



Barium adsorption using extract from *Moringa oleifera* bark and seeds

Adsorção de íons bário a partir de extrato das cascas e sementes de *Moringa oleifera*

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This study investigated the application of *Moringa oleifera* extract as a sustainable and effective solution to remove barium from potable water. The results showed removal efficiencies up to 98.97% in experiments conducted with extract concentrations of 50 mg.L⁻¹. The study utilized fractions derived from *Moringa oleifera* seeds and barks and demonstrated that the adsorption process is significantly influenced by key variables, including pH and temperature, with optimal performance observed under neutral pH conditions. However, the gradual degradation of the biosorbent and the associated increase in chemical oxygen demand (COD) of the treated effluent present challenges to the long-term implementation of this approach. Despite these limitations, the findings underscore the substantial potential of *Moringa oleifera* as an environmentally sustainable and cost-effective solution for the remediation of water contaminated with heavy metals.

Keywords: *Moringa oleifera*, barium adsorption, treated effluent.

Este estudo investigou o uso do extrato de *Moringa oleifera* como uma alternativa sustentável e eficiente para a remoção de bário de águas de abastecimento, atingindo até 98,97% de remoção em experimentos com concentrações de 50 mg.L⁻¹ do extrato. A pesquisa, conduzida com frações de sementes e cascas de *Moringa oleifera*, demonstrou que a adsorção do metal é influenciada por variáveis como pH e temperatura, com maior eficiência em condições neutras. Contudo, a degradação do biossorvente ao longo do tempo e o aumento na demanda química de oxigênio (DQO) do efluente tratado apontam desafios na aplicação contínua desse método. Apesar dessas limitações, a *Moringa oleifera* apresenta um potencial significativo para tratamento de águas contaminadas por metais pesados, destacando-se como uma solução ambientalmente amigável e acessível.

Palavras-chave: *Moringa oleifera*, adsorção de bário, efluente tratado.

1. INTRODUCTION

Ensuring access to quality drinking water is a fundamental global challenge for human health and well-being, especially in regions where the water treatment infrastructure is precarious. Potable water means water free of pathogens and toxic substances, suitable for essential activities such as ingestion, personal hygiene, and food preparation [1]. Conventional water treatment methods involve the sequential processes of coagulation, flocculation, and sedimentation, with chemical coagulants being the most widely used. However, natural coagulants, obtained from various types of biomass, have been proven to be viable for reducing impurities in water, allowing a reduction in water treatment costs [2], which is an interesting factor, especially in underdeveloped countries that still need to improve their basic sanitation infrastructures.

Moringa oleifera, a tree native to South Asia, has gained prominence for its coagulant and adsorptive properties [3, 4]. Its seeds contain active compounds that can agglutinate particles suspended in water, facilitating the impurities removal [5]. *Moringa oleifera*, in addition to its coagulant properties, has high biosorption potential, which makes it a promising alternative for removing heavy metals from water [6].

The combination of natural with chemical coagulants has shown promising results in water treatment, as evidenced by Valverde et al. (2018) [7]. Water was collected from the Pirapó River basin in Maringá, Brazil, specifically at the collection point of the water treatment plant (WTP), after pre-treatment stages that include screening and grit removal, which aim to reduce the presence of coarse solids [7]. The authors observed that the use of *Moringa oleifera* as an auxiliary coagulant led to a reduction of up to 40% in the consumption of polyaluminum chloride (PAC). It is important to emphasize that the efficiency of the coagulation, flocculation, and sedimentation processes was not compromised, with sedimentation times of only 15 minutes being sufficient, considering a water pH ranging from 6 to 8 [7].

Natural coagulants can improve the efficiency of chemical coagulants, allowing for more effective removal of impurities from water, which can result in enhanced treated water quality and a reduction in the need for additional treatment processes [2].

