# EFFECT OF FLOCCULATION-ADSORPTION COMBINATION ON THE PURIFICATION OF SURFACE WATER: A CASE STUDY OF SIDIYACOUB DAM WATER, ALGERIA

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# ABSTRACT

An increase in the amount of Natural organic matter (NOM) was observed over the last 10 to 20 years in raw water supplies, which has a significant effect on the treatment of drinking water. The most common and most profitable disposal methods of the NOM are coagulation and flocculation followed by sedimentation and sand filtration. Nevertheless, there are some organic compounds, which cannot be removed completely in a coagulation step. An alternative for better elimination of NOM was studied in this work. The use of adsorbents such as activated carbon and bentonite as additives for flocculation significantly reduced the organic matter content.

**KEY WORDS** : Aluminum sulphate, Bentonite, Coagulation-flocculation, Powdered activated carbon, Turbidity

#### INTRODUCTION

Natural organic matter (NOM) is defined as a complex matrix of organic matter in natural waters (Matilainen *et al.*, 2010; Papageorgiou *et al.*, 2016). Aquatic NOM consists of a wide variety of organic compounds which are primarily derived from the plants residue and animals decomposition (Summers and Roberts, 1988; Joseph *et al.*, 2012; Papageorgiou *et al.*, 2016). In addition to this, a recent study showed that the concentrations of NOM have increased over the past decade in many rivers, including drinking water sources (Hruška *et al.*, 2009). As a result, the water sources used for drinking water production usually contain NOM (Matilainen *et al.*, 2010).

The presence of NOM in various sources of water is a major concern for scientists and environmental engineers, especially in the treatment of water (Kozyatnik *et al.*, 2010; Khaouane *et al.*, 2017). The NOM often contributes to unpleasant taste and odors in potential sources of drinking water (Matilainen *et al.*, 2005; Yan *et al.*, 2008; Zhan *et al.*, 2010; John C. Crittenden, 2012; Joseph et al., 2012) and acts as a support for metals and various harmful organic chemicals (Jacangelo et al., 1995). In addition, NOM can disrupt various processes in a conventional water treatment installation (Joseph et al., 2012; Ivanèev-Tumbas, 2014). It affects floc formation during the coagulation process, which often generates small flakes (Parsons et al., 2004). NOM is considered a precursor of by-products of the carcinogenic disinfection such as trihalomethanes and haloacetic acids that can be formed during chlorination (Rook, 1977; Weinberg et al., 2002; Singer, 2006; Chowdhury et al., 2009; American Water Works Association, 2011; Kristiana et al., 2011; Mahdavi et al., 2017) and contribute to bacterial regrowth and biofilm formation in water distribution networks (Jacangelo et al., 1995; Lu and Chu, 2005; Qaiser et al., 2014).

The removal of organic matter from aqueous environment has been widely studied (Choo *et al.*, 2008) and the development of technologies for the removal of NOM from water is of great importance (Wang *et al.*, 2011; Mahdavi *et al.*, 2017).

Conventional water treatment processes are mainly composed of coagulation, sedimentation and filtration by gravity. Coagulation is an essential process for the removal of various particles and organic matter (Tomaszewska *et al.*, 2004; Zouboulis *et al.*, 2007; Zhan *et al.*, 2010; Mahdavi *et al.*, 2017). Nevertheless, there are some organic compounds, which cannot be removed completely in a coagulation step (Zhan *et al.*, 2010). In addition, conventional treatment processes are primarily designed for the reduction of turbidity and inactivation of pathogenic microorganisms (Parsons *et al.*, 2004). These processes are regularly faced with difficult water conditions so that the removal of the NOM may be compromised (Jarvis *et al.*, 2005).

In such cases, other methods are required (Zhan *et al.*, 2010). The adsorption on activated carbon and anion exchange are two recommended technologies for the removal of the NOM from water (Graf *et al.*, 2014). A wide variety of adsorbents were studied to eliminate NOM from water, including activated carbon (Bjelopavlic *et al.*, 1999), as well as some new nanoparticles such as carbon nanotubes (Lu and Su, 2007). The use of activated carbon as adsorbent is an effective method to eliminate NOM and has been widespread used (Najm *et al.*, 1998; Jarvis *et al.*, 2005).

