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Behavior of turbidity, pH, alkalinity and color in Sinú River raw water treated by natural coagulants

Comportamiento de la turbidez, pH, alcalinidad y color del agua del río Sinú tratada con coagulantes naturales

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ABSTRACT: Natural coagulants were evaluated in saline *Hylocereus* cf. *trigonus* stems, gum exudate of *Albizia saman*, bark and seeds of *Moringa oleifera* and bark of *Guazuma ulmifolia* in raw water samples from the Sinú river with 56, 104, 200 and 301 Nephelometric Turbidity Units, and Jar tests were conducted to determine the performance removal turbidity of each coagulant, with doses between 5 mg/L to 60 mg/L. pH, color and alkalinity of the water samples were measured before and after treatment to verify the incidence of the coagulant in the purification process. For *H. trigons, A. saman, G. ulmifolia* and bark of *M. oleifera,* removal percentages ranged between 50% and 70% and up to 95% for *M. oleifera* seed. Greater coagulant activity was recorded for applied doses between 20 mg/L to 30 mg/L, independent of raw water turbidity. The pH and total alkalinity had no significant changes for the entire dose range, while the true color slightly increased with extracts of *H. trigons, A. saman*, *G. ulmifolia* and bark of *M. oleifera,* and decreased significantly with *M. oleifera* seeds to values lower than 5 CU, which was the extract with the highest in removing turbidity and color.

RESUMEN: Se evaluaron coagulantes naturales en solución salina de tallos de *Hylocereus* cf. *trigonus*, exudado gomoso de *Albizia saman*, corteza y semilla de *Moringa oleífera* y corteza de *Guazuma ulmifolia*, en muestras de agua cruda del río Sinú, con turbidez de 56, 104, 200 y 301 UNT y se realizaron ensayos de jarras para determinar la remoción de turbidez de cada coagulante, con dosis de 5 mg/L a 60 mg/L. Se midió el pH, la alcalinidad y el color de las muestras de agua, antes y después del tratamiento, para verificar la incidencia de los coagulantes en el proceso de potabilización. Los porcentajes de remoción variaron entre el 50% y 70% para *H. trigons*, *A. saman*, *G. ulmifolia* y corteza de *M. oleífera* y hasta del 95% con semilla de *M. oleífera*. Para las dosis aplicadas entre 20 mg/L a 30 mg/L se registró mayor actividad coagulante, independiente de la turbidez del agua cruda. El pH y la alcalinidad total no tuvieron variaciones significativas para todo el rango de dosis, mientras que el color verdadero aumentó levemente con los extractos de *H. trigons*, *A. saman*, *G. ulmifolia* y corteza de *M. oleífera* y disminuyó significativamente con la semilla de *M. oleífera* hasta valores menores a 5 UC, siendo este el extracto de mayor eficiencia en la remoción de turbidez y color.

1. Introduction

Coagulation-flocculation is an essential process in the treatment of surface water and waste water, including the removal of dissolved organic species and the reduction of turbidity and color of water [1]. Chemical coagulants based on inorganic salts and synthetic organic polymers are conventionally used, however, they have disadvantages associated with harmful effects on human health [2, 3], temperature, relatively high costs for acquisition, high-volume production sludge and significant alteration of pH and water alkalinity after treatment [4-10]. Additionally, it is likely to increase aluminum concentrations in the treated water after application of the coagulant and cause problems in distribution systems, interfering with the disinfection process, due to masking of microorganisms which adhere to the precipitated hydrated aluminum. Another problem is deposition of products from aluminum hydrolysis in the

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pipe walls, thereby decreasing the transmission capacity and generating corrosion problems [11].

An alternative solution to this problem is the development of new coagulants, preferably extracted from natural renewable sources such as microorganisms, animals or plants, which are biodegradable and safe for human health [1, 5, 12-14]. These naturally occurring coagulants have low acquisition costs, high biodegradability, low toxicity and low sludge production [15].

Usually natural coagulants can unlikely alter pH of treated water. It has a large number of charges on the surface that increase coagulation efficiency.

These advantages are greater if the plant from which the coagulant is extracted is native to the region where the raw water is intended to be purified and easily accessible to local communities [1]. Coagulation mechanisms associated with natural coagulants are primarily, adsorption and charge neutralization as well as adsorption and bond between particles [5, 6, 13]. Adsorption and charge neutralization relate to sorption of two particles with oppositely charged ions, whereas a bridge between particles occurs when the coagulant provides a sucking particles polymer chain. Polymer coagulants are usually associated with long-chain structures, especially polymers with high molecular weights, which greatly increase the number of available adsorption sites [13].

