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Comparison of Efficiency between Poly-aluminium Chloride and Aluminium Sulphate Coagulants during Full-scale Experiments in a Drinking Water Treatment Plant

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Abstract: The efficiency of poly-aluminium chloride (PACl) and of aluminium sulphate (alum), two commonly applied coagulant agents, was studied comparatively in this work, during full-scale experiments in a drinking water treatment plant. The removal of suspended solids (turbidity) and the residual aluminium concentrations were carefully monitored and they were used for the evaluation of effectiveness for each coagulant, as well as for the determination of optimal operative conditions. Two alternative treatment processes were examined:

(a) the conventional coagulation-flocculation-sedimentation process, followed by gravity filtration through sand filter beds, and
(b) the direct filtration process, i.e., coagulation-flocculation and sand filtration, but without the intermediate sedimentation step.

PACl proved to be a more efficient coagulant than alum, as lower dosages of PACl, about 1.35 mg Al/L in this case, resulted to the production of treated water with low turbidity and residual aluminium content. In addition, the direct filtration process through dual sand-anthracite filter beds was found to be equally sufficient, as the

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conventional one, i.e., when applying the sedimentation step; in this case, 0.70 mg Al/L of PACl resulted in low turbidity water (around 0.1 NTU) and residual aluminium content (lower than 150 μg/L). In addition, the operation time of filters was extended to more than 24 hours.

Keywords: PACl, alum, coagulation, direct filtration, residual aluminium concentration, drinking water treatment, full-scale experiments

INTRODUCTION

Aluminium sulphate (alum) is among the most widely applied coagulant agents, used mainly for the treatment of drinking water. The conventional treatment processes applied in most large-scale drinking water plants include typically the coagulation–flocculation step, followed by sedimentation and by gravity filtration through granular filter beds (usually consisted from sand). The coagulant agents, such as alum, are usually added during the initial stage of coagulation process, in order to enhance the removal of suspended solids, including colloidal particles, as well as of natural organic matter (NOM) (1). However, the efficiency of coagulants for the production of high quality water is often determined by the operational conditions. Thus, the selection of an appropriate coagulant, its dosage and various other operational parameters, such as the water temperature and the pH control, may significantly affect the coagulation-flocculation process and the subsequent treatment steps of sedimentation and filtration (2).

More recently, the research related to coagulation processes was focused on the production of new and more effective coagulants. Coagulants are chemical substances, carrying a positive charge on their surface. Upon their addition in water and following their hydrolysis, the (usually) negatively charged colloidal impurities will be destabilized by the action of various mechanisms, such as charge neutralisation, due to mutual electrical attraction, or sweep flocculation (3). Therefore, the higher the positive charge of a coagulant, usually based on Al(III) or Fe(III) salts, the highest its efficiency for the removal of suspended solids. However, the corresponding coagulation mechanisms are very complicated, because a large number of mono- and polymeric hydroxyl complexes, formed during the hydrolysis of parent cations in water, may substantially participate in the colloidal destabilization process. Al(III) salts (e.g., $Al_2(SO_4)_3$, usually termed alum) has been used especially for drinking water treatment, as it is considered more effective than Fe(III) for that application (4). Despite its higher efficiency, the residual aluminium concentrations need to be careful controlled, according to the respective safe maximum concentration limits, as set by the legislation, because otherwise higher concentrations of this element have been connected with several health problems (e.g., Alzheimer disease).

Several hydroxyl complexes of aluminium, including $Al(OH)_2^{+}$, $Al(OH)^{2+}$, $Al_2(OH)_2^{4+}$, $Al_3(OH)_2^{5+}$, and $Al_{13}O_4(OH)_{24}^{7+}$ (or "Al_{13}"-Keggin form), may be usually formed during the hydrolysis process of alum; however, this process is greatly affected by temperature, or pH variations (3, 5). For this reason, a polymeric aluminium compound has been proposed for drinking water treatment, as an alternative coagulant agent to alum, termed poly-aluminium chloride (PACl). PACl is a poly-nuclear form of $AlCl_3$ species, containing already highly positive charged species, such as "Al_{13}" in relatively high concentrations, even without the hydrolysis step. Consequently, PACl is considered as a more efficient coagulant than alum, mainly because of its higher overall positive charge. A comprehensive comparison between alum and PACl typical characteristics is presented in Table 1 (3, 6, 7).