The further development of the adsorbents was studied. Many researchers have used materials such as cane bagasse, peanut shell, apple pomace, wheat straw, sawdust, coconut, orange zest, soft banana and pine bark, as adsorbents to remove dyes, heavy metals, nitrate, etc ...(Zhan *et al.*, 2010). These materials showed a good adsorption capacity for different dyes organic acids (Orlando *et al.*, 2002).

Experimental procedures carried on combined treatment (coagulation and adsorption) have shown that this technique can be effective in eliminating NOM from various types of water sources as well as the elimination of other organic contaminants from potential sources of drinking water, and also acts on improving the settling of flocs formed (Zhan *et al.*, 2010; Joseph *et al.*, 2012). Removal percentages obtained with the process of combined treatment vary from 45 to 80% (Najm *et al.*, 1998; Uyak *et al.*, 2007; Álvarez-Uriarte *et al.*, 2010).

In Algeria, the city of Chlef supplies drinking water primarily from a conventional treatment plant which proceeds in the following stages: prechlorination, coagulation, flocculation, sedimentation, gravity filtration through granular bed, and disinfection. Yet a growing number of consumers prefer drinking spring water delivered by tankers or bottled water. The reasons for this are mainly aesthetic and closely related to tastes and smells. Tests carried on water for consumption showed the presence of significant amounts of organic matter.

This work focuses on improving the quality of feed water from the city of Chlef, through a combination of coagulation with aluminum sulphateand adsorption by one of the two materials, activated carbon and bentonite.

Water samples were taken on arrival at the station throughout the study period and transported to the analytical laboratory. Some measurements were taken on site.

# MATERIALS AND METHODS

#### Quality of the raw water

Waters object of this study were extracted on arrival at the treatment plant to provide a range of physical and chemical characteristics of the raw water. Turbidity, pH, electrical conductivity, mineralization, nitrogen compounds, chemical oxygen demand, absorbance  $UV_{254'}$  total organic carbon (TOC) and oxidizabilityto KMnO<sub>4</sub> in water were measured (Table 1).

**Table 1.** Summary of physico - chemical quality<br/>parameters of the raw water from the dam at<br/>the time of the experiment

Parameter	unit	Max - Min
Turbidity	NTU	75.4–7.88
pH	-	8.30 - 7.25
Electric Conductivity	µs/cm	1520 - 1184
TDS	mg/l	1394 - 1193
NO <sub>3</sub> <sup>-</sup>	mg/l	2 - 0
NH <sub>4</sub> <sup>+</sup>	mg/l	0.1 - 0
COD	$mg O_2/l$	68.0-13.0
absorbance UV <sub>254</sub>	cm <sup>-1</sup>	1.847 -0.308
SUVA	l/m.mg C	3.81 - 2.55
Oxidizability to KMnO <sub>4</sub>	$mg O_2/l$	47.0–9.0

SUVA is the ratio of the UV absorbance at 254 nm to the total organic carbon (TOC) content of the sample (Rodier, 2009; American Water Works Association, 2011; Klymenko *et al.*, 2014). Water with high SUVA values of 4 or greater indicate that the NOM is composed mainly of aquatic humic matter. Raw waters with SUVA of 2 to 4 contain a mixture of aquatic humic matter and non-humic NOM. Raw waters with SUVA < 2 contain mainly non-humic

material (American Water Works Association, 2011).

# **Preparation of Solutions**

The aluminum sulphate stock solution 10 g/l of  $Al_2(SO_4)_3$ .18H<sub>2</sub>O, was prepared from powdered commercial aluminum sulphate, used for water treatment. This solution is produced by adding 10g of aluminum sulphate to 1 liter of ultrapure deionized water, and stirred for overnight. The solution was renewed and stored in the refrigerator for the duration of the study.

The adsorbents used, powdered activated carbon (inert microporous carbon, trademark Fluka, where 75% of the particles have diameters  $\pounds$  40 mm) and bentonite (pretreated local powder gray clay) were prepared from adding 100 mg of each adsorbent in 1000 ml of deionized water and stirring the solution on a magnetic stirrer at 600 rotations per minute (r.p.m).

