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Treatment of Synthetic Effluent Containing Azo Dyes by Coagulation and Rapid Filtration

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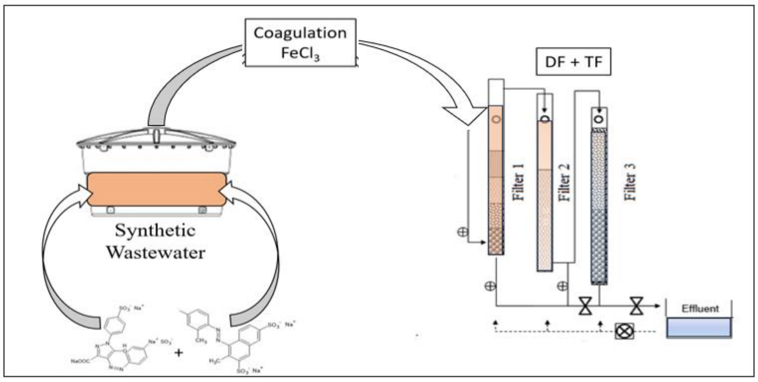
The present study evaluated the treatment of synthetic effluent containing azo dyes using a coagulation system followed by double filtration (DF) and triple filtration (TF). Treatment efficiency was assessed based on the removal of Chemical Oxygen Demand (COD), Biochemical Oxygen Demand (BOD), Linear Alkyl Sulfate (LAS), Total Nitrogen (TN), nitrate, nitrite, turbidity, color, and the dyes Ponceau and Tartrazine. After coagulation and DF, removals reached 76.73% COD, 81.31% BOD, 42.31% LAS, 80.00% TN, 40.96% nitrate, 100.00% nitrite, 99.58% turbidity, and 92.86–100.00% color. Following TF, removals were greater than 77.00% for nitrate, above 98.00% for COD and BOD, and approximately 100.00% for LAS, TN, nitrite, turbidity, and color. Although chloride and conductivity removals were low, the values remained within the effluent disposal limits established by Brazilian legislation. For the dyes, the DF step achieved over 40.00% removal, while TF reduced dye concentrations below the quantification limits for both Ponceau and Tartrazine. The main removal mechanisms were adsorption via the porosity of activated carbon and ion exchange by zeolite. Overall, the combined coagulation, DF, and TF processes demonstrated high removal efficiencies for most evaluated parameters, indicating their suitability for treating effluents containing azo dyes.

Graphical abstract



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1. Introduction

It is estimated that there are approximately 100,000 types of dyes available on the market, with an annual production of around 700,000 tons, a significant portion of which is discarded as domestic or industrial effluents [1]. The main

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sectors generating dye-containing effluents are the textile industry (54.00%), dyeing industry (21.00%), and pulp and paper industry (10.00%) [2]. Textile industries are the largest producers of dye effluents, generating approximately 200.00 liters of effluent per kilogram of fabric produced. The disposal of these dyes into water bodies is a current environmental concern due to their toxicity and the significant risk of ecosystem contamination [3].

Among the different classes, azo dyes are commercially used in food, cosmetics, and textiles, representing over 65.00% of the dyes available on the market. Due to their high toxicity, several countries have banned their production [4,5]. Azo dyes exhibit carcinogenic effects caused by interactions with cytosolic receptors or by the formation of free radicals and arylamines generated during biotransformation [3]·

These dyes contain the azo group (-N=N-) as the chromophore. In monoazo compounds (X-N=N-Y), such as Tartrazine and Ponceau dyes, the X radical represents electron acceptor groups, while Y acts as electron donor groups, which makes them behave as anionic species in aqueous media [6]. The azo groups are mainly linked to benzene or naphthalene rings, as well as heterocyclic or aliphatic aromatic groups. The different absorption wavelengths of these compounds allow their identification in mixtures by spectroscopic methods such as UV-VIS. For example, Tartrazine and Ponceau absorb radiation at 425 nm and 525 nm, respectively [7].

The presence of these dyes in effluents also leads to high biochemical oxygen demand (BOD) values (80.00 to 6000.00 mg/L), chemical oxygen demand (COD) (150.00 to 30,000.00 mg/L), and turbidity (4.02 to 137.00 NTU), representing a challenge for various treatment systems [2,8].

There are various alternatives for treating effluents containing dyes, such as biological processes (bacteria, algae, fungi, yeasts, enzymes) [9-11], advanced oxidation processes (AOPs) [12], electrochemical methods [13], membrane filtration [14], and ion exchange [15]. These technologies achieve significant color removal but face industrial-scale limitations and high costs [2,16].

Coagulation, commonly used for color removal, involves adding iron or aluminum salts to form hydrolyzed products that polymerize and form flakes with suspended material. Ferric chloride ($FeCl_3$) effectively removes color (up to 90.00%) from industrial effluents at a pH range of 5.50 to 8.50 [17]. After coagulation, physicochemical processes like sorption and ion exchange on solid supports are economically viable and efficient [18-20].

