Elimination of organic matter by optimising coagulation treatment: Case of water from the Sidi Yacoub dam, Algeria

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Abstract: The presence of natural organic matter (NOM) in water has a significant influence on water treatment processes. Water industries around the world consider coagulation/flocculation to be one of the main water treatment methods. The chief objective of conventional coagulation-based processes is to reduce the turbidity of the water and to remove natural organic matter (NOM) present in solutions. The aim of this paper is to present some developments in terms of improved coagulation for the drinking water of Sidi Yacoub treatment plant located in the Northwest of Algeria.

The experiments involved studying the effects of the application of two coagulants (ferric chloride and aluminium sulphate) on the removal of turbidity and natural organic matter from water by measuring the chemical oxygen demand (*COD*) and the UV absorbance at 254 nm. The results showed that the rate of turbidity removal increased from 81.3% to 88% when ferric chloride was applied and from 89.91% to 94% when aluminium sulphate was applied. For NOM removal, the maximum removal rates of *COD* and UV_{254} were 48% and 52%, respectively, in the case of ferric chloride. These rates increased to 59% and 65% after optimised coagulation. When aluminium sulphate was used, the rate of removal in water increased from 43% to 55% for *COD* and from 47% to 59% for UV_{254} after optimised coagulation. The combination of the two coagulants at equal dosage shows a slight improvement in the values obtained after optimisation, both in terms of turbidity and the NOM.

Keywords: aluminium sulphate, coagulation, ferric chloride, organic matter, Sidi Yacoub dam, turbidity

INTRODUCTION

Organic matter (OM) is a ubiquitous component of terrestrial (soils, sediments [MacCarthy 2001]), aquatic (surface water [Dittmar, Stubbins 2014] and groundwater [Artinger *et al.* 2000]), and anthropogenic (wastewater treatment plants [Imai *et al.* 2002], landfills [Kang *et al.* 2002]) environments. Its origin and composition remain specific to each environment.

Natural organic matter (NOM) is classified into non-humic and humic substances. All recognisable plant debris and organic compounds, such as polysaccharides, lignin, proteins, and polypeptides, are classified as non-humic substances. Other highly processed, brown to black materials with high molecular weight are considered humic substances [Sparks 2003; Tipping 2002].

The presence of NOM can colour the water, causing aesthetic effects, and react with disinfectants and oxidants, thereby causing the formation of disinfection by-products (DBPs) and other by-products that may have an impact on health [Croue *et al.* 2000; Edzwald 2011; Joseph *et al.* 2012]. NOM can cause contamination and interfere with certain water treatment processes. In addition, NOM can have effects on the quality of water in the distribution network [Edzwald 2011; Jacangelo *et al.* 1995; Lu, Chu 2005; Qaiser *et al.* 2014]. Several water treatment processes focus on the removal of NOM. This means that NOM is important for multiple reasons [Edzwald 2011].

It is generally believed that the most common and economical processes for NOM removal are coagulation and flocculation, followed by sedimentation/flotation, and sand

filtration [JACANGELO *et al.* 1995]. Coagulation-flocculation is a treatment process where colloids in water are destabilised so that they can aggregate and be physically removed, which effectively reduces turbidity [IRFAN *et al.* 2017; SOROS *et al.* 2019]. Unfortunately, conventional coagulation technology has clearly failed to provide water of the required quality [CUI *et al.* 2020]. Therefore, improved and/or optimised coagulation has been suggested as the primary treatment option for better control of NOM and hence disinfection by-product (DBPs) formation [MATILAINEN *et al.* 2010].

In this article, we present the results of coagulation treatment experiments carried out on the water of the Sidi Yacoub dam, in order to assess the importance of removing NOM.

In order to deal with the taste and sometimes unpleasant odour of the water produced by the Sidi Yacoub treatment station, caused mainly by the presence of certain organic matter contents in the raw and treated water, samples of raw water were collected at different times of the year. They subsequently underwent coagulation optimisation tests, which involved manipulating with two coagulants: ferric chloride and aluminium sulphate.