#### **Analytical Methods**

# General parameters of water quality

The sampled pH was measured using a pH-meter with a combination pH-electrode (WTW pH inoLab LEVEL1). The calibration was performed with two buffer solutions having respectively a pH of 6.86 and pH 9.18. A Turbid-meter (2100N model Hach) was used for a direct reading of the turbidity of a sample in nephelometric turbidity unit (NTU).

The electrical conductivity was measured by means of conductivity meter (salt meter conductivity meter LF 197) that measures directly micro Siemens/centimeter ( $\mu$ s/cm). We also use the conductivity meter to measure the sum solids dissolved in water (TDS). The determination of the nitrogen compounds were carried out by the molecular absorption spectrometric method (Rodier, 2009).

Measuring the organic material was evaluated on the basis of the oxidizability to potassium permanganate (KMnO<sub>4</sub>) which consists of a hotchemical oxidation in an acid medium (Rodier, 2009), absorbance UV<sub>254</sub> analyzed using 9200UV/ VS Spectrophotometer, TOC was determined by BioTector B7000-HACH and the chemical demand of oxygen (COD) using the reflux method in an open system. Some matters contained in the water oxidize to 150 °C boilingdue to an excess of potassium dichromate in an acidic medium (Rodier, 2009).

### Jar test

A "JAR TEST pro LABO" variable speed, with six

flat blades agitator was used and which results in six beakers 01 liter of water to be examined, strictly identical and adjustable stirring during the same duration through a central control.

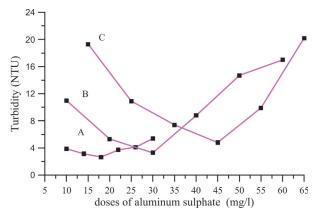
During the experiments, the raw water and coagulant are subjected for 2 min at rapid stirring to 170 r.p.m. The speed is subsequently reduced to 40 r.p.mfor a period of 20 min. After, a 30 min settling (a phase during which destabilized floc is driven towards the bottom of the beaker).

Afterwards, a sample was taken from each beaker for measurements and subsequent trials. The turbidity measurement is made on the water surface fringe at 2 cm.

#### **RESULTS AND DISCUSSION**

#### Coagulation treatment without additives

Three jar tests with aluminum sulphate alone were carried out on raw water arriving at the SidiYacoub treatment plant for different periods of time. The results of the measurement of Turbidity, COD,  $UV_{254}$  and oxidizability to KMnO<sub>4</sub> before and after treatment are shown in Table 2 and presented in Figures 1 and 2.



**Fig. 1.** Turbidity variation according to the aluminum sulphatedose (A : Trial 1 , B : Trial 2 , C : Trial 3)

La Figure 1 montre l'effet de la dose de sulfate d'aluminium sur le traitement de l'eau. Une élimination maximale de la turbidité est obtenue pour différentes doses de sulfate d'aluminium. Turbidity is reduced with increasing dose of flocculant, reaching a minimum where the removal rate reaches 66.24%, 83.50 and 85.50% for the three respective trials. Beyond that it increases, because of exceeding the corresponding optimal dose.

The measure of oxidabilityto KMnO<sub>4</sub> and COD

Trial N°	Turbidity		Natural organic matter (NOM)						
	R. W.	T. W.	$COD (mg O_2/L)$		UV <sub>254</sub> (cm <sup>-1</sup> )		Oxidiz	Oxidizability to	
			R. W.	T. W.	R. W.	T. W.	$KMnO_4 (mgO_2/L)$		
							R. W.	Ť. W.	
1	7.88	2.66	25.0	19.0	0.677	0.545	16.2	11.3	
2	20.0	3.3	35.0	24.5	0.935	0.733	22.0	17.5	
3	33.1	4.8	43.5	31.0	1,202	0,841	28.0	21.4	

**Table 2.** Results of coagulation treatment alone with Aluminium Sulphate

R. W.: Raw water ; T.W.: Treated water

expresses the content of dissolved organic matter. The  $UV_{254}$  absorbance makes it possible to characterize the presence of organic molecules (aromatic nuclei in particular) which absorb ultraviolet light (Figure 2).

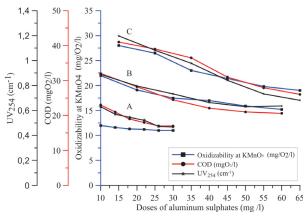


Fig. 2. Variation of the oxidizability to  $KMnO_4$ , COD and  $UV_{254}$  according to aluminum sulphate dose (A : Trial 1, B : Trial 2, C : Trial 3)

These three parameters follow similar paces and show a slight decrease as a function of the dose of aluminum sulphate. They were reduced with rates ranging from 20 to 30% depending on the maximum reduction in turbidity.