Double filtration (DF) and triple filtration (TF) systems have been used for post-treatment of domestic effluents. DF with gravel and sand removes more than 90.00% of parameters such as suspended solids, turbidity, true color (TC), apparent color (AC), and phosphorus, but less than 65.00% for COD and BOD [21]. DF with gravel and clinoptilolite removes over 95.00% of suspended solids, turbidity, BOD above 80.00%, and ammoniacal nitrogen above 98.00% [22]. In TF with gravel, sand, and activated carbon or zeolites, removals of COD, total nitrogen, and ammoniacal nitrogen reach 97.90%, 94.10%, and 99.52%, respectively. These filtration systems are low-cost, easy to operate, and efficient for producing water **reuse** [23,24].

Based on this, the present study evaluates the efficiency of azo dye removal (Tartrazine and Ponceau) from synthetic effluents using coagulation, DF, and TF with gravel, sand, and carbon/zeolite filters.

2. Material and Methods

2.1 Chemical reagents

The Ponceau and Tartrazine dyes, sucrose, soy extract, starch, detergent, and soybean oil were obtained as food-grade standards. Sodium bicarbonate, sodium chloride, magnesium chloride, and calcium chloride were acquired as analytical-grade standards. All solutions were prepared using distilled water.

2.2 Preparation and characterization of synthetic effluent with dyes

The synthetic effluent was prepared based on the characteristics of effluents from the food industry [8]. For the preparation of synthetic effluent, 0.0700 g/L of sucrose, 0.3120 g/L of soy extract, 0.1140 g/L of starch, 0.1140 g/L of detergent, 0.0510 g/L of soybean oil, 0.2000 g/L of sodium bicarbonate, 0.2500 g/L of sodium chloride, 0.0070 g/L of magnesium chloride, 0.0045 g/L of calcium chloride, and 0.0264 g/L of potassium hydrogen phosphate were used in distilled water. The azo dyes Ponceau and Tartrazine were added to this effluent to obtain a final concentration of 6.5000 mg/L. For the study, 300 L of effluent was used.

Table 1 presents the characterization of the synthetic effluent and the methods used for their analysis, developed based on the Standard Methods for the Examination of Water and Wastewater, 23rd edition [25].

Table 1. Physical-chemical parameters for the characterization of the synthetic effluent.

PARAMETERS	SYNTHETIC EFFLUENT	METHOD
BOD (mg/L)	365.20 ± 1.00	Oximetric 5210B
COD (mg/L)	712.20 ± 10.00	Spectrofotometric 5220D
LAS (mg/L)	1.30 ± 0.01	Spectrofotometric (Alfakit)
Conductivity (mS/cm)	0.90 ± 0.00	Conductivity meter 2510 B
pH	7.16 ± 0.23	Potentiometric 4500H$^+$ B
Turbidity (NTU)	210.67 ± 1.53	Nephelometric 2130 B
AC (uC)	625.67 ± 8.62	Spectrometric 2120 B
TC (uC)	8.67 ± 0.58	Spectrometric 2120 B
TN (mg/L)	10.05 ± 0.01	Spectrometric (Spectroquant)
Nitrate (mg/L)	7.91 ± 0.01	Spectrometric (Alfakit)
Nitrite (mg/L)	0.45 ± 0.01	Spectrometric (Alfakit)
Chloride (mg/L)	200.22 ± 21.67	Titrimetric 4500 B
Tartrazine (mg/L)	6.50 ± 0.01	Spectrometric scan
Ponceau (mg/L)	6.50 ± 0.01	Spectrometric scan

Note: BOD: Biochemical Oxygen Demand; COD: Chemical Oxygen Demand; LAS: Linear Alkylbenzene Sulfonate; pH: Hydrogen Potential; AC: Apparent Color; TC: True Color; TN: Total Nitrogen. (Mean (%) ± SD, n = 3).

2.3 Preparation of curves for dye analysis

The determination of Tartrazine and Ponceau concentrations was performed using a UV/Vis Spectrophotometer (DR6000-HACH) in scanning mode within the wavelength range of 200 to 600 nm. Two main wavelengths were selected: 425 nm and 525 nm. For the dye analysis, the external standard method was used with the construction of a curve at concentrations of 1.00, 2.00, 4.00, 6.00, 8.00, 10.00, 12.00, and 14.00 mg/L of the Ponceau and

Tartrazine dyes. Figures 1A and 1B present the typical spectra obtained for the studied dyes.

