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Investigation of the impact of ozone pretreatment and powdered activated carbon addition on the removal of natural organic matter by coagulation

Jasmina Agbaba*, Aleksandra Tubić, Božo Dalmacija, Malcolm Watson, Jelena Molnar, Srđan Rončević, Snežana Maletić

Faculty of Sciences, University of Novi Sad, Trg Dositeja Obradovića 3, 21000 Novi Sad, Serbia, Tel. +381 (21) 450039; Fax: +381 (21) 454 065; email: jasmina.agbaba@dh.uns.ac.rs (J. Agbaba), Tel. +381 (21) 4852724; email: aleksandra.tubic@dh.uns.ac.rs (A. Tubić), Tel. +381 (21) 4852720; email: bozo.dalmacija@dh.uns.ac.rs (B. Dalmacija), Tel. +381 (21) 4852798; email: malcolm.watson@dh.uns.ac.rs (M. Watson), Tel. +381 (21) 4852724; email: jelena.molnar@dh.uns.ac.rs (J. Molnar), Tel. +381 (21) 4852728; email: srdjan.roncevic@dh.uns.ac.rs (S. Rončević), Tel. +381 (21) 4852886; email: snezana.maletic@dh.uns.ac.rs (S. Maletić)

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ABSTRACT

The main goal of this study was to investigate the effects of ozone oxidation pretreatment and powdered activated carbon (PAC) addition on the efficacy of natural organic matter (NOM) removal by coagulation. Relatively high iron chloride doses from 100 to 200 mg $FeCl_3$/L were applied, due to the high dissolved organic carbon (DOC) content (10.27 ± 0.49 mg C/L) of the raw water investigated, with PAC used in doses of 5–30 mg/L. Preozonation was carried out at doses of 0.2–1.3 mg O_3/mg DOC. Jar test results indicate that baseline coagulation with the optimal coagulant dose (200 mg $FeCl_3$/L) achieved was 39% DOC and 54% UV_{254} removals. PAC addition improves coagulation efficacy in NOM removal by up to 9% DOC and 25% UV_{254}. The best results were obtained in combination with 0.6 mg O_3/mg DOC, 5 mg/L PAC and 200 mg $FeCl_3$/L (removals of 58% DOC and 72% UV_{254}), and were a significant improvement compared to coagulation alone.

Keywords: Coagulation; Powdered activated carbon; Preozonation; Natural organic matter

1. Introduction

One of the basic problems for a large number of communities in Vojvodina and the most common cause of the unacceptable drinking water quality, is the high content of natural organic matter (NOM). While NOM itself is not of direct concern in drinking water, it affects water quality in several ways. Apart from the aesthetics of colour, taste and odour that accompany NOM, especially after water disinfection with chlorine, there is the potential health hazard of disinfection by-products (DBPs), the deterioration of water quality in distribution systems because of bacterial growth, and the more difficult oxidation of dissolved iron and manganese to insoluble forms [1–4].

It is well known that through the reduction of NOM content before the disinfection process, it is possible to minimise these negative effects. Generally, the most common and economically feasible processes for NOM removal are considered to be coagulation and flocculation using ferrous or alum salts [5]. Through changes to the physical and chemical characteristics of

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*Corresponding author.

the colloid and suspended particles, their aggregation into solid particles is favoured, which can then be removed from water via sedimentation and/or filtration. The most commonly used coagulants are ferric-based coagulants due to their high efficacy in NOM removal [5–8]. However, it is not unusual for the coagulation process to fail to achieve adequate removal, as the efficacy of NOM removal is affected by many factors, including: the nature and characteristics of the NOM, the type and dose of coagulant, mixing speed, pH, ionic strength, temperature, etc. [5–9]. When conventional coagulation processes fail to achieve satisfactory results, it is necessary to modify them with other technologies to improve NOM removal.