Several studies have demonstrated the effective removal of turbidity in different types of water, with the application of natural coagulants obtained from basil, Senna, Moringa and Bean seeds; Cactus, rubber, and bark mucilage; and Acacia and Campano wood, among other plants and potential sources [4, 5, 8, 9, 13, 16-20].

Removals greater than 90% of turbidity of water treated with use of *Moringa* seed extracts have been reported, without significantly altering pH and alkalinity of the samples after coagulation [21, 22]. Similarly, it was found that the mucilage of different types of Cactus *Opuntia* reached such removal without significantly altering the pH and alkalinity of the treated water [12, 23].

Research with gummy exudate obtained from *Acacia siamea*, *Albizia saman* and *Cedrela odorata*, shows that after the application of optimal doses of these coagulants, a decrease greater than 80% on turbidity levels is reached, with no significant change in total alkalinity and pH; suggesting that the use of gum exudate as coagulating agent does not require the addition of buffer substances for balancing the carbonate in the treated water [11, 14, 24]. In the case of Guácimo, it is known that extracts prepared from the bark have been used to clarify sugarcane juice and soaps [25], aiming to also be effective in removing turbidity of raw water.

This work shows the turbidity removal efficiency of natural coagulants *Hylocereus cf. trigonus, Albizia saman, Pithecellobiun* and *Moringa oleifera* raw water in the Sinú River and its impact on the physical and chemical characteristics such as turbidity, pH, total alkalinity and true color of treated water.

2. Experimental

2.1. Sampling of raw water

Raw water samples were collected on the right bank of the Sinú River at the Mocarí neighborhood, Municipality of Montería, Department of Córdoba, Colombia. The Sinú River is the source of water for most of Cordoba´s population. It supplies water to 16 municipalities and 18 water systems [26]. Four single samples were tested in the morning (from 8 to 8:30) and 100 L of each raw water sample were taken 50 cm deep, between November 27, 2013 and June 4, 2014 during the dry and rainy season.

2.2. Preparation of coagulant extracts

Stems of *Hylocereus cf. trigonus*, gummy exudate of *Albizia saman* and bark of *Guazuma ulmifolia* were collected in the village of Severá, Municipality of Cereté. The bark and seeds of *Moringa oleifera* were collected on the campus of Pontifical Bolivarian University at Montería, Department of Córdoba, Colombia.

Stems, gum exudate and seed were dried in a reflow oven, ground and sieved in a No. 30 mesh according to Tyler series (0.60 mm), while barks were macerated to small fibers [6, 17, 19, 21, 23, 24, 27, 28].

Then, 10.0g of each of the five processed vegetables were dissolved separately in flasks, up to 1.0L saline sodium chloride 1.0% (w/v). The solutions were mixed with magnetic stirring for 1 hour, centrifuged at 3,500 rpm for 10 minutes and filtered under vacuum with cellulose filter paper. The filtrates were labeled as saline coagulants extracts with concentration 10,000 mg/L and kept refrigerated at 4 °C until application.

2.3. Jar tests

The coagulation, flocculation and sedimentation with each coagulant were made in a jar test Flocculator, E & Q F6-330-T model. Twelve doses of each coagulant were used: 5, 10, 15, 20, 25, 30, 35, 40, 45, 50, 55, 60 mg/L to each sample of raw water with starting turbidity of 56, 104, 200 and 301 NTU. A white reference was used for each test, without applying coagulant. The process of rapid mixture was maintained at 200 rpm for 1 minute, followed by a slow mix at 40 rpm for 20 minutes, and a settling time of 20 minutes [29, 30, 31].

2.4. Equipment and parameters of water quality

Quality parameters of water treated by coagulation with five natural coagulants were measured after the sedimentation phase using Standard Methods [32]. Parameters analyzed were: turbidity, pH, alkalinity and color. Turbidity was measured with a HACH 2001P turbidimeter; pH with a potentiometer SCHOTT instruments Lab 850; alkalinity by the potentiometric titration method with 0.02 N sulfuric acid; true color by UV-visible spectrophotometry in a spectrophotometer Thermo Scientific GENESYS 10S UV-Vis, λ = 456 nm (standard solution of Pt/Co).

