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Statistical modelling of turbidity removal applied to non-toxic natural coagulants in water treatment: a case study

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ABSTRACT

An investigation into two non-toxic natural coagulants abundantly growing in different countries, cactus (*Opuntia spp.*) and okra was performed on monthly river water samples (one-year period). The studied case was the Euphrates river/Al-Mashroo canal/Iraq. Six statistical models were interpreted and tested describing the residual turbidity after coagulation-flocculation for the three studied cases (optimum-coagulant-dose, optimum-flocculator-velocity-gradient and optimum-flocculation-time). According to the environmental parameters recorded during the study and the statistical analyses, two facts were concluded. The first fact was that controlling the optimum-flocculator-velocity-gradient of the coagulation-flocculation process gave the highest contribution ratio of the models. The second fact was that the most significant environmental parameter (statistically) in the coagulation-flocculation process was the initial turbidity. This was proved for the two natural coagulants under study. Also, from the results of the study, it was concluded that the two natural coagulants were of similar coagulation-flocculation properties, and they were competent for turbidity removal.

Keywords: Natural coagulants; Coagulation-flocculation; Turbidity removal; Statistical modelling

1. Introduction

The coagulation process, which is a process of destabilising colloids by reducing the zeta potential to agglomerate the colloids together forming small flocs, is a central part of chemical treatment of water and wastewater [1–5]. The addition of chemical coagulants into the aqueous media could be done either chemically (directly addition of solid chemicals) or electrically (electrochemical methods) [6,7]. The direct addition method requires larger amount, of coagulants, than the electrochemical methods [6]. In addition, the direct addition method produces large volumes of sludge [5,6]. Then, the generated flocs will be removed from the solution using one of a number of different methods [8–10]. Although, a large number of chemicals have been used as coagulants, aluminium sulphate (alum) is the commonly used coagulant in many countries [6,11,12]. Recently there have been many rumours about a possible link between high levels of residual aluminium and several health issues, such as the development of Alzheimer's disease [13]. In addition, the traditional chemical coagulants generate a huge amount of sludge, which in

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turn increases the operating cost of treatment plants, because the solid wastes require expensive and complex handling and management systems [5,14,15]. Therefore, these negative effects of synthetic chemical coagulants have initiated global interest in the search for safe and economic natural coagulants [16,17]. For instance, Torres and Carpinteyro-Urban [16] applied *Opuntia mucilage* and *Prosopis galactomannan* to treat the elevated chemical oxygen demand (COD) of municipal wastewaters. The *Opuntia dillenii* solution shows very good performance in terms of clarification of very turbid water (186-418 NTU) [18]. Additionally, the cactus leaf extracts showed a good flocculating activity [19]. Taa, Benyahya and Chaouch [20] successfully extracted and applied a bio-flocculent, from the racket of *Opuntia ficus indica*, in the treatment of synthetic industrial wastewater. Vishali and Karthikeyan [21] utilised cactus *Opuntia ficus-indica* as a coagulant for the treatment of a paint effluent wastewater. Furthermore, a number of researchers have investigated the ability of the natural coagulants to remove dangerous pollutants from water. For example, Young [22] successfully applied cactus *Opuntia ficus-indica* for the removal of arsenic from a synthetic water sample. The literature shows that, besides the cactus, okra powder is an effective natural coagulant, where it shows very good efficiency in terms of turbidity and removal of heavy metals [23–25]. For example, Al-Samawi and Hama [23] used okra to remove turbidity from municipal wastewater and leachate of a solid waste sanitary landfill. The outcomes of this study proved the efficiency of okra as a natural coagulant. It is noteworthy to highlight that the literature shows that okra could achieve the maximum turbidity removal at a pH of 8 for medium turbidity level (≤150 NTU), and at a pH of 7 for higher levels of turbidity [23,26,27].

The current work therefore has been mainly devoted to model the performance of both cactus and okra in terms of turbidity removal from river water taking into consideration the influence of key operating parameters. The specific objectives of the current project are:

1. To prove the competence of cactus and okra for turbidity removal.
2. To study the coagulation-flocculation properties for the two coagulants through a jar tester.
3. To model the residual turbidity at the optimum coagulant dose, optimum velocity gradient and optimum flocculation time, using multiple regression analysis.

It is noteworthy to highlight that the studied water samples were collected from the Euphrates River, at Al-Musayab city, Iraq (this part of the river is known as the Al-Mashroo Canal). The sampling station was located in front of the Al-Musayab water treatment plant (Fig. 1). The samples were collected during the period 29/8/2014 until 23/7/2015.

