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Assessment of Natural Bentonite Efficacy for Dye Removal in Textile Wastewater Treatment: Implications for Mitigating River Citarum Pollution

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Abstract: Textile industries contribute significantly to the economy but release harmful pollutants into the environment, especially rivers. The effluent from the textile industry contains toxic dyes that can harm the river ecosystem. Several studies have been conducted to reduce toxic dyes in a river system using bentonite as an adsorbent to reduce river pollution effectively. However, the effectiveness of bentonite still needs to be tested again using textile liquid waste that has not gone through any waste processing at all. Citarum is one of the main rivers on Java Island, which suffers from textile effluent, especially azo dyes which are toxic, mutagenic, and carcinogenic which can harm the aquatic ecosystem. Therefore, this study aims to implement natural bentonite as an adsorbent to remove dyes from textile wastewater. We performed a laboratory test to adsorption on bentonite and textile wastewater considering the variation of adsorbent weight of 10 g and 20 g in 100 mL of textile wastewater stirred in an Erlenmeyer flask at room temperature for 0-300 minutes. The initial concentration of textile wastewater used was 10%, 30%, and 50%. We found that the maximum dye removal efficiency was 91.25% with 10% initial concentration treatment, 20 g adsorbent weight, and 60 minutes contact time. Longer contact time will increase the removal efficiency and adsorption capacity, while higher adsorbent dosage will decrease the concentration of dyes in wastewater. Efficient textile wastewater treatment has improved water quality, effectively meeting river water quality standards and environmental regulations.

Keywords: Natural Bentonite, Dye removal, Textile wastewater, Adsorption, River pollution, River Citarum

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1. Introduction

The textile sector is a major force behind commerce, innovation, and employment growth on a worldwide scale. Unquestionably, it has fueled economic expansion by supporting millions of people and increasing consumer demand for a wide variety of textile and clothing products (Rahman & Tabassum, 2024). Instead due to the textile industry's rapid expansion and intensification, there are now a lot more environmental risks, especially those related to water contamination. Pollution in aquatic bodies is caused by the release of untreated or improperly treated wastewater, which contains a range of toxins, including synthetic dyes. These dyes are essential for producing vibrant color in textiles, but they pose a significant environmental risk due to their toxicity, persistence, and tendency to bioaccumulate in aquatic habitats (Kuśmierek *et al.*, 2023).

To date, the dye removal process in textile liquid waste has been widely carried out using several processes, namely physics, chemistry, and biology, as well as a combination of these three processes. Some methods that have been developed include coagulation, sedimentation, activated sludge, and adsorption methods. Adsorption is the easiest way to apply (Suryawan *et al*., 2018). In practice, adsorption requires an adsorbent as a binder or contaminant absorber. Bentonite is one of the adsorbents that can be easily found and cheap. Bentonite can reduce dye levels in textile liquid waste. Bentonite content consists of illite, montmorillonite, and quartz, where 85% of the content is montmorillonite (Aichour & Zaghouane-Boudiaf, 2020; Dhar *et al*., 2023; Khan *et al*., 2023). The unique property of natural bentonite is that it has the ability to swell and form colloids when put into water. The swelling ability of bentonite is quite large. This swelling ability makes bentonite an adsorbent with a greater adsorption capacity than other adsorbents (Tahari *et al*., 2022). Therefore, bentonite is suitable for use as an adsorbent in reducing dyes in textile liquid waste. Apart from its low cost, it is also abundant in nature. Research on bentonite used as an adsorbent in degrading dye levels has proven its versatility. Bentonite is able to reduce methylene blue (Boukerroui, 2020) as well as cation and anion dyes Li *et al*., 2018). Natural bentonite is proven to be able to reduce dyes from various kinds of artificial waste such as methylene blue solution with an adsorption capacity of 73.25 mg/g and congo red 73.25 mg/g using natural bentonite from Algeria (Oussalah *et al*., 2019), while natural bentonite from Iraq can reduce methylene blue by 256 mg/g (Jawad *et al*., 2023), reducing dyes from Rodhamine solution by 142.86 mg/L(Priatna *et al*., 2023). Of all these studies, no research has tested natural bentonite directly on the original waste. The condition of textile wastewater in the final reservoir is a collection of several production processes that not only contain one dye but a combination of various types of dyes used during the production process.

