likewise shows that the lower amount of turbidity, TOC, temperature and higher alkaline pH of sample water boost the ozonation effect on removing the variables in question. The results also demonstrate a direct relationship between pH, ozone dosage and contact time, as well as an indirect relationship between temprerature and the removal of parameters.

Key words: disinfection, humic acid, pilot plants, pre-ozonation, surface water treatment.

Paper type: Research article.

1. Introduction

In drinking water refining, disinfection manifests a key procedure (Nabi et al., 2006). Water disinfection plays a major role in public health by combatting the spread of water-related diseases (Collivignarelli et al., 2018). Pre-oxidation of potable water allows eliminating color, taste, odor, and pollutants (Van der et al., 2011). Pre-oxidation also has a significant effect on the natural organic matter (NOM) and increases the biodegradability of organic matter (Rietveld et al., 2008).

In water treatment, ozone represents an effective disinfectant and oxidant used before coagulation as a pre-oxidant (Liu et al., 2006) for enhancing coagulation, oxidation of organic compounds and improving the overall water quality (Carrim, 2006). Masoomi et al. (2019) showed that pre-ozonation and coagulation are effective in removing physical, chemical, organic, and biological parameters of water.

Ozone is a powerful oxidizer (Heng & Guibai, 2009) and is capable of eliminating organic compounds in water (Honarmandrad et al., 2020; Javid et al., 2019). Ozone is commonly used for treating potable water due to excellent reactivity and complete lack of chemical residue (Carlo et al., 2017). Physical, chemical, and biological characteristics are the main factors determining water quality, and ozone has a different effect on each of these properties (Masoomi et al., 2019). The removal of NOM represents one of the main objectives of water treatment (Mahvi et al., 2011). The organic compounds affecting water color include humic and fulvic acids, and soil humus. Often, watercolor levels below 15 TCU are acceptable to consumers (NCBI, 2017). In general, colored water adversely impacts human health and the environment (DFAS, 2004).

In water refining and freshwater recirculating systems, different ozone dosages are applied. Based on the literature, multiple experiments allow evaluating the effects of ozone doses on water quality (Aikaterini et al., 2018). Water ozonation leading to polymer breaking of a humic substance reduces watercolor. Ozonation allows removing up to 80% of color depending on the ozone dose, and increasing ozone dosage reduces color removal (Melin & Odegaard, 2013). Suspended materials such as clay, silt, organic and inorganic materials, as well as other microscopic organisms in water are caused by turbidity (Mark et al., 1981). As per the World Health Organization standards, the maximum permissible turbidity limit for drinking water is 5 nephelometric turbidity units (NTU) (WHO, 2011). High turbidity can significantly reduce the quality of water in lakes and streams and has a harmful impact on human activities such as recreation and tourism. Also, it can increase the cost of potable water treatment (MPCA, 2008). Controlling the amount of algae in water represents one of the main uses of ozone (Carrim, 2006). Chlorophyll a manifests the main photosynthesis pigment in green algae. Its concentration represents the volume of aquatic plants in the water column (Water Quality Parameter Definitions, Chlorophyll a, 2019). Chlorophyll is the primary pigment produced by algae in freshwater lakes, and its content is an important water quality parameter used for evaluating algae performance (US EPA, 2017). Chlorophyll a is present in micro-phytoplankton and can be used for approximating the existing biomass (Agency bbe moldaenke, 2019). All surface waters have NOMs, which the humic material is a significant part of. Various analyses can be used to measure NOMs in potable water. TOC (Total Organic Carbon) and DOC (Dissolved Organic Carbon) are the most important parameters for analyzing NOM in water treatment processes (Sillanpaa et al., 2015). The presence of natural organic substances such as tannic, fulvic, and humic acids in surface water changes its color (OMRC of Iran, 2007). Changes in pH during ozonation may occur due to the production of organic acids or the use of organic acids. In designing chemical disinfection, the CT values (disinfectant concentration * time (mg/l*min)) are considered sufficient to eliminate 99-99.9% of microorganisms (Twort & Ratnayaka, 2000). Lage Filho (2010) showed that the amount of CT was directly related to the ozone dosage and the ozone contact time with water. Morrison et al. (2012) argued that pre and medial ozonation do not affect the pH, DOC, and TOC. Further, chlorophyll a and total chlorophyll spectral absorption coefficient (SAC254) does not change under the influence of pre-ozonation, yet it is influenced by medial ozonation. Also, Torabian et al. (2006) showed that pre-ozonation in the course of water treatment can improve TOC removal. Thus, the main objective of this study was to assess the effects of pre-ozonation on water quality variables. This pilot study was conducted on the water entering the Kuh-Sabz Drinking Water Refinery (DWR) located near Marvdasht City in the southwest of Iran's Fars Province in July 2017.