To determine the behavior of the main physicochemical parameters, they were triply measured before and after the jar tests to compare them and verify if the coagulant application could lead to possible variations of these parameters.

Analysis of Variance was made to determine the quality of pH, alkalinity and color in the different treatments (12 doses and 4 levels of initial turbidity). For cases where there was statistically significant difference between treatments, The Tukey test was performed [33]. The results show the mean ± standard deviation of turbidity, pH, alkalinity and color of samples of the raw water. The removal percent and behavior of pH, alkalinity and color were visualized by column graphs with error bars.

3. Results and discussion

3.1. Physicochemical characteristics of samples of the raw water

Table 1 shows the physicochemical characteristics of the raw water samples.

The turbidity of the collected samples varied in a range of 56 to 301 NTU. These values are within the characteristics levels of raw water from the Sinú River (40- 1200 NTU). pH values were 7.82 to 8.24, close to neutrality within the allowed range according to Colombian regulations (30 to 250 mg $CaCO_3$/L). The Real Color of the raw water showed values from 15.54 to 42.51 Color Units, which were above the allowed values according to Colombian regulations (15 CU) [34].

3.2. Turbidity removal efficiencies

The removal efficiencies from *H. trigonus, A. saman and G. ulmifolia* extracts were between 50% and 70%, regardless of the dose applied and the turbidity of raw water (Figure 1(a), 1(b) and 1(c)). For the samples treated with bark of *M. oleifera*, the removal percentage was 60% to 70% (Figure 1(d)). *M. oleifera* seed had the best performance in removing turbidity of raw water, achieving greater efficiencies by 95% for samples with 200 NTU and 301 NTU with optimal doses between 20 mg/L and 30 mg/L; however, they are not statistically different to other coagulant concentrations, which reach values up to 2 NTU, above the Colombian norm (2NTU) [34]. These results are complementary to the study of [35], where the coagulant activity was evaluated with a response surface design.

The ANOVA "P" values made for pH, total alkalinity and true color of treated water samples are shown in Table 2.

The values of "P" for pH and alkalinity after applying the coagulant extracts to water samples were higher than 0.05, which means that there is no statistically significant difference among the means of these parameters with a confidence level of 95 %. However, the "P" values for the color of the samples to which the *A. saman* gum exudate and *M. oleifera* seed were applied, were less than 0.05, indicating significant difference in the effect of these coagulants on the samples. With the Tukey test, it was found that significant differences for the extract of *A. saman*, occurred between the average doses applied in raw water from 104 NTU to 200 NTU, while for the samples treated with *M. oleifera* seed, significant differences occurred in raw water between 200

Table 1 Physicochemical characteristics of samples of raw water

Parameters	Sample 1	Sample 2	Sample 3	Sample 4
Initial turbidity (NTU)	56.33 ± 3.21	103.67 ± 4.04	200.33 ± 3.21	301.33 ± 5.86
pH	7.82 ± 0.21	8.20 ± 0.03	8.24 ± 0.02	8.19 ± 0.02
Alkalinity (mg$CaCO_3$/L)	46.27 ± 0.70	46.20 ± 0.80	53.53 ± 0.64	35.80 ± 0.20
Color (CU)	15.54 ± 2.03	19.06 ± 2.03	20.23 ± 2.03	42.51 ± 3.52

Mean ± standard deviation

Table 2 ANOVA P Values

Parameters	*H. cf. Trigonus* stems	*A. saman* gum exudate	*G. ulmifoia* bark	*M. oleífera* bark	*M. oleífera* seed
pH	0.476	0.999	0.999	0.999	0.340
Alkalinity	0.999	0.998	0.999	0.999	0.967
Color	0.997	0.021	0.991	0.860	0.031