Heavy metals such as barium (Ba^{2+}) are commonly found in wastewater and water supplies, posing serious health risks due to their toxicity [8]. According to Graça et al. (2022) [9], contamination from natural sources includes the presence of barium in minerals such as barite and witherite, which occur in the Earth's crust. Anthropogenic sources are of greater concern and include industrial emissions, for example, the combustion of coal and diesel oil, mining and refining of minerals, the use of fertilizers and pesticides in agriculture, and the improper disposal of industrial and urban waste [9].

The barium presence in high concentrations is associated with sources of contamination, such as the inadequate disposal of industrial sewage, which can lead to serious toxicological problems for living beings, due to its capacity for bioaccumulation in the trophic chain [10]. Although previous studies have demonstrated the effectiveness of *Moringa oleifera* in removing various contaminants [11, 12], exploration of its specific potential for removing barium is still limited.

Given the problem raised, the current study evaluated the efficiency of *Moringa oleifera* extract in removing barium from water supplies, offering a sustainable and effective alternative for the water contaminated by heavy metals treatment.

2. MATERIAL AND METHODS

The study was carried out at the Environmental Sanitation Laboratory and the Magnetic and Superconducting Materials Laboratory of the Federal University of Triângulo Mineiro (UFTM). The macerated extract of *Moringa oleifera* seeds and bark was used, collected from a tree located in a public space in the city of Uberaba (MG), in July 2022.

To simulate a water supply contaminated with Ba^{2+} ions, a solution was prepared by diluting 30 mg of anhydrous barium chloride ($BaCl_2$) in 1 L of distilled water (single-element solution). The stock solution of *Moringa oleifera* extract was prepared according to methodology proposed by Madrona et al. (2010) [13], which consisted of adding 1g of macerated fractions to 100 ml of 1 M sodium chloride (NaCl) solution. Previous studies indicate that the solvent choice for extracting *Moringa oleifera* significantly affects the activity of various functional groups in plant coagulation. Specifically, NaCl has been found to be particularly effective in recovering proteins and polysaccharides, which are associated with enhanced turbidity removal [14]. From the stock solution, three different solutions were produced, with extract concentrations equal to 50, 75, and 100 $mg.L^{-1}$ [13].

The Ba^{2+} ion adsorption tests were performed in batches (duplicate), adding 100 mL of the standard *Moringa oleifera* extract solutions to 2 L of the solution containing the metal. The solutions were continuously stirred in a magnetic stirrer (200 rpm) for 30 minutes and then left to rest. Five samples were collected at residence periods of 0, 1, 7, 23, and 28 days to determine the remaining concentrations of the analyzed metal. Observing adsorption in this way allows for effective monitoring of the removal of remaining metals, since most of the extraction occurs in the first few days, with efficiency gradually decreasing after this period, allowing accurate assessment of the performance of the biosorbent over time [15].

To determine barium concentrations in the liquid medium, after adsorption tests, acid digestion was initially performed at 150°C (Tecnal® Te-008/50-04 Digester Block) for 30 minutes, as

recommended by Melo et al. (2021) [16]. The liquid fractions were then sent for microwave-induced plasma atomic emission spectrometry analysis (MP-AES 4200 –Agilent).

Turbidity, temperature, COD concentration and pH were also monitored at intervals of seven days, following procedures from Standard Methods for the Examination of Water and Wastewater (2023) [17]. For the barium removal, turbidity, temperature, and pH data, two-way analysis of variance (ANOVA) followed by Tukey's test was applied ($p < 0.05$).

3. RESULTS AND DISCUSSION

Table 1 presents the results of barium removal efficiency in tests with different concentrations of *Moringa oleifera* extract. The highest removal efficiency (98.97%) was obtained on day 0 with an extract concentration of 50 mg.L⁻¹ (Table 1). All extract concentrations demonstrated satisfactory removal and were similar to each other, if the built-in experimental error was considered. Duplicate sampling was performed for all experiments, which showed identical results.

Table 1: Initial concentration, final concentration, and barium removal throughout the experimental period.

