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# A biosorption coupled ozonation treatment system towards decolorization/removal of a textile dye (reactive blue 19)

**Iqra Shahzadi, Gulfam Abid, Anam Khalid*, Azhar Ali, Sana Islam**

*College of Earth & Environmental Sciences, University of Punjab, Lahore, Pakistan*

## Abstract

The present study aims to assess the efficiency of a biosorption coupled with ozonation treatment system for the removal of RB-19 dye from textile effluent. The removal efficiency of biosorption was observed to be 73.2 % for 100 ppm in 150 min at 1 g; whereas ozonation increased the decolorization of high concentration textile dyes up to 91.6% for 100 ppm in 35 min at 70 g/m$^3$ dose. Nevertheless, the process is time-consuming along with the formation of carcinogenic by-products. The pollution indicators showed a high pollution load in treated water, which reflects that the biosorption and ozonation could be a good choice for the color removal from dye solution with removal efficiency of 96.9% and reduced ozonation time (from 35 min to 10 min) and dosage ( from 70 g/m to 40 g/m ). On the contrary side, pollution load was increased beyond the textile industry standards, i.e., national environmental quality standards (NEQs), which must be considered for future studies. Anyhow, it is concluded that the biosorption followed by the ozonation may bring effective solutions for the decolorization of textile dye Reactive Blue 19.

**Keywords:** Contact Time; Ozone Dye; Removal Efficiency; TDS; Turbidity

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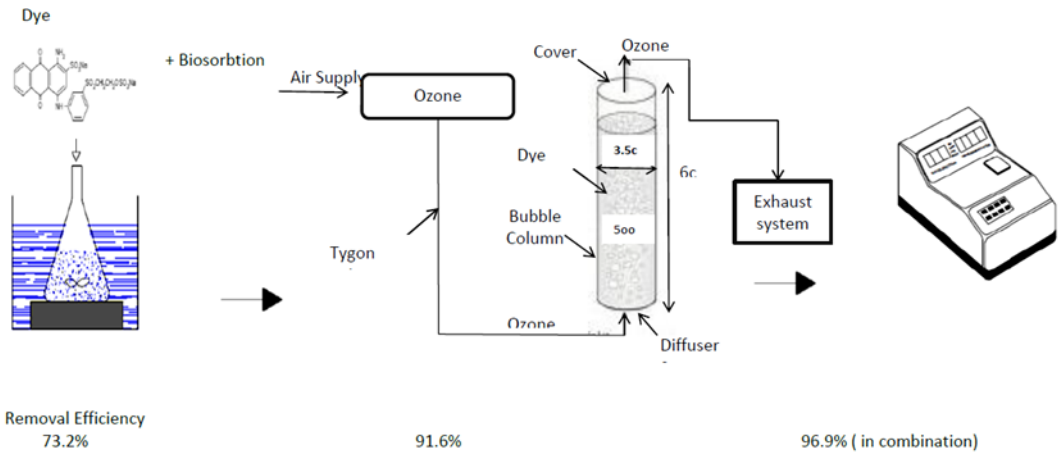
***Correspondence:**
Anam Khalid
University of the Punjab, Lahore, Pakistan
Email: meetanam@yahoo.com

## Graphical Abstract




## Introduction

Textile business is one of an emerging business worldwide. It uses high quantities of water and produces enormous volumes of waste products originating from various processes such as dyeing, finishing, printing, etc. (Babu et al., 2007). The waste includes potentially hazardous compounds and is, therefore, has been considered as one of the most pollution causing industries. Composition and volume of textile liquid by products vary greatly and depends on chemicals being used and treatment processes (Arslan-Alaton et al., 2002 and Tapalad et al., 2008).

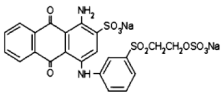
These chemicals lead to a strong color, high chemical oxygen demand (COD), dissolved and suspended solids, noxious organic

compounds (e.g. phenols), heavy metals (e.g. Cr, Cu, Ni, etc.), and total organic carbon (TOC); together with fluctuating pH and temperature of the effluent (Wu and Wang, 2001). All of the compounds settle their fate into the water bodies as of discharged effluent (Galindo et al., 2001). The direct discharge renders the color of receiving water body being undesirable and displeasing aesthetically (Fahimi et al., 2010).

