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Turbidity removal and pH of raw water treated with natural coagulants

Remoção de turbidez e pH de água bruta tratada com coagulantes naturais

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ABSTRACT: Concerns regarding water pollution and scarcity have led researchers to investigate new sustainable water treatment technologies, such as the use of natural coagulants, that can be used for water treatment in decentralized locations at small rural communities, as these materials can be used in compact water treatment plants. Thus, the present study evaluated the use of natural coagulants, drumstick tree seeds and tannins extracted from black acacia, in water treatment to remove turbidity. To determine the ideal coagulant dosage, jar tests were performed, followed by filtration to remove the formed sludge. pH and turbidity were analyzed to compare the coagulants efficiency. The natural coagulants achieved satisfactory removal of suspended solids, completely removed turbidity, and causing no significant changes in pH, unlike aluminum sulfate, hence, these natural materials can be used as an alternative to chemical coagulants.

Keywords: *Acacia mearnsii*. Aluminum sulfate. Decentralized water treatment. *Moringa oleifera*. Tannins.

RESUMO: A preocupação com a poluição e a escassez da água aumentam a cada dia; com isso, pesquisadores estudam novas tecnologias para o tratamento de água como o uso de coagulantes naturais que podem ser utilizados para o tratamento de água descentralizado em pequenas comunidades rurais, já que esses materiais podem ser utilizados em Estações Compactas de Tratamento de Água. Sendo assim, o presente trabalho avaliou o uso dos coagulantes naturais, sementes de Moringa e taninos extraídos de Acácia Negra, para a remoção de turbidez. Para determinar a dosagem ideal de coagulante, foram realizados testes de jarro, seguidos de filtração para remover o lodo gerado. Posteriormente foram analisados parâmetros físico-químicos como pH e turbidez, para a comparação da eficiência entre os coagulantes. Os coagulantes naturais obtiveram remoção satisfatória de sólidos suspensos, removendo completamente a turbidez e não causando alterações significativas nos valores de pH, como ocorreu para o sulfato de alumínio, sendo assim o estudo mostra que podem ser utilizados como alternativa aos coagulantes químicos.

Palavras-chave: *Acacia mearnsii. Moringa oleífera.* Sulfato de alumínio. Taninos. Tratamento de água descentralizado.

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INTRODUCTION

Water is a fundamental resource for human survival and development at all economic, social and environmental levels. Although water is abundant, its available fraction, that which is easily accessible and has good quality for consumption, is limited and negligible compared to the total water available on the planet. Given the increasing demand for this natural resource, the waste and misuse of water, which further reduce its availability, must be minimized (ZHANG *et al.*, 2010; FAO, 2015; PIRATOBA *et al.*, 2017).

Anthropogenic interventions in the environment that lead to the misuse of water resources are causing water pollution to increase, which represents a real threat to water quality, health and the environment. Therefore, the treatment of water prior to human drinking and consumption is essential. For this reason, physicochemical parameters are used to analyze and classify water quality, and the values of these parameters are established by regulations (SILVA *et al.*, 2016). In Brazil, the Ministry of Health, through Ordinance No. 888/2021 (BRAZIL, 2021), regulates water quality for human drinking and consumption.

Chemical coagulants, such as aluminum sulfate, are typically used in water treatment; however, treatments using these coagulants require alkali addition due to hydroxyl consumption and generate sludge that has great pollution potential and, when improperly disposed of, can cause damage to the environment (LIMA; ABREU, 2018).

As a result, organic coagulants obtained from plants have emerged as an alternative solution to this problem. These coagulants have plant origins and are biodegradable. Thus, their sludge, after dewatering, can be used on soil, where it will be decomposed by microorganisms, thus reducing environmental damage in addition to avoiding health risks.

Due to low population density and a lack of financial resources, conventional water treatment units are seldom installed in small rural communities. Thus, members of these communities often consume poor-quality water. Under such conditions, compact treatment plants (CTPs) using organic compounds as coagulants are easier to operate and incur lower costs than conventional systems because organic compounds are cheaper than synthetic coagulants and offer an alternative for water treatment (ARANTES *et al.*, 2015).

