

The Effect of Ozonation, Coagulation and Adsorption on Natural Organic Matter Removal

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ABSTRACT

For the treatment of surface water, the coagulation process was used with highly alkaline polyaluminium chloride PAX-XL19F and coagulation supported by ozonation and adsorption on powdered activated carbon CWZ-30 for reducing the level of surface water pollution with organic substances. In addition to the typical indices used to assess the content of organic compounds (total organic carbon TOC, oxidisability OXI, ultraviolet absorbance UV_{254}), the study also evaluated colour, turbidity, and the potential of trihalomethanes formation THM-PF. Reduction in the content of TOC in water after coagulation ranged from 22 to 24%, OXI - in the range of 34–36%, and UV_{254} absorbance - from 52–55%. The turbidity and colour of the water was reduced by 70–73% and 56–60%, respectively. The use of preliminary ozonation and activated carbon-assisted coagulation increased the efficiency of water treatment. Changes in the values of TOC, OXI, UV_{254} absorbance, turbidity and colour were 28–33%, 45–46%, 69–73%, 72–79% and 89–100%, respectively. In the water purified by coagulation and then chlorinated, THM-PF was 37–38% lower than in untreated surface water. The use of additional pre-ozonation and activated carbon during coagulation increased the reduction of THM-PF by 9–12%.

Keywords: ozone, activated carbon, polyaluminium chloride, trihalomethane precursors.

INTRODUCTION

Strong oxidants such as chlorine, chlorine dioxide and ozone have been used in the process of water disinfection. Their use can lead to the formation of compounds called disinfection by-products (DBPs). The safest way to prevent the formation of DBPs is to remove their organic and inorganic precursors from water before the contact with a chemical oxidant. Trihalomethanes (THMs) and haloacetic acids (HAA) are the two main classes of DBPs that can be present in the waters disinfected with chlorine [Tang et al. 2020]. The permissible concentration of THMs in drinking water, which include $CHCl_3$, $CHBr_3$, $CHCl_2Br$, $CHClBr_2$, is 100 $\mu\text{g/L}$ [Council Directive 1998, Regulation 2017]. In modern water treatment technology, the processes that prevent the formation of trihalomethanes are more important than the processes that lead to the removal

of the already formed THMs [Nawrocki, 2010]. Preventing the formation of THMs means the removal of the natural organic matter (NOM) from water, especially humic and fulvic acids.

Treatment processes, such as coagulation and adsorption can be employed to reduce the NOM content in water. In recent years, the use of pre-hydrolysed salts such as polyaluminium and polyiron(II) chlorides and sulphates for coagulation has become more and more popular. Pre-hydrolysed coagulants, unlike non-hydrolysed ones, have hydroxyl groups, which give them higher basicity. The studies on water treatment with different composition have demonstrated the effectiveness of the coagulation process in the removal of NOM with the use of pre-hydrolysed salts [Sillanpää et al., 2018; Zhang et al., 2018]. The coagulation process is generally effective in removing high molecular weight organics, but less so in removing smaller

molecular weight fractions. Furthermore, the simultaneous use of coagulation and adsorption can also be effective in removing organic substances. According to Zainudin et al. [2018], activated carbon is effective in both minimising odorants and acts as an effective adsorbent to eliminate the THM precursors. The location for the introduction of the powdery activated carbon (PAC) can be a raw water pipeline or a quick stirring tank, even before the application of the coagulant. This results in long contact time between PAC and water. PAC can also be applied with a delay in relation to the coagulant, which prevents carbon build-up in the formed agglomerates. The coagulation process is often preceded by oxidation, e.g. through the introduction of ozone [Yan et al., 2007; Tubić et al., 2016; de Vera et al., 2019]. Pre-ozonation has been demonstrated to facilitate coagulation for decreasing the coagulant dose, destabilizing the aggregation of particles and increasing the length of filter runs [Jin et al., 2017]. Ozone used in small doses may act as an agent that destabilizes NOM present in a colloidal form. Ozonation may also cause the fractionation of compounds difficult to adsorb to the forms susceptible to adsorption and/or biodegradation. The most important effect of ozonation is the growth of biodegradable organic matter [Zainudin et al., 2018]. Ozonation has been reported to be an effective treatment for THM reduction in drinking water, even without a significant decrease in the initial total organic carbon [Sadrnourmohammadi et al., 2020].