According to this table, as well as to previously reported laboratory experimental results (8), PACl is considered as more efficient than alum, presenting higher suspended solids removal capacity. However, its efficiency is greatly affected by several parameters, such as composition of raw water, specific operational conditions, etc. Additionally, although the respective results are referred to small-, laboratory- or even pilot-scale experiments, very few results have been presented in the literature, regarding the comparative full-scale application of these two coagulants. As a result, during the full-scale application of PACl significant concerns may arise, regarding the benefits that would be eventually deduced by the replacement of alum with PACl. The aim of this study was to evaluate comparatively the efficiency of alum

Table 1. Comparison of basic characteristics between alum and PACl, based on information collected from the literature

Criteria	Alum	PACl
Temperature	Hydrolysis of alum and consequently the production of the positively charged hydroxyl complexes, which are essential for the destabilization of colloidal impurities of natural waters, are affected by temperature.	Less impact of temperature, as it contains pre-polymerised forms of aluminium.
pH	The range of pH controls which hydroxyl specie of aluminium will be produced.	Less impact of pH is expected, as it contains pre-polymerised forms of aluminium.
Aluminium species	The majority of aluminium species are monomeric hydroxyl complexes with a cationic charge varying from +1 to +3.	Monomeric and polymeric forms of aluminium are present. Poly-nuclear 'Al_{13}' (7+) is present in relatively high concentrations.
Kinetics	Slower	Faster

and PACl for the treatment of surface (river) water in a full-scale drinking water facility, the determination of optimum main operational conditions, i.e., coagulant dosage and pH, and the assessment of the overall performance of the respective treatment plant.

EXPERIMENTAL PART

Materials

Two commercial coagulant reagents were examined in this study: solid aluminium sulphate (alum) with very small basicity (0.2%, connected with the ratio [OH]/[Al]) and poly-aluminium chloride (commercially named PAC-18), containing 16.6% (w/w) Al_2O_3, with medium basicity (about 40%) and density of $1.34\ g \cdot cm^{-3}$. Both chemicals were obtained from the Greek Chemical Industrial Plant "Phosphates Fertilizers Industry SA".

In addition, a high molecular weight anionic polyelectrolyte, Magnafloc LT25, used for drinking water production, was examined as a commonly applied flocculation reagent (obtained from CIBA). It is noted that polyelectrolytes are water soluble organic polymers that are used mainly as coagulant aids. They are generally classified as:

i. anionic (i.e., ionize in solution to form negative sites along the polymer molecule),
ii. cationic (i.e., ionize to form positive sites), and
iii. non-ionic (i.e., showing very slight ionization). They function primarily through inter-particle bridging and enhance the flocculation process.

Plant Description

The main processes used for the treatment of surface (river) water in the Thessaloniki (N. Greece) drinking water treatment plant, are presented in Fig. 1. The overall treatment process of the full-scale plant includes a pre-ozonation step, followed by adjustment of pH from the initial values of 8.0–8.3 to about 6.9–7.2; the addition of coagulant at concentrations 1.7–2.5 mg Al/L (or 15–21 mg $Al_2(SO_4)_3$), followed by the polyelectrolyte addition at the concentration of 0.04–0.1 mg/L in the flocculation basin; the sedimentation, the filtration through sand bed filters, the post-ozonation, the granular activated carbon filters, the pH readjustment (to 8) by lime addition and finally, the chlorination for the control of residual disinfection of drinking water.

It should be noted that the most important water quality parameters (e.g., pH, turbidity) were monitored online and the dosage of chemicals was controlled by specific dosimetric pumps through the established SCADA system of the full-scale water treatment plant.