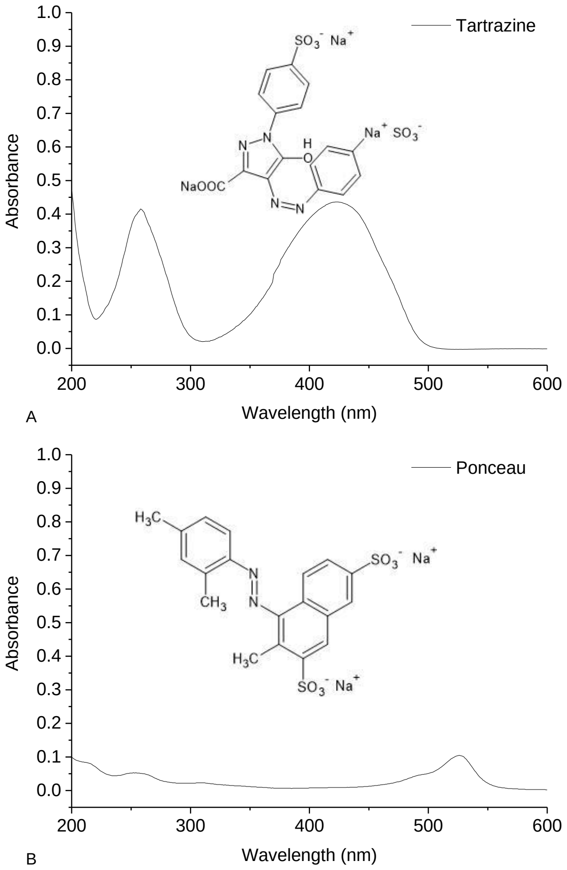

Fig. 1. A: Tartrazine dye absorption spectrum (Band at 425 referring to the chromophore group); B: Ponceau dye absorption spectrum (Band at 525 referring to the chromophore group).

2.4 Coagulation test

The optimal concentration of $FeCl_3$ used in the coagulation process was determined through a Jar-Test assay using synthetic effluent. The control parameters employed during this stage were turbidity and color. The coagulant concentration varied from 29, 58, 116, 174, to 232 mg/L, with a rapid mixing time of 30 seconds and the coagulation pH ranged from 7.00 to 7.50 [23].

2.5 Evaluation of the efficiency of azo dye removal by the DF and TF system

In the evaluation of dye removal efficiency by DF and TF, three concentrations of the dye mix were prepared in the effluent using a water tank to achieve concentrations of 10, 30, and 50 mg/L, representing low, medium, and high levels, respectively. To assess dye removal, samples were collected from the initial solution, after the settling tank, after the DF filter, and after the TF filter. The samples were analyzed for pH, color, turbidity, and dye removal efficiency. In this stage, the filter operated for five hours. Whenever dye breakthrough occurred from DF to TF, a backwash was performed in DF to remove the excess dye, after which filtration resumed. The number of backwashes in DF was recorded for final evaluation. This step was crucial for evaluating the performance of the DF and TF filters in response to variations in dye concentrations, allowing the selection of a final concentration to proceed with the subsequent tests.

2.6 Treatment of synthetic effluent by DF and TF

The filtration system consists of three filters. The first filter (F1) is an upward-flow gravel filter with four layers, each with a height of 300 mm, containing different grain sizes within the following ranges: 19.10 to 38.10; 9.52 to 19.10; 4.80 to 9.52; and 2.00 to 4.80 mm. The second filter (F2) is a downward-flow sand filter consisting of a single layer with a height of 800 mm and grain size ranging from 0.42 to 2.36 mm. The third filter (F3) comprises a layer of clinoptilolite zeolites (900 mm and 0.0455 cm^3/g average pore volume) and a layer of bituminous activated carbon (700 mm and 0.0600 cm^3/g average pore volume). Figure 2 illustrates a schematic representation of the system in a DF configuration (F1+F2) or TF (F1+F2+F3).

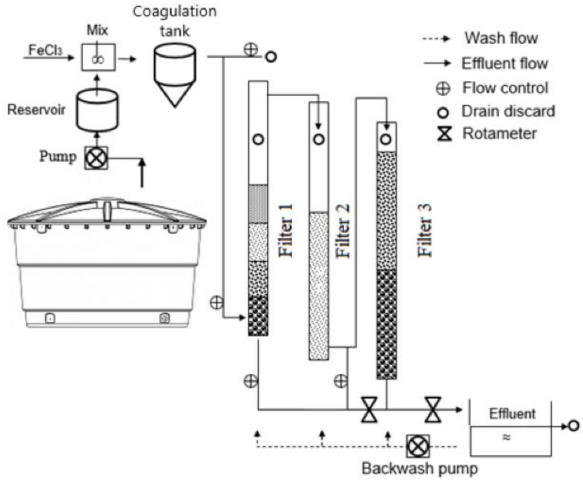

Fig. 2. Flowchart of the pilot double and triple filtration system.

The synthetic effluent was pumped into a 300 L water tank installed at a height of 3.50 m (Figure 1). Subsequently, the effluent was poured through the system, where it received the dosage of ferric chloride previously determined in the Jar-Test tests. It was then directed to a clarifier and subsequently permeated through the filter beds. For DF, only F1 and F2 were used, and for TF, F1, F2, and F3 were employed. The filtration rates adopted were 120 m^3/m^2/day for all filters [23].

3. Results and Discussion

3.1 Coagulation test

The assays to determine the concentration of $FeCl_3$ used in the effluent coagulation are shown in Figure 3.

Based on the results obtained (Figure 3), there is a significant increase in the removal of color and turbidity from the synthetic effluent up to a 116.00 mg/L dosage of the coagulant. In the range of 29.00 mg/L to 116.00 mg/L, turbidity removal increases from 24.52% to 82.88%, while color removal increases from 13.75% to 62.50%, respectively. Within this range, an exponential increase in the removal of both parameters is observed in a small concentration interval. This effect diminishes for the range of 116.00 mg/L to 232.00 mg/L, where it can be seen that after 116.00 mg/L of the coagulant, an increase of approximately 10% in the removal of turbidity and color requires an increase in concentration of about 50.00 mg/L of the coagulant. Thus, considering that this would be a preliminary stage to treatment by DF and TF, this concentration was chosen for subsequent studies. Other studies employing coagulation with $FeCl_3$ followed by DF and TF for the treatment of domestic effluents show a turbidity removal of >80,00% from the effluent by the coagulation process [22, 23].