This study investigated the effect of pre-ozonation process, coagulation with usage polyaluminium chloride and adsorption on powdered activated carbon in reducing the level of surface water contamination. The effectiveness of these processes was evaluated by measurement of turbidity, colour, total and dissolved organic carbon, oxidisability, ultraviolet absorbance, and trihalomethane formation potential.

MATERIAL AND METHODS

The surface water collected in April and November 2020 from the Stradomka River in Częstochowa (Poland) was used in the study. The ozonation, coagulation and adsorption processes were performed in laboratory settings. High-alkaline polyaluminium chloride PAX-XL19F (produced by the Kemipol company) was used

as a coagulant. It is characterized by basicity of 85% and content of Al_2O_3 16%. Powdered activated carbon with the commercial name CWZ-30 (produced by the company Gryfskand) was used to support coagulation. The specific surface area of the activated carbon was of $1134 \text{ m}^2/\text{g}$. Coagulant and powdered activated carbon were selected on the basis of the previously conducted studies [Dąbrowska, 2018; Dąbrowska, 2019].

In the first stage, the water ozonation process was carried out. Bottles with a capacity of 2 L were used as reactors. High purity oxygen was supplied with a flow rate of 1.5 L per minute to an ozone laboratory generator (Model L20 SPALAB). The ozonation time was 3, 6, 10 minutes. The ozone dose introduced into the water within 1 minute amounted to $1.2 \text{ mgO}_3/\text{L}$. The unreacted ozone in the off-gas was discharged into the potassium iodide solution.

In the second stage, the coagulation process was carried out for the coagulant doses: 2; 3; 4 mg Al/L, using fast and slow mixing and sedimentation (the course described in the third stage). The adsorption process was carried out in glass beakers with a capacity of 1 L, into which 0.5 L of the analysed water was measured and powdered activated carbon was introduced in a form of suspension in doses of 15; 30; 45 mg/L. The samples were mixed with a mechanical stirrer for 15 minutes at 500 rpm, and then filtered through filter papers.

The third stage was carried out twice. The following processes were used for water purification:

- 1) coagulation (PAX-19),
- 2) adsorption-assisted coagulation (PAX-19 + CWZ-30),
- 3) coagulation preceded by ozonation (O_3 + PAX-19),
- 4) coagulation preceded by ozonation and enhanced by adsorption (O_3 + PAX-19 + CWZ-30).

This stage was started by water ozonation (time 5 minutes) as described in the first stage. Then, 1.5 L of non-ozonised water was measured into two glass beakers with a capacity of 2 L, and 1.5 L of water after the ozonation process into the next two. The coagulant solution was introduced at a dose of 3 mgAl/L to the water in all beakers and fast stirring was performed for 2 minutes (applying 300 rpm). Powdered activated carbon in a form of suspension was then introduced into the second and fourth beakers. The carbon dose was equal to 30 mg/L of water and was adopted based on the research results from the second stage.

Quick stirring was continued for 2 minutes, followed by slow stirring for 15 minutes (30 rpm). The samples were subjected to 1 hour sedimentation. Afterwards, 0.3 L of water was decanted from each beaker and analysed.

The quality indicators of raw and purified water were determined using the following methods: pH - potentiometric, turbidity – nephelometric, colour – colorimetrically with platinum-cobalt scale, oxidisability OXI – by potassium permanganate, total and dissolved organic carbon TOC and DOC – by infrared spectrophotometry (Vario Cube organic carbon analyser), UV absorbance at 254 nm wave length (UV 5600 spectrophotometer).