One glaring illustration of the detrimental impact of textile effluent pollution is the Citarum River in Indonesia. Thousands of businesses along the Citarum River watershed in the Bandung area of West Java are engaged in the textile industry (Susanti *et al*., 2023). The river used to have clear water and abundant biodiversity but has now been severely contaminated due to the overflow of wastewater from the surrounding companies. The wastewater from these companies contains many contaminants such as heavy metals, dyes, and other chemicals, that are discharged directly into the river without treatment. As a result, the Citarum River has poor water quality, which has resulted in the destruction of aquatic habitats, deterioration of human health, and disruption of community livelihoods (Prayoga *et al*., 2022). The urgency in conducting this research is to address the underlying causes of contamination and implement effective remediation procedures to restore the integrity and health of the Citarum River. This urgency is further emphasized by Presidential Regulation No. 15/2018, which underscores the government's commitment to controlling pollution and protecting watersheds.

Based on the urgent need to mitigate textile wastewater pollution in the Citarum River, the objectives of this study are to determine the performance of bentonite as a natural adsorbent in adsorbing textile effluent dyes in terms of removal efficiency, adsorption capacity, and its effect on water quality. This study's results will serve as a reference for the textile industry in adopting waste treatment technologies that minimize environmental impact, thus promoting sustainable practices and reducing pollution discharges into the Citarum River and similar water bodies. Therefore, while the primary focus of the study may not be directly on the Citarum River, its findings have significant implications for mitigating pollution in this critical waterway and addressing the broader environmental challenges associated with textile wastewater contamination.

2. Materials and Methods

2.1. Materials

This research was conducted at the Testing Laboratory in the Research Center for Limnology and Water Resources, BRIN, Bogor, from January to March 2023. The tools used include a rotary shaker, UV-Vis spectrophotometer Shimadzu 1800, analytical balance, volumetric pipette, Erlenmeyer, 100 mL bottle sample, aspirator, filter paper, test tube, 100 mesh sieve, tray, beaker glass and water quality checker (WQC). The materials used include powdered bentonite, textile liquid waste, and dyestuff analyzers. Untreated textile liquid waste was taken from PT. X in Bandung, West Java. PT. X is one of the large-scale textile industries, with tens of millions of meters of fabric produced annually. Waste from the production process has been partially processed, but it is still thick in color (Figure 1), which, if discharged directly into the river, will pollute the waters.