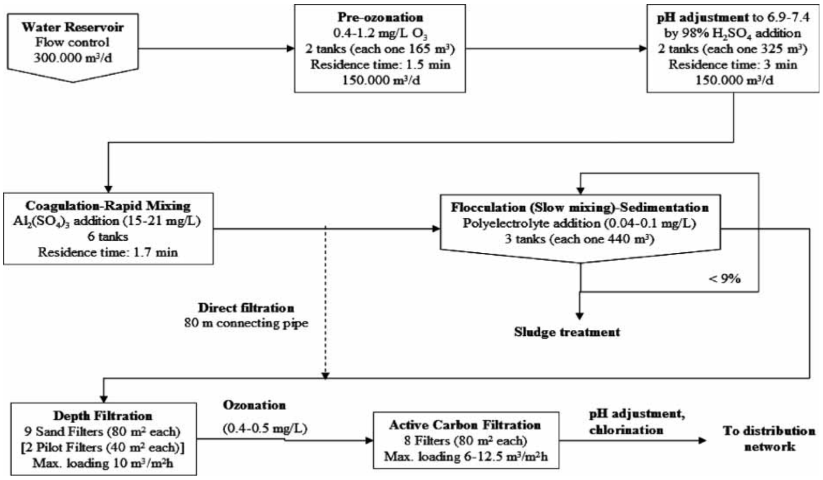

Figure 1. Flow diagram of the Thessaloniki's drinking water treatment plant.

Experimental Set-up

Two alternative treatment processes were examined in order to compare the alum and PACl performance: the conventional coagulation-flocculation-sedimentation process followed by filtration, and the direct filtration process, i.e., without the sedimentation stage, through either single medium, or dual media sand filter beds. Dual media filter beds consisted from the sand layer extended up to 60 cm from the bottom of the filter bed and from the anthracite layer, placed over the sand layer with additional height of about 40 cm. The filtration was conducted in a system of two similar filter beds (mono- or dual-bed), each one having surface area of about 40 m^2. Therefore, the total depth of each filter bed was 100 cm, whereas the inlet flow rate was 375 m^3/h; hence, resulting in the filtration rate (velocity) of around 9.4 m/h.

The mixing of coagulants during the conventional treatment method took place in an appropriate mixing tank; however, during the examination of direct filtration the coagulant was added to the raw water at the beginning of treatment process. Effective mixing was obtained by using an appropriate connecting pipeline (ID 20 cm, total length 80 m), used to transfer raw water from the pH adjustment unit directly to the sand filters and specifically constructed to by-pass the flocculation-sedimentation tank; this is known as "pipe" or "in-line" flocculation process. The respective retention time of water and coagulant in this pipe was estimated to 96 s; this time period is quite similar to the residence time of coagulation stage during the conventional treatment process.

The filtration medium of the twin filter beds used in this study was river sand with an effective size (D_{10}) of 0.64 mm and uniformity coefficient

(D_{60}/D_{10}) in the range of 1.75–1.79. Grain size distribution tests of coarser anthracite layer resulted to an effective size D_{10} of 1.0–1.1 mm. According to the literature, an effective size in the range of 2–4 mm and a uniformity coefficient in the range of 1.3–1.8 are usually recommended for sand filters with a depth of 1–2 m, such as those examined (9).

Analytical Determination Methods

The comparable evaluation of alum and PACl coagulation and the monitoring of water quality were carried out by continuous measurements of turbidity, using on-line sensors, because this parameter constitutes a primary indicator of treated drinking water quality (10). In addition, on-line measurement of pH, conductivity, and temperature took place by appropriate sensors and the experimental data were closely monitored and handled by the appropriate SCADA software (Supervisory Control and Data Acquisition), installed for the overall remote control of drinking water treatment plant. The residual aluminium concentration was measured at the outlet of filter beds, by using the Eriochrome Cyanine R (Riedel-de-Haan, Sealze, Germany) photometric standard method (11). In order to determine the total aluminium concentration (soluble and particulate), the filtration of the samples through a 0.45 μm membrane filter was avoided.