A variety of physical, chemical and biological methods have been used for the treatment of wastewater effluents (Guendy, 2007). Integrated processes involving several combinations such as ultra (membrane) filtration, reverse osmosis, ion exchange, and adsorption are widely employed to remove the color from the dye-contaminated effluents (Inaloo et al., 2011; Siddique et al., 2011). Though many of these methods are promising and non-destructive, however, need of intensive capital investment for variety of dyes makes them economically inefficient. So cheap process for textile color removal is biosorption that is why a lot of work is being done in field of biosorption many researchers worked to find out biosorption potentials of algea (Hamdy, 2000 and Seki and Suzuki, 1998), fungi (Guibal et al., 1992; Kapoor et al), bacteria (Ozturk, 2007; Pumpel et al; Fein et al., 1997) plants (Joshi et al., 2003; Rahman et al., 2005) and agricultural waste (Sud et al., 2008) for metal removal and attained favorable results compared to conventional technologies.

In recent years, application of ozonation has been increased drastically for the decolorization of textile dyes. It is because the process has great potential to eliminate high COD and color pigments while increasing biodegradability which further help in recovering heavy metals (Adams and Gorg , 2002; Hsinget al., 2006; Zhou and Smith, 2002). Biosorption, on the other hand, is a method in which a particular biomolecules or biomass (inactive or dead) adsorb and concentrate certain ions (metallic) or molecules from aqueous solution to clean the water (Igwe, J.C. and Abia, 2006). Although, the biosorption is a cheap technique for the treatment of textile dyes but it is less efficient in removing dyes as compared to ozonation for high dye concentration. To compensate the drawback of both the processes, this study was designed on the combined treatment of biosorption followed by subsequent ozonation. The primary aim was to investigate the combined effect of biosorption and ozonation on pollution indicators namely pH, turbidity, electrical conductivity (EC) and TDS (total dissolved solids).

**Table 1:** Physical and chemical characteristics of BR-19 dye

| Commercial Name | Remazol Brilliant Blue Special |
|---|---|
| Generic name | Reactive blue-19 |
| Chemical Structure |  |
| Chemical class | Anthraquinone |
| Chemical name | 2-(3-(4-Amino-9,10-dihydro-3-sulpho-9,10-dioxoanthracen-4-yl) aminobenzenesulphonyl) vinyl) disodiumsulphate |
| Formula | $C_{22}H_{16}O_{11}N_2S_3Na_2$ |
| λmax (nm) | 592 |
| Molecular weight | 626.5 g/mol |
| Biodegradability | <10 % |
| Toxicity to fishes, CL 50 | 500-1,000 mg/L |

# Materials and Methods

## Preparation of The Biosorbent

The cotton balls, used as biosorbent, were obtained from a farmland of Rampur Jahangir Lahore, Pakistan. These cotton balls were dipped in 1 N HCl solution for five hours, followed by washing in distilled water thrice to get rid of impurities. The balls were oven dried at 105 °C until a constant weight was obtained in order to get powdered form of biosorbent, the balls were crushed and then sieved by 100 nm ASTM D 4607-86, 1990.

## Preparation of Dye Solution

After the optimization of dose, adsorption experiments were conducted by mixing 0.5 g of dry adsorbent into five different concentration of dye solution (Ismail et al., 2012).

## Experimental Procedure

**Contact Time:** The equilibrium time is an important parameter of agitation and, therefore, flasks of volume 250 ml were taken and added 0.75 g of the adsorbent in 100 ml of dye solution of 100 ppm. These flasks then put on the shaker and were agitated at 250 rpm on an orbital shaker at 32±2 °C. During the elapsed time flasks were taken out one by one in order to check how much of the dye in the solution was adsorbed (Suteu and Zaharia. 2011; Ignat et al., 2012).

**Adsorbent Dose:** To achieve an optimum dose for biosorption, different adsorbent dosage was added into flasks. Then, the absorbance of each treated sample was measured using the spectrophotometer to check removal efficiency after given 2.5 hours of time (Barka et al., 2013).

