

# Advanced Oxidation of Segregated Streams for Effective Color Removal from Denim Processing Effluents

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**Abstract:** The study introduced footprint analysis as a new methodology and focused on differentiating wastewater streams with the highest color content and optimizing the advanced oxidation process for the segregated streams for effective color removal from denim processing. Experiments were implemented to four segregated streams rather than the entire plant effluent. A flow proportional composite mixture of segregated streams was used for color removal experiments using the advanced oxidation process with ozone and hydrogen peroxide and Fenton oxidation as other alternatives. The latter yielded the best results achieving total removal of color below visual detection limit after an optimum reaction time of 10 minutes. The Fenton oxidation process was also applied to a representative sample from the plant effluent after the physical-chemical treatment sequence, where color absorbance levels were lowered at all wavelengths below  $1.0 \text{ m}^{-1}$ . The merit of the new footprint approach was confirmed by the results, which provided a conclusive indication that color treatment at *source*, implemented on selected segregated wastewater streams, presented concrete advantages over the *end of pipe treatment* of the overall effluent.

**Keywords:** color removal; denim processing; stream segregation; advanced oxidation.

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## 1. Introduction

The traditional way of wastewater management for industrial effluents prescribes an end-of-pipe approach, which would treat the overall plant effluent. While this approach may be acceptable for conventional parameters such as organic carbon (COD), suspended solids, it should be avoided to the extent possible, especially in textile plants, because it does not permit to identify and remove specific pollutants such as color at the source, without being diluted in the overall waste stream. This study specifically focused on color removal in plants manufacturing denim materials, a key branch in the textile industry, possibly with the highest market share.

Denim processing relies on a variable sequence of various steps, each utilizing different processes and chemicals; dyeing and release of dyes into the waste stream occurs only in a few of these steps. Consequently, this study defined and implemented an innovative methodology of an experimental in-plant assessment, which uncovered the color footprint, i.e., the color fingerprint of all individual wastewater streams. Recently, pollution footprints were suggested

as an effective approach to identify the source and the relative magnitude of specific pollutants of interest among different steps of industrial activities [1]. Indexes were suggested for a more effective footprint assessment [2, 3]. Recently, this approach was successfully implemented to manage the wastewater of a textile dye house handling different types of fabrics [4, 5] and for sludge generated in a municipal wastewater treatment plant for the optimization of net useful energy [6].

Color is one of the most easily recognized properties of wastewaters because it directly addresses visual perception. Therefore, it immediately raises aesthetic concerns, and it is usually interpreted as a sign of serious pollution. Sewage has an inherent brown color usually associated with the mixture of organics it contains [7]. However, the major color problem is mostly related to dyeing and washing operations in industrial activities, especially in textile operations, where a fraction of the dye is not exhausted on the fiber of fabric and released into the effluent and causes deep color [8]. Since the presence of a small fraction of dye remains visible, even after a high dilution ratio, discharge of these effluents decreases the beneficial aesthetic use of streams and other water bodies [9, 10]. Aside from aesthetic effects, most dyes with intricate aromatic structures resist biodegradation during treatment and in the environment; if not properly removed, dyes may significantly reduce photosynthetic activity in aquatic environments as they impair light penetration [11].

Generally, the textile generates high volumes of wastewater in the range of 20-380 m<sup>3</sup>/ton of product, depending on the type of fiber/fabric, applied processes, and technologies [12-14]. Textile activities also consume a large variety of dyes and processing chemicals. It is generally estimated that a significant fraction of chemicals are wasted, and around 10 to 30% of the dye is lost to the effluent during the dyeing process depending on the fixation ratios fixation in the production processes [15, 16]. These facts have triggered efforts to minimize the use of water and chemicals and encouraged reuse options within the production steps [17, 18].