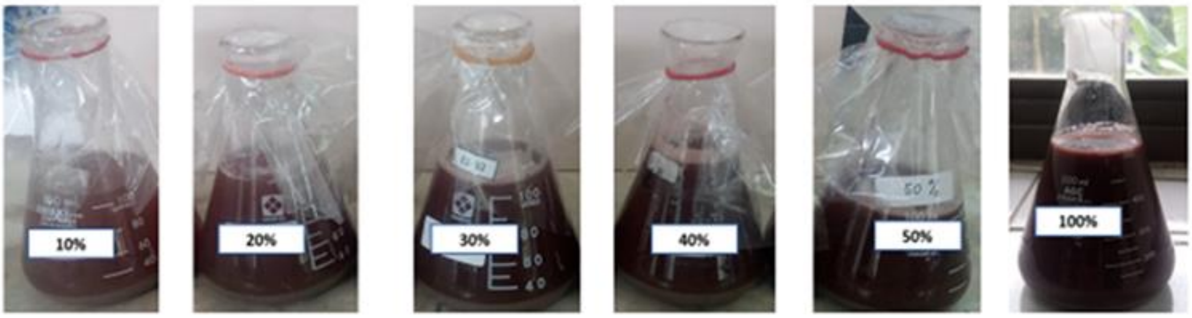

Figure 1. Visualization of textile wastewater from PT. X in Bandung, West Java

2.2. Batch Adsorption

Testing for dye reduction was carried out in batches by adding 10 g and 20 g of bentonite to 100 ml of diluted wastewater with three variations, namely 10%, 30%, and 50%. The solution was stirred at 125 rpm for intervals of 10, 30, 60, 90, 120, 150, 180, 210, 240, 270 and 300 minutes. The suspension was then filtered using a cellulose nitrate filter with a pore size of 0.45 µm and the dye concentration in the supernatant with Pt-Co standard solution using a UV-Vis Spectrophotometer at a wavelength of 456.2 nm. The literature reports on dye adsorption were the basis for selecting these process variables and value ranges (Jamil *et al.*, 2023). The schematic of the batch adsorption process is illustrated in Figure 2. The dye removal efficiency was calculated using Equation 1 and the adsorption capacity was calculated using Equation 2.

$$\%R = \frac{C_i - C_t}{C_i} x\ 100\ \% \quad ....(1)$$

$$q = (C_i - C_t)\frac{V}{m} \quad ....(2)$$

where Ci and Ct are the initial concentration of dye in the waste and at time t (mg/L), %R is the absorption efficiency, q is the adsorption capacity (mg/g), m is the adsorbent mass (g), and V is the wastewater volume (L).

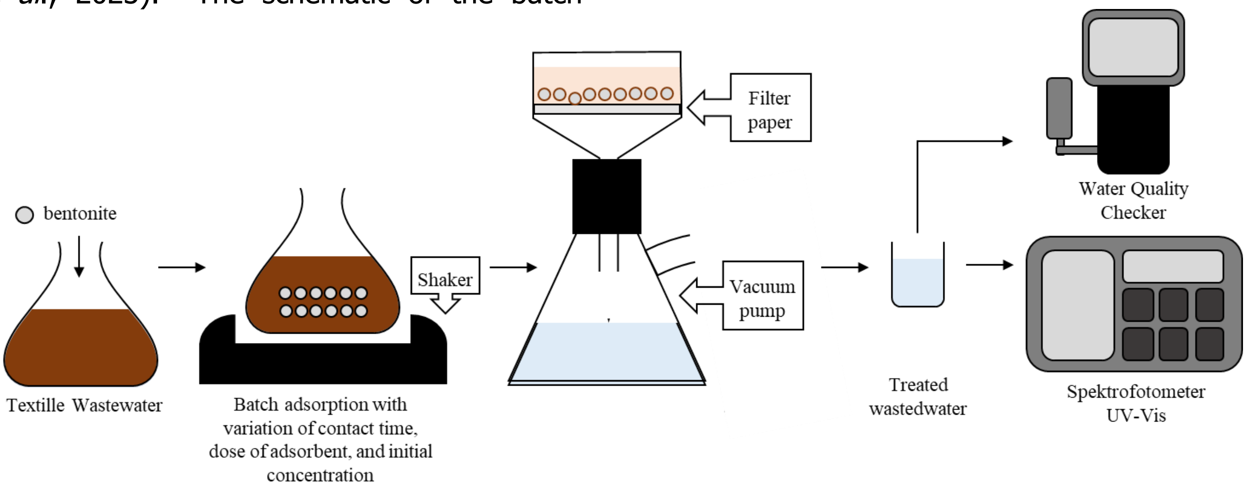

Figure 2. Schematic of dyes adsorption process using bentonite

The study in this research limits the testing of color reduction from textile effluents at a laboratory scale. Batch adsorption studies provide useful insights at laboratory scale but may not directly translate to large-scale industrial applications. Limitations in this research include not accounting for the effects of pH changes on bentonite adsorption capacity and not addressing bentonite regeneration and reuse. Variations in pH can significantly affect adsorption efficiency, and the lack of consideration for bentonite reuse affects the assessment of the feasibility and economic sustainability of the process in real-world applications.