2. Materials and method

2.1. Materials and water samples

To simulate turbidity, clay and kaolinite were used; and to simulate TOC, humic acid was added to raw water. Watercolor and chlorophyll a were simulated by adding green algae. Also, the algae solution of $NaNO_3$ was used to supply the nitrate required by algae. The ozone was produced by the OPW27 Ozone Generator with the nominal capacity of 5.0 gr/hr fitted with an air supply pump.

2.2. Analytical method

The Kuh-Sabz DWR is located in the south of Iran near Kuh-Sabz Village at 52°41´29.16´´ E and 29°55´16.79´´ N. To investigate the effects of ozone on water quality parameters, initially the sample (20 liters) was taken from the water entering the DWR, and its turbidity, color, chlorophyll a, TOC, pH, and temperature were measured at 8.8 FTU, 17.5 TCU, 1.3 mg/l, 8.7 mg/l, 7.5 and 21.5°C, respectively. The sample underwent the simulation of turbidity, color, chlorophyll a, and TOC.

Consequently, the turbidity, color, and TOC were regulated based on the trial and error method. The parameter values (turbidity, color, chlorophyll a, TOC, and pH) in the simulated water sample equaled 21.8 FTU, 19.6 TCU, 1.6 mg/l, 9.5 mg/l, and 7.5 (Table 1.). To study the effects of ozonation on the variables, the ozone dosages of 1.0 to 5.0 gr/hr and ozone-water contact time of 5 to 20 min were selected. To increase the ozone-water contact time, the ozone injection site with 4 diffusers was placed at the bottom of the reactor cell. The sample pH underwent daily calibration by the Swiss-made Metrohm device and buffer solutions. For measuring the TOC, the DR 4000 Spectrophotometer was used based on Method 10129; and the Algaturch device was used for monitoring turbidity and chlorophyll a concentration. The sample color was measured as per Standard Method 5910B. Fig. 1 shows the schematic plan of the plug flow reactor used in this study. The sample water's pH was adjusted by hydrochloric acid and sodium hydroxide from 4.0 to 8.6. After that, the 20-liter sample solution was prepared as per the specifications in Table 1. below.

Table I. Specifications of simulated water sample.

Parameter	pH	Temperature (°C)	Chlorophyll a ($\mu g/m^3$)	Turbidity (FTU)	Color (TCU)	TOC (mg/l)
Amount	7.6	21.9	1.6	21.8	19.6	9.5

The required ozone was supplied by the OPW27 Ozone Generator with the nominal capacity of 5 gr/hr) via air supply pump. A 20-liter glass tank (40*25*20 cm) was used for operating the pre-ozonation process, which was divided into 4 cells by 3 baffles (20*20 cm). The ozone was injected into the reactor via four diffusers from the cell bottom with very fine bubbles (Fig. 1.). The pre-ozonation of water samples was conducted based on different scenarios: ozone-water contact times of 5, 10, 15, and 20 minutes; and ozone doses of 1.0 up to 5.0 gram(s) per hour. Finally, the variation of water quality parameters in different scenarios was investigated. The rates (percentage) of removal for each of the variables in each scenario are presented below.