Over the years, extensive research effort was devoted to test and develop effective technology for color removal, which may be briefly reviewed in four different categories: (i) physical and/or physicochemical methods such as adsorption, chemical settling, ion exchange, etc. [2, 19-25]. This group defines the traditional but continuing studies on color removal practice, mainly focusing on coagulation/flocculation. The general consensus is that a particular coagulant is only suitable for a certain type of dye. The study of [1] is interesting in the sense that it tested *polygorskite clay* as a strong adsorbent to remove indigo dye from synthetic wastewater; the adsorbed dye was used to synthesize the *Maya Blue pigment*, a valuable product. (ii) Advanced oxidation processes (AOP)s, such as ozonation, H<sub>2</sub>O<sub>2</sub>/UVC photochemical oxidation with hydrogen peroxide), Fenton, photo-Fenton, anodic oxidation, and electro-Fenton [26-34]. In a study, Fenton oxidation was tried before the activated sludge process, and chromium-induced advanced oxidation was recommended as a novel process, which does not require additional metal (Fe<sup>2+</sup>) supply [35]. (iii) Biological processes based on pure and mixed cultures operated in aerobic and/or anaerobic conditions [36-39]. Biological processes were even tested as a pre-treatment in a combined process configuration involving coagulation/flocculation and chemical settling; electrochemical oxidation steps for effective color removal [40, 41]. (iv) Membrane processes [42-44]. This application involved ultrafiltration (UF) and reverse osmosis (RO) after biological treatment, essentially for reuse purposes. The outcome of the extensive information available for color removal while testing all possible processes, is mostly case-specific and does not offer a generally applicable guideline for this purpose.

In denim production, dyeing and washing processes are the main sources of wastewater generation. Wastewater includes high levels of indigo blue ( $C_{16}H_{10}N_2O_2$ ), a dye belonging to the vat group, and alkaline chemicals. Indigo blue is a synthetic organic dye, and it is considered recalcitrant due to its complex chemical structure [14, 21, 45]. Approximately 30% of the applied indigo blue dye remains unfixed and ends in the wastewater [21]. Wastewater containing indigo blue dye also includes a very high level of suspended solids due to pumice stone used in the bleaching process, a high amount of chemical oxygen demand (COD), dissolved solids, and a distinct dark blue color [46-48]. Earlier works mostly investigated the biodegradation characteristics and applicable treatment technologies of denim effluents [37, 49-51]. A few recent studies also evaluated color removal, using both physicochemical and advanced oxidation technologies, but they focused on effective color removal from the entire plant effluent, prescribing an end-of-pipe approach [52, 53].

Thus, the study aimed to identify wastewater streams with the highest color content and optimize the advanced oxidation process for the segregated streams for effective color removal from denim processing. For this purpose, an innovative approach for the assessment of the *color footprint* of the plant was adopted, which involved a full inventory of wastewater and polluting sources in the plant. Advanced oxidation experiments using ozonation and Fenton oxidation were carried out on segregated wastewater streams differentiated based on the color footprint. Results indicated effective color removal and offered a new perspective for evaluating wastewater reuse potential.

## 2. Materials and Methods

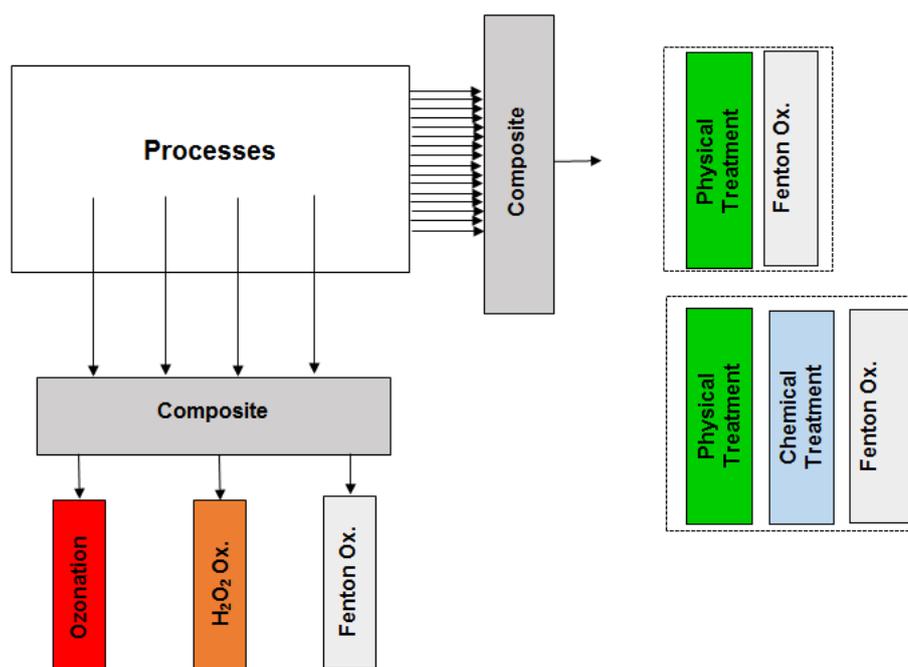
### 2.1. Research rationale.