MATERIALS AND METHODS

SAMPLING

The experiments were conducted on raw water used to supply the drinking water production plant and originating from the Sidi Yacoub dam.

It is known that the character and concentration of NOM in raw water sources vary regionally and seasonally [FABRIS *et al.* 2008; SHARP *et al.* 2006]. In order to assess the quality of the water, both in the dry and in the rainy season, seasonal samples of raw water were collected on their arrival at the treatment plant. The samples were collected and stored according to standard methods for examining drinking water. The main physicochemical characteristics of these water samples were presented in Table 1.

PREPARATION OF SOLUTIONS

Metal salts such as ferric chloride and aluminium sulphate are widely used in coagulation, due to their high efficiency for water treatment. In solutions, these salts are characterised by their high electropositivity, which is responsible for the neutralisation of particle charges.

The solutions of ferric chloride $FeCl_3$ and aluminium sulphate $Al_2(SO_4)_3$ were obtained from the commercial sub-stances $FeCl_3 \cdot 6H_2O$ and $Al_2(SO_4)_3 \cdot 18H_2O$. We dissolved 10g of each substance in distilled water. This solution was freshly prepared and stored at 4°C to preserve it in the same state. The pH of the samples was adjusted using either strong acid (HCl) or strong base (NaOH).

ANALYTICAL METHODS

The coagulation test was performed using "JAR TEST pro LABO" jar test equipment. The samples were kept at room temperature and then shaken carefully to prevent solids from settling. We poured 500 cm^3 of the test sample into six beakers, which were then stirred simultaneously. The raw water and the coagulant were subjected to rapid stirring at 170 rpm for 2 minutes. The speed was then reduced to 40 rpm for a period of 20 min. After that, 30 min was allowed for settling (phase during which the destabilised floc is driven to the bottom of the beaker). Then, a sample was taken from each beaker for subsequent measurements and testing. The turbidity measurement was made on the water's edge, at 2 cm below the surface.

The pH of the samples was measured using a pH meter with a combination of pH electrodes (WTW pH inoLab LEVEL1). The calibration was carried out with two buffer solutions with pH of 6.86 and 9.18, respectively. A turbidity meter (model 2100N Hach) was used for a direct reading of the turbidity of a sample in a nephelometric turbidity unit (NTU). The electrical conductivity was measured with a conductivity meter (salt conductivity meter LF 197) which directly measures the microsiemens per centimetre. The measurement of organic matter was evaluated on the basis of chemical oxygen demand (COD) using the reflux method in an open system. Some materials in water oxidise at 150°C, due to an excess of potassium dichromate in an acidic medium [RODIER 2009]. The UV_{254} absorbance was determined using the 9200UV/VS spectrophotometer.

RESULTS AND DISCUSSION

COAGULATION WITH FERRIC CHLORIDE (FECL$_3$) AND ALUMINIUM SULPHATE (AL$_2$ (SO$_4$)$_3$)

ELIMINATION OF TURBIDITY

The dosage of the coagulant is one of the most important parameters to consider in the process of coagulation and flocculation. Insufficient dosage or overdose result in poor performance of the coagulant during flocculation. Therefore, the optimal dosage of the coagulant should be determined to

Table 1. Summary of physicochemical quality parameters of raw water from the dam at the time of experiment

Parameter	Unit	Sample 1	Sample 2	Sample 3	Sample 4
Turbidity	NTU	7.88	20.0	33.1	68.4
pH	–	7.2	7.54	8.0	7.7
Electric conductivity	$\mu S \cdot cm^{-1}$	1473	1516	1506	1385
COD	mg $O_2 \cdot dm^{-3}$	25.0	35.0	43.5	47.5
Absorbance UV_{254}	cm^{-1}	0.677	0.935	1.202	1.278

Source: own study.

minimise the cost of coagulant and achieve optimal elimination rates [ABU HASSAN et al. 2009].