RESULTS AND DISCUSSION

The overall assessment of the performance of the examined drinking water treatment plant can be shown in Table 2, comparing the mean (average) values of various online, as well as of laboratory taken measurements after the optimisation of each examined (conventional or direct filtration) process. It is worth noting that PACl could have an even lower residual aluminium concentration, in the case it would be applied at lower pH values (around 7.2), i.e., comparable with the respective best conditions found for alum. However, the higher pH value (7.5) was selected in order to show that supplementary amount of sulphuric acid, used for pH control of raw (river) water (having initial pH value 8.0–8.3), can be also preserved, when using PACl instead of alum.

Comparison of Alum and PACl Performance during the Conventional Coagulation-Sedimentation-Filtration Treatment Process

The evaluation of alum and PACl efficiency was carried out during the conventional treatment scheme consisted of coagulation-flocculation, followed

Table 2. Comparison of the mean (average) values for various online and laboratory measurements after the optimisation of each process

	Online and laboratory measurements	Mean value Alum	Mean value PACl
Conventional treatment scheme	pH	7.2	7.5
	Laboratory pH	7.2	7.5
	Turbidity at the outlet of flocculator basins, NTU	1.15	1.10
	Laboratory turbidity at the outlet of flocculator basins, NTU	1.22	1.09
	Outlet turbidity of dual-bed filter, NTU	0.06	0.05
	Laboratory outlet turbidity of dual-bed filter, NTU	0.07	0.06
	Outlet turbidity of single filter bed, NTU	0.05	0.06
	Laboratory outlet turbidity of single filter bed, NTU	0.07	0.06
	Conductivity, μS cm^{-1}	444	436
	Laboratory conductivity, μS cm^{-1}	442	431
	Residual Al concentration at the outlet of dual filter bed, μg L^{-1}	25	77
	Residual Al concentration at the outlet of single filter bed, μg L^{-1}	28	71
Direct filtration scheme	pH	7.2	7.4
	Laboratory pH	7.2	7.5
	Outlet turbidity of dual-bed filter, NTU	0.45	0.11
	Laboratory outlet turbidity of dual-bed filter, NTU	0.58	0.14
	Outlet turbidity of single filter bed, NTU	0.25	0.15
	Laboratory outlet turbidity of single filter bed, NTU	0.21	0.16
	Conductivity, μS cm^{-1}	441	435
	Laboratory conductivity, μS cm^{-1}	415	420
	Residual Al concentration at the outlet of dual-filter bed, μg L^{-1}	176	115
	Residual Al concentration at the outlet of single filter bed, μg L^{-1}	47	131

by sedimentation and then by filtration through the sand filter beds. The experimental procedure included the addition of PACl solution to the first flocculation tank, while the other two tanks were operated with alum, according to the treatment plant common operational schedule.

Various dosages of PACl were examined for the treatment of surface water and for the removal of suspended solids, ranging from 0.60 up to