This study was carried out within a large project organized for the optimal management of wastewater streams generated in a denim plant located in *Çorlu, Turkey*. The first part of the project was focused on a detailed assessment of pollution footprint, covering all operational steps with a major contribution to the production scheme of the plant.

This database indicated all individual wastewater streams with selected pollutants and color and identified the sources with the highest impact on the color level in the wastewater. Representative wastewater samples were taken from the four segregated sources with the highest color input and analyzed for major parameters. The same analyses were also conducted on their flow-proportional composite mixture.

Then, a complete evaluation of the plant effluent was carried out in terms of its color content. For this purpose, daily composite samples were collected for 30 days, during the period of 19.02.2018 – 05.04.2018, and analyzed for all color components. For each color component, all samples' results were evaluated in terms of a statistical distribution yielding percentile values of corresponding measurements. Results were compared with those associated with four segregated sources with the highest color input.

Samples characterizing the four selected segregated sources and a representative sample from the plant effluent were subjected to laboratory-scale evaluation for color removal. Oxidation with ozone and hydrogen peroxide ( $H_2O_2$ ) and Fenton oxidation were tested on the flow-proportional composite mixture of segregated streams. Based on best removals achieved, Fenton oxidation alone was tested using the plain settling effluent and the effluent physical-chemical treatment sequence applied to composite plant effluent. A schematic description of the adopted experimental program is displayed in Figure 1.



**Figure 1.** Schematic display of the experimental program.

## 2.2. Experimental setup.

Wastewater samples were oxidized by ozone in a borosilicate glass column with a volume of 1500 mL. The effective volume of the reactor was 1000 mL. For the oxidation process, the  $O_3$  + air mixture was supplied to the reactor's bottom at a flow rate of 1 L/min through a coarse sintered gas dispersion disc. An Arcbull Meo-20 model ozone generator, with a capacity between 5-25 mg/L, was used to obtain ozone from oxygen. Oxygen was separated from air with a Health time OC-5 oxygen concentrator. The ozone generator was connected to the reactor using teflon tubing. A typical run was initially started by filling with wastewater; then it proceeded with continuous ozonation in a semi-batch mode concerning ozone gas feeding. Samples were taken at regular time intervals from the reactor and analyzed for color, COD, and pH. Ozone concentrations were measured via iodometry [54], and the utilized ozone doses after ozonation experiments were determined using the difference between the inlet and the off-gas ozone concentrations. Ozone treatment was carried out by passing ozone gas through a 750 mL sample volume at 5, 10, and 15-minute exposure times. The ozone dosage was adjusted to 45 mg/min in the experiments. Sample volume was set as 250 mL, and the initial  $H_2O_2$  concentration was calculated as 7140 mg/L. At the original pH of 5.89 of the sample, the effect of  $H_2O_2$  oxidation was observed after 5, 10, 15, and 30 minutes.

Fenton oxidation experiments were conducted in the range of 5-30 minutes in parallel 250 mL glass beakers that were continuously stirred. The stoichiometric oxygen equivalent of the wastewater, computed based on the initial COD concentration ( $H_2O_2 = 2.12 \times COD$ ), was used to determine the initial  $H_2O_2$  concentration [55].  $Fe^{2+}$  catalyst was fed to the solution while the parallel reactors (beakers) were continuously stirred at 100 rpm from a  $FeSO_4 \cdot 7H_2O$  stock solution (10% w/v, Merck).