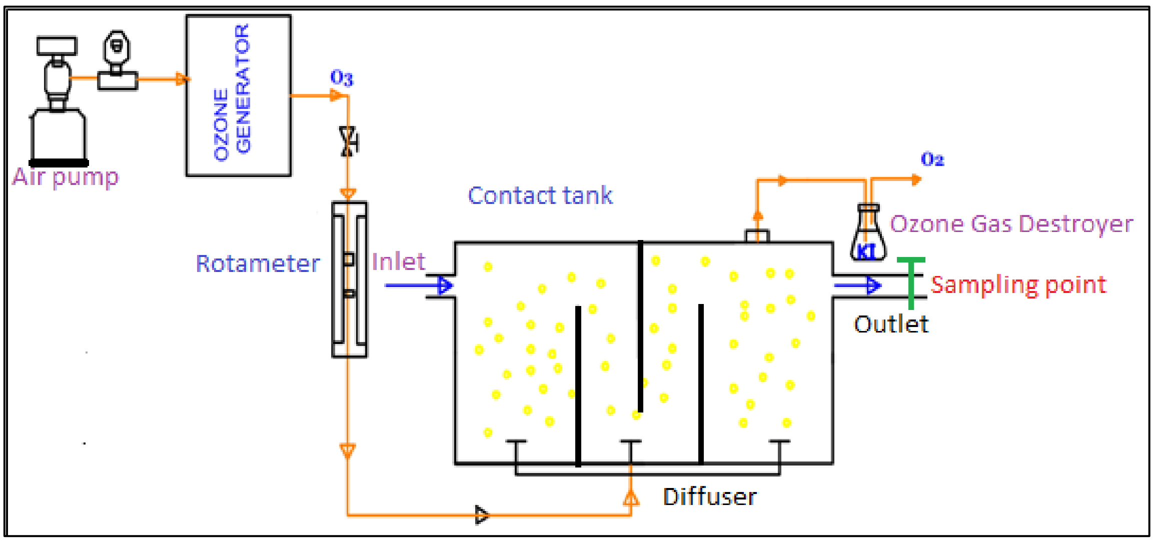

Figure 1. Schematic plan of experimental pre-ozonation reactor.

2. Results

The results for the 4 scenarios tested within the study's framework are presented in Fig. 2. below.

3.1. Scenario 1

The simulated water sample was first ozonized for 5 min at different ozone doses of 1.0-5.0 gr/hr, with the pH ranging from 4.0 to 5.6, and approximate temperature of 23.9°C. Fig. 2A. shows the corresponding changing trend in the quality parameters of the ozonized water sample; and Table 2. shows the elimination rate for the variables. Based on the results, the removal rate for turbidity, TOC, color, and chlorophyll a were 8.3, 0.0, 3.06 and 34.5%, respectively, at 5-min contact time, 1.0 gr/hr ozone dose, pH 4.0 (acidic), and 24.35°C temperature. Increasing the ozone dosage to 5.0 gr/hr, pH to 5.6, temperature to 23.8°C, and contact time by 5 min, raised the removal rate for the parameters of turbidity, TOC, color, and chlorophyll a up to 17.4, 2.1, 7.7, and 49.1%, respectively. Therefore, the alteration of water temperature, pH, ozone dosage and ozone-water contact time had a positive impact on the ozonation.

3.2. Scenario 2

Fig. 2B. shows the change trend in the quality of the ozonized water sample at 10 min ozone-water contact time and different ozone dosages. As shown, with the increasing of the contact time from 5 to 10 minutes at pH 5.6, ozone dosage of 1.0 gr/hr and 23.79°C, the elimination rate for the parameters of turbidity, TOC, color, and chlorophyll a equaled 2.1, 18.3, 9.2 and 49.1%, respectively. By increasing the ozone dose from 1.0 to 5.0 gr/hr at pH 6.8 (acidic), the removal rate for turbidity, TOC, color, chlorophyll a at 23.86°C amounted to 26.6, 6.3, 16.3, and 70.9%, respectively. Generally, reducing temperature and increasing pH along with increasing the ozone dose injected into the system had a positive effect on water ozonation.