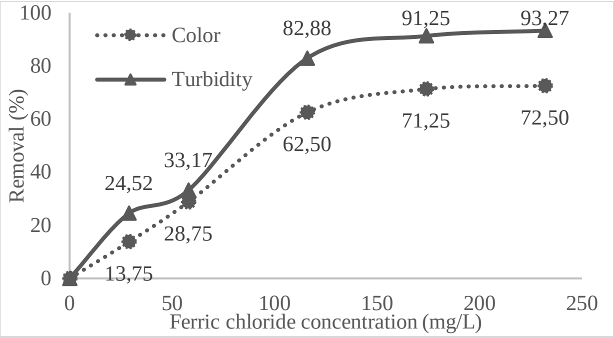

Fig. 3. Removal of turbidity and color from the synthetic effluent, during the Jar-Test coagulation. Ferric chloride concentration used: 29.00, 58.00, 116.00, 174.00 and 232.00 mg/L; stirring time: 30.00 s. Effluent characteristics: pH: 7.16 ± 0.03; turbidity: 210.67 ± 0.29 NTU; and apparent color: 652.67 ± 2.68 uC.

Studies reported similar efficiency in color removal during the coagulation of effluents containing dyes using $FeCl_3$ [17]. At concentrations ranging from 100.00 to 350.00 mg/L of the coagulant, the authors achieved 90.00% removal of the parameter in effluents from textile factories. The use of $FeCl_3$ was found to be more effective than other coagulants such as $Al_2(SO_4)_3$, which achieved color removal of 50.00 to 70.00% at equivalent concentrations. Other authors also suggest that $FeCl_3$ exhibits lower toxicity for the final effluent compared to aluminum-based coagulants. Additionally, $FeCl_3$ aids in the removal of COD, total suspended solids, and Total Nitrogen (TN) [26].

At this stage, the hydrolysis of the coagulant is the main factor in the coagulation process and can be influenced by the characteristics of the effluent, such as pH. Studies describe that coagulation mechanisms using $FeCl_3$ are efficient at a pH range close to 7.16 [23]. This occurs due to the formation of positive species such as $Fe(OH)^{2+}$ and $Fe(OH)_2^+$, which enhance the sweep flocculation mechanism [27].

The color of the effluent, indicative of the presence of dyes, is also removed during this process due to the formation of radicals. Ferric hydroxides have an affinity for dyes in the effluent due to charge differences, promoting mechanisms of electrostatic adsorption. Once adsorbed, the formed flocs tend to be larger and denser, being easily removed by the sedimentation process [27]. Therefore, the removal of dyes begins in the coagulation process through their interaction with the formed iron hydroxides.

3.2 Azo dye removal by the DF and TF system

Table 2 presents the results of dye removal at different concentrations, highlighting the efficiency of the DF and TF filters in relation to the initial dye levels

The removal of the evaluated parameters using DF was efficient across all tested concentrations, with values exceeding 95.00% for color, turbidity, and dye removal. Throughout the analyses, the pH remained stable and close to neutral (ranging from 6.50 to 6.80) under all conditions and for both treatment methods, indicating that the processes do not significantly alter the chemical equilibrium of the effluent.

Table 2. Results of pH analysis and removal of color, turbidity, and dyes after treatment by DF and TF.

	Concentration of the dye mix (50:50) (mg/L)					
	10		**30**		**50**	
	DF	**TF**	**DF**	**TF**	**DF**	**TF**
pH	6.80 ± 0.10	6.80 ± 0.10	6.50 ± 0.05	6.60 ± 0.05	6.50 ± 0.05	6.50 ± 0.05
Color (Removal %)	97.00 ± 0.80	>99.99	95.00 ± 1.00	98.00 ± 0.90	96.00 ± 1.20	98.00 ± 0.90
Turbidity (Removal %)	98.00 ± 0.60	>99.99	95.00 ± 0.80	100.00 ± 0.00	95.00 ± 0.80	98.00 ± 0.70
Dyes (Removal %)	98.00 ± 0.70	>99.99	95.00 ± 1.10	99.00 ± 0.80	96.00 ± 1.00	98.00 ± 0.90
Descargas de fundo (n)	5.00	-	17.00	-	33.00	-

Note: Initial pH: 7.60 ± 0.10, Initial Color: 8.67 ± 1.00 uC, Initial Turbidity: 210.00 ± 0.80 NTU, Filtration time: 5 hours. (Mean (%) ± SD, n = 3).

In terms of color removal, the TF system achieved 100.00% efficiency at the 10.00 mg/L dye concentration, maintaining 98.00% efficiency at both 30.00 and 50.00 mg/L. The DF system also showed high performance, with removal efficiencies of 97.00%, 95.00%, and 96.00% at concentrations of 10.00, 30.00, and 50.00 mg/L, respectively.