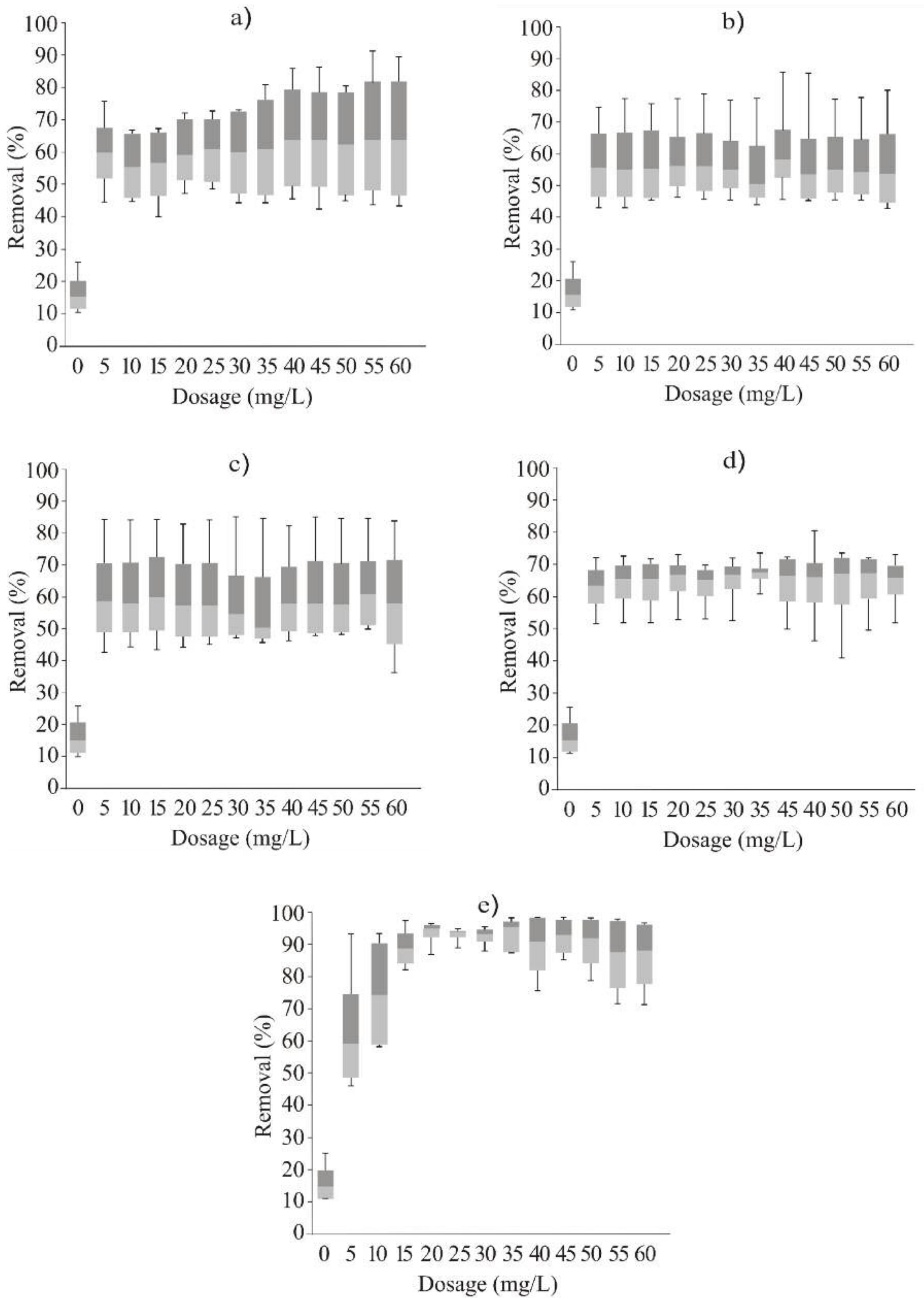

Figure 1 Removal of turbidity of raw water from the Sinú River treated with: a) H. trigonus, b) A. saman, c) G. ulmifolia, d) M. oleifera bark, e) M. oleifera seed

NTU and 301 NTU. The color increased significantly when gummy exudate of *A. saman* was applied and decreased with doses of *M. oleifera* seed. The color did not change significantly, with a confidence level of 95%, when extracts of *H. trigonus, G. ulmifolia* and bark of *M. oleifera* were used, since the "P" values were greater than 0.05.