**Initial Dye Concentration:**After the optimization of dose, adsorption experiments were conducted by mixing 0.5 g of dry adsorbent into five different concentration of dye solution (Ismail et al., 2012).

## Experimental Method for Biosorption + Ozonation

The 1 g dose of biosorbent was added into into 100 ml of dye solution of different concentrations concentration (20, 40, 60, 80,100, 150, 200, 250, 300 and 350 ppm). Each dye solution was agitated at 250 rpm for 150 minutes. After biosorption, the ozonation applied on a biosorpted solution.

# Results and Discussion

## Color Removal by Biosorption

**Effect of Contact Time:** The effect of contact time on the removal of reactive dye Reactive Blue-19 by biosorption has been shown in Table 2. The contact time has a noteworthy effect on the adsorption potential of biosorbent. The rate of sorptive removal of

**Table 2:** Effect of contact time on color removal by biosorption

| | Time (min) | Initial dye concentration (ppm) | Color removal (%) |
|---|---|---|---|
| 1. | 30 | 100 | 63.2±0.4e |
| 2. | 60 | 100 | 65.6±0.2cd |
| 3. | 90 | 100 | 66.2±0.7c |
| 4. | 120 | 100 | 69.6±0.9b |
| 5. | 150 | 100 | 73.2±0.9a |
| 6. | 180 | 100 | 60.1±0.5ef |
| 7. | 210 | 100 | 54.0±0.1g |

Values presented in table 2 are the averages of three independent replicates. Values with the ±signs show standard error between replicates of the same treatment. Small letter represent level of significance as governed by ANOVA at (P>0.05) (At constant concentration and same amount of biosorbent dose).

this reactive dye was slow in first 60 minutes, which later increased quickly achieving the maximum of 73.2% of decolorization in 150 minutes. The equilibrium was established at about 150 minutes of the running time: however, further increasing the shaking time decreased the removal efficiency. After this time high removal rate of the adsorbate in the start could be due to the large surface area of the adsorbent available for the adsorption of dye ions (Khattri and Singh, 2009). Nevertheless, after a certain period of time, a slight increase in the dye uptake might be attributed to the availability of a few active sites on the surface of sorbent (Tan et al., 2010).

**Effect of Biosorbent Dose:** The results of the impact of biosorbent dosage are expressed in Table 3. It can be seen that, by increasing biosorbent dosage, removal efficiency was increased initially up to a certain limit, and then remain stable. Because, as biosorbent dosage increase, active sites also increase but further addition of adsorbent in the dye solution could limit the transportation of dye ions to the active adsorption sites and hence reducing the adsorption efficiency (Tanet al., 2010; Gode and Pehlivan, 2005). The maximum uptake of the Reactive Blue-19 was observed at biosorbent dosage of 1 g.

**Effect of initial dye concentration:** The effect of initial dye concentrations on the efficiency of decolorization is presented in Table 4. The results showed that the color removal efficiency was increased by increasing the dye concentration, but up to a certain limit. It could be due to the fact that fewer dye molecules were available to be adsorbed onto the biosorbent surface. The higher dye concentration helps the development of the stronger driving forces of the concentration gradient leading to the high adsorption capacity (Feng et al., 2012). Subsequently, the system attained equilibrium, which reflected, the adsorption of the maximum of dye by the adsorbent (Lutpi et al., 2011). In the experimental setup, concentration equilibrium was established between the dye molecules and available adsorbent site at 100 ppm of dye; which is why the maximum removal efficiency of 73.9% is observed. Afterward a stable trend in the color removal was observed for higher dye content. This is due to the saturation of the sorption sites on the adsorbents as the concentration of the dye increased (Tan et al., 2010; Ismail et al., 2012).