Regarding turbidity, TF maintained an efficiency greater than 99.99% at 10.00 and 30.00 mg/L, with a slight decrease to 98.00% at 50.00 mg/L. DF exhibited removal rates of 98.00%, 95.00%, and 95.00% for the same concentrations. Dye removal followed a similar pattern, with TF reaching >99.99% at 10.00 mg/L and 99.00% and 98.00% at 30.00 and 50.00 mg/L, respectively, while DF achieved 98.00%, 95.00%, and 96.00% at the corresponding concentrations.

In DF, the predominant removal mechanism is adsorption, where suspended particles adhere to the filtration media. Over time, the filtration kinetics decrease due to progressive clogging of the media, leading to its gradual saturation. A similar mechanism occurs in TF, although removal also involves charge-based interactions in addition to adsorption [23].

Based on these characteristics, it can be inferred that the filtration kinetics of DF and TF are comparable, though not identical, especially at higher concentrations. As dye concentration increases, the saturation of filtration media occurs more rapidly, leading to a higher frequency of backflush discharges needed to restore filtration capacity. This effect results in elevated operational costs due to more frequent cleaning cycles and the potential need for earlier replacement of filter materials, increasing system complexity. Furthermore, the volume of excess sludge generated from frequent backflush operations presents an additional management challenge.

Considering the results obtained for color, turbidity, and dye removal, and taking into account the operational burden associated with backflushing, a maximum recommended concentration of 10.00 mg/L is suggested for the filtration system. At this level, both DF and TF demonstrate excellent performance (DF ≥ 97.00%; TF >99.99%) with minimal operational disruption, thus enhancing sustainability and cost-effectiveness. At concentrations above 10.00 mg/L, the accelerated saturation of filtration media and increased backflush requirements compromise system efficiency and economic feasibility, especially in large-scale applications. Under these conditions, it is advisable to implement a preliminary physicochemical or biological treatment stage to reduce dye concentration before filtration.

3.3 Physicochemical analysis of the effluent after treatment by DF and TF

Figure 4 shows the removal efficiency of physicochemical parameters from the synthetic effluent using DF and TF systems.