3.3. pH behavior

All samples of raw water showed an average value of 8.11 pH units. In samples treated with extracts of *H. trigonus*, from 35 mg/L, a slight decrease of pH up to 7.9 units compared to untreated water (Figure 2(a)) was presented. Likewise, this behavior was observed with samples treated with *M. oleifera* seed where the pH decreased depending on the dose applied up to values of 7.6 (Figure 2(e)) consistent with that reported by [21]. In contrast, samples treated with bark extract of *M. oleifera* increased the pH to values of 8.6 (Figure 2(d)). In samples treated with *A. saman* gum exudate, the pH increased to 8.4 with 5 to 30 mg/L; for doses above 35 mg/L, pH was kept constant (Figure 2(b)). The bark extract of *G. ulmifolia* did not influence pH variation, regardless of the dose and of the turbidity of raw water (Figure 2(c)). However, all variations of pH, according to the ANOVA results (Table 2), are not statistically significant, due to the buffering capacity of the raw water and natural extracts. Similar results were obtained when treating water with coagulant extract from *Cactus lefaria* wherein the pH ranged from 6.5 to 8.5 [19]. Although the pH of the Cactus extract is acidic (4.90), it does not significantly alter the pH of the treated water, it can be due to the low doses applied [36]. In [24] was reported that doses applied at different levels of turbidity, use of gummy exudate does not alter the pH of the water, which is consistent with what was found in this research.

The optimum pH for coagulation with *M. oleifera* should be slightly basic, since for values greater than 7, negative charges dominate on the colloidal particles. This allows adsorption to occur between colloidal dispersions and *Moringa* cationic polyelectrolytes, producing charge neutralization. In a pH less than 7, the colloidal particles are less negative, causing further repulsion effect between colloids and polyelectrolytes. In the case of *Cactus opuntia*, the effect of pH on coagulation is unclear due to the nature of their coagulants, although it has been suggested that (key component *Opuntia* mucilage) galacturonic acid plays a key role in the coagulation of particles due to dissociation providing $-COO^-$ adsorption sites that may act as a bridge between the coagulant and colloidal particles [13].

In treatability tests, the water pH for turbidity removal and color was not controlled in order to not alter the characteristics of the raw water and thus to be able to identify the behavior of the physicochemical parameters which were studied after applying the five natural coagulants.

3.4. Alkalinity behavior

The alkalinity of raw water samples from Sinú River ranged from 35 and 53 mg$CaCO_3$/L. In samples treated with *H. trigonus,* for doses between 5 to 25 mg$CaCO_3$/L, alkalinity remained constant and from 30 mg$CaCO_3$/L it tended to increase to values of 52 mg$CaCO_3$/L (Figure 3(a)). For samples treated with bark extracts of *A. saman, G. ulmifolia*, and seeds of *M. oleifera*, alkalinity showed no statistically significant variations, it remained constant across the range of doses applied, regardless of the turbidity of raw water (Figure 3(b), 3(c), 3(d) and 3(e)), with values less than acceptable by the Colombian Norm, 200 mg $CaCO_3$/L [34]. The effect of the tested doses of coagulants on the alkalinity of the water samples is shown in Figure 3.

The use of natural coagulants extracts does not add substances that alter pH and alkalinity of raw water from the Sinú River, so it is not necessary to condition the water to buffer the acidity caused by iron or aluminum salts in the treatment process [32, 37]. The buffering capacity of the water depends on the alkalinity, which is primarily contributed by bicarbonate ions (HCO_3^-) in natural waters, contributing to the ability of water to neutralize acid and regulate hydrogen ion concentration [38].

Very low values of alkalinity make water corrosive for distribution networks, which are leached and protect microorganisms from disinfectants, representing an obvious threat to public health [11, 24].

3.5. Color behavior

The real color of raw water averaged 24.33 CU. For samples treated with extracts of *H. trigonus*, *G. ulmifolia* and bark of *M. oleifera*, color increased proportional to the dose applied, up to 87 CU, these variations were not statistically significant (Figure 4(a), 4(c) observed and 4(d)). Samples treated with *A. saman* extract showed a significant increase in the color of water depending on the applied dose, with values between 8.50 and 84.40 CU (Figure 4(b)), whereas *M. oleifera* seed evidenced a significant reduction in this parameter (Figure 4(e)), with values below the method detection limit (3.82 CU) and the allowable limit by the Colombian norm (less than 15 CU); results similar to those reported in references [21, 27]. According to the above, the extract obtained from *M. oleifera* seed has greater advantage in the treatment of waters than other coagulants tested.

Coagulants of *Opuntia cochenillifera* and *Cactus lefaria* have shown effectiveness in treating drinking water and they remove turbidity significantly, but were ineffective for removal of color generated by humic substances in samples with low turbidity and alkalinity, regardless of dose but directly proportional to the residual turbidity of raw water [19, 39].