In the segregated stream experiments, the initial dosage of 2352 mg/L of  $Fe^{2+}$  dropped the original pH of the sample from 5.89 to 5.20, which was further dropped to 2.81 after the addition of 7140 mg/L of  $H_2O_2$  (35% w/w) to approach the optimum operational pH of around 3.0. The molar ratio of  $H_2O_2:Fe(II)$  was adjusted to 5:1.

A similar procedure was adopted when evaluating plain settled and chemically treated effluents in the plant effluent surveys: The experiment was started at an initial pH of 6.74 for plain settling effluent; the sample's pH was adjusted to 3.0 by adding 423 mg/L of Fe<sup>2+</sup> and 1284 mg/L of H<sub>2</sub>O<sub>2</sub>. In the experiments conducted on chemically treated effluent with an initial pH of 4.1, the pH was reduced to 3.0 with the addition of 370 mg/L of Fe<sup>2+</sup> and 1122 mg/L of H<sub>2</sub>O<sub>2</sub>. The experiments were terminated by increasing the samples' pH to the range of 7.0-7.5 with 6N NaOH to ensure the formation of iron hydroxide flocs, which were removed by filtration using 0.45 μm membrane filters.

All experiments were carried out duplicated at room temperature (20 ± 1°C); average values were taken when presenting the results.

### 2.3. Analytical measurements.

Color measurements were carried out using a spectrophotometer (Hach Lange DR 5000), both as absorbance at different wavelength 436, 525, 620 nm, and Pt-Co units [56]. The COD analyses were conducted by open reflux-titration according to the procedure defined in ISO 6060 method [57]. All other analyses were conducted following the procedures defined in Standard Methods [56]. pH values were measured with a digital pH meter (Thermo Scientific Orion Star A211, Waltham, MA). Analyses involved at least triplicate measurements: additional measurements were performed when necessary until the observed variance decreased below 5%.

### 2.4. Statistical analyses.

The probability distribution of different color levels measured in 30 plant effluent samples was established using a simple approach, which also proved successful in other similar studies [58, 59]. Essentially, the method relied on arranging measured color, C, from the smallest to the highest and estimated pC<sub>i</sub> as:

$$pC_i = i/n + i$$

Where, pC<sub>i</sub> is the probability of observing the C value equal or smaller than C<sub>i</sub>; C<sub>i</sub>, the color level in the i<sup>th</sup> rank; and n, the total number of observations. Then, pC<sub>i</sub> values were plotted using a probability scale to yield the 50 and 70 percentile levels for different color measurements.

## 3. Results and Discussion

### 3.1. Pollution profile.

The first part of the experimental work started with a detailed survey of the pollution profile for the plant; it was conducted on 16 different processes representing the core of the production scheme in the plant. Different processes included 3 to 11 successive steps, and the analytical survey covered each and every existing step taking part in the processes. The results give a clear indication that the utilization frequency of each process greatly varies depending on the scheduled activities of the plant. Consequently, the wastewater flow fluctuates between 3600 m<sup>3</sup>/year and 196000 m<sup>3</sup>/year, and the COD load from 3400 kg/year to 265000 kg/year between different processes. The survey enabled us to compute average values of

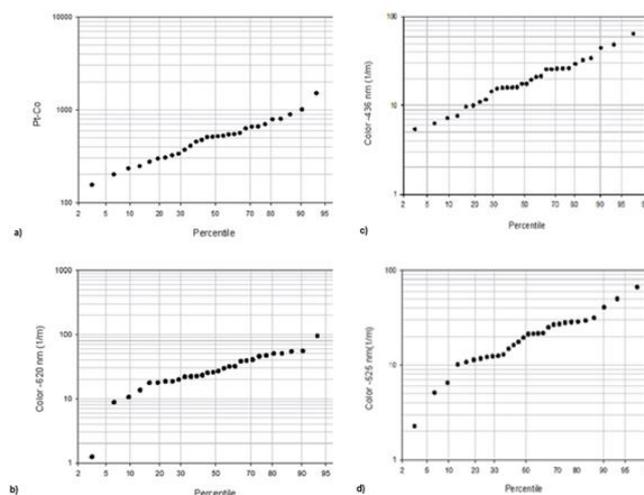
approximately 2000 m<sup>3</sup>/day and 1200 mg/L for the daily flow rate and the overall plant effluent's COD content.