2. Materials and methods

2.1. Standard solutions

Cactus and okra solutions were prepared according to the standard methods recommended by; Shokralla [28]; [29] and [30]. Standard (0.1%) solutions were prepared for both coagulants daily. The dose was calculated as 1 ml of solution to be equivalent to 1 mg of cactus or okra solution.

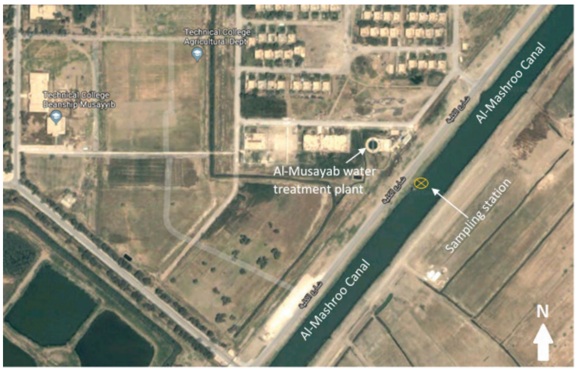

Fig. 1. Sampling station.

2.2. Coagulation-flocculation in water treatment

Coagulation-flocculation properties studied here in this investigation consist of:

1. Optimum-coagulant-dose (DO).
2. Optimum-flocculator-velocity-gradient (GO).
3. Optimum-flocculation-period (TO).

Here, DO refers to the coagulant dose which produces maximum turbidity removal for a constant velocity gradient ($G1$ = 25 1/s) and constant flocculation period ($T1$ = 20 min.). GO refers to the flocculator velocity gradient which produces maximum turbidity removal for a coagulant dose = DO, $G1$ = 25 1/s and variable $T1$. TO refers to the flocculation period which produces maximum turbidity removal for a coagulant dose = DO, $T1$ = 20 min and variable $G1$. Coagulation and flocculation in water treatment have been well defined in the literature; the following references may be consulted for further knowledge about elementary principles [31–33].

2.3. Optimum dose

For both alum and cactus solutions, the optimum dosage for the studied coagulants has been calculated according to the following procedures [28,30]:

1. Preparing six beakers (1,000 ml each) with raw water from the inlet chamber of the sedimentation tank that delivers raw water from Al-Mashroo Canal.
2. Measuring the initial turbidity, pH level, and temperature.
3. Adding six different doses of the coagulant.
4. Placing the six beakers in the floc-tester ET-750 (jar tester) and rapid mixing for G = 250 (1/s), T = 120 s, and according to Table 1, decide the speed of the mixers (n) according to the raw water temperature.
5. Slow mixing for G = 25 (1/s), T = 20 min, and according to Table 1, decide the speed of mixers (n) according to the raw water temperature.
6. Stopping the mixing and setting the beakers aside for settling for 15 min.
7. Taking samples from the top 30 ml of the beakers and measuring the residual turbidity. This is the clear water turbidity.
8. Drawing the curve between the coagulant dose and residual turbidity to decide the optimum dose that gives the minimum residual turbidity.

Table 1
Mixer speed (*n*) in terms of velocity gradient (*G*) for different raw water temperatures for LOVIBOND FLOC TESTER ET-750

G (1/S)	*n* (rpm)[a]					
	Raw water temperature (°C)					
	10	15	20	25	30	40
25	60	57	55	53	51	47
35	75	71	68	66	63	59
45	88	84	81	78	75	70
55	101	96	92	89	86	80
65	113	108	103	99	96	90
250	277	265	254	244	235	220

[a] rpm = revolution per minute.

For a detailed explanation of the experimental procedure and how to decide the DO, GO and TO, it is recommended to refer to [28,30].

2.4. Experimental work

The appropriate device required for jar testing during the study was the LOVIBOND FLOC TESTER ET-750. Laboratory analysis was performed during the period 29/8/2014 until 23/7/2015. Every point represents results for pH, temperature (TMP), initial turbidity (N_1), residual turbidity (N_2), DO, GO and TO during a month.

Turbidity removal efficiency *R* (%) was calculated according to the following equation:

$$R(\%) = \left(\frac{N_1 - N_2}{N_1}\right) \times 100 \tag{1}$$

where $N_1$and N_2 represent the initial river water turbidity (NTU) and residual turbidity after coagulation-flocculation (NTU), respectively.