Organic coagulants include the seeds of the drumstick tree (*Moringa oleifera*) and tannins extracted from the bark of black acacia (*Acacia mearnsii*); according to several studies, these materials have good color and turbidity removal capacity and, in contrast to aluminum sulfate, which is the chemical product most widely used in water treatment, do not cause significant changes in water pH and alkalinity (KATALO *et al.*, 2018; VALVERDE *et al.*, 2018; LEONE *et al.*, 2016; SKORONSKI *et al.*, 2014, SENGUPTA *et al.*, 2012).

Based on the above, the aim of this study is to evaluate the use of natural organic coagulants derived from *M. oleifera* seeds and *A. mearnsii* bark tannins in the treatment of raw water by evaluating turbidity removal and pH and comparing the results to those obtained with the chemical coagulant aluminum sulfate.

2 MATERIAL AND METHODS

Water samples were collected from the Restinga Stream, located in the community of Baiões, a district of Formiga City, Minas Gerais, Brazil. Twenty liters of water was obtained on a rainy day in November 2016, to have a naturally high turbidity water sample. The samples were stored in four 5 L polyethylene containers. The pH and turbidity values were 7.57 and 103.5 NTU, respectively.

The coagulation tests were performed using only this raw water to avoid influences in the process of clarification due to the quantity and nature of the suspensions. In samples with high turbidity, the high density of particles provides nucleation sites for dense flake formation, therefore increasing the sedimentation speed and the efficiency of turbidity removal. However, waters with lower turbidity yield a low clarification efficiency, producing less desirable results.

The *A. mearnsii* tannin-based coagulants Tanfloc SG and SL were obtained in liquid form at a 2% concentration from the manufacturer TANAC SA. *M. oleifera* seeds were bought at a local market. At the Water and Waste Analysis Centre (Centro de Análises de Águas e Resíduos - CENAR), Formiga University Centre (Centro Universitário de Formiga - UNIFOR), Minas Gerais State, the seeds were peeled, and oven dried at approximately 100 °C for 48 h. The preparation of the *M. oleifera* solution was based on the method of Folkard *et al.* (1993), and a 20 g L^{-1} solution was produced from *M. oleifera* seed powder. The powder was obtained following the method of Arantes et al. (2014), who processed seeds using a pestle, followed by sieving (5/16 mesh). These authors indicated that this processing method resulted in a coagulant solution with a high turbidity removal capacity. Due to the experimental procedures, the prepared *M. oleifera* solution also had a concentration of 2%.

To prepare 2% aluminum sulfate solution, a reagent (pro analysis) from Dinâmica Química Contemporânea LTDA was used. For this step, 20 g of aluminum sulfate was weighed and subsequently diluted in 1 L of distilled water. To determine the optimal dosage, coagulation and flocculation assays were performed using a jar test apparatus (Q305M, Quimis).

The tests were performed based on the recommendations of Richter (2009), in which simulated rapid mixing was performed at approximately 300 rpm for 7 s. The flocculation process was started by applying a rotation speed of 100 rpm for 7 minutes. After this time, the speed was reduced to 60 rpm for 4 minutes, and to finish the flocculation step, the rotation speed was reduced to 30 rpm for 4 minutes. Subsequently, the sedimentation process started and proceeded for 5 minutes.

Two hundred milliliters of sample were transferred to the jar test apparatus to perform assays with solutions of 1 mg L^{-1}, 1.5 mg L^{-1} and 2 mg L^{-1} to 20 mg L^{-1}, with 2 mg L^{-1} intervals, to determine the optimal dosages of the coagulants. After the jar test assay, the samples were filtered to remove the sludge that had formed.

Turbidity and pH analyses were performed after the filtration process to determine the efficiency of the evaluated coagulants. Turbidity was determined using a portable microprocessor turbidimeter (Alfakit) with a measurement range of 0 to 1000 NTU, resolution of 0.01 NTU and emission wavelength of 880 nm.

The pH was obtained with an Alfakit AT 315 microprocessor digital pH meter with automatic temperature compensation and a waterproof keyboard. The reading range of the device was 0.0 to 14.0 with a resolution of 0.01 and an accuracy of ±1%.