It is thus noted that the results of the measurements show that there is a strong correlation between the COD,  $UV_{254}$  and the oxidability to KMnO<sub>4</sub> in the treated water.

Finally, we deduce that the coagulationflocculation process with aluminum sulphate achieves a good performance turbidity removal but still not enough to reduce organic matter.

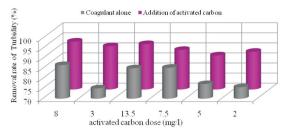
# Combined coagulation and adsorption using activated carbon and bentonite for NOM removal

Raw water samples taken regularly during the study period are first subjected to coagulation tests with aluminum sulphate, and then to coagulation treatments associated with adsorption: after having optimized the dose of coagulant for each sample following the jar test protocol, a second jar test is performed in order to determine the optimal dose of the adsorbent.

The results obtained are shown in Table 3.

# Turbidity elimination rate by coagulation associated with adsorption

The evolution of the turbidity is practically identical for the two adsorbents: By using the coagulant alone, the turbidity of the raw water has been considerably reduced. Levels ranging from 75.74 to 86.73% are recorded depending on the increase in the dose of the coagulant. After the addition of the adsorbents, a marked increase in the turbidity removal rates was observed for both adjuvants which reached 12.43%, except that the lowest turbidity after treatment was obtained for lower doses of activated carbon and this for all the tests carried out (Figures 3 and 4).



**Fig. 3.** Variation of the rate of elimination of turbidity as a function of the dose of activated carbon.

# NOM elimination rate by coagulation associated with adsorption

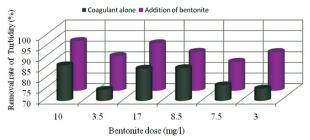
#### **Activated carbon Employment**

Figure 5 shows NOM removal rates as a function of activated carbon doses associated with aluminum sulfate coagulation.

The test jars used resulted in doses of activated carbon varying between 2 and 13.5 mg/l, depending

 Table 3. NOM removal and turbidity from various dam water samples using coagulation and combined adsorption using activated carbon and bentonite.

Samples					1	2	3	4	5	6
Raw	Turbidi	ty		NTU	63.3	15.3	75.4	28.1	20.7	16.9
water	COD			mg O <sub>2</sub> /l cm <sup>-1</sup>	44.0	17.5	68.0	36.5	32.0	13.0
	absorba	absorbance UV <sub>254</sub> Oxidizability to KMn O <sub>4</sub>			1.314	0.543	1.847	0.988	0.781	0.308
	Oxidiza				30.0	11.5	47.0	28.0	24.5	9.0
	SUVA		*	l/mg.m	-	3.81	-	-	2.9	2.55
Treated	first jar	Optimal		mg/l	40	20	45	30	25	16
water	test	dose of Al. S								
		Optimal Tur	bidity	NTU	8.4	3.8	11.2	4.1	4.7	4.1
		SUVA		l/mg.m	-	3.76	-	-	2.78	2.33
	second	Association	optimal	mg/l	8	3	13.5	7.5	5	2
	Jar test	of activated	dose of							
		carbon	activated							
			carbon							
			COD	$mg O_2/l$	11	6.5	15	9.5	11	5.5
			absorbance	cm <sup>-1</sup>	0.381	0.218	0.419	0.301	0.285	0.130
			UV <sub>254</sub>							
			Oxidizability	$mg O_2/l$	9.0	4.6	12.2	9.0	9.8	4.1
			to KMnO <sub>42</sub>							
			SUVA	l/mg.m	-	2.66	-	-	2.11	2.09
			Turbidity	NTU	3.9	1.3	5.6	2.9	2.7	1.9
		Association of	of							
		Bentonite								
			optimal dose	mg/l	10	3.5	17	8.5	7.5	3
			of bentonite							
			COD	$mg O_2/l$	16.28	8.40	22.44	14.24	12.80	6.76
	•		absorbance UV	V <sub>254</sub> cm <sup>-1</sup>	0.533	0.270	0.583	0.450	0.367	0.169
			Oxidizability	$mgO_2/l$	12.6	6.2	16.9	13.4	11.8	5.0
			to KMnO <sub>4</sub>							
			SUVA	l/mg.m	-	2.9	-	-	2.3	2.18
			Turbidity	NTU	4.2	2.1	5.8	3.3	3.4	2.0



**Fig. 4.** Variation of the rate of elimination of turbidity as a function of the dose of bentonite.

on the amount of organic matter in the raw water. By going further from optimal doses, the elimination of NOM is reduced due to desorption.