3. Results and discussion

In this research, we focused on the analysis of textile waste obtained from the local textile processing industry because it has a characteristic of a striking red solid color, representing a significant problem in industrial waste management. We managed to measure the concentration of dyes in the waste, with results reaching 2,720.67 Pt-Co units. The intense red color observed in textile wastewater indicated the presence of intense and possibly persistent dyes in the environment. Some textile dyes that often produce red liquid waste include reactive, acidic, direct, and azo dyes, which can significantly negatively impact the environment and human health (Al-Tohamy *et al.*, 2022).

The high concentration of this liquid waste required dilution measures to facilitate further analysis and testing. This study showed textile effluent dilution ranging from 10% to 50%, as illustrated in Figure 1. Textile wastewater exhibits varying characteristics depending on the dilution concentration (Table 1). Dilutions in the 10-100% range at relatively the same temperature and pH conditions in each condition have very high color concentrations. Therefore, three concentrations were chosen that represent the performance of bentonite in reducing dyes, namely at dilution concentrations of 10%, 30%, and 50% with concentration values of 445.67 Pt-Co, 1,122.33 Pt-Co, and 1,774 Pt-Co, respectively. Wastewater with a concentration of 100%—meaning the original waste without additional solvent (distilled water)—was not tested because it is highly concentrated, with a dye concentration of 2,720.67 Pt-Co, and may require pre-treatment to avoid clogging and damage to the adsorbent. The pH conditions of the solution can affect the adsorption process between bentonite and dyes in effluent (Priatna *et al.*, 2023). In the pH range of 6.87 to 8.67 observed in this study, these conditions are generally favorable for dye adsorption by bentonite. At higher pH, there is usually an increase in the number of hydroxyl groups on the bentonite surface, which facilitates interaction with dyes that tend to be acidic. In addition, higher pH can also increase the ionization of dyes, which can increase the electrostatic attraction between dyes and bentonite surfaces (Patanjali *et al.*, 2022).

Table 1. Characteristics of initial wastewater

Dilution Concentration (%)	Dyes (Pt-Co unit)	T (°C)	pH
10	445.67	25.02	8.67
20	529.00	25.10	8.50
30	1,122.33	25.31	8.27
40	1,430.67	25.88	6.64
50	1,774.00	25.97	6.87
100	2,720.67	26.00	6.03

3.1. Effect of Contact Time on Dye Removal Efficiency in Textile Wastewater

At the start of the contact time, the effectiveness of removing dye from liquid waste textiles grows exceptionally quickly. It then keeps getting better throughout each phase of the contact time until it reaches its peak, after which it starts to decline. Adsorption increases with increasing contact time due to increased interaction between adsorbent and adsorbate so that the allowance efficiency value can increase to the equilibrium point (Marella 2019). The equilibrium condition is usually characterized by the point at which the absorption or adsorption of a particular substance by the adsorbent reaches its maximum point before reaching saturation (Parlindungan *et al.*, 2019; Vithalkar & Jugade, 2020). In this study, it was seen that the reduction efficiency rose steadily until 150 minutes, after which the concentration slowly continued to fall. The rapid change in the initial contact time is due to the large number of active sites on the bentonite adsorbent surface. The slowing down of the adsorption process as contact time increases due to the active sites on the adsorbent beginning to be occupied by dye molecules so that the adsorption process becomes constant and tends to decrease (Chauhdary *et al.*, 2022; Huang *et al.*, 2017).

Adding 20 g of bentonite gives a higher efficiency value than 10 g (Fig. 3), with a maximum value of 91.25% in 60 minutes. The increase in color removal efficiency value with increasing adsorbent weight occurs due to a larger surface area and the availability of more adsorption sites (Guezzen *et al.*, 2023; Jawad *et al.*, 2023). Relatively low removal effectiveness resulted from the adsorbent's surface becoming saturated with dyes when its weight is low, even when the concentration of dyes in textile liquid waste remains high.