In order to improve the efficacy of the coagulation process in NOM removal and DBPs formation control, especially in water which is rich in NOM, before the coagulation process, an oxidative pretreatment with ozone can be applied [10,11]. As well as ozonation, coagulation may also be combined with powdered activated carbon (PAC) addition. Therefore, the effect of these processes must be examined in the context of coagulation/flocculation processes. Despite extensive investigations over the last 30 years, the effects of ozonation on technology-related parameters, e.g. biodegradability and flocculation, are still of interest. The effects of ozone water pretreatment can be roughly classified as direct (direct removal of NOM, removal of colour, taste and odour) or indirect (increased removal of NOM in coagulation, flocculation and direct filtration processes). Studies have shown that the preozonation of water using doses of up to 1.5 mg O_3/mg TOC, directly removes NOM via mineralisation and evaporation and\or stripping. Based on the TOC value, 6–10% NOM removal can thus be achieved, or 45% based on UV_{254} absorbance [3,12].

Considering the complexity of the water matrix which we have investigated previously [3], we decided to include PAC in order to investigate its potential to improve the pre-ozonation/coagulation treatment. PAC may be successfully applied in drinking water treatment for odour and taste control [13], as well as for the removal of different organic contaminants and DBP precursors [14]. PAC can also be an effective adsorbent of low molecular weight organic matter [8] which can show significant potential to form DBP in reaction with chlorine [15]. PAC has the advantage of being an inexpensive material, easily applied, with minimal capital expenditure required for PAC feeding and contacting equipment. PAC is most commonly added in the fast mixing phase of water treatment, together with the coagulation agent, and is then removed by settling during the sedimentation phase [14]. However, the application of PAC in drinking water treatment is influenced by the PAC characteristics, and by the nature of the water being treated [8]. Due to the complexity of the microflocculation and adsorption effects, the specificity of water matrices, and differences in the nature and structure of NOM in water, detailed investigation of the ozone and PAC application conditions is required, in order to define the optimal NOM removal working conditions for each water source.

The aim of this work was to determine the efficiency of combined processes of coagulation, preozonation and/or adsorption onto PAC, in particular for the treatment of NOM rich groundwater from Vojvodina, and to define the working conditions which result in the optimal NOM removal by coagulation combined with oxidative pretreatment and/or adsorption onto PAC.

2. Experimental

The laboratory investigation used groundwater from Central Banat (Province of Vojvodina, Serbia), originating from two water-bearing layers at depths of 40–80 and 100–150 m, which is used for water supply to the town of Zrenjanin. It is sodium-hydrocarbonate type water formed in Pliocene-Pleistocene sediments [16]. The detailed general characteristics of the raw groundwater are given by Molnar et al. [17].

In order to determine the effects of the coagulation process combined with preozonation and PAC adsorption on NOM removal, and define the optimal coagulation working conditions, five series of jar tests were performed (shown in Fig. 1): (1) basic coagulation, (2) coagulation combined with preozonation, (3a) coagulation combined with preceding PAC adsorption (PAC–$FeCl_3$), (3b) coagulation combined simultaneously with PAC adsorption ($FeCl_3$ + PAC) and (4) coagulation combined with a pretreatment of ozonation and PAC adsorption. In order to evaluate the effects of the combined coagulation processes with different pretreatments, the results obtained during series 2, 3 and 4 were compared with the results from series 1. The parameters used to evaluate the changes in NOM content during the above processes were: UV absorption at 254 nm (UV_{254}) and non-purgeable dissolved organic carbon (DOC).

2.1. Raw water characteristics

During this investigation, the general characteristics of the groundwater supplied varied (Table 1), with DOC content 10.27 ± 0.49 mg C/L. As expected, high NOM content is followed by high value of DBPs formation potential such as trihalomethanes (THMFP)