Color enhancement of water samples treated with gummy exudate of *A. saman* is probably due to increased doses of natural coagulant, sediment resuspension and the own color provided by the coagulant [14]. Figure 4 shows the behavior of water color treated with natural coagulants.

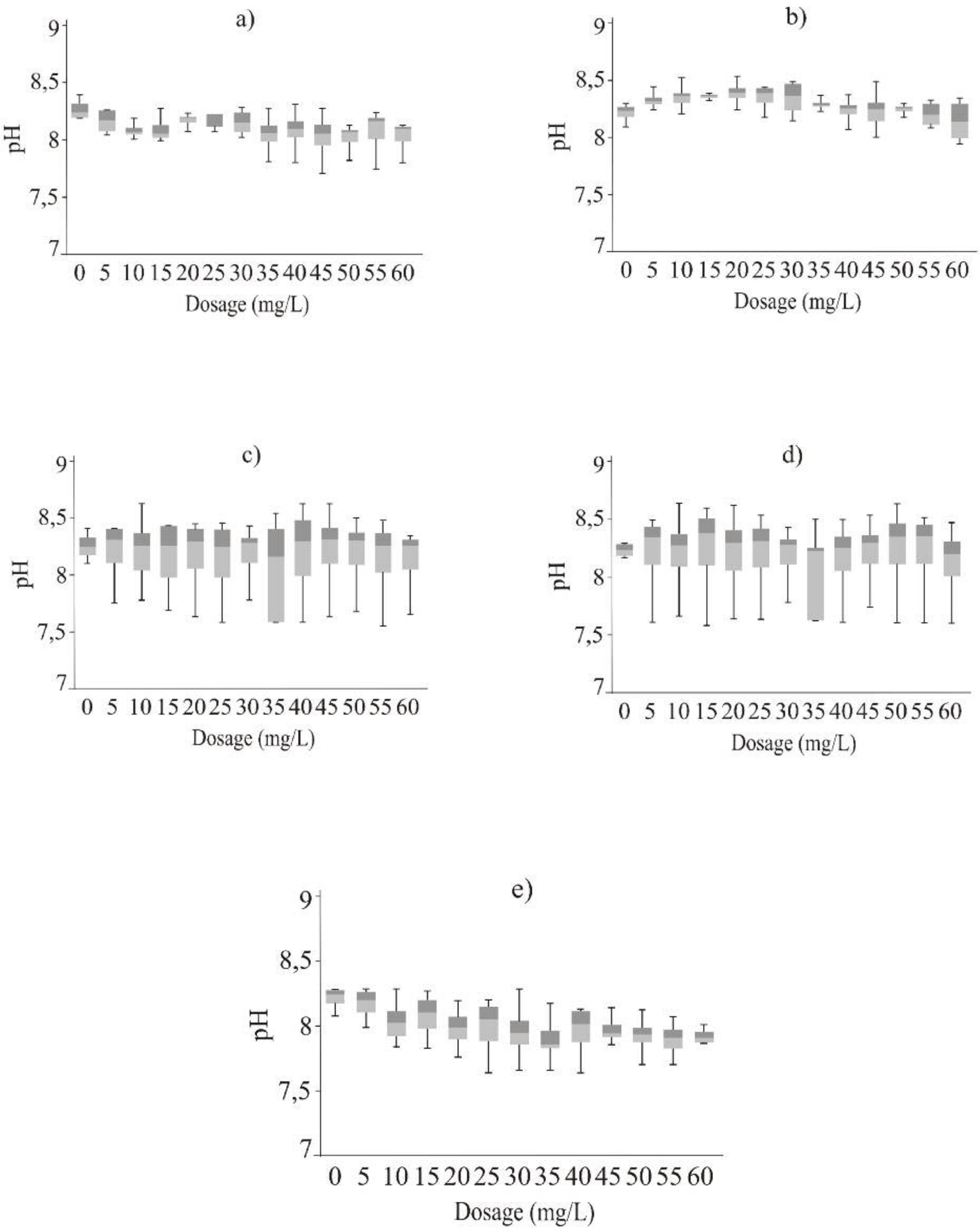

Figure 2 Behavior of pH of raw water treated from the Sinú River: a) H. trigonus, b) A. saman, c) G. ulmifolia, d) M. oleifera bark, e) M. oleifera seed