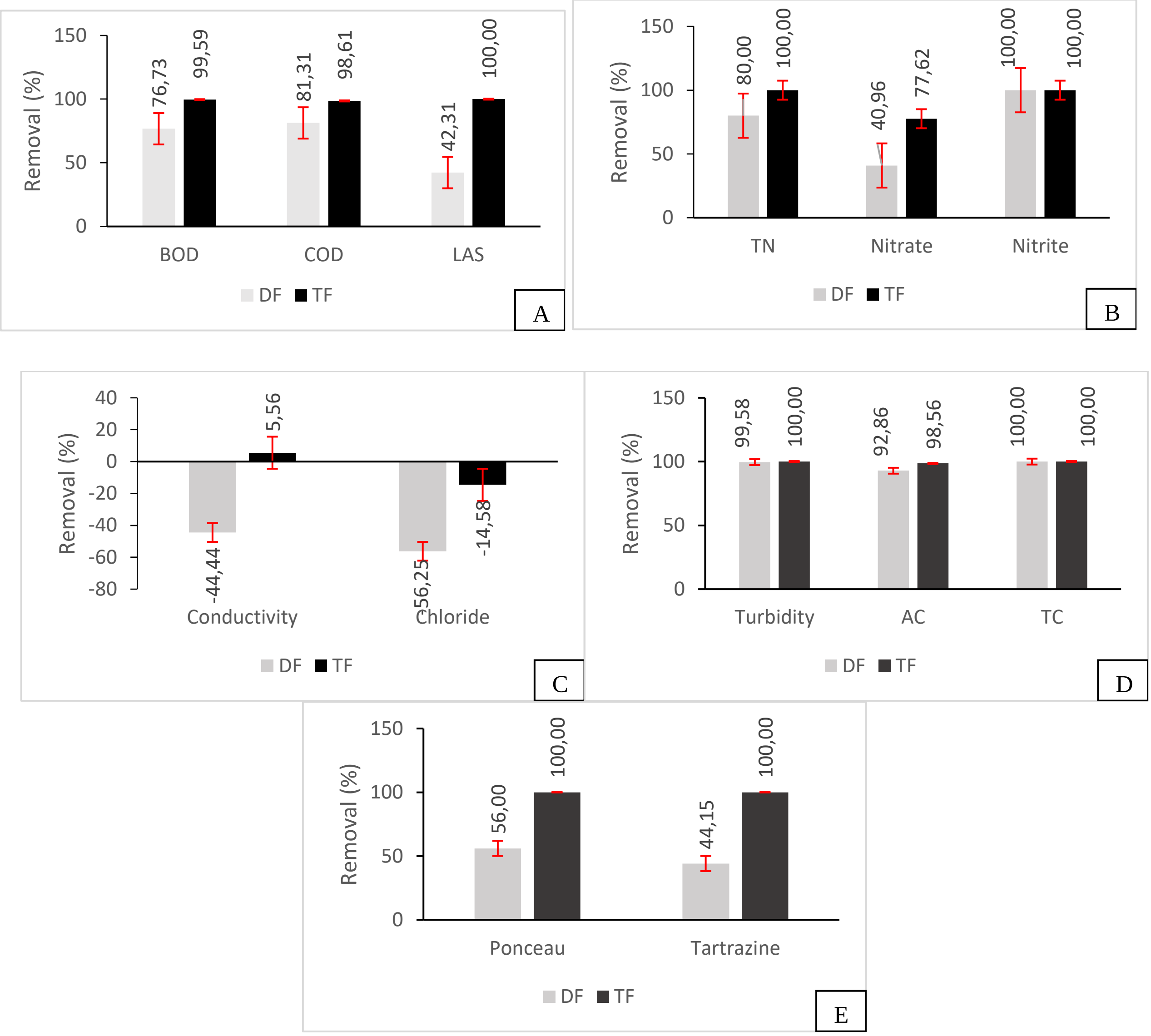

Fig. 4. Removal of A - Biological Oxigen Demand (BOD), Chemical Oxigen Demand (COD), Linear Alquil Sulfonate (LAS); B - Total Nitrogen (TN), Nitrate, Nitrite; C - Conductivity, Chlorine; D - Turbidity, Aparente Collor (AC), True Collor (TC); E - Ponceau and Tartrazine Dyes, from the synthetic effluent, by the DF and TF. Ferric chloride concentration used: 116.00 mg/L; stirring time: 30 s. (Mean (%) ± SD (in red), n = 3).

The removal of COD reached 81.31% with DF and exceeded 98.61% with TF, decreasing from 712.20 mg/L in the raw effluent to less than 10.00 mg/L after TF. This high efficiency in the TF system suggests that COD removal is primarily attributed to the characteristics of the filter media, where the increased surface area enhances interaction with organic matter (OM) through adsorption processes—particularly in media with fine particles, such as activated carbon and zeolite [23,28]. These results are comparable to those achieved by aerobic membrane filtration systems, which can remove over 80.00% of COD, although they are hindered by irreversible fouling on the membrane surface [29,30] and higher maintenance costs [31]. In contrast, sand, carbon, and zeolite filters are low-cost, easy to maintain, and offer extended operational lifespans [32].

BOD removal was 76.73% for DF and 99.59% for TF, with concentrations decreasing from 365.20 mg/L in the raw effluent to 1.50 mg/L after TF. In other studies, sand filtration achieved approximately 75.00% BOD removal, while activated carbon reached around 38.00%, and coagulation even led to a slight increase in BOD levels [5]. The high efficiency observed in TF is largely attributed to the presence of clinoptilolite, which exhibits excellent performance due to its high porosity and ion exchange capacity, promoting electrostatic adsorption and a strong adsorption potential for OM [22-24].

These results are comparable to those obtained with membrane bioreactors (MBR), nanofiltration (NF), and reverse osmosis (RO) systems, which have demonstrated BOD removal efficiencies exceeding 70.00% in combined treatment configurations [33].

Linear Alquil Sulfonate (LAS) removal was 42.31% for DF and nearly 100.00% for TF, with concentrations reduced to below the quantification limit (<0.01 mg/L). LAS, commonly used in fabric washing processes, is frequently discharged into effluents along with dyes. Similar removal efficiencies have been reported in the literature, although such levels typically require advanced oxidation processes (AOPs), which can be complex and costly to implement on a large scale [34]. The presence of an effective treatment system for LAS is essential, as surfactants can interfere with dye adsorption mechanisms and reduce overall treatment efficiency [35].

The results show that TN removal was 80.00% for DF and approximately 100.00% for TF, with concentrations decreasing from 10.05 mg/L in the raw synthetic effluent to below the quantification limit (<0.01 mg/L) after TF. DF achieved a nitrate removal rate of 40.96% (reducing from 2.94 mg/L), while TF reached 77.72% (down to 1.77 mg/L). Nitrite removal was close to 100.00% for both DF and TF, with final concentrations also falling below the quantification limit (<0.01 mg/L).

Previous studies treating textile effluents containing dyes with 1.00 g/L of coagulant at pH 9, followed by adsorption and filtration, reported TN removal of up to 88.10% after passing through a zeolite filter [26]. In contrast, the present study achieved nearly 100.00% TN removal using a coagulant concentration ten times lower and operating at a pH close to neutral (~7.00). These findings are consistent with results from anaerobic/aerobic photobioreactor studies, which reported TN removal greater than 87.00%, with nitrate and nitrite removal rates of 40.40% and 32.40%, respectively. However, those processes required a treatment time of 10 days—significantly longer than the timeframe in the current study [36].

Zeolite's significant effectiveness in removing nitrogen species such as nitrate and nitrite is attributed to its ion exchange capacity [37]. As illustrated in Figure 4, the inclusion of zeolite in the TF process enabled the removal of over 98.00% of TN and nitrite, surpassing previously reported removal rates of approximately 36.00% in the literature [5].

After coagulation and DF, the concentrations of both parameters increased in the effluent. Conductivity decreased from 0.91 mS/cm in the raw synthetic effluent to 0.85 mS/cm after TF. Conversely, chloride levels increased following TF, reaching a final concentration of 29.02 mg/L. These values are typical in dye treatment systems, with the primary explanation for these changes attributed to the amount of $FeCl_3$ used during the coagulation process [8,38].