The experiment followed a completely randomized design in a 4 x 12 factorial arrangement with two replicates, with the sources of variation being the coagulants (four levels: Tanfloc SG, Tanfloc SL, *M. oleifera* seeds and aluminum sulfate) and the coagulant doses (12 levels: 1, 1.5, 2, 4, 6, 8, 10, 12, 14, 16, 18 and 20 mg L^{-1}).

Analysis of variance was performed using the F test, and because the qualitative "coagulant" factor was significant, the Scott-Knott test was used to compare the means. For the significant quantitative "dose" factor, linear regression analysis was performed to determine the interaction between the factors.

3 RESULTS AND DISCUSSION

According to Ministry of Health Ordinance No 888/2021 (BRAZIL, 2021), the maximum permitted limit for turbidity is 5 NTU. All tested coagulants (Tanfloc SG and Tanfloc SL (*A. mearnsii* tannin-based); *M. oleifera* seeds; and aluminum sulfate) completely removed turbidity at all tested concentrations and in all replicates, yielding values of 0 NTU. Corroborating the results in this study, Sánchez-Martín *et al.* (2009) also obtained a turbidity removal efficiency of 100% using a low concentration (2 – 6 mg L^{-1}) of *Acacia mearnsii* coagulant.

Reducing turbidity provides aesthetic benefits to water and reduces the risk of the presence of microorganisms and microbial resistance to disinfectants. Muniz *et al.* (2015) evaluated the turbidity of treated water after clarification and filtration and reported that the turbidity removal efficiency was positively related to the dosage of *M. oleifera* (in that study, from 250 mg L^{-1} to 500 mg L^{-1}), which was not observed in the present study, since from the lowest used concentration, we obtained total turbidity removal. This finding may be related to the initial high turbidity of the raw water. Nevertheless, according to previous authors, the use of M. oleifera seed extract enhances the decantation of suspended solids.

Valverde *et al.* (2018) reported that the use of *M. oleifera* with a synthetic coagulant at a ratio of 80/20 or 60/40 *M. oleifera*/aluminum polychloride (PAC) was efficient according to the limits established by the Brazilian Potable Water Ordinance. Valverde *et al.* (2015) found that the combination of *M. oleifera* with PAC reduces the sedimentation time of colloids, thus making the coagulation/flocculation process more efficient.

The core of *M. oleifera* seeds has a relevant quantity of high-molecular-weight water-soluble proteins. These proteins are positively charged in water solution and thus destabilize negatively charged particles (*e.g.*, clay and bacteria). As a result of this process, large flocs are formed under intermittent agitation, as applied in water treatment systems, and then the flocs can be removed by sedimentation or filtration (AHO; LAGASI, 2012), reducing or completely removing water turbidity.

Sánchez-Mártin *et al.* (2009) conducted an experiment using tannins derived from *A. mearnsii* as a coagulant for the treatment of surface waters with turbidity in the range of 20 to 120 NTU and obtained turbidity removal rates of 99% with dosages of up to 10 mg L^{-1}, values comparable to those obtained in this experiment.

Skoromski *et al.* (2014) evaluated the potential application of tannins as coagulants for the treatment of water and obtained complete turbidity removal at a dosage of 2.5 mg L^{-1}. The authors stated that sludge generated by treatment using tannins has potential for use as fertilizers or in biodigestion, increasing its value. Mangrich *et al.* (2013) found that the generated sludge from the use of tannins as coagulants was within normal specification limits, and contained no aluminum or iron salts, as observed for chemical coagulants. The final pH values of the treated water samples were analyzed by ANOVA, and the results are summarized in Table 1.

Table 1. Summary of ANOVA results for water pH after the use of several coagulants at different concentrations

Source of Variation	Degrees of Freedom	Sum of Squares	Mean Squares
Coagulant (C)	3	7.575754	2.525251**
Concentration (Co)	11	9.909363	0.900851**
C x Co	33	14.141946	0.428544**
Error	48	2.923700	0.060910
Corrected total	95	34.550763	

**Significant at 1% probability by the F test.

Based on Table 1, both the simple effect of treatments and the interactions between them were significant. The mean pH values due to the simple effect of the coagulants used are shown in Table 2.