2.5. Statistical modelling

To manipulate the results, statistical models were built using multiple regression analysis. All theoretical aspects of the above analysis may be found in different statistical textbooks, such as [34]. Within this article, SPSS 20 package was used to perform the stepwise multiple regression analysis. The general multiple regression equation adapted was:

$$Y = a_0 + b_1X_1 + b_2X_2 + b_3X_3 + b_4X_4 + e \tag{2}$$

where *Y* is the Dependent variable, α_0 is the Intercept, b_1,b_2,b_3,b_4 are the Partial regression coefficients, X_1,X_2,X_3,X_4 are the Independent variables, and *e* is the Error term (residuals). It is noteworthy to mention that *e* must be normally and independently distributed (NID) with mean μ = 0 and standard deviation σ = 1, (it is written as NID (0, 1)).

In this study, the dependent variable is the residual turbidity (N_2) which will be predicted for three cases:

1. Optimum-coagulant-dose (DO).
2. Optimum-flocculator-velocity-gradient (GO).
3. Optimum-flocculation-period (TO).

As two coagulants will be investigated, then six models will be explored (3 models for cactus + 3 models for okra) concerning the above cases. For all the models, the dependent variable will be $Y = N_2$, so that the analysis will be performed for the following cases:

1. Modelling N_2 for the DO
   Here the independent variables for both coagulants will be: X_1 = pH, $X_2 = N_1$, X_3, = TMP and X_4 = DO
2. Modelling N_2 for the GO
   Here the independent variables for both coagulants will be: X_1 = pH, $X_2 = N_1$, X_3 = TMP and X_4 = GO
3. Modelling N_2 for the TO
   Here the independent variables for both coagulants will be: X_1 = pH, $X_2 = N_1$, X_3 = TMP and X_4 = TO

3. Results and discussion

Table 2 shows the variation of DO and turbidity removal efficiency *R* (%) for both coagulants for a constant flocculator-velocity-gradient (*G*1), constant flocculation-period (*T*1) and variable coagulant-dose (*D*1). Table 3 shows the variation of GO and residual turbidity for both coagulants at DO and *T*1 = 20 min., while Table 4 shows the variation of TO and residual turbidity for both coagulants at DO and *G*1 = 25l/s.

3.1. Modelling residual turbidity (N_2) for optimum-coagulant-dose (DO)

Applying the data which resulted from Table 2 in the multiple regression model represented by Eq. (2), and analysing using SPSS version 20, the appropriate stepwise multiple regression model for cactus was:

$$N_2(\text{Cactus}) = -6.765 + 0.234(N_1) + e \tag{3}$$

This means that the independent variables of pH, TMP and DO were insignificant statistically. The above regression gave F-ratio = 26.543 compared with the theoretical F (0.01, 1, 10) = 10.04 (which resulted from the tables); this means that regression Eq. (3) was highly significant. The obtained R^2 value, 0.726, indicates that the regression contribution ratio was very acceptable [35]. Additionally, the one-sample Kolmogorov-Smirnov test gave the calculated

Table 2
Variation of DO and R (%) for $G1$ = 25 1/s, $T1$ = 20 min and variable $D1$

Date	River water properties			Cactus coagulation			Okra coagulation		
	pH	N_1 (NTU)	TMP (°C)	DO (mg/l)	N_2 (NTU)	R (%)	DO (mg/l)	N_2 (NTU)	R (%)
29/8/2014	8.20	900	34	2.0	296	67.1	10.0	348	61.3
25/9/2014	8.19	659	29	8.0	59	91.0	12.0	69	89.5
16/10/2014	7.73	339	25	0.5	27.57	91.9	0.5	32.3	90.5
26/11/2014	8.14	319	17	1	33.45	89.5	8.0	35.29	88.9
24/12/2014	8.09	12.18	14	1	0.00	100	1.0	0.00	100
23/1/2015	8.07	18.36	13	8.0	9.16	50.1	10.0	8.43	54.1
26/2/2015	8.18	30.72	15	6.0	15.33	50.1	14.0	12.21	60.3
27/3/2015	8.19	38.27	19	12.0	13.64	64.4	20.0	12.57	67.2
30/4/2015	8.09	26.41	26	4.0	3.33	87.4	2.0	3.52	86.7
28/5/2015	7.82	18.27	30	12.0	11.13	39.1	12.0	11.75	35.7
11/6/2015	7.80	24.68	30	1.0	7.09	71.3	8.0	7.8	68.4
23/7/2015	7.81	13.40	35	16.0	4.97	62.9	16.0	2.23	83.4
Average						72.1%			73.8%