The effects of the coagulant dosage on the removal of turbidity using ferric chloride and aluminium sulphate are shown in Fig. 1. The tests were carried out by varying the dosage of the coagulant at a constant pH value. The optimum aluminium sulphate concentrations obtained in our study were 18, 25, 35, and 55 mg·dm^{-3}, respectively, for the sample 1, 2, 3 and 4. With ferric chloride, these concentrations were adjusted respectively to 16, 20, 30 and 55 mg·dm^{-3} for the same samples. These doses were previously optimised. However, the maximum turbidity removal rates for the four samples varied depending on the quality of the raw water and the type of the coagulant used.

In the case of ferric chloride, the highest percentage of turbidity removal was observed when raw water turbidity was low. Elimination rate decreases as the turbidity of the raw water increases, due to the higher dosage of the coagulant, which gives its own brownish colour to the treated water. Compared to ferric chloride, aluminium sulphate achieves lower removal rates at low levels of raw water turbidity. However, removal rates are more important for high turbidity values. Therefore, the turbidity removal rates for ferric chloride and aluminium sulphate were 63 at 81.3% and 66.24 at 89.91%, respectively.
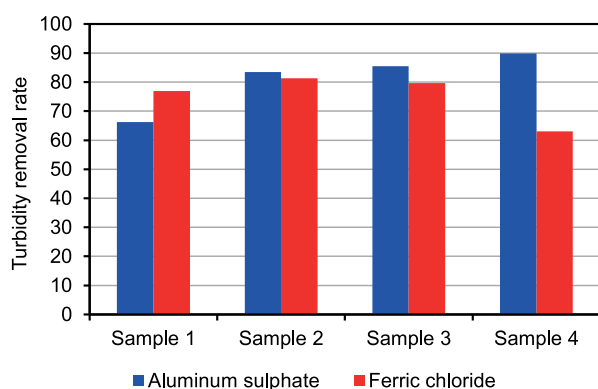

Fig. 1. Evolution of the turbidity elimination rate; source: own study

Elimination of organic matter

Historically, coagulation has been used in water treatment to reduce turbidity, neutralise colour, and eliminate pathogens. However, the optimal conditions for turbidity or colour removal are not always the same as those for NOM removal [BUDD et al. 2004; YAN et al. 2008; YU et al. 2007]. In basic coagulation, the coagulation conditions are optimised for the removal of turbidity, while the optimised coagulation refers to dose and pH conditions optimised in particular for the reduction of organic matter.

Organic matter is generally defined by the measurements of total organic carbon, dissolved organic carbon, UV absorbance at 254 nm, or by the oxidability in an acidic and hot medium [LEFEBVRE 1995; YAMADA et al. 2019]. In this study, we rely on the measurements of the chemical oxygen demand (COD) and the UV absorbance at 254 nm as parameters for organic matter quantification.

Figure 2 illustrates the effects of the dosage of ferric chloride and aluminium sulphate on the elimination of NOM. For ferric chloride, the NOM elimination rates corresponding to the optimal dosages varied from 28 to 48% and from 21.42 to 52%,

respectively, with COD and UV absorbance at 254 nm. The use of aluminium sulphate shows a variation between 24 at 43% and 19.5 at 47%, respectively, with COD and UV absorbance at 254 nm.

Compared to aluminium sulphate, ferric chloride showed the greatest reduction in UV$_{254}$ absorbance and COD in all four samples studied.
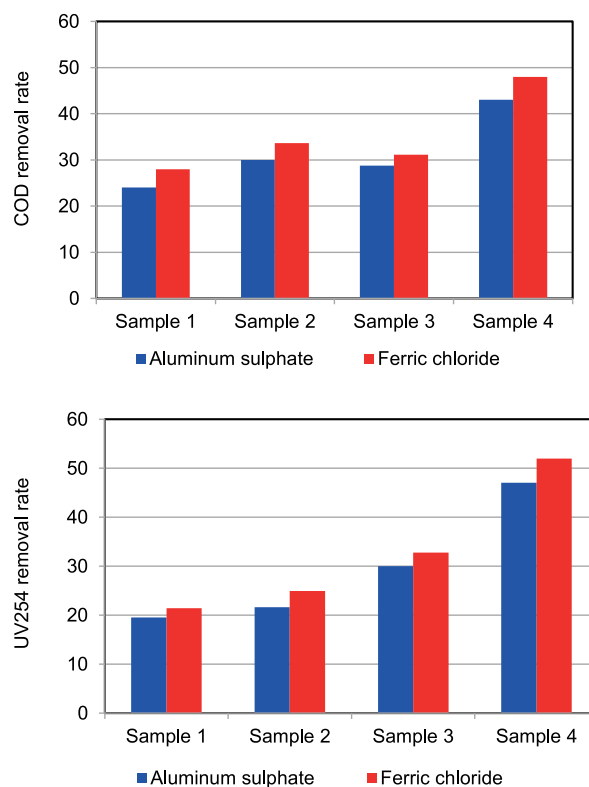