It is important to highlight that, given the levels of chloride and conductivity removal achieved, the effluent treated through the DF and TF processes complies with Brazilian regulations for discharge and reuse [39]. According to the United States Environmental Protection Agency, water with conductivity ranging from 30.00 to 540.00 mS/cm and chloride concentrations between 106.50 and 355.00 mg/L can be considered suitable for restricted agricultural reuse, unrestricted urban use, and applications in cooling towers and boiler feed systems [40].

The treatment process consisting of coagulation followed by DF and TF demonstrated high efficiency in removing turbidity, AC and TC. The main mechanisms involved are charge destabilization and particle formation during coagulation, followed by adsorption and ion exchange during filtration. Turbidity removal reached 99.58% with DF and 100% with TF, decreasing from 210.67 NTU in the raw synthetic effluent to <0.01 NTU after TF. AC removal reached 98.62% by the end of the treatment, with a final value of 9.00 uC after TF, while TC removal was 100%, with values below the detection limit (<0.01 uC) after TF.

Turbidity removal is essential for maintaining effluent quality, as it is associated with the elimination of suspended and dissolved solids through interactions between the filter media and particulate matter [23]. Filtration mechanisms that contribute to turbidity removal include diffusion, Van der Waals forces, electrostatic interactions, adsorption, and chelation [41].

When comparing turbidity removal from textile industry effluents containing dyes, studies using pressurized sand and activated carbon filtration systems reported removal efficiencies above 95% for both media [5]. The results of the present study confirm that both DF and TF systems are effective in removing turbidity.

Moreover, the turbidity removal achieved here is comparable to that of other systems, such as biofiltration. Research on laundry effluents, which typically contain high concentrations of dyes, reported a turbidity removal rate of 99.22% [42], further confirming the effectiveness of filtration in this context.

The color in synthetic effluents originates from dyes, pigments, paint residues, chemicals, and other organic compounds [42]. Dyes alone contribute to approximately 50.00% of the total color in textile industry effluents [2]. The results of this study show high removal rates of both AC and TC (>96.00%) through DF, aligning with findings from sand filtration studies, in which $FeCl_3$ also played a role in color removal [21].

Other studies have shown that activated carbon filtration contributes to color removal in textile effluents, achieving a 65.00% reduction in AC from a raw synthetic effluent with an initial color of 180.00 uC. This efficiency was attributed to the porosity of the carbon, although the mechanisms behind the removal of large molecules, such as aromatic groups, remain nuclear [43].

Activated carbon derived from coconut shells has also demonstrated color removal efficiency, with a 74.00% reduction by the end of treatment. In this case, the observed efficiency was attributed to a reduction in dye solubility and polarity during filtration [37].

In the TF system, clinoptilolite contributed to color removal through the electrostatic adsorption and ion exchange, particularly by interacting with aromatic compounds and anionic dyes due to its anionic character, thereby ensuring effective removal [44, 45].

The efficiency achieved in this study is comparable to other treatment systems, such as horizontal and vertical constructed wetlands. Studies have shown that horizontal wetlands can remove up to 90.00% of color from synthetic dye-containing effluents, whereas vertical wetlands are less effective, with removal rates around 10.00% [46].

The dye Ponceau exhibited a removal efficiency of 56.00% in DF, reaching 100.00% in TF. For the dye Tartrazine, the removal was 44.15% in DF, also increasing to 100.00% in TF. This high efficiency is attributed to the multiple interactions between the filter media and the dyes, including retention, adsorption, physical attraction, and ion exchange processes

[5].

In DF, the sand's high surface area, combined with its favorable adsorption and retention potential, stability, and kinetic equilibrium properties, contributed to dye removal [1].

However, the greater number of active sites and higher porosity of the activated carbon and clinoptilolite used in TF significantly enhanced the removal of both dyes. It is assumed that the main removal mechanisms in this filtration stage include adsorption onto activated carbon and electrostatic adsorption interactions with clinoptilolite.

According to the literature, activated carbon effectively removes both cationic and anionic dyes via adsorption [47,48], while clinoptilolite generates electrostatic attraction or repulsion forces depending on the dye's charge. When modified, clinoptilolite may interact differently with various dye types [44]. Since the azo dyes used in this study are anionic and the clinoptilolite applied is cationic, attractive forces are established between the filter medium and the dyes, promoting efficient removal [45,48].

These hypotheses are supported by the structural analysis of the dye molecules, which reveals that the presence of axial negative charges in aqueous solution promotes electrostatic interactions with clinoptilolite. Additionally, the presence of bulky atoms such as sulfur and nitrogen, along with the dyes' cyclic structures, results in high molecular density, enhancing their adsorption. This may explain the removal rates exceeding 40.00% observed in DF. However, due to the greater porosity of the TF filter media, the adsorption of these compounds is further facilitated, leading to even higher removal efficiencies.