3.3. Scenario 3

Fig. 1C. illustrates the trend of changes in the quality of the ozonized water samples at 15 min ozone-water contact time for different ozone doses at pH 6.8, 23.76°C temperature, and the initial ozone dosage of 1gr/hr. The results indicate that the removal rates for the parameters were clearly higher compared to the previous Scenarios 1 and 2, specifically for turbidity, TOC, color, and chlorophyll a they amounted to 26.6, 8.4, 17.9, and 56.4%, respectively. Increasing the ozone dose up to 5.0 gr/hr at pH 8.6 and 23.60°C, the elimination rates came up to 36.7, 14.7, 27.6, and 78.2%, respectively. Generally, reducing temperature and increasing pH along with increasing the ozone dose and contact time had a positive impact on the water ozonation process.

3.4. Scenario 4

Fig. 2D. displays the trend of quality changes in the sample ozonized at 20 min contact time and different (from 1.0 to 5.0 gr/hr) ozone dosages; and Table 5. indicates the removal rates for the parameters. The sample had alkaline pH and average temperature of 23.6°C. Compared to the prior scenarios, the removal rates for the parameters were considerably higher, specifically, for turbidity, TOC, and color at 5.0 gr/hr ozone dose, alkaline pH (10.6) and temperature of 23.8°C they amounted to 44.07, 32.6, 39.8, and 92.7%, respectively. The temperature was gradually reduced from Scenario 1 to Scenario 4 (from 24.35°C to 23.8°C). Considering the higher removal of the water quality parameters in Scenario 4, it can be argued that the ozone-water contact time as well as temperature change affect the pre-ozonation process. The pH was changed from acidic (4.0) in Scenario 1 to alkaline (10.6) in Scenario 4. The removal percentage for the parameters in Scenario 4 was higher than in the other three scenarios. Therefore, it can be alleged that alkaline environment obviously influenced the pre-ozonation process, as the ozone breakdown rate raised with the increased pH due to the production of radical hydroxyl (•OH). Thus, the most important parameters affecting water ozonation include: ozone injection site, ozone dosage, ozone-water contact time, water quality, temperature, and pH. Increasing the temperature limits the dissolution of ozone in water, and water ozonation does not take place completely. Increasing the pH leading to the production of hydroxyl radicals, water ozonation performs better.