Fig. 2. Evolution of the chemical oxygen demand (COD) and UV$_{254}$ elimination rate; source: own study

Influence of pH ON COAGULATION

Studying the effect of pH on coagulation is essential for determining the optimum pH for elimination. A pH range of 4 to 9 was chosen for the experiments. The doses of ferric chloride and aluminium sulphate were held at their optimal turbidity removal levels. The effects of pH on the removal of turbidity and NOM using ferric chloride and aluminium sulphate are shown in Figure 3a, b, c, d.

Turbidity elimination

The elimination of turbidity using the two coagulants, ferric chloride and aluminium sulphate increased with increasing acidity until reaching a maximum at pH value close to 5 in the case of ferric chloride and close to 6 in the case of aluminium sulphate. The elimination rate decreased above these levels.

The highest percentage of turbidity removal relative to each test reached 88% and 94% for ferric chloride and aluminium sulphate, respectively. The pH range was 4.5 to 5.5 for ferric chloride and 5.5 to 6.5 for aluminium sulphate. The rate of removal by ferric chloride was lower for high turbidity values in raw water, due to the increased dosage of the coagulant which induces its own colour.
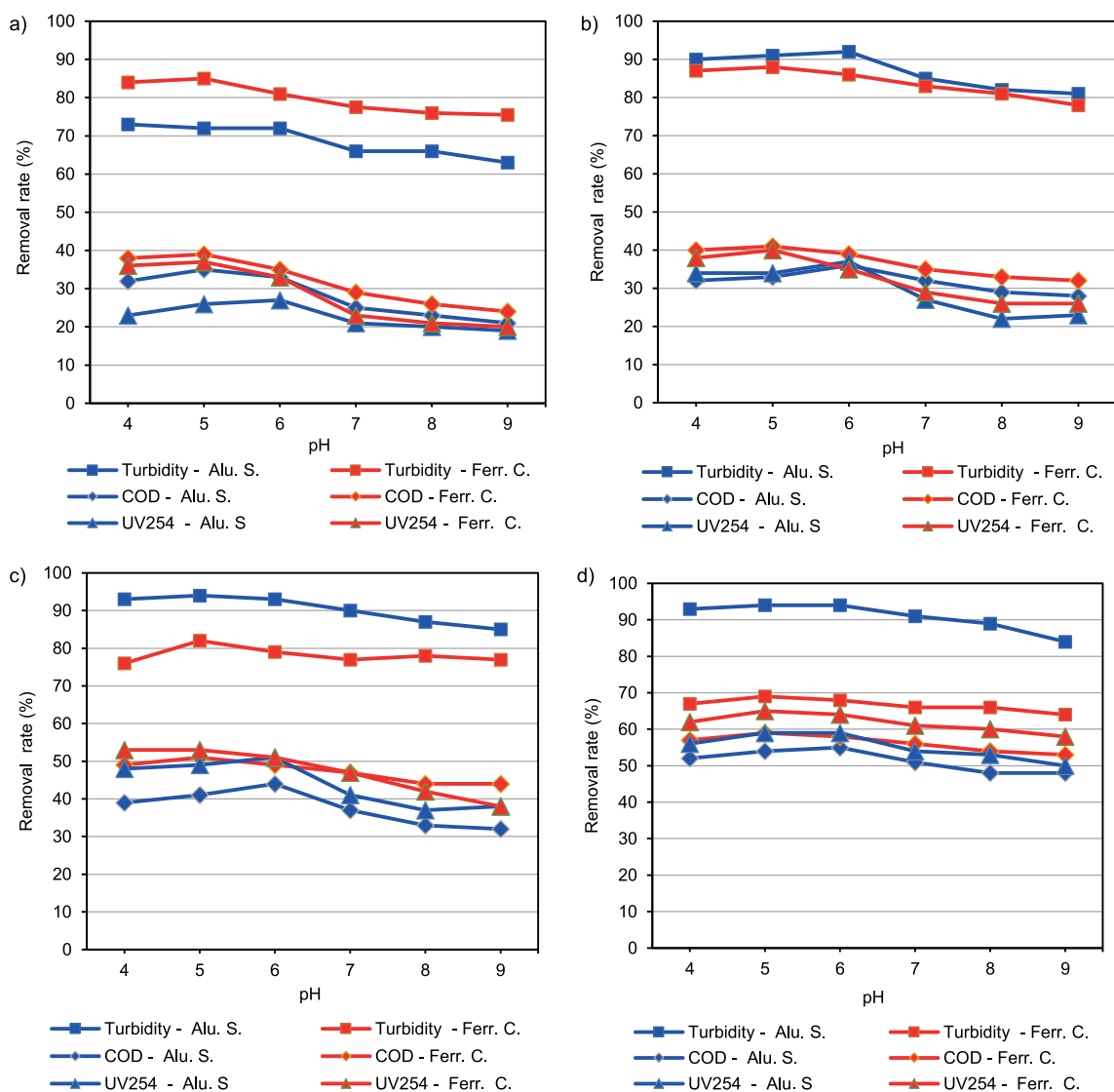
Fig. 3. Variation of removal rate of turbidity, chemical oxygen demand (*COD*) and UV$_{254}$ as a function of the pH: a) sample 1, b) sample 2, c) sample 3, d) sample 4; Alu. S. = aluminium sulphate, Ferr. C. = ferric chloride; source: own study