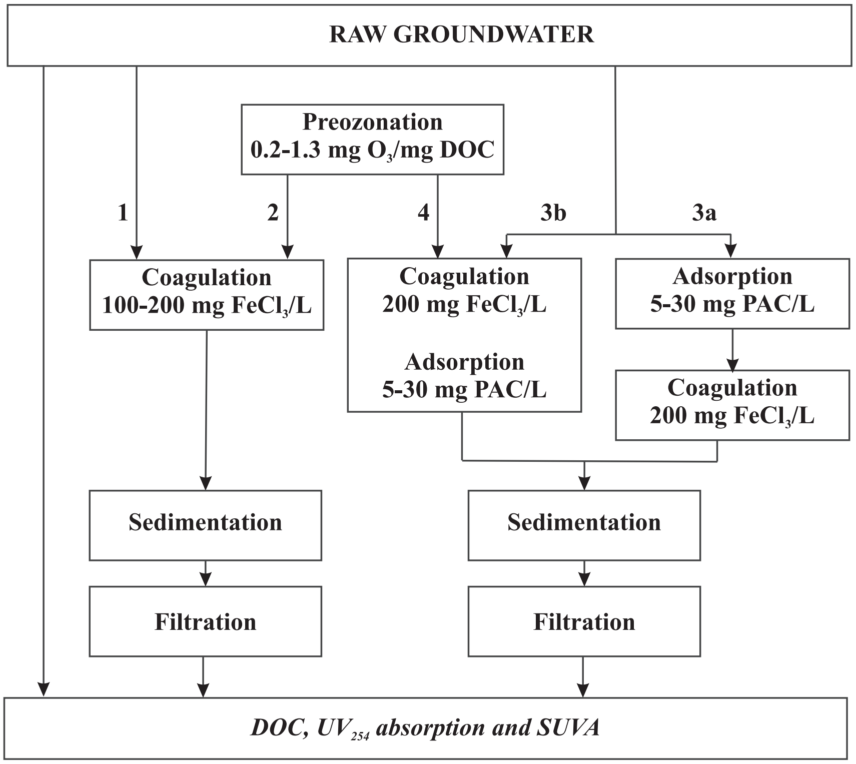

Fig. 1. Plan of the experiment.

Table 1
Raw water characteristics

Parameter	Unit	Mean value ± σ
pH	–	7.46 ± 0.14
Conductivity	μS/cm	1,189 ± 7.17
DOC	mg C/L	10.27 ± 0.49
UV_{254}	cm^{-1}	0.495 ± 0.015
SUVA	$L\,mg^{-1}\,m^{-1}$	4.82 ± 0.49
THMFP	μg/L	555 ± 128
HAAFP	μg/L	131 ± 20.7

Note: σ: Standard deviation based on 10 measurements.

and haloacetic acids (HAAFP) (Table 1). Using a NOM fractionation procedure, Molnar et al. [17] showed that the NOM in the groundwater mostly consists of the fulvic acid fraction (65%), while the humic acid fraction is 14% of the total DOC. There are significantly less hydrophilic substances in the NOM structure, accounting for only 21%. These results are in agreement with NOM classification based on the high specific UV absorbance at 254 nm value (SUVA, 4.82 ± 0.49 $L\,mg^{-1}\,m^{-1}$) which is characteristic of hydrophobic high molecular NOM. Edzwald and Tobiason [18] note that NOM from water with a high SUVA (>4 $L\,mg^{-1}\,m^{-1}$) can be removed significantly during the coagulation process.

2.2. Ozonation as pretreatment

Preozonation of raw water was carried out in a glass column (80 mm in diameter, 1.5 m long, with a sampling tap at the bottom) into which ozone was introduced through a ceramic diffuser at the bottom. The reactor was continuous with respect to ozone and discontinuous with respect to water. The ozone generator used was an Argentox type GLX 1. Water samples (6 L/sample) were ozonated, with ozone doses during the preozonation process ranging from 0.2 to 1.3 mg O_3/mg DOC.