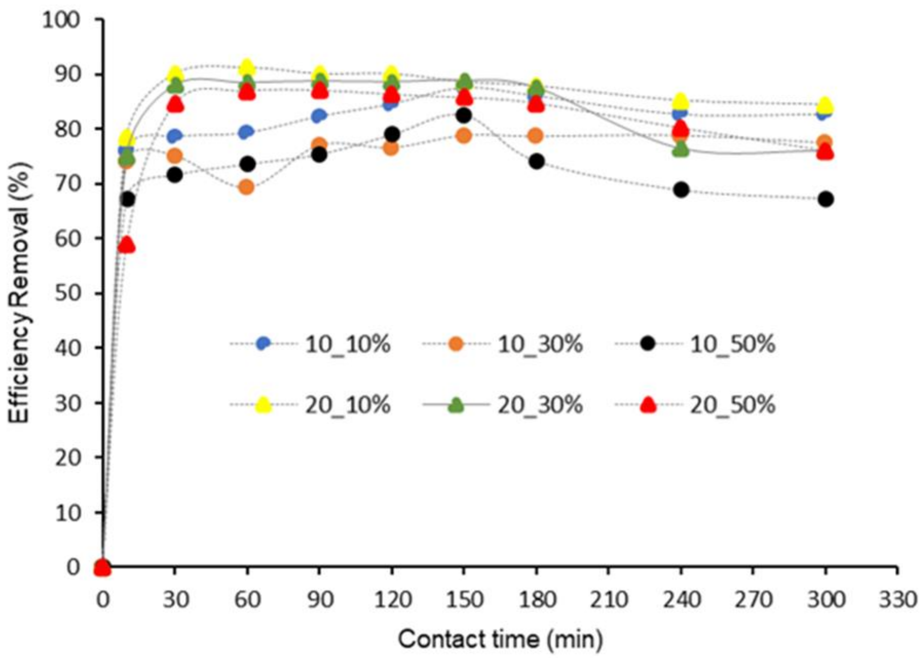

Figure 3. Effect of contact time for dye efficiency removal in textile wastewater

3.2. Effect of Dose for Dyes Adsorption Capacity by Bentonite

The adsorption capacity decreases when larger adsorbent weights are used. The addition of bentonite by 10 g showed that an increase in solution concentration had a significant impact on the rise in bentonite adsorption capacity in absorbing dyes, while the addition of weight by 20 g presented that the increase in solution concentration did not significantly affect the adsorption capacity (Figure 4). This occurred because increasing the amount of adsorbent minerals led to particle aggregation, resulting in a decrease in surface area and an increase in diffusion path length. In addition, increasing the amount of adsorbent causes the number of saturated sites per unit adsorbent to decrease, decreasing adsorbent capacity (Majiya et al., 2023). This implied that a portion of the adsorbent surface remained exposed.