Color contents of different steps in all processes were also assessed. Obviously, color measured in terms of Pt-Co unit and/or absorbance at different wavelengths is not amenable to mass balance. Instead, measurements indicated four segregated wastewater streams with much higher color input compared to others. Representative waste wastewater samples were taken from the four sources with the highest color input for further evaluation in lab-scale experiments. Table 1 summarizes major characteristics of these four segregated wastewater streams and their flow-proportional mixture; it clearly shows that the selected wastewater streams and the composite mixture indeed represent highly colored and strong effluents with an average COD content of 3380 mg/L, approximately three times higher than the average value of 1200 mg/L in the plant effluent.

**Table 1.** Major characteristics of the four selected wastewater steam and their mixture.

Process	pH	Total COD (mg/L)	Soluble COD (mg/L)	Color			
				Pt-Co	436 nm (m-1)	525 nm (m-1)	620 nm (m-1)
Heavy stone washing	-	6150	3330	3240	105.6	114	190
Dark stone washing	-	4670	2770	2530	177.2	163.6	226.4
Rinse washing / pre-washing	-	2270	1350	4870	123.6	133.2	133.6
Hot stone washing	-	1580	1100	1310	42.4	55.6	100.4
Composite	5.89	3380	2440	2340	64	73.2	115.2

Furthermore, the color content of the plant effluent was also assessed in 30 different daily composite samples representing an observation period of 45 days. The plant effluent always involved a high color level, which was determined using both Pt-Co units and absorbance at 436, 525, and 620 nm. Figure 2 reflects the statistical distribution of color measurements. Similar statistical evaluations were also carried out for other major parameters. As displayed in the figure, the distribution of all color measurements exhibited significant daily variations depending on the plant's daily production activities. Corresponding 50 percentile values (C<sub>50</sub>) for Pt-Co, 436 nm, 525 nm and 620 nm were determined as 517, 17.46 m<sup>-1</sup>, 21.14 m<sup>-1</sup> and 26.64 m<sup>-1</sup>, respectively. These values were identified as 654, 25.85 m<sup>-1</sup>, 27.09 m<sup>-1</sup> and 40.45 m<sup>-1</sup> for 70 percentile values (C<sub>70</sub>). Comparison of the statistical color levels in the plant effluent with those given in Table 1 reflect exceedingly high color levels of the selected four wastewater streams both in terms of Pt-Co unit and absorbance as three wavelengths.



**Figure 2.** Statistical distribution of color measurements as (a) Pt-Co units (b) absorbance at 620nm (c) absorbance at 435 nm (d) absorbance at 525 nm.

### 3.2. Color removal experiments.

#### 3.2.1. Segregated wastewater streams.

The first part of the experiments on color removal was started with the flow-proportional composite mixture of the segregated wastewater streams. Relevant characteristics of the composite mixture were outlined in Table 1. Experiments on color removal involved ozone treatment, H<sub>2</sub>O<sub>2</sub> oxidation, and Fenton oxidation.

In the first step, ozone treatment was carried out at 5, 10, and 15-minute exposure times. After 675 mg of ozone dosage in 15 minutes, the off-gas ozone was measured as 482 mg, corresponding to 193 mg of ozone utilized in the experiment. As indicated by the results summarized in Table 2, ozone treatment could only yield partial color removal; even after 15 min exposure time, color reduction remained at 46% in Pt-Co units and 36% as absorbance at 436 nm. However, reductions in absorbance at 525 nm and 620 nm reached 86% and 94%, respectively. It is noteworthy to mention that the intense blue color of the sample faded away, and it was substituted by a light brown-yellow color (Figure S1).