The kinetics of dye removal in DF are rapid due to the vertical flow, which limits the contact time between the effluent and the filter media to approximately 3 to 5 minutes. Despite this short contact time, significant dye removal rates ranging from 40.00% to 50.00% were achieved. This performance is attributed to the adsorption potential of sand, which possesses a relatively high surface area. The adsorption kinetics in DF are influenced by the physical and chemical characteristics of the filter media, including sand particle size, porosity, and overall adsorption capacity. Additionally, the process is driven by a relatively fast sorption equilibrium, in which physical and electrostatic interactions between dye ions and the sand surface play a key role.

In TF, adsorption kinetics are further enhanced by the use of activated carbon and clinoptilolite—materials with higher porosity and a greater number of active sites [47]. These materials enable layered adsorption and more complex retention mechanisms, facilitating complete dye removal within similarly short contact times. The role of activated carbon is particularly significant due to its microporous structure, which increases adsorption capacity by trapping smaller dye molecules and offering specific binding sites [49]. The high efficiency observed in TF is therefore directly related to the rapid kinetics afforded by these highly porous filter materials, which optimize dye retention even under limited contact time.

When comparing the dye removal efficiency of the coagulation, DF, and TF systems with those reported in other studies, it is observed that sand filtration systems typically achieve dye removal rates between 10.00% and 20.00% when treating textile industry effluents. In one such study, the effluent remained in contact with the filter media for approximately 30 minutes [1]. In contrast, the DF system used in this study operates with a contact time of only 3 to 5 minutes due to its downward flow configuration, yet removal efficiencies exceeding 40.00% were achieved. This enhanced performance can be attributed to the pre-treatment by coagulation. Indeed, other studies have demonstrated that the use of coagulants such as $FeCl_3$ and $Al_2(SO_4)_3$ in the treatment of industrial effluents can result in dye removal efficiencies of 85.00% and 75.00%, respectively [17]. Additional sources corroborate that the majority of dye removal often occurs during the coagulation stage [5].

In another study, three types of activated carbon—with an average pore volume of 0.57 cm^3/g—were used for dye removal from aqueous solutions [43]. After contact times ranging from 5 to 240 minutes, dye removal efficiencies between 70.00% and 99.00% were obtained. The authors concluded that increasing the carbon's porosity had a more pronounced effect on dye removal than extending contact time. In the TF system of the present study, the activated carbon had a pore volume of only 0.06 cm^3/g and a contact time of 3 to 5 minutes, yet achieved complete (100.00%) removal of both dyes. This result supports the hypothesis that earlier treatment stages contributed to dye removal and highlights the high efficiency of the activated carbon used in TF.

Other studies have reported dye removal efficiencies of approximately 16.30% for anionic dyes using clinoptilolite alone, with a pore volume of 0.42 cm^3/g and a contact time of 360 minutes [50]. The authors attributed this performance to the material reaching its adsorption capacity. In contrast, the clinoptilolite used in the TF system had a smaller pore volume of 0.04 cm^3/g and a much shorter contact time (3 to 5 minutes), yet still contributed to 100.00% dye removal. This suggests that while clinoptilolite plays a supportive role, the activated carbon is the primary contributor to the high removal efficiency observed in TF. These findings also emphasize the importance of the combined coagulation and DF processes in enhancing dye removal from synthetic effluents.

The performance observed here is comparable to that of more complex treatment technologies, such as electrochemical and UV-based processes, which have been shown to remove over 90.00% of azo dyes from industrial effluents [32]. However, compared to those systems, the filtration media used in this study—gravel, sand, activated carbon, and zeolite—offer advantages in terms of lower operational complexity, reduced maintenance requirements, and lower costs.

Based on these considerations, the integrated coagulation, DF, and TF systems proved to be effective for the removal of azo dyes from synthetic effluents with a concentration of 6.50 mg/L. Additionally, they contributed to the overall improvement of final effluent quality, as demonstrated by the removal rates achieved across the analyzed parameters.

4. Conclusions

The results demonstrate that double filtration (DF) and triple filtration (TF) effectively treat azo dye-containing effluents when using $FeCl_3$ as a coagulant at an optimal dosage of 116.00 mg/L. Increased dye concentrations (30.00 and 50.00 mg/L) led to faster saturation of the filtration media, highlighting the importance of incorporating pre-treatment steps to improve sustainability and cost-efficiency. Both DF and TF processes achieved significant removal efficiencies, with approximately 60.00% color reduction and 82.33% turbidity removal, thereby enhancing overall filtration performance. Specifically, DF removed over 40.00% of nitrate and more than 75.00% of chemical oxygen demand (COD) and total nitrogen (NT), whereas TF surpassed 75.00% nitrate removal and achieved greater than 95.00% removal for COD, NT, and azo dyes. However, reductions in conductivity and chloride concentrations were limited. The treated effluents met EPA standards for safe disposal and restricted agricultural reuse, indicating that DF and TF represent cost-effective, low-maintenance treatment options with considerable industrial applicability. This study is the first to evaluate the combined application of DF and TF for azo dye-laden wastewater treatment, demonstrating promising efficacy in removing both physicochemical contaminants and dye molecules. Future research should focus on optimizing pre-treatment strategies and investigating the performance of these filtration processes across different dye types and wastewater sources.

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Author Contributions

Matos, D. G. S.: Selection of articles, organization, and discussion of the writing. Campos, S. X.: Review and organization of the writing.

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