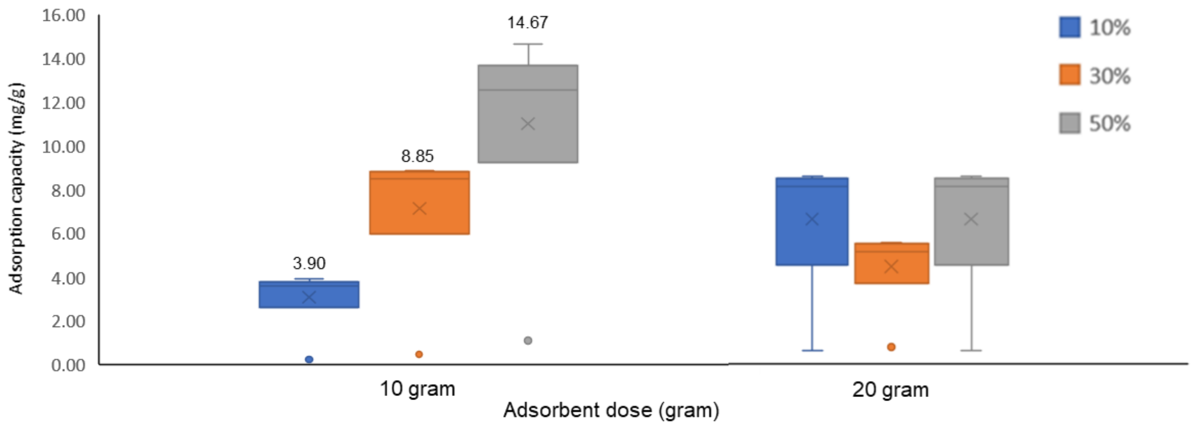

Figure 4. Effect of adsorbent dose to dyes adsorption capacity by bentonite

At equilibrium conditions, namely within 150 minutes, it can be seen how the initial concentration of waste affects the reduction efficiency and adsorption capacity. The two graphs in Figure 5 indicate that the greater the percentage value of the initial concentration of textile liquid waste, the increased adsorption capacity value while the allowance efficiency value decreases. The adsorption capacity increases to a specific concentration along with the increase in the initial concentration of effluent, which aligns with previous research (Albadarin *et al*., 2017; De Gisi *et al*., 2016). The increase in adsorption capacity revealed that active sites are still available on the adsorbent surface that can adsorb textile liquid waste dye molecules. Decreased obstruction of mass transfer between adsorbents and adsorbates when there is a greater concentration difference can also be a cause (Moosavi *et al*., 2020). At the beginning of the adsorption process, the number of adsorbate molecules competing to reach the active side on the surface is very high. While at low concentrations, the adsorption yield (the amount of substance successfully absorbed by the sorbent in the adsorption process) becomes higher due to the small ratio between the dye molecules and the available active side. Moreover, at high concentrations, the pushing force of the adsorbate molecules is higher, so the number of adsorbed molecules is greater.

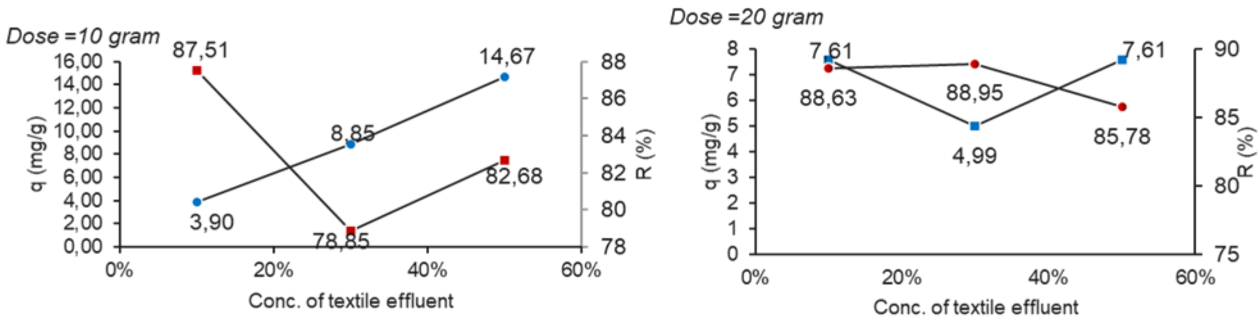

Figure 5. Effect of concentration on removal efficiency and adsorption capacity

3.2. Enhancement of Water Quality

The results revealed that wastewater quality improved after being treated with bentonite. Water quality parameters after treatment were measured under maximum efficiency conditions, namely 10% solution concentration, 20 g dose, and 60 minutes. The analysis showed that the color concentration decreased from 445.67 Pt-Co units to 39 Pt-Co units (%R= 91.25%), so water turbidity also reduced from 648 NTU to 330 NTU. The dissolved oxygen content also rose to 9.77 mg/L, TDS to zero. These parameter values follow class 3 water quality standards intended for fishery activities based on Government Regulation 22 of 2021(Table 2).