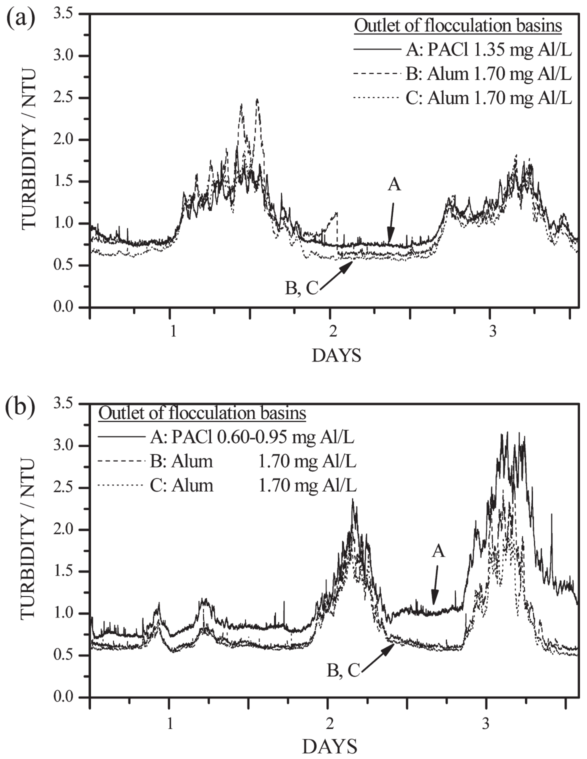

Figure 2. Turbidity of water at the outlet of the flocculation-sedimentation basins. Conditions: alum dosage 1.70 mg Al/L, PACl dosage: (a) 1.35 mg Al/L and (b) 0.60–0.95 mg Al/L, polyelectrolyte dosage 0.06 mg/L, pH 7.4.

1.35 mg Al/L. The turbidity of effluent from the sedimentation tanks as a function of operation time and under different dosages of alum and PACl, is given in Fig. 2. Small dosages of PACl, about 0.6–0.95 mg Al/L, resulted in poor coagulation-sedimentation performance and consequently, a higher turbidity was observed at the outlet of the tank, where PACl had been added, than in the outlet from the sedimentation tanks working with alum. However, the application of slightly increased PACl dosages, i.e., 1.35 mg Al/L, resulted in treated water with a turbidity similar to the water turbidity produced from the alum tanks; nevertheless, as shown in Fig. 2, similar water quality was obtained as by the addition of higher alum dosage, i.e., 1.70 mg Al/L.

Single and dual bed filtration processes were not significantly influenced by the application of various PACl dosages. The treated water at the outlet of filter beds presented very low turbidity (below 0.1 NTU) for all PACl and alum dosages. In addition to turbidity, the feeding with different aluminium dosages may affect the concentration of this element at the outlet stream, considering that the concentration of residual aluminium is of specific importance for the determination of most appropriate coagulant agent. The residual aluminium concentration at the outlet of single and dual filter beds is shown in Fig. 3, as a function of the filter's operation time. Sufficiently small residual aluminium concentrations were observed both from single or dual filter beds, varying around 60–90 μg/L, i.e., well below than the recent limit of 200 μg/L, as set by national and international legislation guidelines. Furthermore, the operation of single and dual filter beds, i.e., the time between subsequent backwashing stages, was not significantly affected by the addition of PACl.

Comparison of Alum and PACl during the Direct Filtration Operational Scheme

The direct filtration process has been examined by several researchers during the treatment of drinking water and it has been found that the main drawback of this method is the relatively rapid clogging of filter medium, due to increased rate of solid build-up upon their removal (10, 12). Direct filtration was examined in this work, by using various dosages of the two coagulants, alum and PACl, with and without the addition of polyelectrolyte. The aim of this study was to optimize the filter beds performance for the production of high quality effluent, presenting low turbidity and low residual aluminum content, while the operational period of filters should be simultaneously extended.

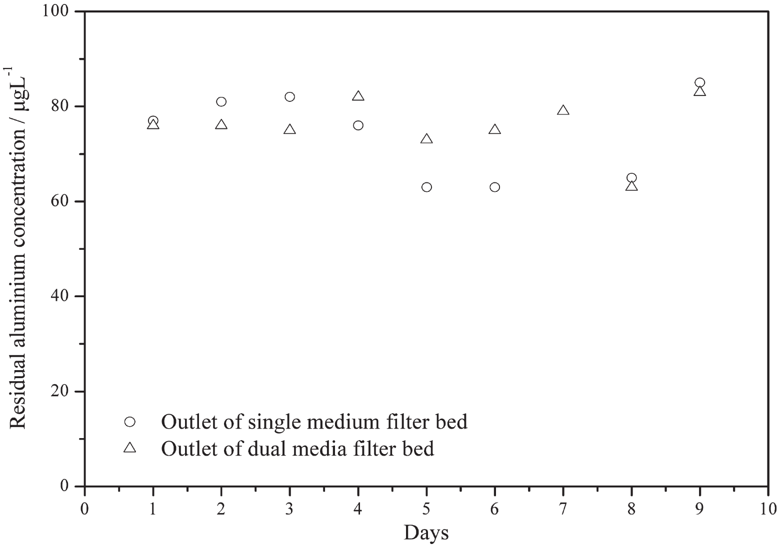

Figure 3. Residual aluminium concentration of filtrated water, treated with PACl.