The combined technique of coagulationadsorption with activated carbon led to a considerable elimination of NOM with rates ranging from 55 to 78%.

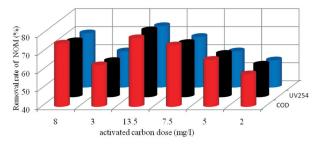
Slight increases in NOM elimination with increasing doses of activated carbon were observed.

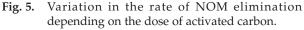
# The bentonite Employment

Figure 6 shows NOM removal rates as a function of bentonite doses associated with aluminum sulfate coagulation.

The test jars used resulted in doses of bentonite varying between 3 and 17 mg/l.

The NOM removal percentages were low in this case. They vary between 44 and 67%. Likewise, an increased rate of NOM removal was observed with increasing doses of bentonite.





#### CONCLUSION

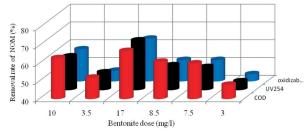
Experimental procedures were performed to investigate the individual use and the one associated with coagulation adsorption, in order to assess the effectiveness in improving the produced water quality by the drinking water station of Chlef (Algeria) and mainly on the elimination of NOM.

It was found that both treatment methods have high removal efficiency of turbidity (66.24 to 86.73 % in the individual employment and 86.96 to 93.84 % through the associated use with the activated carbon and 83.57 to 93.36 % with Bentonite). This also indicates that the combination of coagulation and adsorption can enhance the removal of the turbidity of the treated water and also acts on the improvement of the flocs formed settling.

In terms of the organic matter removal, we find out that the reduction of the organic material during the use of the individual coagulation was only 20 to 30 %, whereas coagulation combined with the adsorption was more effective. The best results were recorded with activated carbon. The reduction of the organic material was 58 to 78% according to the COD measurement and 55 to 74 % according to the oxidizability to KMnO<sub>4</sub> and this for an activated carbon dosage varying from 2 to 13.5 mg/l. Nevertheless, Bentonite also improves the removal efficiency of the organic material up to 67 % as measured by COD, and 64 % compared to the oxidizability to KMnO<sub>4</sub>, for a dose ranging from of 3 to 17 mg/l of bentonite. Beyond these values, there is desorption.

The NOM removal efficiency based on  $UV_{254}$  absorbance measurements evolved in almost a similar fashion as that of COD and oxidizability to KMnO<sub>4</sub>, for both adsorbents used. The organic matter removal rates were 58 to77% for activated carbon and 45to 68% for bentonite.

Another important parameter describes the nature of NOM in the water sample in terms of hydrophobicity and hydrophilicity is the specific



**Fig. 6.** Variation in the rate of NOM elimination depending on the dose of bentonite

ultraviolet absorption (SUVA). The raw water SUVA average was 3.09 l/m.mg C, reduced to 2.96 after aluminum sulphate coagulation, became 2.29 and 2.46 l/m.mg C, after combining the activated carbon and the bentonite respectively. This indicates that the raw water was constituted of a varied mixture of hydrophobic (humic) and hydrophilic (non-humic) organic compounds. The treated water is dominated by non humic substances.

We see also a small reduction in SUVA following the coagulation treatment. This is due to the low value of SUVA of the raw water on the one hand and on the other hand; because water treatment processes such as coagulation and adsorption will preferentially suppress aquatic humic matter.

It is also interesting to note that in this study, activated carbon has somewhat higher affinities for hydrophobic humic substances compared to bentonite.

Finally, the combination treatment with activated carbon showed a considerable improvement in the treatment effectiveness. The use of adsorbents characterized by clay properties, such as bentonite, may also be an attractive solution to replace the carbon if required.

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