Elimination of organic matter

The effect of pH on the operation of the two coagulants was highly noticeable. Removal rates were found to be limited under alkaline conditions as well as at neutral pH. When the pH value was reduced below 7, the drop in *COD*, and UV$_{254}$ was remarkable, and it was found to be most effective in acidic pH values. The maximum reduction of *COD* and UV$_{254}$ measured was 59% and 65% for ferric chloride and 55% and 59% for aluminium sulphate. The dosage of the two coagulants was high.

It was established that the optimum pH level in ferric chloride-based coagulation is between 4.5 and 6 and the optimum pH for coagulation using aluminium sulphate is between 5.0 and 6.5.

Finally, in terms of average values, the turbidity removal rate increased from 81.29% to 88.25% for aluminium sulphate and from 75.23% to 81.00% for ferric chloride after optimised coagulation. However, the *COD* removal rate was not very high and increased from 31.43% to 42.50% for aluminium sulphate and from 35.19% to 47.50% for ferric chloride. As far as the UV$_{254}$ absorbance is concerned, the values of this parameter were similar to those found in the case of *COD* and increased from

29.53% to 43.5% when aluminium sulphate was used and from 32.78% to 48.75% when ferric chloride was used. Our results are consistent with the study carried out by VOLK *et al.* [2000] on surface water coagulation.Volk found that there was an increase in an average removal rate from 29% to 43% for *COD* while turbidity removal rates with the use of aluminium and ferric salts reached 93% and 82% respectively, after optimised coagulation. In optimised coagulation on river water [SILLANPÄÄ *et al.* 2018], the average removal rates of *COD* and UV$_{254}$ with aluminium sulphate were 43.2% and 41.2%, respectively. When ferric chloride was used, the removal rates for *COD* and UV$_{254}$ were 57.9% and 47.6%, respectively.