**Table 2.** Color removal performance of ozone oxidation on the mixture of segregated wastewater streams.

Time (min)	Total COD (mg/L)	Soluble COD (mg/L)	Color			
			Pt-Co	436 nm (m <sup>-1</sup> )	525 nm (m <sup>-1</sup> )	620 nm (m <sup>-1</sup> )
0	3380	2440	2340	64	73.2	115.2
5	3240	2430	1870	58.8	23.2	20.0
10	3170	2590	1520	49.2	13.2	8.4
15	3180	2600	1260	41.2	10.0	7.2

The next step involved H<sub>2</sub>O<sub>2</sub> oxidation started with an initial concentration of 7170 mg/L calculated as the theoretical equivalent of 2.12 COD. The experiments indicated that H<sub>2</sub>O<sub>2</sub> remained ineffective during all the reaction periods (5, 10, 15, and 30 minutes). As shown in Table 3, the achieved color removal was only 5% as Pt-Co units and stayed in the range of 6-19% as absorbance at different wavelengths.

The Fenton process is oxidation by H<sub>2</sub>O<sub>2</sub> and Fe<sup>2+</sup>. The reaction times for Fenton oxidation were again selected in the range of 5 to 30 minutes. As displayed in Table 4, the start of the experiment reflected the very high color levels in the mixture of segregated wastewater streams, namely 2340 Pt-Co units together with 64 m<sup>-1</sup>, 73.2 m<sup>-1</sup>, and 115.2 m<sup>-1</sup> absorbance levels at 436 nm, 525 nm, and 620 nm, respectively; a 5 minutes contact time did not prove sufficient, as the color removal remained limited to the range of 69-89%. However, 10 minutes of contact time induced very effective color reduction down to 33 Pt-Co and below 1.0 m<sup>-1</sup> for absorbance at 436 nm and 620 nm. The optimum contact time for absorbance at 525 nm was 15 minutes, also securing color reduction down to 0.68 m<sup>-1</sup>. As also displayed in Figure S2, the color disappeared completely below the visual detection limit after an optimum reaction time of 10 minutes.

#### 3.2.2. Plant effluent.

The merit of an integrated physical-chemical scheme as an appropriate pre-treatment for denim processing wastewaters was investigated in the earlier part of the study; it indicated that the selected pre-treatment, first involving plain settling followed by chemical treatment, was quite effective in providing adjusting the major parameters in wastewater to a level suitable for biological treatment [52]. In this context, color removal from the overall plant effluent was also tested to offer a benchmarking against a selection of segregated wastewater streams. Fenton

oxidation, which proved to be the most effective advanced oxidation method, in this case, was applied to the effluent sample after (i) plain settling and (ii) chemical treatment. Performance of physical-chemical treatment, i.e., effluent characteristics after plain settling and chemical treatment, are summarized in Table 4, which shows quite a limited color reduction, despite significant COD removals achieved through these pre-treatment steps. Therefore, the biological treatment units will receive an influent with a strong color level, which would be likely to by-pass this treatment step and persist in the effluent without an additional color removal mechanism.

**Table 3.** Color removal performance of H<sub>2</sub>O<sub>2</sub> and Fenton oxidation on the mixture of segregated wastewater streams.

Time (min)	Total COD (mg/L)	Soluble COD (mg/L)	H <sub>2</sub> O <sub>2</sub> Oxidation				Fenton Oxidation				
			Color				Soluble COD (mg/L)	Color			
			Pt-Co	436 nm (m <sup>-1</sup> )	525 nm (m <sup>-1</sup> )	620 nm (m <sup>-1</sup> )		Pt-Co	436 nm (m <sup>-1</sup> )	525 nm (m <sup>-1</sup> )	620 nm (m <sup>-1</sup> )
0	3380	2440	2340	64	73.2	115.2	2440	2340	64	73.2	115.2
5	-	2470	2030	54.6	56.8	82.6	825	675	19.96	14.28	13.6
10	-	2515	2250	60.6	63.8	93.8	660	33	0.76	4.56	0.1
15	-	2315	2300	61.8	65.4	96.4	450	55	1.8	0.68	0.21
30	-	2250	2230	60	63.6	93.4	470	73	2.28	1.08	0.56