Table 2. Mean pH values obtained according to coagulant in the treatment of raw surface water

Coagulant	pH
Tanfloc SG	7.92 a
Moringa oleifera	7.73 b
Tanfloc SL	7.77 b
Aluminum sulfate	7.18 c

Means followed by the same letter do not differ significantly by the Scott-Knott test at 5% probability.

According to Table 2, the use of aluminum sulfate as a coagulant resulted in the lowest pH among the coagulants used due to its hydrolysis process (which does not happen for polymeric coagulants), and the highest pH was obtained using Tanfloc SG. However, it should be noted that all means obtained were in accordance with the potability standard established by Ministry of Health Resolution No. 888/21 (BRAZIL, 2021), which indicates that treated waters must have a pH between 6.0 and 9.5.

Ndabigengesere and Narasiah (1998) observed that the use of aluminum sulfate as a coagulant promoted a significant change in pH, reducing the pH from 7.6 to 4.2, a condition that was not verified in this study, probably due to the experimental conditions applied, such as the dosage of coagulant and the initial quality of the treated water; this observation is contrary to the effect of *M. oleifera*, which did not cause significant changes in the pH of the treated water. Skoronski *et al.* (2014) found that when using a coagulant for water treatment, it is necessary to ensure that the added product does not increase the concentration of dissolved substances, which, among other effects, would cause a significant change in the pH of the treated water and lead to difficult sedimentation. The use of aluminum sulfate did not cause a marked reduction in the water pH after treatment due to the low dosage; however, its use resulted, on average, in the lowest pH measured.

Zaman *et al.* (2017) developed and tested a water purification system using *M. oleifera* seed powder as a coagulant and scallop shell powder as an antibacterial agent, followed by biofiltration, to treat raw water from different surface sources. The evaluated raw water had pH values varying from 7.62 to 8.01, but after purification, values from 7.37 to 7.89 were measured, similar to the results observed in this research. Beyond the pH evaluation, these authors stated that the resulting water was drinkable, considering the United States Environmental Protection Agency (USEPA, 2015) standard for drinking water.

The effects of the various coagulant concentrations used on the pH of the treated raw water are shown in Figure 1.

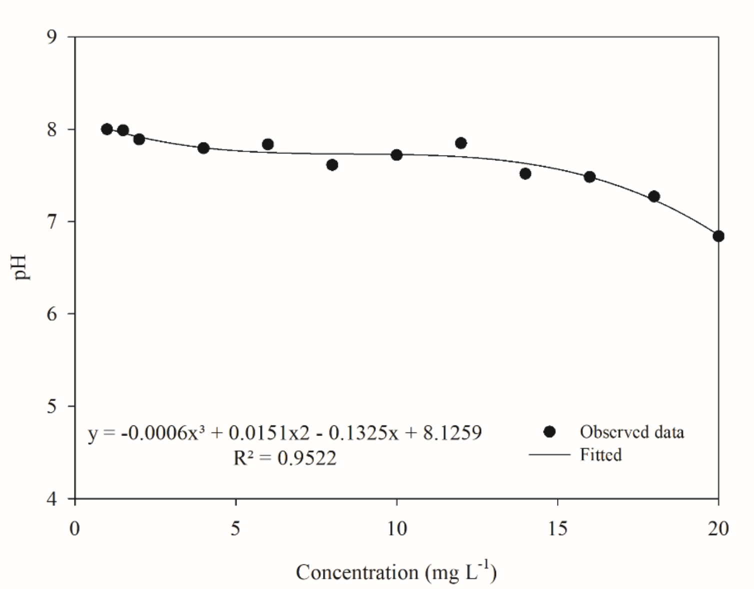

Figure 1. Simple effect of the concentration of different coagulants in the treatment of raw water on the pH value of the treated water.