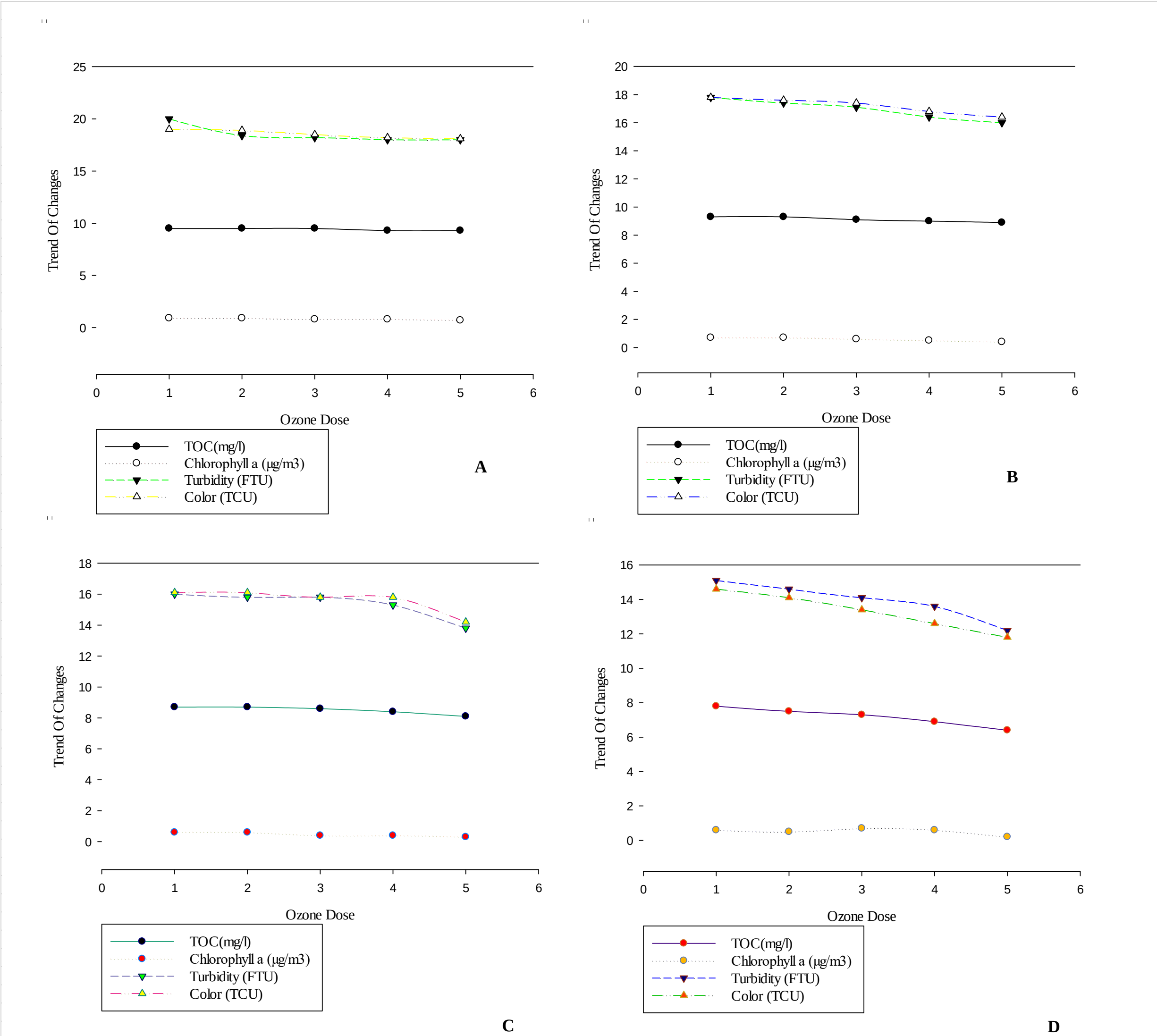

Figure 2. The trends of changing quality parameters of ozonized water at different ozone dosages and ozone contact time with water (A: 5 min, B: 10 min, C: 15 min, D: 20 min).

3.5. Assessment of removal efficiency for water quality parameters

Fig. 3. compares the removal rates for turbidity, color, chlorophyll a and TOC at different ozone dosages (gr/hr) and contact time (Fig. 3A: contact time = 5 min, pH = 4.0-5.6, T = 24.35-23.8°C), (Fig. 3B: contact time = 10 min, pH = 5.6-6.8, T = 23.86-23.88°C), (Fig. 3C.: contact time = 15 min, pH = 6.8-8.6, T = 23.86-23.89°C), (Fig. 3D: contact time = 20 min, pH = 6.8-8.6, T = 24.35-23.8°C).

Fig. 3A. indicates the removal rates for the parameters at 5 min contact time, different ozone doses (1.0 to 5.0 gr/hr), pH levels and temperatures. The mean removal rates for turbidity, TOC, color and chlorophyll a were 15.04, 0.84, 5.4, and 40.4%, respectively. Also, it was revealed that increasing the ozone dose up to 5.0 gr/hr, ozone-water contact time up to 20 min, pH up to 8.6, and temperature up to 23.8°C demonstrated the best efficiency in removing the variables.

Fig. 3B. indicates the removal rate for the parameters at 10 min contact time, different ozone dosages (1.0 to 5.0 gr/hr), pH levels and temperatures. The mean removal rates for turbidity, TOC, color and chlorophyll a amounted to 22.28, 4.0, 12.3 and 57.8%, respectively.

Fig. 3C. shows the removal rates for the variables at 15 min contact time, different ozone dosages (1.0 to 5.0 gr/hr), pH levels and temperatures. The mean removal rates for turbidity, TOC, color, and chlorophyll a were calculated at 29.62, 10.52, 20.44 and 66.56%, respectively.

Fig. 3D. indicates the removal rates for the parameters at 20 min contact time, ozone dosages of 5gr/hr, different pH levels and temperatures. The mean removal rates for turbidity, TOC, color, and chlorophyll a amounted to 36.13, 24.4, 32.13 and 79.6%, respectively.