For the determination of the THM-PF trihalomethane formation potential, 50 mL water samples before and after treatment were chlorinated using the chlorine water prepared from sodium hypochlorite. Such dose was used to achieve, after a period of 24 hours the concentration of residual free chlorine in the range of 3 to 5 mgCl₂/L [Standard methods 1998]. Then, 10 mL of water after chlorination was taken into

test tubes, 1 mL of n-pentane was added and it was intensively shaken for 2 minutes. After separation of the layers, the compounds were analysed by gas chromatography with detector ECD (chromatograph Agilent 6890 N).

The colour was determined in two repetitions, whereas the remaining determinations were made in three repetitions.

RESULTS AND DISCUSSION

Surface water was characterized by turbidity of 5.1–7.7 NTU and colour of 30–45 mgPt/L. TOC content was 7.8–10.5 mgC/L, DOC was 6.6–9.7 mgC/L and OXI was 5.6–7.5 mgO₂/L. Absorbance UV₂₅₄ varied from 17.2 to 22.9 1/m. The water pH in April was slightly acidic, whereas in November it was slightly alkaline.

The results of the effect of ozone, coagulant PAX-XL19F or powdered activated carbon CWZ-30 dose on the change of selected parameters are presented in Table 1.

Table 1. Mean values of colour, oxidisability, total and dissolved organic carbon and UV₂₅₄ absorbance of surface water and water after ozonation or coagulation or adsorption process

Parameter	Unit	Surface water	Water after ozonation			Water after coagulation using PAX-19			Water after adsorption using CWZ-30		
			3 min	6 min	10 min	2 mg/L	3 mg/L	4 mg/L	15 mg/L	30 mg/L	45 mg/L
Colour	mgPt/L	45.0	35.0	28.0	22.0	25.0	18.0	15.0	30.0	25.0	20.0
OXI	mgO ₂ /L	7.5	7.1	6.4	6.1	5.3	4.3	3.9	7.1	6.8	6.6
TOC	mgC/L	10.5	10.1	9.7	9.4	7.8	7.1	6.7	9.7	9.2	8.8
DOC	mgC/L	9.7	9.5	9.3	9.1	7.3	6.7	6.4	9.2	8.9	8.4
UV ₂₅₄	1/m	22.9	18.9	16.1	14.8	13.4	10.9	9.8	17.6	17.0	16.7

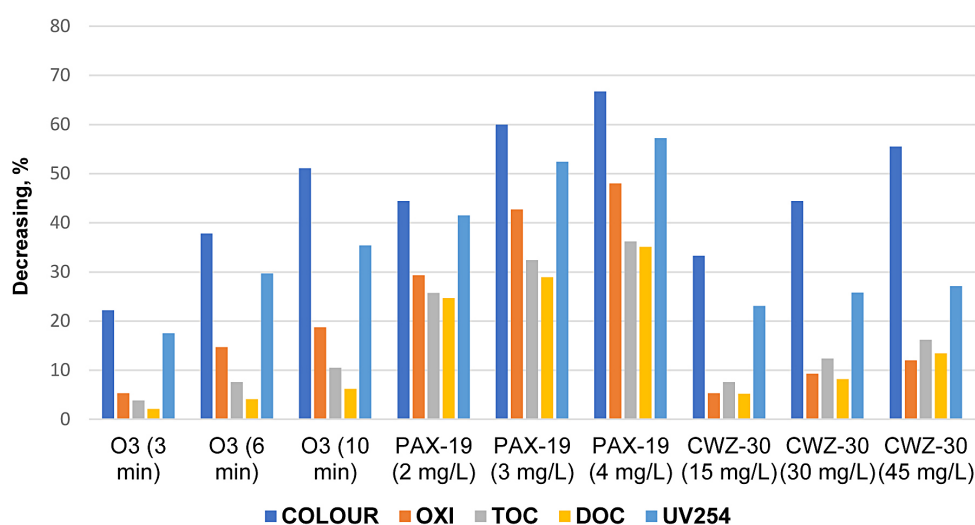


Figure 1. Efficiency of colour, total and dissolved organic carbon removal and decreasing oxidisability, UV₂₅₄ absorbance value during the ozonation or coagulation or adsorption process