Extract concentration (mg.L ⁻¹)	Final concentration (mgBa. L ⁻¹)	Barium Removal (%)	Time (days)
50	0.31	98.97	0
	0.50	98.33	1
	0.45	98.50	7
	0.56	98.13	23
	0.70	97.67	28
75	0.37	98.77	0
	0.47	98.43	1
	0.42	98.60	7
	0.64	97.87	23
	0.63	97.90	28
100	0.41	98.63	0
	0.59	98.03	1
	0.55	98.17	7
	0.71	97.63	23
	0.81	97.30	28

A slight reduction in barium removal was observed over time (Table 1). This fact can be explained by the possible degradation of *Moringa oleifera* due to changes in the medium over time, a process that resulted in the release of adsorbed barium into the liquid medium. This behavior was observed by Araújo et al. (2013) [3], where metals such as lead, cadmium, and nickel, with reported removal values of between 50% and 80% from contaminated water, were released after the degradation of *Moringa oleifera*. In their study, Araújo et al. (2013) [3], reported that thermal or chemical degradation of *Moringa oleifera* biosorbents, such as leaves or seeds, can result in the release of metals previously adsorbed into the aqueous medium, due to the deterioration of the biosorbent structure over time.

When analyzing the adsorption curves for zeolites obtained in the study of Jimenez et al. (2004) [18], it can be observed that the metals cadmium (II), manganese (II), and nickel (II) present greater retention as the pH of the solution increases, due to the solution having a lower concentration of free H⁺ ions. In the ion exchange process in zeolite, more acidic solutions favor more intense competition between H⁺ ions and transition metal ions for adsorption, making retention of these metals difficult [18]. Furthermore, when comparing adsorptions at different temperatures, Jimenez et al. (2004) [18] observed that the adsorption rate increased considerably with temperature, since this is a common behavior for metals due to the ionic radius of hydrated species.

The increase in barium concentration over the experimental period is depicted in Figure 1.

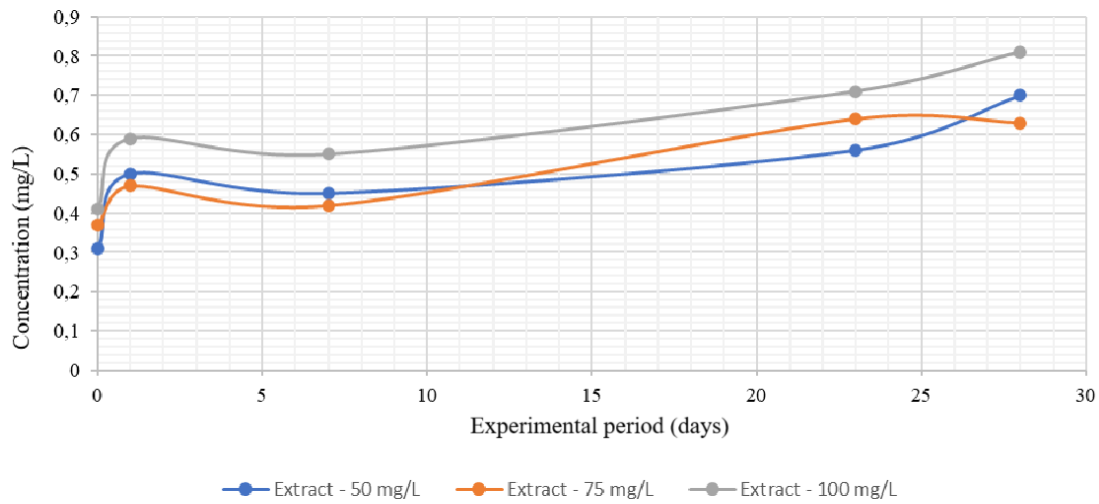


Figure 1: Variation in barium concentration over the 30 days of the experiment.

The statistical evaluation of the barium removal efficiencies obtained throughout the experimental period by the Tukey test ($p < 0.05$) showed no significant differences between the extract concentrations used. Figure 2 presents the interquartile range of barium removal percentages in each of the three *Moringa oleifera* extracts.