The simple effect of the concentration of the different coagulants on the pH of the treated water can be explained by a cubic equation model (Figure 1), which predicts a maximum pH of 8.01 for a coagulant concentration of 1.0 mg L^{-1} and a minimum pH of 6.72 for a concentration of 20 mg L^{-1}. Thus, because all pH values agreed with those proposed by Brazil (2021), the best coagulant dose to use would be 1 mg L^{-1}. The mean pH values of the treated water according to the coagulant and concentration levels are shown in Table 3.

Table 3. Effect of coagulants at each evaluated concentration level on the mean pH of treated raw water

Concentration (mg L^{-1})	Aluminum sulfate	*M. oleifera*	Tanfloc SG	Tanfloc SL
1	7.93 a	7.96 a	8.19 a	7.93 a
1.5	7.99 a	7.98 a	8.08 a	7.91 a
2	7.92 a	8.00 a	7.90 a	7.75 a
4	7.63 a	7.85 a	7.75 a	7.95 a
6	7.80 a	7.82 a	7.95 a	7.78 a
8	7.68 a	7.62 a	7.57 a	7.59 a
10	7.46 a	7.70 a	7.92 a	7.78 a
12	7.37 b	7.78 b	8.47 a	7.78 b
14	6.86 c	7.43 b	8.13 a	7.66 b
16	6.60 b	7.83 a	7.81 a	7.71 a
18	6.30 b	7.54 a	7.55 a	7.70 a
20	4.60 b	7.33 a	7.74 a	7.70 a

Means followed by the same letter in a row do not differ significantly by the Scott-Knott test at 5% probability.

Between concentrations of 1 and 10 mg L^{-1}, the effects of all coagulants were not significant in relation to pH, as shown in Table 3. At a 12 mg L^{-1} concentration, Tanfloc SG had a significant positive effect on pH, in contrast to the other coagulants.

At a concentration of 14 mg L^{-1}, aluminum sulfate and Tanfloc SG provided a significant change in pH relative to the results for the other coagulants, and the pH was maintained below 7 and above 8, respectively. At concentrations of 16 mg L^{-1} and 18 mg L^{-1}, aluminum sulfate caused the water pH to dip below 7 (Table 3). Although significant differences in pH were obtained depending on the coagulants used, all values were within the standards established by Ministry of Health Resolution No. 888 (BRAZIL, 2021).

At a 20 mg L^{-1} concentration, aluminum sulfate caused a large reduction in the pH of the raw water after the coagulation process, which decreased to 4.6 (acidic), as shown in Table 3. This value is not in accordance with the recommendation by Brazil (2021), which makes the use of this coagulant without alkali unfeasible at this concentration. The effects of different aluminum sulfate concentrations on the coagulation of raw surface water are shown in Figure 2.

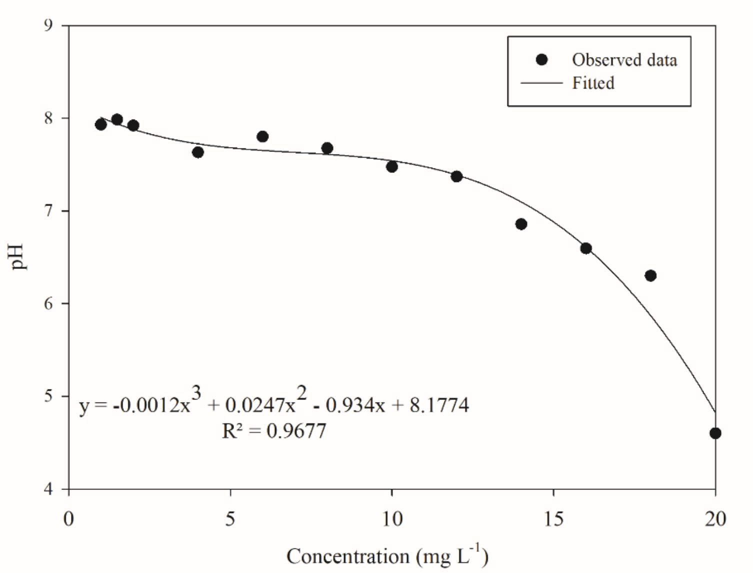

Figure 2. pH values of treated water due to the use of aluminum sulfate as a coagulant at different concentrations.