2.3. Jar tests

The jar test experiments were performed with six-paddle stirrer (FC6S Velp Scientific), in 1 L beakers at room temperature (22–25°C). To 0.6 L samples of raw and preozonated water (0.2–1.3 mg O_3/mg DOC), coagulant ($FeCl_3$) and PAC (Norit W 35) were added. Characteristics of PAC Norit W 35 as given by the producer: apparent density, tamped—450 kg/m^3, average particle diameter—20 μm, Iodine No.—600 and Moisture—4% by mass. After coagulant addition, the water samples underwent fast mixing (120 rpm for 2 min), flocculation with addition of anionic flocculant A110 (30 rpm for 30 min) and settling for 60 min. After clarification, the water samples were filtered through glass fibre filters (0.6 μm).

The doses of coagulant applied during the experiment were in the range of 100–200 mg $FeCl_3$/L, and PAC doses were from 5 to 30 mg/L (added as slurry to the water). The flocculant dose used in this research was 0.5 mg/L.

The study was performed on water without pH correction (pH raw water 7.46 ± 0.14). During the investigation into the efficiency of the adsorption-coagulation system in NOM removal, two different approaches were applied: (1) The coagulant was dosed after 10 min of adsorption on PAC with mixing of 30 rpm (PAC–$FeCl_3$) and (2) PAC was added simultaneously with the coagulant ($FeCl_3$ + PAC).

Baseline coagulation without oxidative pretreatment or PAC addition was performed in order to compare the effects of the combined coagulation method.

2.4. Analytical methods

Water samples were analysed before and after coagulation for DOC content using a Thermo HiPerToc after filtration through a 0.45 μm membrane filter. UV_{254} absorbance measurements were performed in accordance with standard methods [19] by UNICAM SP600 UV spectrophotometer at a wavelength of 254 nm with a 1 cm quartz cell. The specific UV absorbance (SUVA, $L\,mg^{-1}\,m^{-1}$) was obtained by dividing the UV_{254} absorbance by the DOC value and multiplying by 100. pH was measured by portable instrument WTW InoLab pH. The conductivity metre used was a WTW Cond 330i. DBPs formation potential (trihalomethane and haloacetic acids formation potential) was determined in accordance with standard method for THMFP determination [19]. Analysis of THMs and HAAs was performed by GC/μECD (Agilent 6890N) [19,20].

The ozone content in the inlet and outlet gases was determined by the iodometric method [19]. The ozone dose was calculated from the difference between them. All experiments were performed in triplicate and are presented as average values.

3. Results and discussion

In Table 2, the percentage removal of NOM based on DOC and UV_{254} absorbance in water during the coagulation process is shown, depending on the applied coagulant dose (baseline coagulation) and ozone dose (coagulation combined with preozonation). In addition, changes in the NOM structure, during coagulation and combined treatment with ozone preoxidation, was monitored using calculated SUVA values.

Table 2
NOM removal by coagulation and combined ozonation and coagulation

Water type	DOC (mg/L)	UV (cm^{-1})	SUVA ($L\,mg^{-1}\,m^{-1}$)	DOC reduction (%)	UV reduction (%)
Raw water	10.27	0.495	4.82	–	–
Coagulated water					
100 mg $FeCl_3$/L	7.22	0.345	4.78	30	30
150 mg $FeCl_3$/L	6.79	0.290	4.27	34	41
170 mg $FeCl_3$/L	7.60	0.260	3.42	26	48
190 mg $FeCl_3$/L	7.17	0.230	3.21	30	54
200 mg $FeCl_3$/L	6.30	0.230	3.65	39	54
Ozonated water					
0.2 mg O_3/mg DOC	9.62	0.425	4.42	7	14
0.6 mg O_3/mg DOC	9.47	0.350	3.70	8	29
0.9 mg O_3/mg DOC	9.42	0.310	3.29	8	37
1.3 mg O_3/mg DOC	8.64	0.260	3.01	16	48
Coagulated (200 mg $FeCl_3$/L) ozone-pretreated water					
0.2 mg O_3/mg DOC	5.93	0.215	3.62	42	56
0.6 mg O_3/mg DOC	4.82	0.173	3.59	53	65
0.9 mg O_3/mg DOC	7.30	0.149	2.04	29	70
1.3 mg O_3/mg DOC	5.64	0.134	2.38	45	73