Combination of ferric chloride with aluminium sulphate

In this part, the third and fourth samples were each subjected to a jar test, with equal doses of ferric chloride and aluminium sulphate. The doses applied corresponded to the doses of the coagulant that achieved maximum turbidity elimination in the baseline coagulation test. This translates into doses of

17.5 mg·dm^{-3} and 27.5 mg·dm^{-3} for each coagulant in the combination test made on the third and the fourth sample, respectively. The pH was reduced and kept at 5.5 for both tests.

Figure 4 illustrates the results obtained. Optimal turbidity removal shows an average rate of 96%. The corresponding *COD* was reduced by an average of 56.5% and UV$_{254}$ absorbance by 64%. Compared to the basic coagulation, the recorded rates indicate an efficient removal of turbidity and the NOM.
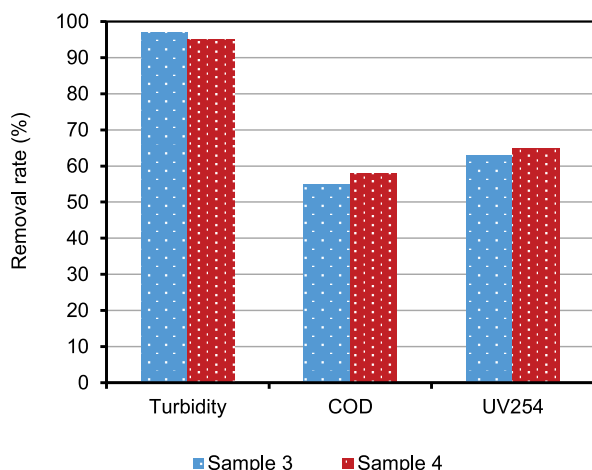

Fig. 4. Removal rate of turbidity, chemical oxygen demand (*COD*) and UV$_{254}$ in combination of ferric chloride with aluminium sulphate; source: own study

CONCLUSIONS

In this study, the coagulation process was used to treat water samples collected on the arrival of water at the classic treatment station of Sidi Yacoub. Coagulation and, in particular, improved or optimised coagulation, is considered to be the main treatment option applied to prevent the deterioration of the quality of raw water. This process involves the destabilisation of the colloidal particles by neutralising the forces which keep them away from each other. Cationic coagulants impart positive electrical charges to reduce the negative charge of colloids. As a result, the particles collide to form macro-particles called flocs.

The experiments were carried out on four samples collected throughout the year in order to determine the optimum conditions for coagulation using two conventional coagulants: ferric chloride and aluminium sulphate. These two coagulants behaved differently; ferric chloride was found to be much more efficient at reducing *COD* and UV$_{254}$, while aluminium sulphate was efficient at reducing turbidity.

The study of the effect of pH on coagulation demonstrated that, in general, elimination increases when the pH is lowered in the presence of multivalent cations. The optimum range for this elimination was 4.5 to 5.5 for ferric chloride and 5.5 to 6.5 for aluminium sulphate. The maximum turbidity removal rate reached 94% with the use of aluminium sulphate and 88% with ferric chloride.

As far as the elimination of NOM is concerned, both coagulants have also shown that the reduction in NOM increases, as the acidity increases until it reaches a pH of around 4.5. Ferric chloride achieved a slightly greater reduction in *COD* and UV$_{254}$ compared to aluminium sulphate.

The maximum *COD* and UV$_{254}$ removal rates amounted to 48% and 52%, respectively. In the baseline coagulation in the case of ferric chloride, the values were 59% and 65%. Aluminium sulphate evolved from 43% and 47% in baseline coagulation to 55% and 59% for *COD* and UV$_{254}$ respectively, in optimised coagulation.

The combination of the two coagulants at equal dosage shows a slight improvement in relation to the values obtained after optimisation. However, it can be a good option for coagulation treatment in the case of high turbidity of raw water, as it would ensure a sufficient NOM removal rate.

Finally, for raw water with moderate or high NOM concentrations, optimised coagulation can be an efficient and inexpensive tool for controlling NOM in water. Therefore, it can be considered a good option for the treatment of the water examined in the study.

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