**Table 3:** Effect of biosorbent dosage on color removal by biosorption

| | Biosorbent Dosage (g) | Initial dye concentration (ppm) | Color removal (%) |
|---|---|---|---|
| 1. | 0.25 | 100 | 52.8 ±0.5g |
| 2. | 0.5 | 100 | 58.0±0.4f |
| 3. | 0.75 | 100 | 64.8±0.2e |
| 4. | 1 | 100 | 73.6±0.3a |
| 5. | 2 | 100 | 72.3±0.6ab |
| 6. | 3 | 100 | 67.2±0.1c |
| 7. | 4 | 100 | 65.8±0.3cd |

Values presented in table 3 are the averages of three independent replicates. Values with the ±signs show standard error between replicates of the same treatment. Small letter represent level of significance as governed by ANOVA at (P>0.05) (At constant concentration and time).

Values presented in table 4 are the averages of three independent replicates. Values with the ±signs show standard error between replicates of the same treatment. Small letter represent level of significance as governed by ANOVA at (P>0.05) (At constant time and equal amount of biosorbent dose).

**Table 4:** Effect of initial dye concentration on color removal by biosorption

| | Initial dye concentration (ppm) | Color removal (%) |
|---|---|---|
| 1. | 20 | 60.2±0.9e |
| 2. | 40 | 63.0±0.2d |
| 3. | 60 | 69.7±0.3bc |
| 4. | 80 | 70.1±0.4b |
| 5. | 100 | 73.9±0.6a |
| 6. | 150 | 49.5±0.8f |
| 7. | 200 | 40.7±0.2g |
| 8. | 250 | 15.1±0.7h |
| 9. | 300 | 5.5±0.1i |

## Color Removal by Ozonation

**Effect of contact time:** The impact of reaction time on color removal is displayed in Table 5. The color reduction of dye was low (55.7%) for 5 min, and then enhanced quickly achieving 91.3% decolorization in 35 min (Table 5). The optimum contact time for effective reduction was observed at 35 minutes. The results of the present work showed that the degradation of RB-19 in aqueous solution by ozonation involved the destruction of dye chromophore components (Fanchiang and Tseng, 2009). Because the longer Ozonation durations increases liquid phase $O_3$ content resulting in raised dye removal by degrading aromatic structures and double bond (Hsing et al., 2007; Wu and Wang, 2001).

**Effect of Ozone Dosage:** The results of the impact of applied ozone dosage on color removal are expressed in Table 6. The result depicts that, with increasing ozone dosage, dye removal efficiency was also increased. Briefly, by increasing the ozone dosage from 10 g/m$^3$ to 70 g/m$^3$, the decolorization efficiency enhanced from 17.4-92.7 %. The optimum dose was found to be 70 g/m$^3$ as no significant increase in color removal is observed

**Table 5:** Effect of contact time on color removal by biosorption

| | Time (min) | Color removal (%) |
|---|---|---|
| 1. | 5 | 55.7±1.2$^{h}$ |
| 2. | 10 | 63.3±0.9f$^{g}$ |
| 3. | 15 | 69.2±1.6$^{f}$ |
| 4. | 20 | 73.6±1.3$^{e}$ |
| 5. | 25 | 81.7±1.7$^{d}$ |
| 6. | 30 | 86.7±1.5$^{c}$ |
| 7. | 35 | 91.3±1.4$^{a}$ |
| 8. | 40 | 91.3±1.1$^{ab}$ |

Values presented in table 5 are the averages of three independent replicates. Values with the +signs show standard error between replicates of the same treatment. Small letter represent level of significance as governed by ANOVA at (P>0.05) (At constant concentration and equal amount of Ozone).

**Table 6:** Effect of ozone dosage on color removal by ozonation

| | Ozone dosage ($g/m^3$) | Color removal (%) |
|---|---|---|
| 1. | 10 | 17.4±0.4$^{g}$ |
| 2. | 20 | 25.2±0.6$^{f}$ |
| 3. | 30 | 65.6±0.1$^{e}$ |
| 4. | 40 | 72.0±0.3$^{d}$ |
| 5. | 50 | 82.1±0.6$^{c}$ |
| 6. | 60 | 89.2±0.2$^{b}$ |
| 7. | 70 | 91.4±0.7$^{a}$ |

Values presented in table 6 are the averages of three independent replicates. Values with the +signs show standard error between replicates of the same treatment. Small letter represent level of significance as governed by ANOVA at (P>0.05) (At constant concentration and equal amount of Ozone).

after this dosage because of the structural arrangement of dye atoms and its configuration in solution form.