Various dosages of each coagulant were added in the filter influent stream, ranging from 0.1 up to 0.75 mg Al/L, whereas their effect to the bed operation and to the turbidity of effluent was monitored; representative results for alum and PACl are given in Fig. 4, as a function of operational time. Small dosages of both coagulants, ranging from 0.1 to 0.3 mg Al/L, resulted to the production of effluent with rather high turbidity values, i.e., exceeding 0.5 NTU in most cases, although the duration of filtration cycles was not significantly affected. However, the addition of higher dosages of alum, up to 0.4–0.5 mg Al/L and 0.03 mg/L of polyelectrolyte, resulted to the production of effluent with decreased turbidity, around 0.1–0.2 NTU; additionally, the increased coagulant dosages reduced the active operational time of filters, as the filtration cycle was decreased significantly to about 4.5–6 h.

The filtration process was operated more efficiently during the operation of dual filters. The addition of relatively high alum dosage, up to 0.40 mg Al/L, resulted to water with increased turbidity values, reaching up to 0.5 NTU, which however was further decreased after prolonged operation time. In addition, extended operation times of the filters were observed, around 13–18 h. The addition of relatively high dosages of PACl resulted to more efficient operation, than for the case of alum, as the addition of 0.75 mg Al/L PACl (even without the presence of polyelectrolyte), produced water with

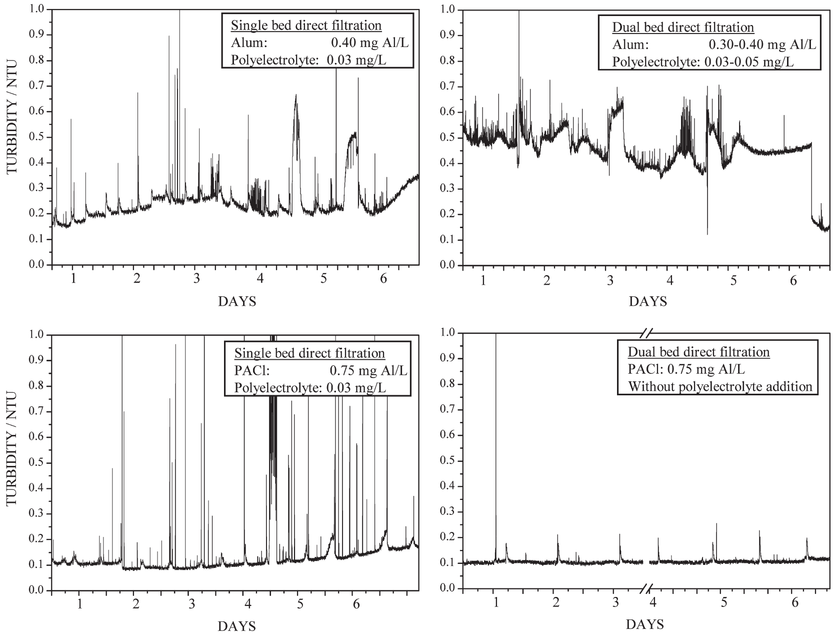

Figure 4. Turbidity of water at the outlet of filter beds.

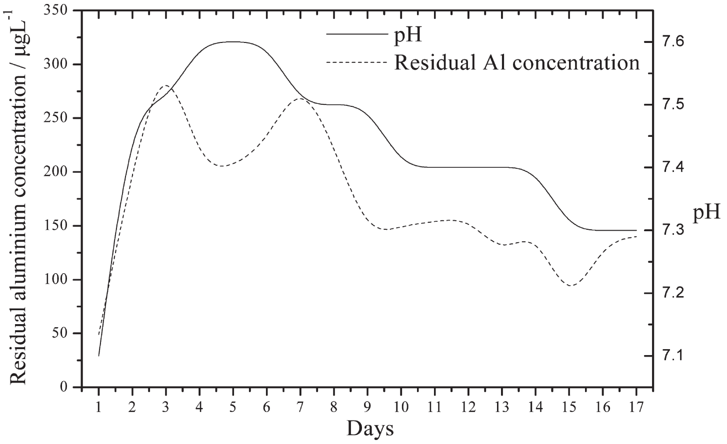

Figure 5. Effect of pH on the residual aluminium concentration of filtrated water at the outlet of dual filter bed; direct filtration with PACl.