With the application of ozonation, depending on the dose the colour removal was reported by 22 to 51% (Fig. 1). The value of the OXI index decreased by 5–19%. Slight changes in the content of TOC and DOC by 2–10% were found. The UV_{254} absorbance value decreased by 17 to 35%. A significant reduction of the UV_{254} absorbance proves that the qualitative composition of organic compounds, the organic carbon fraction characterised by a high content of aromatic components, is changing. An additional advantage of applying ozone oxidation, resulting from the decomposition of humic substances, is the reduction of the colour intensity of the water subjected to ozonation.

It was found that the water purification effect increased along with the dose of coagulant PAX-XL19F and powdered activated carbon CWZ-30. For the highest coagulant dose of 4 mgAl/L, 36; 34; 48% removal of TOC, DOC and OXI was obtained, while by 16; 13 and 12% during adsorption on the powdered carbon (Fig. 1). The colour removal for the highest dose of coagulant and

powdered activated carbon was equal to 67 and 56%.

The average values of turbidity and colour of water, OXI, TOC, DOC and UV_{254} absorbance in surface and treated water are presented in Tables 2 and 3. Comparison of the percentage reduction of the analysed parameters in the treated water is presented in Figures 2 and 3.

Using pre-hydrolysed polyaluminium chloride PAX-XL19F in the dose of 3 mgAl/L for coagulation, turbidity was removed by 70 and 73%. The colour reduction was 56–60%. Supporting the coagulation with adsorption with the use of activated carbon CWZ-30 caused the colour reduction by an additional 7–11%, and enhancing coagulation with ozonation by 22–23%.

The content of organic compounds after the coagulation process, determined as OXI and TOC, decreased by 34–36% and 22–24%, respectively. The use of pre-ozonation resulted in a further reduction of OXI and TOC by another 7–9% and 4–5%, respectively. Matilainen et

Table 2. Mean values of turbidity, colour, oxidisability, total and dissolved organic carbon and UV_{254} absorbance of surface water collected in April and water after ozonation, coagulation and adsorption processes

Parameter	Unit	Surface water	Treated water using			
			PAX-19	O ₃ + PAX-19	PAX-19 + + CWZ-30	O ₃ + PAX-19 + + CWZ-30
pH	-	6.87	6.73	6.84	6.80	6.89
Turbidity	NTU	7.72	2.05	1.55	1.92	1.63
Colour	mgPt/L	45.0	20.0	10.0	15.0	5
OXI	mgO ₂ /L	6.9	4.4	3.9	4.3	3.8
TOC	mgC/L	8.5	6.5	6.1	6.3	5.7
DOC	mgC/L	7.6	6.0	5.5	5.8	5.3
UV_{254}	1/m	21.9	9.8	6.6	7.5	5.9
SUVA	L/mg·m	2.88	1.63	1.29	1.20	1.11

$$SUVA = UV_{254} / DOC$$

Table 3. Mean values of turbidity, colour, oxidisability, total and dissolved organic carbon and UV_{254} absorbance of surface water collected in November and water after ozonation, coagulation and adsorption processes

Parameter	Unit	Surface water	Treated water using			
			PAX-19	O ₃ + PAX-19	PAX-19 + + CWZ-30	O ₃ + PAX-19 + + CWZ-30
pH	-	7.10	6.68	6.80	6.75	6.86
Turbidity	NTU	5.10	1.54	1.37	1.82	1.42
Colour	mgPt/L	30.0	12.0	5.0	10.0	0
OXI	mgO ₂ /L	5.6	3.7	3.2	3.5	3.0
TOC	mgC/L	7.8	6.1	5.8	6.0	5.6
DOC	mgC/L	6.6	5.9	5.6	5.8	5.2
UV_{254}	1/m	17.2	8.3	6.1	6.8	5.4
SUVA	L/mg·m	2.61	1.46	1.21	1.13	1.06