3.1. Baseline coagulation

During the investigation of baseline coagulation with $FeCl_3$ (100–200 mg/L, Table 2), it was determined that the coagulation process enables significant removal of DOC content (26–39%). With increasing coagulant dose, the removal percentage of organic matter which absorbs UV light at 254 nm also increases (30–54%). It can be noticed that DOC value does not fall evenly with increasing coagulant dose, as in the case of UV_{254}. The maximum removal of NOM content was obtained by coagulation with 200 mg $FeCl_3$/L. These results are in accordance with our previously published results [21], where the investigated dose range was 18–300 mg $FeCl_3$/L. In this previous work, the coagulant showed a significant decrease in efficacy at doses lower than 180 mg $FeCl_3$/L (which achieved a maximum DOC removal of 38%, relative to raw water). According to the NOM characterisation based on SUVA value, coagulation with 100–150 mg $FeCl_3$/L did not result in the significant removal of hydrophobic NOM structures (SUVA >4 $L\,mg^{-1}\,m^{-1}$). With further increase of coagulant dose from 170 to 200 mg $FeCl_3$/L, the hydrophobic fraction of NOM was removed to a higher extent and the residual NOM in the coagulated water represented a mixture of hydrophilic and hydrophobic NOM structures (3.21–3.65 $L\,mg^{-1}\,m^{-1}$).

On the basis of the performed experiments, a coagulant dose of 200 mg $FeCl_3$/L was chosen for further investigation, the main goals of which were the optimisation of the ozone dose during preozonation and the PAC dose for improving the coagulation process.

3.2. Coagulation combined with preozonation

The effects of the combined processes of preozonation and coagulation on the UV_{254} absorbance and DOC, are given in Table 2. For the sake of comparison, the results of preozonation alone are also given in Table 2. With increasing ozone dose during preozonation from 0.2 to 1.3 mg O_3/mg DOC, a gradual growth in the efficacy of coagulation in reducing UV_{254} absorbance was observed (from 56 to 73%). It is known that the effect of ozone on the coagulation process during drinking water treatment, as a consequence of ozone-NOM interaction, is significantly affected by the nature and content of NOM in water [5]. As can be seen in Table 2, preozonation leads to the partial oxidative degradation of NOM molecules (UV_{254} absorbance decreases by 14–48%, relative to the raw water), and a reduction in the SUVA value (3.1–4.42 $L\,mg^{-1}\,m^{-1}$) leading to the formation of molecules with more hydrophilic characteristics. These effects are a consequence of ozone attack, mainly upon the unsaturated bonds of molecules which are responsible for the absorption of UV light at 254 nm [10]. This also explains the high level of UV_{254} absorbance removal observed during the combined coagulation with preozonation experiment.

On the other hand, changes in the DOC during the coagulation treatment combined with preozonation show greater fluctuation depending on the applied ozone dose. As can be seen from the data in Table 2, a maximum NOM removal based on DOC value (53%) was achieved using 0.6 mg O_3/mg DOC combined with 200 mg $FeCl_3$/L. Preozonation alone had a negligible effect on DOC removals (7–16%, relative to the raw water), as expected from the literature [2,12]. Generally, compared to the conventional coagulation processes, the introduction of an oxidation pretreatment with ozone doses (0.2–1.3 mg O_3/mg DOC) commonly applied in drinking water treatment [10,21], improves the efficiency of the NOM removal expressed as DOC. The SUVA values above support the assumption that the obtained results are probably a consequence of the changes in the NOM surface chemistry and increase in the polarity and acidity of their molecules after ozonation. This increases the NOM absorptivity on the surface of the metal-hydroxide formed after the addition of the coagulant and therefore increases the efficacy of the combined process compared to the baseline coagulation [5,22,23]. Our previous results [15] have also shown that preozonation improves efficacy of NOM removal by coagulation for similar water type.

Many authors point out the different effects of ozone on organic material content removal in coagulation processes [3,10,12]. These authors indicate that by preozonation within the range 0.8–1.5 mg O_3/mg DOC, flocs obtained in the presence of NOM may be more negatively charged, and/or form stable flocs, which in some cases could results in a lower degree of NOM removal by coagulation, relative to non-ozonated samples.