low turbidity, about 0.1 NTU, while extended filtration cycles were achieved, i.e., higher than 24 h. The faster kinetics of PACl than alum, taking place within the by-pass connecting pipe, probably can be attributed to the higher rate of coagulation-flocculation process, due to the formation of bigger and hence, more filterable flocs (6).

The variation of dual filter bed effluent pH values and the residual aluminium concentrations as a function of operational time are shown in Fig. 5, using PACl or alum. The effluent pH values were ranged between 7.3 and 7.6; these values did not affect significantly the process efficiency, i.e., the duration of filtration cycle and the effluent turbidity. However, the pH values were related to the content of residual aluminium. At relatively higher pH values (exceeding 7.5), increased residual aluminium concentrations were measured, i.e., higher than 200 μg/L, due to the improved hydrolysis of coagulant and the more dissolved aluminium formed in this pH range (8, 13, 14). However, at pH values lower than 7.5, the residual aluminum concentrations were lowest than the threshold value of 200 μg/L permitted by the legislation guidelines. The lower residual aluminum values were measured at pH values lower than 7.4.

CONCLUSIONS

The objective of this work was to examine the efficiency of two coagulants, alum and poly-aluminium chloride (PACl), for the treatment of surface (river) water in a full-scale drinking water plant. The addition of both coagulants was studied by applying a conventional treatment system, including coagulation, sedimentation, and sand filtration (filter bed), as well as the alternative treatment system of direct filtration, i.e., coagulation and sand filtration, without the intermediate sedimentation; furthermore, the performance of single and dual filter beds was comparatively evaluated. PACl proved to be slightly more efficient coagulant than alum. During the conventional treatment scheme, the addition of lower dosages of PACl than alum, i.e., 1.35 mg Al/L as compared to 1.70 mg Al/L respectively, resulted in an effluent water of similar quality in terms of residual turbidity. Although this difference in the respective concentrations is not high enough, considering the high volume of raw (surface) water to be treated, the difference between them in terms of quantities used from the respective reagents become substantially higher, resulting also to smaller sludge production in the case of PACl use. Additionally, PACl can work efficiently in terms of residual turbidity and aluminium concentrations for slightly higher pH value (i.e., 7.4, instead of 6.9–7.2 for the case of alum), resulting to smaller acid consumption for pH correction. Also, PACl solution is expected to behave rheologically better than alum during the lower winter temperatures.

Direct filtration process was more efficient during the addition of PACl, than by the addition of alum. The effluent from the dual filter bed presented substantially lower turbidity, about 0.1 NTU, when PACl was used; in addition, the operational time of filter beds, i.e., the time between subsequent backwashing stages, was extended to a prolonged duration period of more than 24 h. The dual filter beds presented in this case a slow clogging rate in comparison to the single bed, resulting to the higher duration of filtration cycles and reaching up to 3 times the filtration cycle period of alum-using filter beds.

The examination of dual bed effluent pH values revealed that pH values higher than 7.5 resulted in an effluent with a residual aluminium concentration exceeding 200 μg/L, which is the maximum concentration limit permitted by the corresponding drinking water legislation. However, small variation of pH values were not found to affect particularly the system operation, or the turbidity of the effluent.

Summarizing, PACl has similar efficiency as alum in slightly lower dosages during the conventional treatment scheme. Furthermore, optimization of direct filtration process when using alum cannot be achieved, whereas PACl can offer better and stable operation of direct filtration than alum in terms of filtration cycles and turbidity.

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