$$SUVA = UV_{254} / DOC$$

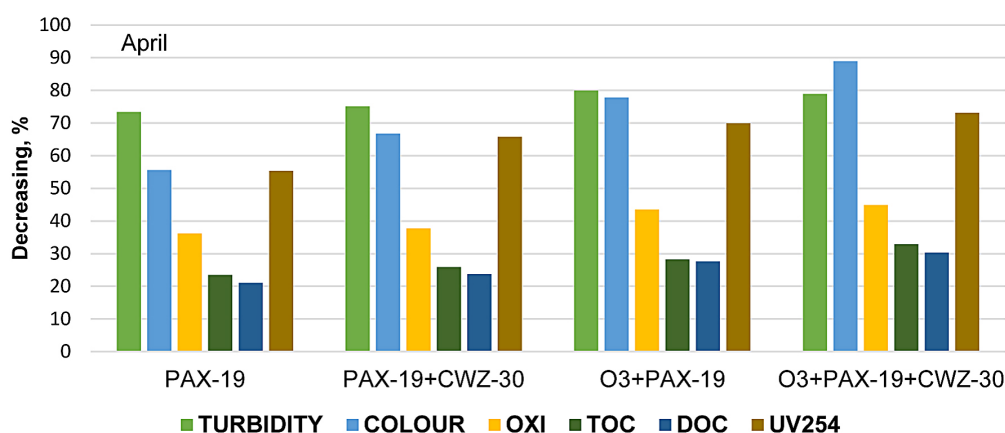


Figure 2. Efficiency of turbidity, colour, OXI, TOC, DOC removal and decreasing UV_{254} absorbance value in April during ozonation, coagulation and adsorption processes (ozonation time – 5 min, dose of PAX – 3 mgAl/L, dose of CWZ – 30 mg/L)

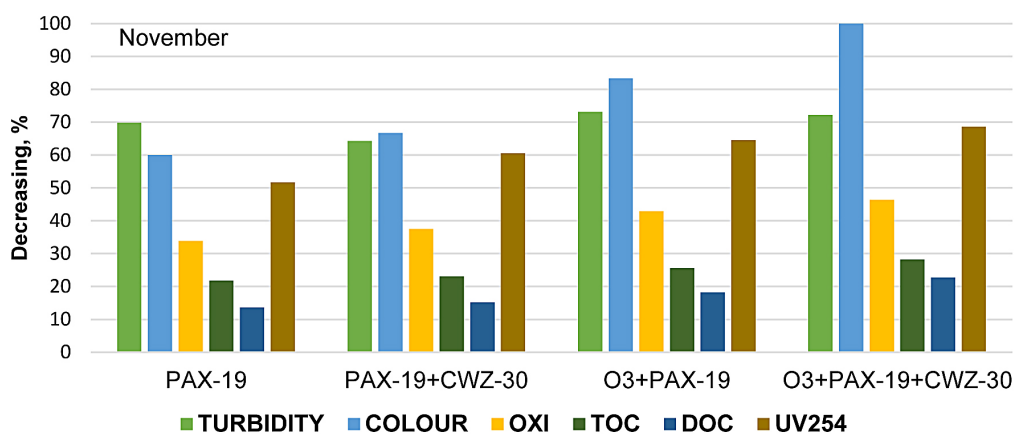


Figure 3. Efficiency of turbidity, colour, OXI, TOC, DOC removal and decreasing UV_{254} absorbance value in November during ozonation, coagulation and adsorption processes (ozonation time – 5 min, dose of PAX – 3 mgAl/L, dose of CWZ – 30 mg/L)

al. [2010] showed that the reduction in DOC content in the coagulation process can range between 25 and 67%, whereas the UV_{254} absorbance values range between 44 and 77%. Compared to DOC, the UV_{254} absorbance has been noted to reduce more, suggesting that aromatic materials are removed more effectively than other NOM fractions.