The results are incoherent with mass transfer theories. According to these theories, the driving force for the transfer of ozone to the dye solution and the ozone consumption per volume of dye solution increases with increasing inlet ozone dose. It augments dye oxidation and color removal (Konsowa, 2003). The previous research by (Konsowa, 2003; Sares et al., 2006; Yasar et al., 2007 and Fahimi et al., 2010) also support the findings of the study. (Tehrani-Bagha et al., 2010) also found the similar result i.e. color removal increases by increasing the ozone dose.

**Effect of initial dye concentration:** It has been observed that a substantial decrease in color removal was observed for higher dye contents i.e. 40.7 % for 350 ppm dye solution and at 20 ppm concentration, the dye removal was found up to 91.8%as shown in Table 7. This could be due to the fact that, under the applied conditions, the ratio of ozone molecules to dye molecules in the solution decreases with the increase of the dye concentration (Tehrani-Bagha et al., 2010). So thecolor removal efficiency of the ozonation process is dependent on the initial dye concentration in an inverse manner.

Conclusively, increasing initial dye concentration may have caused more ozone to be consumed (Sevimliand Kinaci, 2002).It is also observed in advanced oxidation processes that, with an increase in the dye concentration, various intermediates are formed upon degradation of the parent dye that may interfere with the overall oxidation success. Such suppression would be more pronounced in the presence of an elevated level of degradation intermediates formed upon an increased dye concentration (Mahmoodiand Arami, 2006). Thus, the time for complete decolorization would be longer for higher initial dye concentrations (Thrani-Baghaand Aminib, 2010).

## Color removal by biosorption+ozonation

The main aim was to reduce the ozonation time, dose and cost by coupling it with biosorption. Moreover, the experiment is conducted to enhance the decolorization efficiency by combining biosorptionwith ozonation.

The results showed; at higher dye concentration, the color removal efficiency of the biosorbent was decreased (Table 8) (Tan and Rozaini, 2010; Saad et al., 2010; Deniz and Karaman, 2011). For lower concentration the efficiency was significant. The removal success was 73.9% at 100 ppm, which was reduced to 49.5% to at 150 ppm, 40.7% at 200 ppm, and 5.5% at 300 ppm of the dye solution. The removal efficiency of 150 ppm enhanced from 49% to 95.4% just after 10 min duration of ozonation at the rate of 40 $g/m^3$ of ozone flow. The ozone dose has been reduced and removal

**Table 7:** Effect of initial dye concentration on color removal by ozonation

| | Initial dye Concentration (mg/l) | Color removal (%) |
|---|---|---|
| 1. | 20 | 96.7±1.2$^{a}$ |
| 2. | 40 | 96.0±1.4$^{ab}$ |
| 3. | 60 | 94.4±1.6$^{c}$ |
| 4. | 80 | 95.9±1.3$^{cd}$ |
| 5. | 100 | 91.6±1.8$^{e}$ |
| 6. | 150 | 86.6±1.3$^{f}$ |
| 7. | 200 | 73.0±1.7$^{g}$ |
| 8. | 250 | 65.4±1.9$^{h}$ |
| 9. | 300 | 45.1±1.2$^{i}$ |
| 10. | 350 | 40.7±1.1$^{j}$ |

Values presented in table 7 are the averages of three independent replicates. Values with the +signs show standard error between replicates of the same treatment. Small letter represent level of significance as governed by ANOVA at (P>0.05) (At constant concentration and equal amount of Ozone).

**Table 8:** Biosorption + Ozonation

| | Initial dye concentration (mg/l) | Color removal by Biosorption plus Ozonation (%) |
|---|---|---|
| 1. | 20 | 65.7±0.4$^{j}$ |
| 2. | 40 | 73.8±0.6$^{i}$ |
| 3. | 60 | 83.3±0.8$^{h}$ |
| 4. | 80 | 90.5±0.1$^{g}$ |
| 5. | 100 | 97.9±0.7$^{a}$ |
| 6. | 150 | 95.4±0.5$^{ab}$ |
| 7. | 200 | 93.7±0.9b$^{c}$ |
| 8. | 250 | 92.9±0.2$^{d}$ |
| 9. | 300 | 90.5±0.1$^{e}$ |
| 10. | 350 | 85.6±0.3$^{f}$ |

Values presented in table 8 are the averages of three independent replicates. Values with the +signs show standard error between replicates of the same treatment. Small letter represent level of significance as governed by ANOVA at (P>0.05) (At constant concentration and equal amount of Ozone).