Also, the results show that the removal rates increased with decreasing temperature. The pH changed from 4.0 in Scenario 1 (Fig. 3A.) to 8.6 in Scenario 4 (Fig. 3D.). The removal rates for the parameters in Scenario 4 were higher than in the other three scenarios. Thus, it can be alleged that by increasing the ozone-water contact time and temperature, and decreasing the pH, the removal efficiency for the parameters increased. The results likewise indicate that the removal rate for the variables (Fig. 3.) increased along with increasing the injected ozone dose and contact time. The removal of turbidity, TOC, color, chlorophyll a increased significantly due to enhanced contact time from 5 to 20 minutes. Therefore, the duration of ozonation, ozone dosage and concentration constitute the most important factors in ozone disinfection performance. Further, the results indicate that increasing ozone dosage and pH from acidic to alkaline, combined with reducing temperature, positively affect the ozonation of potable water. Thus, ozone decomposition is a function of pH and is directly related to changes in water temperature.

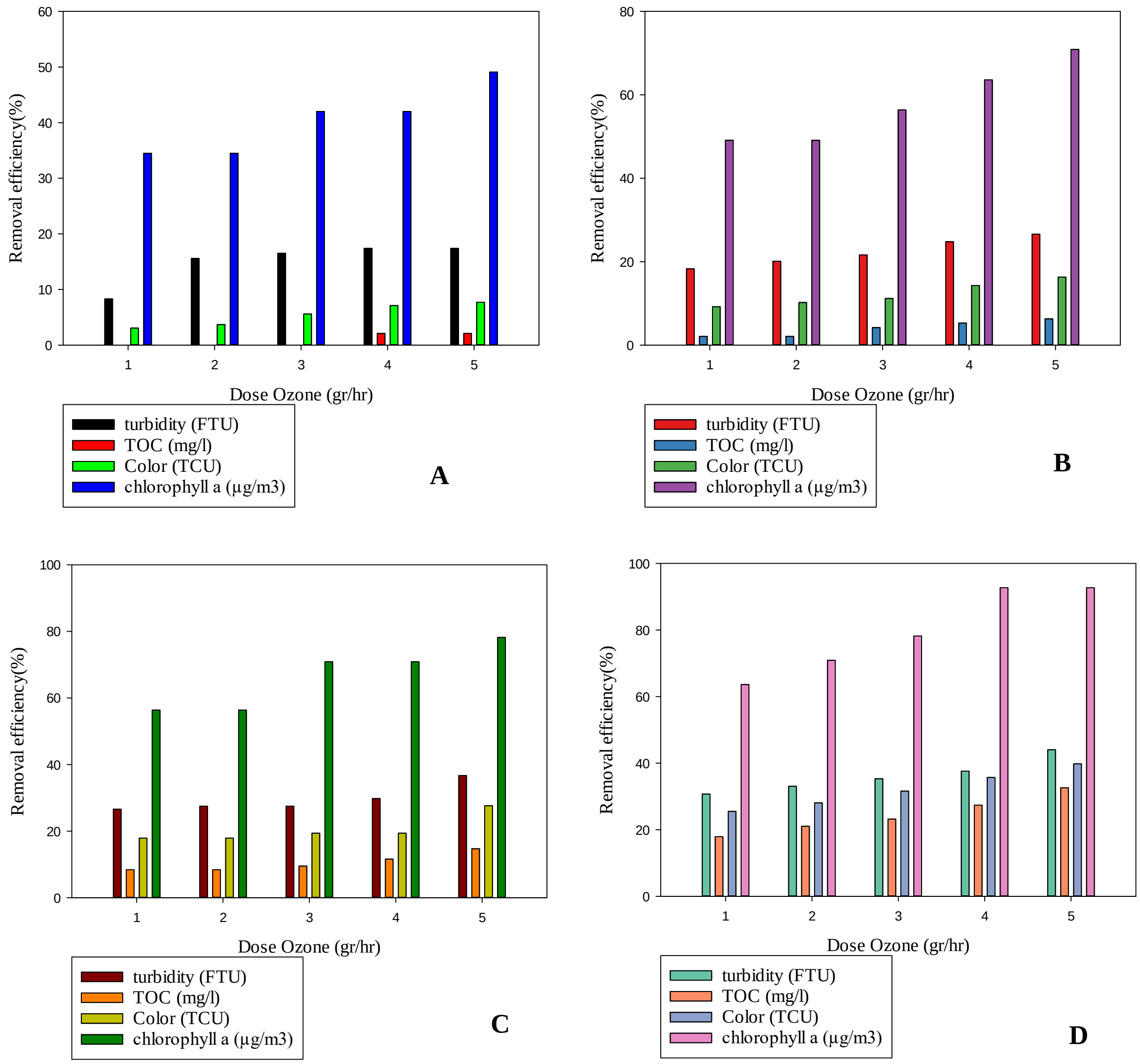

Figure 3. Comparison of the removal rates for turbidity, color, chlorophyll a and TOC at different ozone doses (gr/hr) and contact times (Fig. 3 A: contact time = 5 min, pH = 4.0-5.6, T = 24.35-23.8°C; Fig. 3 B: contact time = 10 min, pH = 5.6-6.8, T = 23.86-23.88°C; Fig. 3 C: contact time = 15 min, pH = 6.8-8.6, T = 23.86-23.89°C; Fig. 3 D: contact time = 20 min, pH = 6.8-8.6, T = 24.35-23.8°C).

4. Discussion

This pilot study aimed to investigate the effects of pre-ozonation on the drinking water quality parameters under different conditions. The research has shown that such parameters as temperature, pH, ozone dosage, and ozone-water contact time represent the most effective factors in water ozonation. Specifically, temperature and pH affected the rate of ozone decomposition in water – increasing the temperature reduced the rate of ozone decomposition, while the rate of ozone decomposition grew with increasing