The decrease in the value of UV_{254} absorbance in the pre-ozonation process and coagulation amounted to 65–70%. Reactions of ozone with humic substances (fulvic acids are predominant and more reactive than humic acids) under neutral pH conditions lead to a slight decrease in the TOC content, but a significant decrease in the value of UV_{254} absorbance, a decrease in the amount of fractions with high molecular weights as well as an increase in fractions with lower molecular weights and a significant increase in the number of carboxyl groups in the molecules of humic substances.

It is difficult to explain the small effect of the adsorption process without additional information on the type of organic substances present in analysed water.

The use of pre-ozonation and supporting the coagulation with powdered activated carbon CWZ-30 improved the efficiency of water purification. The reductions in OXI, UV_{254} absorbance, and TOC and DOC contents in pre-ozonated water which was then treated in the coagulation and adsorption processes were 45–46%, 69–73%, 28–33% and 23–30%, respectively. Water turbidity and colour decreased by 72–79% and 89–100%, respectively. Therefore, the greatest increase in the efficiency of water purification was observed in relation to the decrease in colour and the UV_{254} absorbance index. The additional reduction in the values of these indices compared to the effects of water purification

only in the coagulation process was 33–40% for the colour and 17–18% for UV₂₅₄ absorbance.

While conducting research, Wang et al. [2017] obtained a reduction in DOC and UV₂₅₄ absorbance by 45 and 51% in the coagulation process with polyaluminium chloride, and in coagulation and subsequent adsorption with PAC by 76 and 81%, respectively. In turn, in coagulation preceded by ozonation and ended with adsorption, the reduction of DOC and UV₂₅₄ absorbance amounted to 85 and 91%. These results were significantly better than those presented in this study (especially with regard to adsorption; however, carbon was not introduced during coagulation, but afterwards, as a separate stage of water purification).

No trihalomethanes were found in the collected surface water and in the water after ozonation, coagulation and adsorption. However, their presence was demonstrated in the waters after the chlorination process. The concentrations of THMs and THM-PF values in chlorinated surface water and treated water are shown in Table 4 and percentage decreases in Figure 4.

In the tested samples of surface water after the chlorination process, the presence of trihalomethanes CHCl₃, CHCl₂Br, CHClBr₂ was found, whereas no CHBr₃ was observed. Depending on the month of collecting water, the CHCl₃ concentration ranged from 152 to 338 µg/L, whereas the concentrations of CHCl₂Br and CHClBr₂ were 19–27 and 1–9 µg/L, respectively. Higher values were obtained for the water collected in April. The concentration of CHCl₃ was 88–90% of the total THM content in the chlorinated waters.

The concentration of CHCl₃ in the chlorinated water after treatment in the coagulation process ranged from 88 to 208 µg/L, whereas in the water after pre-ozonation, coagulation and adsorption, this value ranged from 72 to 170 µg/L. These values were by 50–53% lower than the concentrations of CHCl₃ recorded in untreated surface water subjected to chlorination. This proves the high efficiency of removing organic components of water, which are precursors of the by-products of chlorination

Table 4. Concentrations of THMs and THM-PF values in surface water and treated water after 24 hours of contact with chlorine

THM	Unit	Surface water (April)	Treated		Source water (November)	Treated	
			PAX-19	O ₃ + PAX + CWZ		PAX-19	O ₃ + PAX + CWZ
CHCl ₃	µg/L	338.0	208.0	170.0	152.0	88.0	72.0
CHCl ₂ Br	µg/L	27.1	24.5	23.2	19.3	19.0	19.1
CHClBr ₂	µg/L	8.6	7.1	4.6	1.2	1.8	4.7
CHBr ₃	µg/L	nd	nd	nd	nd	nd	nd
THM-PF	µgCHCl ₃ /L	362.6	229.9	185.9	166.8	102.8	88.6

nd – not detected.