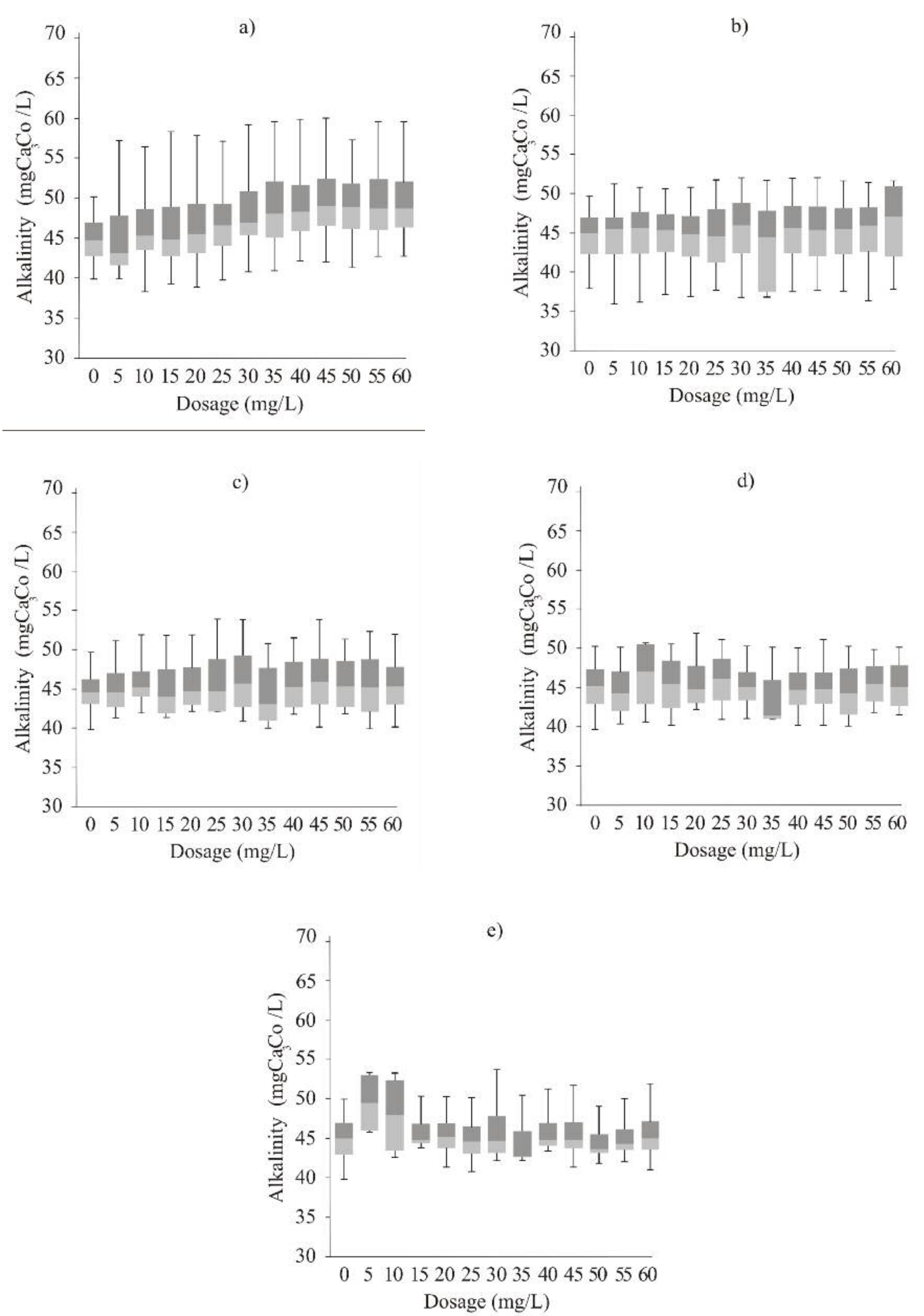

Figure 3 Behavior of total akalinity of raw water from the Sinú River treated with: a) H. trigonus, b) A. saman, c) G. ulmifolia, d) M. oleifera bark, e) M. oleifera seed