the pH. Temperature changes corresponded to 4 different scenarios (from 23.8 to 24.35°C), and the pH was increased (from 4.0 to 8.6). Kevin et al. (2013) and Ershov et al. (2009) showed that ozone decomposition and solubility in water are a function of pH and temperature. With the increasing pH, the formation speed of hydroxyl radicals (˚OH) also grew leading to more efficient water ozonation. The pH during Scenario 1 was 4.0, and was increased up to 8.6 during Scenario 4, which led to the highest percentage of removal for the variables in question (turbidity, color, chlorophyll a, and TOC).

The ozone decay rate increased with the increasing ˚OH, and as a result the effect of ozone on other elements also increased. Thus, the study results indicate that different ozone dosages are required for different pH levels. The decay of pollutants during pre-ozonation can take the direct oxidation pathway using ozone molecules, as well as the indirect oxidation pathway using ozone radicals – indirect oxidizing of a system due to the presence of ˚OH radicals. The oxidation power of this radical is higher compared to other ozone molecules. More radicals emerge with the increasing ozone decomposition rate due to alkaline pH. Thus, the ozone decay rate and the reaction of ozone with other elements also grows at higher pH (Gardoni et al., 2012).

Melicia et al. (2018) showed the efficiency of temperature and pH parameters during ozone decomposition in water. Also, the results of the scenarios under this study demonstrate that ozone dosage and contact time with water represent the main factors governing water ozonation. Liu et al. (2006) pointed to O_3 dosage as one of the major factors influencing its performance on coagulation, and bringing down color, turbidity, and TOC. Other studies have shown that the efficiency of eliminating parameters in an ozonation system grew with increasing the dose and contact time. Previous water treatment research has shown that ozone can reduce color, turbidity, total organic carbon depending on the dose of ozone applied (Liu et al., 2006).