**Table 4.** Performance of plain settling and chemical treatment on the plant effluent.

Characteristics of the wastewater	Total COD (mg/L)	Soluble COD (mg/L)	Color			
			Pt-Co	436 nm (m <sup>-1</sup> )	525 nm (m <sup>-1</sup> )	620 nm (m <sup>-1</sup> )
Raw wastewater	1250	680	970	33.2	37.6	56.4
Plain settled	790	630	870	25.6	31.2	55.2
Chemical treated	485	370	316	8.16	10.4	18.24

Fenton oxidation's performance on the effluent of plain sedimentation is quite impressive, as given in Table 5: Soluble COD drops down to 180 mg/L and 130 mg/L after reactions times of 5 min. and 30 min., respectively. Color is reduced to 58 (93% removal) and 37 (96% removal) Pt-Co units at the end of the same reaction periods. A 30 min reactor phase lowers absorbance level at all wavelengths below 1.0 m<sup>-1</sup>. The color was lowered below the visual detection limit after a reaction time of 5 min.

**Table 5.** Color removal from plant effluent after plain settling with Fenton oxidation.

Time (min)	pH	Total COD (mg/L)	Soluble COD (mg/L)	Color			
				Pt-Co	436 nm (m <sup>-1</sup> )	525 nm (m <sup>-1</sup> )	620 nm (m <sup>-1</sup> )
0	6.74	790	630	870	25.6	31.2	55.2
5	3.0	-	180	56	1.64	2.88	1.40
10	3.0	-	195	34	1.12	1.84	0.68
15	3.0	-	185	48	1.36	1.04	0.88
30	3.0	-	130	37	1.04	0.80	0.64

In the next phase, the effluent of chemical treatment carried out with 1.0 mL/L of PE and 1.0 mL/L of liquid PAC, was used for Fenton oxidation to achieve color removal. Table 4 summarized related characteristics of the chemical treatment effluent. At the start of the experiment, the initial color was only 12-15% of the levels ascertained in the mixture of segregated streams due to substantial dilution within the overall plant effluent. Fenton oxidation's performance was essentially very similar, reducing remaining color levels below 1.0 m<sup>-1</sup> absorbance, i.e., below the visual detection limit, after a 10 minutes contact time. A reaction time of 30 min reduced the soluble COD down to 110 mg/L, a level hard to obtain by

biological treatment due to soluble residual COD and metabolic products [60]. Additional information about this test was reported elsewhere [53].

First of all, this study advocated a new methodology for effective color control in denim processing effluents. Extensive research efforts testing different advanced oxidation techniques on a large spectrum of dyes have so far failed to provide a sustainable answer to solving the color problem in textile wastewaters. The reason because this failure is quite simple: (i) It generally involved an end-of-pipe approach on the overall plant, which dilutes the colored streams; (ii) the advanced oxidation techniques as a pre-treatment the biological stage before remained partly effective due to the interference of organic matter; (iii) biological treatment suffered from adverse effects of dyestuff on the microbial culture; (vi) advanced oxidation was mostly applied as a post-treatment on the biological treatment effluent.

In this context, the study advocated a novel approach of applying effective color removal on segregated waste streams carrying the major fraction of color as a pre-treatment before biological removal of organic matter. It essentially involved segregation of wastewater streams generated by dyeing operations in the plant and treating these streams as a source, before being mixed with and diluted in the overall effluent. This methodology is sustainable and equally applicable to all industrial applications and all selected pollutants. Denim processing and finishing involve a complex array of steps and operations, each generating wastewaters with different characteristics. Therefore, *end of pipe* inspection of wastewater quality may prove meaningless and even misleading for controlling key pollutants such as color.

Consequently, the study adopted a much more sustainable in-plant analysis approach, which created a database covering all production steps and identifying all processes using dyes. Essentially this analysis yielded water and pollutant footprinting for the plant. The footprint approach that was used in a previous study essentially identified the relative magnitude of parameters such as water use and organic load (COD) in each production step in the plant [4]. A numerical/quantitative footprint cannot be established for the color, which is measured in terms of indexes such as absorbance, but these measurements enabled to identification and segregate of the four major wastewater streams to be treated at the source as indicated in Table 1.