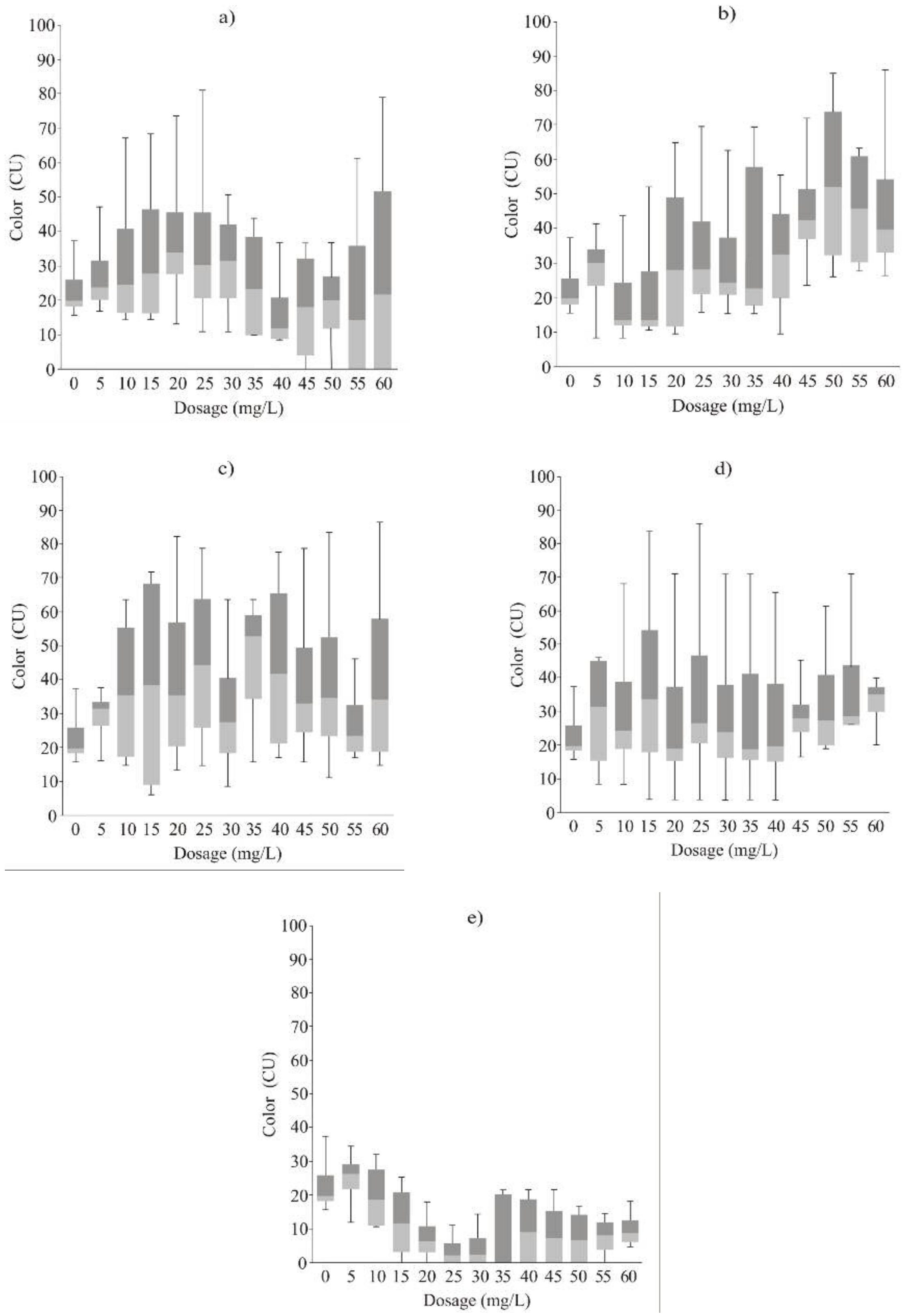

Figure 4 Behavior of the true color of the Sinú River raw water treated with: a) H. trigonus, b) A. saman, c) G. ulmifolia, d) M. oleifera bark, e) M. oleifera seed

4. Conclusion

The applied doses of the tested natural coagulants showed, on average, higher percentages of turbidity removal without significantly altering pH and alkalinity of water after treatment. This represents an advantage over synthetic coagulants, since it is unnecessary to apply buffering substances to adjust the pH. *M. oleifera* seed extract was the only one that managed to reduce the turbidity until 4 NTU and color of treated water to lower values than those allowed by the Colombian sanitary standards. Saline extracts of *H. trigonus, A. saman, G. ulmifolia and M. oleifera* showed great coagulating potential in the treatment of raw water. They are environmentally sustainable and are an alternative of water treatment in low-income communities that do not have access to drinking water.

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