The removal efficiency for the parameters (turbidity, color, chlorophyll a, and TOC) during ozonation at 5, 10, 15 and 20 min contact time (ozone dosage of 1.0 to 5.0 gr/hr) observed during the study are as follows. The removal efficiency for turbidity amounted to 15.0, 22.0, 30.0 and 36.2%; for color – 5.0, 12.0, 2.5 and 31.8%, respectively. The removal efficiency for chlorophyll a were 40.5, 58.0, 67.0 and 80.0%; and for TOC – 0.85, 4.2, 11.0 and 25%, respectively. The study results show that with the increasing contact time and ozone dosage injected into the system, the removal rates for the parameters in question also grow. During ozonation, chlorophyll a showed the highest removal rate at 20 minutes and ozone dosage of 5.0 gr/hr. The TOC removal rate was lower than for the other parameters, and at 20 min contact time and 5.0 gr/hr ozone doze equaled 25.0%. In the optimal state of the ozonation process (Scenario 4), the highest percentage of turbidity and color removal amounted to 36.2 and 31.8%.

Ghadimkhani et al. (2006) showed that pre-ozonation had a stable effect on surface water treatment with low impact on TOC and very low molecular weight of humic substances. Mamba et al. (2009) showed that the percentage of TOC reduction in water sources after ozonation amounted to approx. 28%. Also, Carlos Amor et al. (2019) showed that the percentage reduction of TOC was about 36% by an advanced oxidation process. In the course of water ozonation, ozone can reduce color by breaking down polymers and organic matter in water (Melin et al., 2000). The findings of this study demonstrate that ozone had the highest impact on such water quality parameters as chlorophyll a, turbidity, color and TOC (corresponding to later scenarios) with the lowest impact on TOC. Whereas Morrison and et al. (2012) showed that medial ozonation had a positive impact on chlorophyll a, Geldenhuys et al. (2000) stated that ozone alone had no effect on the removal of chlorophyll-a. Table 2. compared the results of this pilot study with the findings of similar thematic studies.

Table II. Comparison of results of this study with other target studies.

Row	Author(s) name	year	Agree	Disagree
1	Mahmoudi et al.	2015	*	
2	Melicia et al.	2018	*	
3	Jung et al.	2017	*	*
4	Khadre et al.	2001	*	
5	Gardoni et al.	2012	*	
6	Kim et al.	2002	*	
7	Zhang, J.	2014	*	*
8	Spiliotopoulou	2018	*	
9	Ashfaq, S.M.	2017	*	
10	Athanasios et al	2016	*	
11	Tercero Espinoza,	2009	*	
12	Samios, S.	2007	*	
14	Kevin et al.	2013	*	
15	Ershov et al.	2009	*	
16	Liu et al.	2006	*	
17	Ghadimkhani et al.	2006	*	
18	Mamba et al.	2009	*	
19	Carlos Amor et al.	2019	*	
20	Morrison et al.	2012	*	
21	Geldenhuys et al.	2000	*	

5. Conclusion

The study results show that the quantity of ozone to remove organic compounds, color, and turbidity depends on the input water quality. As per the results of this pilot study, water parameters got increasingly removed by increasing the dose of ozone injected into the reactor (from 1.0 to 5.0 gr/hr) corresponding to 4 different scenarios; with the 20 min ozone-water contact time and ozone dosages of 5.0 gr/hr demonstrating the highest percentage of removing the parameters at pH 8.6.

The analysis of the study outcomes points to the greatest impact of ozone on water quality parameters: foremost on chlorophyll a, turbidity, and color, and TOC at later stages, with the least impact on TOC. The study results indicate that different ozone doses are required for different pH levels. The pH in Scenario 1 was 4.0, and was increased up to 8.6 in subsequent scenarios, with the highest removal efficiency occurring in Scenario 4. Likewise, the study results point to •OH as the key initiator of ozone decay. Ozone dissolves in aqueous solutions with pH below 7.0; otherwise it cannot react with water and exists in the O3 molecular form. As pH increases, decomposition takes place automatically and consequently different reactive free radicals such as •OH emerge. The results indicate that the highly efficient removal of the variables in question occurred at alkaline pH (8.6), which corresponds to the findings of other similar research. Also, ozone-water contact time plays a major role in water disinfection with oxidizing materials like ozone. With the longer contact time, the amount of dissolved ozone in water grows enhancing the removal rate for variables. Thus, while designing ozonation tanks the contact time should be considered one of the basic design parameters. To ensure proper contact time, ozonation contactors should be fitted with baffles.

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