The data on chemical use for color removal from the selected segregated streams allows a comparison with the case where the same processes would be applied to the overall plant effluent with an end-or pipe approach. They indicate a chemical reduction of 20-22% for Fenton oxidation in favor of segregated stream treatment. Obviously, an economic feasibility study is well beyond the study's scope because it highly depends on the specific site's characteristics. It involves several factors such as dyes used in the manufacturing processes, type of treatment units, energy and personnel costs, etc. Nevertheless, the comparison of chemical use may be accepted as a reliable yardstick for the merit of stream segregation for color removal because the end of pipe treatment would necessitate a much larger footprint for the same treatment steps, with similarly higher costs for auxiliary factors.

Furthermore, the database derived from the in-plant analysis detected all dyeing operations involving different dyestuff, such as direct, sulfur, acid, reactive dyes aside from the common indigo dye, so that the experimental color removal options tested real-case scenarios. This approach should be contrasted from most studies in the field, which were conducted either on a specific dye, using synthetic wastewater, or used effluents without specifying the textile

facility's characteristics that generated it and applied several processes for color removal on a trial and error basis.

#### 4. Conclusions

The study's main message was to advocate and experimentally test a new approach of looking inside the plant, i.e., the footprint approach allowing a thorough analysis of selected key pollutants for defining and optimizing the sustainable wastewater management strategy.

The significance and the merit of this new approach were underlined by the results of the study. In fact, they offered conclusive experimental evidence that Fenton oxidation was the most suitable advanced oxidation process for achieving complete color removal at source from selected segregated waste streams with the highest color content in denim processing effluents.

Color treatment at source presented distinct technical and economic advantages over the end of pipe treatment of the overall effluent: Complete color removal from segregated waste streams by Fenton oxidation could be achieved with approximately 20% of chemicals that would be spent when the entire flow of plain settling effluent was treated for the same purpose. This ratio was slightly increased to 22% compared to the chemicals when the Fenton oxidation was applied to the entire chemical settling effluent flow. Furthermore, the separate treatment of segregated flows also benefits from advanced treatment of dyeing effluents for onsite reuse by suitable technologies.

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#### Conflicts of Interest

The authors declare no conflict of interest.

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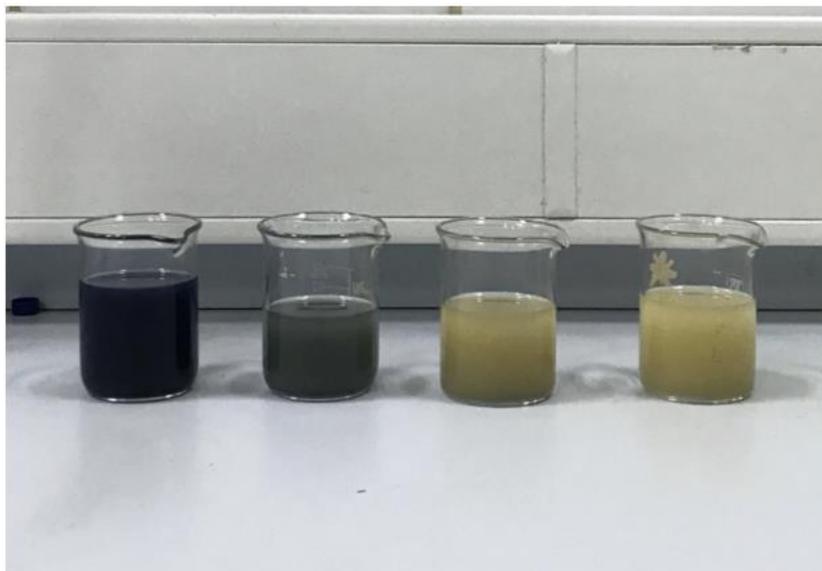
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## Supplementary materials



**Figure S1.** Visual display of remaining color in the mixture of segregated sample after ozone treatment.



**Figure S2.** Visual display of color disappearance with Fenton oxidation at 10 minutes.