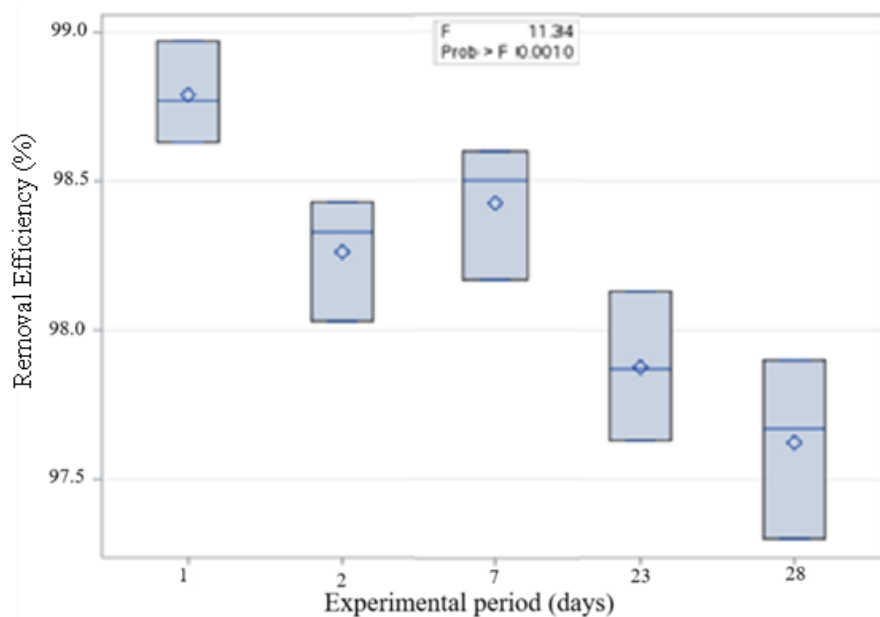


Figure 2: Distribution of barium removal percentage by statistical tests.

In addition to the degradation of the biosorbent, caused by changes in temperature and pH, Shan et al (2017) [19] observed an increase in the COD of the treated effluent after using *Moringa oleifera* as a coagulant, as we observed in this study (Table 2). The COD increase is attributed to release of soluble organic compounds and nutrients from crude plant extracts into the treated water [19]. The organic matter excess can adversely affect water quality by causing discoloration, unpleasant odors, and taste alterations. Additionally, it can promote bacterial proliferation during storage and facilitate the formation of carcinogenic trihalomethanes upon subsequent chlorination

[20]. To address this issue, the literature recommends purifying the extract by isolating the active agents and removing proteins and other organic constituents from the seed that are not essential for adsorption [21].

For the pH, a small increase was observed with the increase in residence time, from the seventh day onwards (Table 2).

Table 2: Mean values of COD, turbidity, pH, and temperature throughout the experimental period.

Extract concentration (mg.L ⁻¹)	COD (mg.L ⁻¹)	Turbidity (uT)	pH	Temperature (°C)	Time (days)
50	33.79	1.67 ± 0.23	6.00	22.0	0
	41.88	1.01 ± 0.13	6.00	22.0	1
	20.63	1.61 ± 0.09	6.09	22.2	7
	53.01	0.56 ± 0.01	6.15	22.4	23
	122.18	0.72 ± 0.11	6.45	21.2	28
75	20.63	3.37 ± 0.05	6.00	22.0	0
	23.95	3.11 ± 0.04	6.00	22.0	1
	25.60	4.78 ± 0.1	6.35	22.1	7
	32.16	1.51 ± 0.03	6.63	21.8	23
	67.01	0.79 ± 0.03	7.05	21.9	28
100	56.15	5.24 ± 0.08	6.00	22.0	0
	38.66	7.38 ± 0.33	6.00	22.0	1
	22.29	6.91 ± 0.74	6.18	22.4	7
	115.28	2.63 ± 0.49	6.79	21.9	23
	99.73	1.76 ± 0.07	6.95	22.5	28

Statistical evaluation of turbidity, temperature, and pH values obtained throughout the experimental period using Tukey test ($p < 0.05$) showed no statistically significant differences. Two-way analysis of variance (ANOVA) followed by Tukey test ($p < 0.05$), did not demonstrate any statistically significant differences in turbidity, temperature, and pH data.