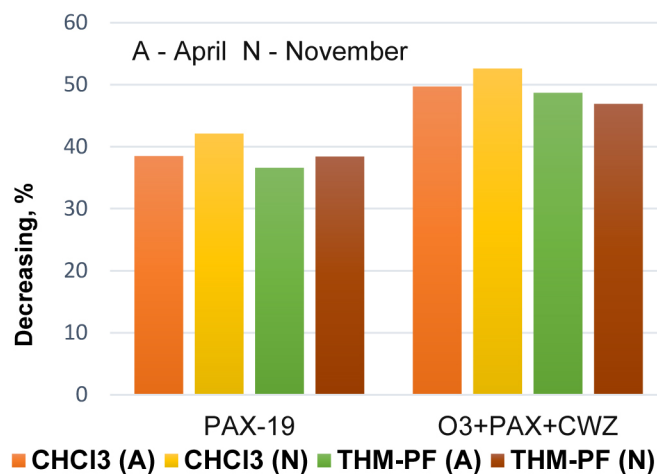


Figure 4. Decreasing of trichloromethane concentration and of trihalomethanes formation potential in waters treated by ozonation (O₃)-coagulation (PAX)-adsorption (CWZ)

(THMs) in the process of coagulation, as well as the opportunities for increasing this efficiency through the processes of ozonation and adsorption on CWZ-30. The concentration of CHCl_2Br in treated waters after chlorination ranged from 19 to 25 $\mu\text{g/L}$. The CHClBr_2 content was at a level of 2 to 7 $\mu\text{g/L}$.

In the water purified by coagulation and subjected to chlorination for 24 hours, the potential for THM formation was 37–38% lower than the values obtained in chlorinated surface water. Application of additional ozonation and adsorption processes resulted in a further reduction in the THM-PF value by 9–12%. The reduction of THM-PF value during water treatment by means of pre-ozonation and coagulation assisted with powdered activated carbon was confirmed by other studies [Wang et al., 2017; Zainudin et al., 2018].

The total concentration of trihalomethanes and the formation of individual THMs in chlorinated water depends on the composition of raw water, chlorine dose, contact time, operating parameters and the presence of residual chlorine in the distribution system [Golea et al., 2017; Niu et al., 2017; Padhi et al., 2019]. The results of research on the reduction of THM-PF value during water treatment by means of ozonation and coagulation are presented, among others, in publications [Chiang et al., 2009; Sadrnourmohamadi and Gorczyca 2015]. The authors achieved the efficiency of reducing THM-PF by 55–60%. They found that an increase in ozone dose caused a decrease in DOC (2–26%) and THM-PF (4–58%).

Ozonation has been reported to be an effective treatment for THM reduction in drinking water, even without a significant decrease in the initial TOC. In these cases, ozone transforms high molecular weight organic compounds into smaller ones, which are resistant to the reactions involving chlorine as a disinfectant [Sadrnourmohammadi et al., 2020].

On the other hand, Wang et al. [2017], using coagulation and adsorption on PAC, obtained a reduction of THM-PF by 70% and no improvement in efficiency in the case of introducing additional preliminary ozonation.

According to Deedomwongsa et al. [2017] pre-treatment of water with ozonation leads to the oxidation of NOM to hydrophilic organic substances with lower molecular weight. In the samples ozonated with doses ranging from 1–5 mgO_3/L , aromatic compounds, measured by UV absorbance, were removed in 23–60%, but the THM-PF value reduction was only 10%.

CONCLUSIONS

The experiment showed the effect of the dose of coagulant, ozone and activated carbon on the effectiveness of removing colour, total organic carbon, oxidisability of water and UV_{254} absorbance. The use of pre-ozonation process and coagulation with polyaluminium chloride supported by powdered activated carbon allowed for a reduction by TOC, OXI, UV_{254} absorbance by 28–33%, 45–46%, 69–73%, respectively. Compared to the effects of the coagulation as a stand-alone process, this was an increase of 6–9%, 9–13%, and 17–18%, for TOC, OXI and UV_{254} absorbance, respectively.

The study confirmed the usefulness of the coagulation process in the removal of organic matter, which represents a precursor for the formation of trihalomethanes. The potential for THMs formation in treated water in the coagulation process was by 37–38% lower than in the untreated surface water after chlorination. The supporting the coagulation with activated carbon and ozonation process resulted in an additional reduction of THM-PF by 9–12%.

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