In accordance with earlier observations, the oxidative attack of ozone, which is primarily directed towards unsaturated molecules, resulted in a significant reduction of UV absorbance relative to DOC. The SUVA decreased during the combined preozonation and coagulation treatment, depending upon the applied ozone dose to 2.04–3.62 $L\,mg^{-1}\,m^{-1}$. The obtained results indicate that with the preozonation of water, the structure of NOM is changed, directly influencing the removal of these components by coagulation. The residual NOM after treatment (0.6–1.3 mg O_3/mg DOC combined with 200 mg $FeCl_3$/L) presents a more hydrophilic mixture of NOM compared to coagulation alone, in accordance with Karanfil [24].

According to the results obtained, the optimal ozone dose for the combined coagulation process is 0.6 mg O_3/mg DOC, which is in agreement with the range of ozone doses proposed in the literature [2,22,23].

3.3. Coagulation enhanced with PAC adsorption

Fig. 2 shows the effects of the application of different doses of PAC, and different dosing approaches on the decrease of organic matter content in water, after coagulation using 200 mg $FeCl_3$/L. It has been determined that the addition of PAC at 5 mg/L does not increase the efficiency of organic material removal regardless of the dosing approach applied. Using PAC doses in range 10–30 mg/L, improvements in UV_{254} reduction (up to 25%) compared to coagulation alone were achieved, while there was slight improvements of DOC removal (up to 9%).

The total decrease in organic material content in water during the combined coagulation process, depending on the applied PAC dose, for the PAC–$FeCl_3$ dosing approach, was: 54–65% UV_{254} and 38–44% DOC. For the $FeCl_3$ + PAC dosing approach, removal was 53–65% UV_{254} and 35–44% DOC. These results suggest that there was no difference between coagulation followed by PAC and simultaneous dosing of PAC and $FeCl_3$.

In all investigated cases (Table 2 and Fig. 2), the reduction in UV absorbance between raw and treated water was more significant than the decrease in DOC. This is in accordance with the findings of other authors such as Tomaszewska et al. [25] and Uyak et al. [8] who reported that aromatic compounds, which are responsible for UV_{254} absorption, are better removed by coagulation processes than other NOM fractions.

These findings are reflected in the SUVA values obtained during the coagulation and PAC adsorption experiment, which are given in Fig. 3. When the combined coagulation process is compared with the baseline coagulation, the additional change in SUVA (0.12–0.77 L mg^{-1} m^{-1}) is relatively small compared to change already achieved by baseline coagulation (1.17 L mg^{-1} m^{-1}).

On the basis of the results obtained, it can be concluded that for combined coagulation processes with NOM adsorption onto PAC, a PAC dose of 10 mg/L is sufficient.

3.4. Coagulation combined with multiple pretreatments —preozonation and PAC adsorption

In earlier experiments, it was shown that combined coagulation, with either preozonation or adsorption onto PAC, improves NOM removal effects in water. Preozonation of water significantly changes the structure of NOM (above all, the size and polarity of the molecules) [1,3,6,26]. Furthermore, one of the most important limiting factors of adsorption on activated carbon is the size of the particles adsorbed [8,27]. Therefore, in the next experiment, the effects of a combined pretreatment of preozonation and PAC