The mathematical model that best explains the effect of different concentrations of aluminum sulfate on the pH of the treated water is a cubic equation (Figure 2). In this case, 20 mg L^{-1} aluminum sulfate provides the lowest pH value, 4.59, while 1 mg L^{-1} leads to a pH of 8.01.

Based on the trend of pH in the treated water samples, the use of aluminum sulfate caused the pH to decrease, especially starting at a concentration of 14 mg L^{-1} (Figure 2). At concentrations higher than 18 mg L^{-1}, the pH was lower than that recommended by Brazil

(2021). When in contact with the natural alkalinity of raw water, aluminum sulfate forms aluminum hydroxide, which causes the formation of flocs, sulfuric acid, and carbonic acid, which are responsible for water acidity (SANEP, 2017). Figure 3 shows the effects of different concentrations of *M. oleifera* on water coagulation.

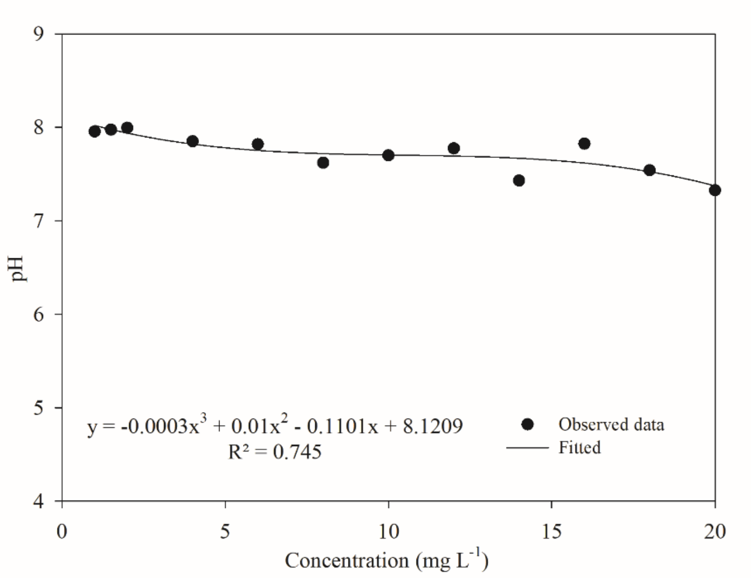

Figure 3. pH values of treated water with respect to the concentration of *M. oleifera* seeds used as a coagulant.

The effect of different *M. oleifera* concentrations on the pH of the treated water can be explained by a cubic equation model, as shown in Figure 3. The maximum pH (7.96) was obtained for 1 mg L^{-1} coagulant, whereas the lowest pH (7.45) was obtained for 20 mg L^{-1} *M. oleifera* seeds.

According to Ndabigengesere and Narasiah (1998), the use of *M. oleifera* seeds instead of aluminum sulfate as a coagulant does not significantly alter the pH of the treated water. In this study, there was a significant difference in pH due to *M. oleifera*-based coagulant concentrations, however, there was no reduction below 7.0 pH value. Additionally, as presented by the same authors, the performance of *M. oleifera* seeds as a coagulant is not influenced by the initial pH of the raw water. The pH values for different Tanfloc SG dosages are shown in Figure 4.