Jimenez et al. (2004) [17] identified that a pH close to 6 is more favorable for metals adsorption, which may represent the greater barium retention using *Moringa oleifera* as a biosorbent. Regarding temperature, Jimenez et al. (2004) [17] observed that adsorption efficiency fell to 70% at an average temperature of 20 °C, which may explain barium release during the experimental period (Table 2).

In any case, results attained in this study reveal high efficiency in barium removal using *Moringa oleifera* extract as a biosorbent. This finding is in line with previous studies that demonstrated *Moringa oleifera* potential for removal of other heavy metals from water supplies, such as iron and aluminum [22, 23]. Barium removal efficiency observed in our work (Table 1) exceeds those reported for other traditional treatment methods. As evidenced by Melo et al. (2021) [16], *Moringa oleifera* presents the potential to adsorb high fractions of metal in contaminated aqueous media. However, the degradation of the coagulant over time and consequent increase in COD represent challenges that need to be overcome to enable the continued use of this biosorbent in water supply treatment. Abatal et al. (2021) [24] demonstrated that *Moringa oleifera* is capable of removing up to 95% of lead, cadmium, and nickel from aqueous solutions. Although the study focused on different metals, the high removal rates found in both cases highlight the versatility and effectiveness of *Moringa oleifera* as a biosorbent in different contexts.

Sharma et al. (2006) [25] investigated the removal of cadmium using *Moringa oleifera* seed powder. The authors observed a maximum adsorption efficiency of 97% [25]. Reddy et al. (2010) [26] studying cadmium, lead, and copper, observed removal efficiencies higher than 90% when using *Moringa oleifera* bark as a biosorbent. Although the removal efficiency of these metals is high, the results of the present study with barium are higher still, which may indicate a particular affinity of *Moringa oleifera* with barium compared to other metals. Hadidi et al. (2022) [14], Mageshkumar and Karthikeyan (2016) [27], and Redha (2020) [28] used NaCl extraction to

obtain a coagulant from *Moringa oleifera*. FT-IR analysis showed that amide, hydroxyl, carboxyl, and carbonyl groups - positively charged soluble proteins with isoelectric points between 10 and 11 - aid in adsorbing and neutralizing particle charges and metal ions. This likely explains *Moringa oleifera*'s strong affinity for barium observed in our study.

Finally, the study by Sajidu et al. (2005) [29] on the removal of lead from effluents also reinforces the effectiveness of *Moringa oleifera* as a coagulant in the treatment of industrial wastewater. Compared to the present study, where barium removal reached almost 99%, it is evident that *Moringa oleifera* is a promising option not only for treating the water supply but also for wastewater containing heavy metals.

4. CONCLUSIONS

Based on results presented, current study confirmed the extract of *Moringa oleifera* bark and seeds efficiency in the removal of barium from water supplies, in between 0- and 7-days residence period. In all experiments, barium adsorption exceeded 95%, and maximum removal was achieved with an extract concentration of 50 mg.L⁻¹, resulting in an extraction efficiency of 98.97%.

However, in addition to biosorbent degradation, caused by changes in temperature and pH, an increase in COD of treated effluent was observed after the use of *Moringa oleifera* as a coagulant. This increase suggests that biosorbent degradation can lead to organic compounds release into the liquid medium, consequently increasing the treated effluent organic load. These high COD values are attributed not only to the material degradation, but also to organic matter presence in the plant structure itself.

Furthermore, the adsorption of heavy metals using *Moringa oleifera* can intensify or reduce depending on temperature and pH variables, which may alter the biosorbent structure, leading to its degradation and compromising the integrity of adsorbent that may result in the release of adsorbed metals into the liquid medium.

Therefore, the main challenge is to develop strategies to mitigate degradation of biosorbent extract and maintain its efficiency in the long term. Despite this challenge, *Moringa oleifera* demonstrates great potential for barium removal from water, being a promising and sustainable alternative for treatment of water contaminated by heavy metals.

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