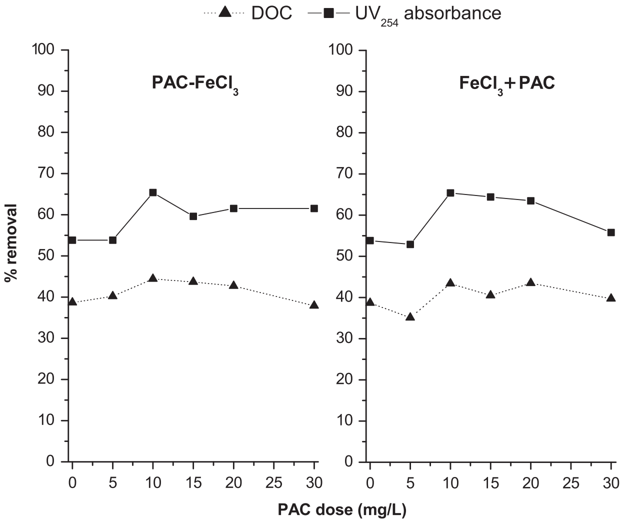

Fig. 2. Effect of PAC dose on the percentage removal of NOM during coagulation (200 mg $FeCl_3$/L) for two dosing approaches: (1) adsorption before coagulation (PAC–$FeCl_3$) and (2) simultaneous PAC and coagulation addition ($FeCl_3$ + PAC).

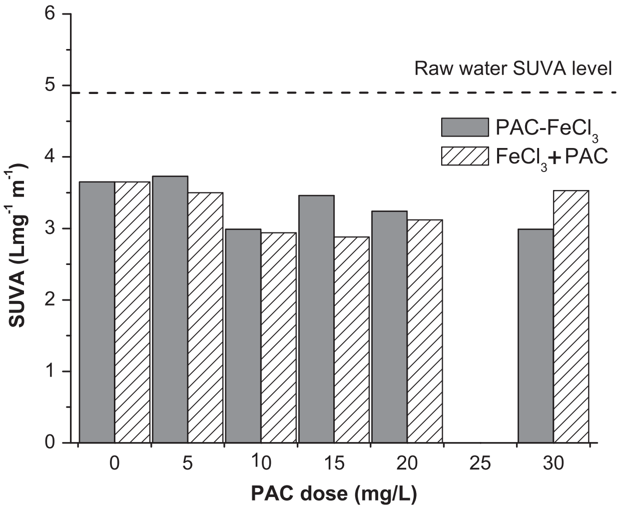

Fig. 3. SUVA vs. PAC dose in two combined coagulation procedures.

adsorption on the removal of NOM by combined coagulation were investigated (Fig. 4). The applied doses of ozone (0.6 mg O_3/mg DOC) and coagulant (200 mg $FeCl_3$/L) were chosen as the optimal ones based on the previous investigations, with simultaneous dosing of PAC and $FeCl_3$ applied ($FeCl_3$ + PAC).

In accordance with expectations, the addition of PAC to preozonated water increased the NOM removal effects of the combined coagulation process, compared to the coagulation and adsorption alone. Research obtained by Badawy et al. [22] also suggests that preozonation combined with conventional coagulation/flocculation and activated carbon filtration, improves organic content removal and certain THMs precursors.

The content of UV_{254} absorbing material, in relation to the applied PAC doses, is up to 24% lower than ozonated coagulated samples without PAC. Compared to coagulation with PAC adsorption, ozonation increased UV_{254} removal from 12 to 40%. The total removal of UV absorbing organic material with applied ozone and depending on the PAC dose is 65–73%. These observed changes can be explained by the easier adsorption onto PAC particles of the lower molecular mass organic compounds formed during the preozonation process, and their further removal during coagulation [8]. However, the effects of the applied combined treatment are less pronounced with respect to DOC, with the addition of PAC resulting in an additional reduction in DOC of up to 11%. Relative to the raw water, the total removal of DOC after preozonation, coagulation and PAC adsorption is 54–58%, depending on the PAC dose. This indicates that the PAC dose was not a limiting factor in this investigation.