Table 3
Variation of GO and N_2 at DO (which resulted from Table 1) and $T1$ = 20 min

Date	River water properties			Cactus coagulation		Okra coagulation	
	pH	N_1 (NTU)	TMP (°C)	GO (l/s)	N_2 (NTU)	GO (l/s)	N_2 (NTU)
29/8/2014	8.20	900	34	45	158	45	170
25/9/2014	8.19	659	29	25	59	25	69
16/10/2014	7.73	339	25	25	27.57	25	32.3
26/11/2014	8.14	319	17	25	36.73	25	24.8
24/12/2014	8.09	12.18	14	25	0	55	0
23/1/2015	8.07	18.36	13	25	9.16	25	8.43
26/2/2015	8.18	30.72	15	25	15.33	25	12.21
27/3/2015	8.19	38.27	19	25	13.64	25	12.57
30/4/2015	8.09	26.41	26	25	2.29	25	1.84
28/5/2015	7.82	18.27	30	35	7.2	35	6.72
11/6/2015	7.80	24.68	30	55	4.98	35	3.32
23/7/2015	7.81	13.40	35	35	3.02	35	2.23

statistic $|D_{\max}|$ = 0.286 which was insignificant compared to $D_{12}^{0.05}$ = 0.375 (from tables at α = 0.05), indicating that e was normally distributed. The Durbin-Watson ratio was d = 1.784, indicating that e was serially uncorrelated (independently distributed). This proved that error term was NID (0, 1). The appropriate regression model for okra was:

$$N_2(\text{Okra}) = -10.271 + 0.278(N_1) + e \qquad (4)$$

This means that the independent variables of pH, TMP and DO were insignificant statistically.

The above regression gave F-ratio = 27.048 compared with the theoretical F (0.01, 1, 10) = 10.04 (which resulted from the tables); this means that regression Eq. (4) was highly significant. The regression contribution ratio was R^2 = 0.73. Also, the one-sample Kolmogorov-Smirnov test gave the calculated statistic $|D_{\max}|$ = 0.301 which was insignificant compared with $D_{12}^{0.05}$ = 0.375 (from tables at α = 0.05), indicating that e was normally distributed. The Durbin-Watson ratio was d = 1.796, indicating that the e was serially uncorrelated (independently distributed). This proved that residuals were NID (0,1).

3.2. Modelling residual turbidity (N_2) for optimum-flocculator-velocity-gradient GO

Applying the data which resulted from Table 3 in the multiple regression model represented by Eq. (2), and analysing using SPSS version 20, the appropriate stepwise multiple regression model for cactus was:

$$N_2(\text{Cactus}) = -0.155 + 0.137(N_1) + e \qquad (5)$$

The above regression gave F-ratio = 59.805 compared with the theoretical F (0.01, 1, 10) = 10.04 (which resulted

Table 4
Variation of TO and N_2 at DO (which resulted from Table 1) and $G1= 25$ 1/s

Date	River water properties			Cactus coagulation		Okra coagulation	
	pH	N_1 (NTU)	TMP (°C)	TO (min.)	N_2 (NTU)	TO (min.)	N_2 (NTU) (NTU)
29/8/2014	8.203	900	34	23	253	23	257
25/9/2014	8.198	659	29	20	59	23	69
16/10/2014	7.734	339	25	20	25	20	32.3
26/11/2014	8.145	319	17	30	28.18	30	29.18
24/12/2014	8.090	12.18	14	20	0	20	0
23/1/2015	8.075	18.36	13	20	9.16	20	8.43
26/2/2015	8.183	30.72	15	26	13.68	20	12.21
27/3/2015	8.192	38.27	19	20	13.64	20	12.57
30/4/2015	8.091	26.41	26	20	3.33	26	3.33
28/5/2015	7.82	18.27	30	30	8.94	30	10.61
11/6/2015	7.80	24.68	30	30	4.26	30	4.11
23/7/2015	7.816	13.40	35	26	3.87	20	2.23

from the tables); this means that regression Eq. (5) was highly significant. The regression contribution ratio was $R^2 = 0.857$. In addition, the one-sample Kolmogorov-Smirnov test gave the calculated statistic $|D_{\max}| = 0.214$ which was insignificant compared with $D_{12}^{0.05} = 0.375$ (from tables at $\alpha = 0.05$), indicating that e was normally distributed. The Durbin-Watson ratio was $d = 1.609$, indicating that e was serially uncorrelated (independently distributed). This proved that residuals were NID (0, 1). The appropriate regression model for okra was:

$$N_2\left(\text{Okra}\right) = -1.792 + 0.152(N_1) + e \tag{6}$$

The above regression gave F-ratio = 72.989 compared with the theoretical F (0.01, 1, 10) = 10.04 (which resulted from the tables); this means that regression Eq. (5) was highly significant. The regression contribution ratio was $R^2 = 0.880$. In addition, the one-sample Kolmogorov-Smirnov test gave the calculated statistic $|D_{\max}| = 0.241$ which was insignificant compared with $D_{12}^{0.05} = 0.375$ (from tables at $\alpha = 0.05$), indicating that e was normally distributed. The Durbin-Watson ratio was $d = 1.593$, indicating that e was serially uncorrelated (independently distributed). This proved that residuals were NID (0,1).

3.3. Modelling residual turbidity (N_2) for optimum-flocculation-period (TO)

Applying the data which resulted from Table 4 in the multiple regression model represented by Eq (2), and analysing using SPSS version 20, the appropriate stepwise multiple regression model for cactus was:

$$N_2\left(\text{Cactus}\right) = -5.625 + 0.204(N_1) + e \tag{7}$$

The above regression gave F-ratio = 30.189 compared with the theoretical F (0.01, 1, 10) = 10.04 (which resulted from the tables); this means that regression Eq. (7) was highly significant. The regression contribution ratio was $R^2 = 0.751$. In addition, the one-sample Kolmogorov-Smirnov test gave the calculated statistic $|D_{\max}| = 0.286$ which was insignificant compared with $D_{12}^{0.05} = 0.375$ (from tables at $\alpha = 0.05$), indicating that e was normally distributed. The Durbin-Watson ratio was $d = 1.722$, indicating that the residuals were serially uncorrelated (independently distributed). This proved that residuals were NID (0, 1). The appropriate regression model for okra was:

$$N_2\left(\text{Okra}\right) = -5.896 + 0.213(N_1) + e \tag{8}$$

The above regression gave F-ratio = 36.839 compared with the theoretical F (0.01, 1, 10) = 10.04 (which resulted from tables); this means that regression Eq. (8) was highly significant. The regression contribution ratio was $R^2 = 0.880$. In addition, the one-sample Kolmogorov-Smirnov test gave the calculated statistic $|D_{\max}| = 0.289$ which was insignificant compared with $D_{12}^{0.05} = 0.375$ (from tables at $\alpha = 0.05$), indicating that e was normally distributed. The Durbin-Watson ratio was $d = 1.742$, indicating that the residuals were serially uncorrelated (independently distributed). This proved that residuals were NID (0, 1).

A close analysis of the results of statistical models given by Eqs. (3)–(8), together with the results of Tables 1–3 revealed the following:

1. That in terms of the optimum-coagulant-dose, optimum-flocculator-velocity-gradient and optimum-flocculation-period, when modelling the residual turbidity it was found that initial turbidity was the only significant variable for both cactus and okra.
2. That model (8) for cactus (in terms of optimum-flocculator-velocity-gradient) gave the highest coefficient of determination ($R^2 = 0.857$).
3. That model (9) for okra (in terms of optimum-flocculator-velocity-gradient) gave the highest coefficient of determination ($R^2 = 0.880$).
4. That DO ranged between 0.5 and 12 mg/l for cactus and between 0.5 and 14 mg/l for okra, according to Table 2.
5. That GO ranged between 25 and 55 1/s for cactus and okra, according to Table 3.

6. That TO ranged between 20 and 30 min. for cactus and okra, according to Table 4.
7. That models (8) and (9) were the most reliable models that can predict residual turbidity of the Euphrates river/Al-Mashroo canal/Al-Mussaib/Iraq, given the initial turbidity.

4. Conclusion

From the results of the study, it was concluded that cactus (*Opuntiaspp.*) and okra were of similar coagulation-flocculation properties, and they were both competent for turbidity removal, which in turn indicates the possibility of using these coagulants in large-scale field applications. Additionally, according to the environmental parameters recorded during the period of study and the statistical analysis, two facts were concluded. The first fact was that controlling the Optimum Velocity Gradient of the Coagulation-Flocculation process gave the highest contribution ratio of the models, as reported by Eqs. (5) and (6). The second fact was that the most significant parameter (statistically) in the flocculation process was the initial turbidity, as shown by Eqs. (3)–(8).

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