Table 2. Water Quality of textile wastewater before and after treatment

Parameter	Unit	Before treatment	After treatment	Threshold*)
Dyes	Pt-Co unit	445.67	33.00	100
Turbidity	NTU	648.00	330.00	400
Total Dissolved Solid (TDS)	mg/L	198.00	0.00	1,000
Dissolved Oxygen (DO)	mg/L	5.13	9.77	3
ORP	mV	20.00	-69.00	-

*)Class 3 water quality threshold based on Government Regulation No. 22 of 2021

Based on this water quality analysis, we can also predict the mechanism of dye absorption by bentonite. Bentonite is a clay mixture of montmorillonite, kaolinite, smectite, and mica. Montmorillonite is one of the main minerals found in natural bentonite, and it has many hydroxyl (OH) and oxygen (O) ions as well as interchangeable cations(Kumari & Mohan, 2021). The high content of hydroxyl and oxygen groups in bentonite provides several advantages in its application as an adsorbent or in cation exchange processes (Dehghani *et al*., 2018). The change in ORP (Oxygen Reduction Potential) value from 20 mV to -69 mV after textile wastewater treatment with bentonite can be explained by bentonite's ability as a reducing agent that reduces oxidative compounds in the effluent, either through cation exchange processes, adsorption of these compounds, or a decrease in the concentration of ions that tend to increase the ORP value, such as heavy metal ions (Daily, 2019). Therefore, treatment with bentonite effectively reduces the oxidation ability of textile wastewater and improves the overall quality of wastewater by lowering pollution levels. The increased dissolved oxygen concentration in water after being treated with bentonite can be caused by the large number of oxygen ions in its structure that can interact with compounds dissolved in wastewater (Ma *et al*., 2021; Oktaviyani *et al*., 2023). This process may cause the release of dissolved oxygen into the water due to reactions between hydroxide ions on the bentonite surface and organic or inorganic compounds in the waste or due to adsorption and cation exchange processes that occur during treatment. Therefore, the rise in dissolved oxygen in water after treatment with bentonite may result from a complex interaction between the oxygen and hydroxide ions in the bentonite structure and compounds in the wastewater. The results of this study, especially regarding the Citarum River, have important implications for the remediation of pollutants from textile effluent. The pre-treatment wastewater quality indicated that dissolved oxygen levels were within acceptable limits for Class 3 water, which is deemed suitable for agricultural and fishery activities according to Indonesian Government Regulation Number 22 of 2021 on Environmental Protection and Management. However, the concentrations of dyes, turbidity, and total dissolved solids (TDS) significantly exceeded the permissible thresholds. Following treatment, all water quality parameters improved to favorable levels for use in fisheries and agriculture.

Natural bentonite can effectively eliminate dye contaminants from wastewater, providing a cost-effective and environmentally friendly solution to reduce river pollution. To maximize the efficiency of bentonite-based treatment systems in industrial settings, addressing various challenges and considerations is essential. These include overcoming issues related to wastewater reuse and adsorbent regeneration and optimizing operational parameters such as pH, temperature, and adsorbent dosage. Additionally, further research through pilot-scale studies and economic analyses is necessary to evaluate the scalability and cost-effectiveness of bentonite-based treatments.

4. Conclusion

The performance evaluation of bentonite as a natural adsorbent to adsorb textile effluent dyes resulted in 91.25% removal efficiency, and the highest adsorption capacity is 14.667 mg/g. Post-treatment analysis showed that the wastewater met river water quality standards and complied with relevant environmental regulations. This research highlights the importance of natural bentonite's function as an effective adsorbent for dye removal in textile wastewater, thus offering a sustainable and cost-effective solution to reduce pollution in rivers such as the Citarum River. The results of this study provide valuable insights for real-world applications, highlighting the need for further optimization of operational parameters and exploration of synergies with other treatment technologies.

Data availability statement

The data included and used in this study is not confidential and is available upon request.

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Conflict of interests

Every author has stated that there is no conflict of interest to the manuscript's writing or submission.

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Author Contributions

AFN and **FZ,** as the main contributors, conceptualized the study and data analysis and wrote the original article. **Z** and **HW** participated in the manuscript review process. All the writers approved the final manuscript.

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