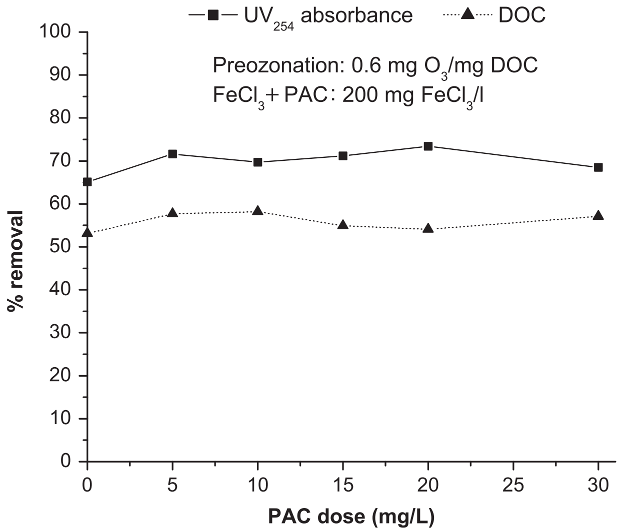

Fig. 4. Effect of simultaneous PAC dosing on the percentage removal of NOM during coagulation with the combined pretreatment.

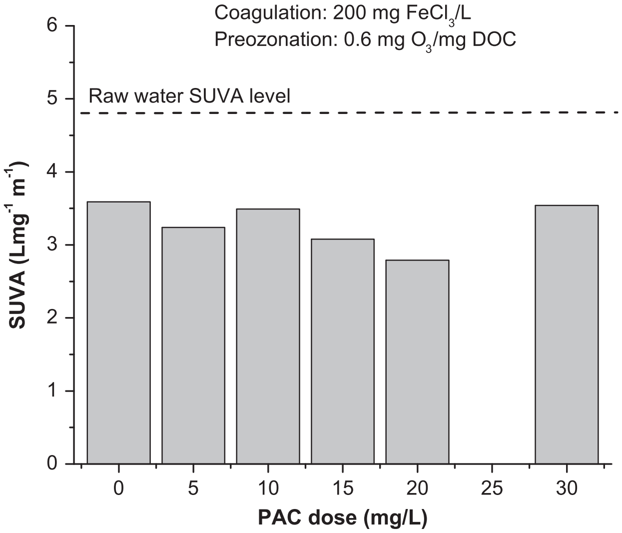

Fig. 5. SUVA vs. PAC dose during coagulation ($FeCl_3$ + PAC) with combined pretreatment.

Based on the SUVA values, which ranged from 2.79 to 3.54 L mg^{-1} m^{-1} (Fig. 5), the residual NOM in the treated water has a mixed hydrophobic and hydrophilic character [24], and the addition of PAC into this process did not result in the preferential removal of the NOM fraction responsible for UV_{254} absorption. These findings are in accordance with Kristiana et al. [28].

In general, the combination of the preozonation, adsorption and coagulation processes was shown to be more efficient than all the other investigated processes for the removal of DOC, as well as the reduction in UV_{254} absorption. Similar results were obtained by Mosteo et al. [29] who concluded that for drinking water treatment the most appropriate application of ozone is in preoxidation treatment, followed by adsorption by PAC, coagulation–flocculation and final post-chlorination.

From a treatment optimisation standpoint, it can be concluded that the best DOC removal as well as satisfactory effects on the reduction of UV_{254} absorbance, were achieved with an applied dose of 0.6 mg O_3/mg DOC and 5 mg/L PAC (removal of 58% DOC and 72% UV_{254}).

4. Conclusion

This work investigated organic matter removal from raw groundwater rich in the hydrophobic NOM, using DOC, UV_{254} and SUVA to identify the most effective pretreatment and coagulation combination. It was shown that preozonation changed the structure of NOM in the water, which directly improved the efficiency of coagulation for the removal of these components. PAC addition alone slightly improved coagulation efficacy in NOM removal, with no difference observed between coagulation followed by PAC and simultaneous dosing of PAC and $FeCl_3$. The ability of preozonation to change the NOM structure, when combined with the adsorption characteristics of PAC, was found to provide the most effective coagulation treatment applied during this investigation, with 58% DOC and 72% UV_{254} removals whilst applying 0.6 mg O_3/mg DOC, 5 mg/L PAC and 200 mg $FeCl_3$/L. It can be concluded that for NOM-rich groundwater, optimised coagulation combined with preozonation and PAC adsorption can be an efficient tool for controlling NOM in water.

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