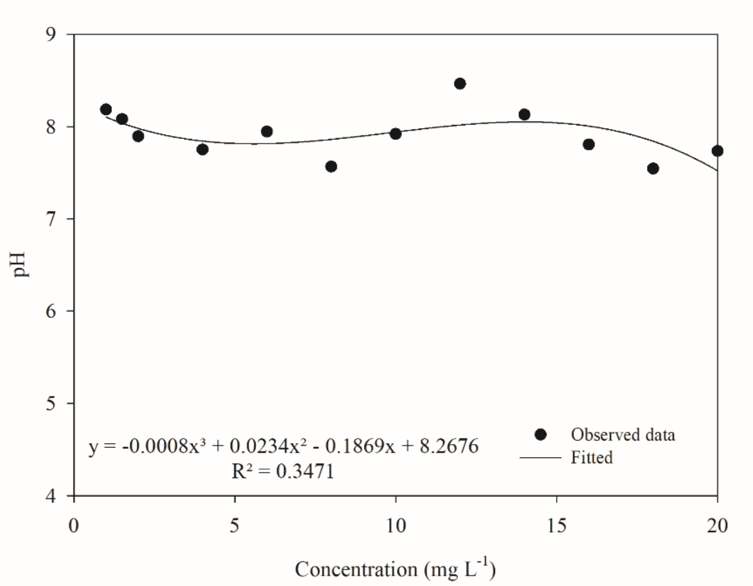

Figure 4. pH values of treated water with respect to the concentration of Tanfloc SG used as a coagulant.

The effect of different Tanfloc SG concentrations on the pH of the treated water can be explained by a cubic equation, as shown in Figure 4. According to this mathematical model, the lowest expected pH is 7.49 at a Tanfloc SG concentration of 20 mg L^{-1}, whereas the highest pH is 8.10 for a concentration of 1.0 mg L^{-1}.

Although the pH underwent significant changes as a function of the concentration of Tanfloc SG used (Figure 4), the obtained values remained within the potability standards in Ordinance No. 888/2021 (BRAZIL, 2021). Sousa (2015) investigated the influence of time and sodium sulfite concentration on the efficiency of tannins in the coagulation process; the pH decreased more in treatments using aluminum sulfate than in treatments using tannins from *Anadenanthera peregrina*; the latter coagulant resulted in a reduction of only 0.35, similar to the results observed in the present study.

In contrast to the other coagulants, the use of Tanfloc SL for the coagulation of surface raw water at concentrations between 1.0 and 20 mg L^{-1} did not cause significant changes in the pH of the treated water. Thus, the use of this coagulant is feasible at the lowest concentration because even at this concentration, turbidity was completely removed from the raw water. Additionally, all pH values obtained were in accordance with the recommendations of Brazil (2021). This result corroborates with the presented by Barradas (2004), who found that tannin-based coagulants do not cause changes in the pH of treated water because they do not consume alkalinity in the medium. Despite the significant differences in pH observed with the use of the other tannin-based coagulant (Tanfloc SG), these differences were minimal, which agrees with the findings of the abovementioned study.

Aluminum or iron sulfate hydrolyzes when added to water, releasing $3H^+$, which leads to a reduction in pH. Thus, in the water treatment process, when these coagulants are used, it is necessary the use alkalinizing solutions to for neutralizing treated water (as can be seen by analyzing Figure 2). Such amendments can make the treatment process more expensive; furthermore, when soda is used for this, Na is added to the sludge, which is another disadvantage of using chemical coagulants. Tannins and *M. oleifera* seeds are polymers, and they do not change when dissolved in water; that is, their original structure is retained in sludge, and therefore, there is no need to use alkali, as shown by the pH values obtained in this work.

4 CONCLUSIONS

Based on the jar tests performed, complete turbidity removal was achieved with the use of the coagulants *M. oleifera* seeds, aluminum sulfate, Tanfloc SG and Tanfloc SL at all concentrations and in all replicates. Therefore, the values remained within the parameters established by Ministry of Health Resolution No. 888/21 (Brazil, 2021), which indicates that the turbidity should be lower than 5 NTU.

The use of aluminum sulfate as a coagulant caused a decrease in water pH starting at a dosage of 14 mg L^{-1}; however, the resulting pH values were within the potability standards established by the Brazilian Ministry of Health. At dosages higher than 18 mg L^{-1}, the pH reached values that do not comply with the established ordinance.

The use of natural coagulants for water treatment was found to be efficient at all coagulant dosages used, as they were able to achieve complete turbidity removal while maintaining the pH of the treated water within the optimal range.

Because there was complete removal of turbidity and acceptable pH values at all dosages of the natural coagulants used, it would be convenient to use the lowest dosage, which was 1 mg L^{-1}; the recommended concentration for aluminum sulfate is also 1 mg L^{-1}.

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