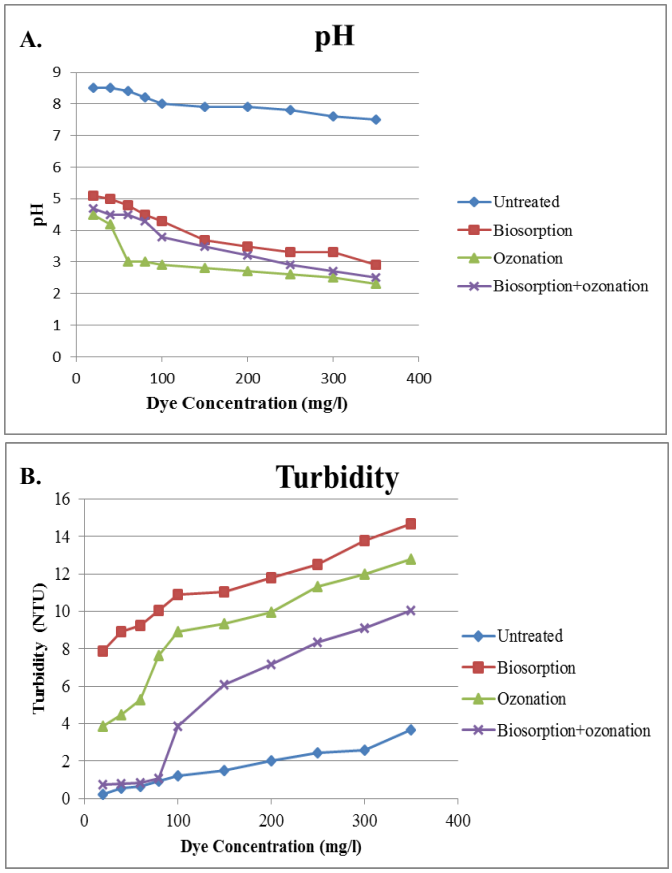
efficiency increased with biosorbtion combination because at first stage the dye from solution was removed by biosorbtion that lower concentration of dye solution. When this pre-treated dye solution feeds for ozonation is enhanced removal efficiency, lowers ozone dose and time as well.

### Pollution indicators values

The effect of biosorption and ozonation were investigated on pollution indicators including pH, turbidity, electrical conductivity (EC) and total dissolved solids (TDS). The results for pollution indicators show a high pollution load in treated water which suggest that the biosorption and ozonation can be a good choice for the removal of color from dye solution but on the other hand, it increase the pollution load exceeding the requirements of textile industry standards. Therefore, it can be stated that ozonation was found to be a major contributor to the increasing pollution load as compared to the biosorption (Figure 1 a, b, c and d). The toxicity of ozonation was decreased when it was coupled with biosorption. The toxicity of the combination of biosorption along with the ozonation was found between the ozonation and the biosorption.

## Conclusions

In light of the entire research work performed, all the results obtained and their comparison and discussion, it is concluded that although other substances are in use for advanced oxidation process, yet chlorine has all the potential to become a powerful source of OH radicals to oxidize in the AOP. Moreover, the use of UV has a very crucial role and a significant effect on the removal of 1,2-dichloroethane using chlorine. Using other different concentrations of aqueous chlorine can give some more insight into this research and these avenues must be searched for. The range of contaminant concentration spiked in water can also be varied and worked on. For this particular research work, the range was a bit below required for study. It is recommended that the lower limit should be defined more close to the maximum contaminant level (MCL) of 1,2-dichloroethane and the number of concentrations should also be increased. Another important constraint that can be varied is the intensity of the UV lamp. The use of ground water for the similar procedure would be of much higher significance and interest.



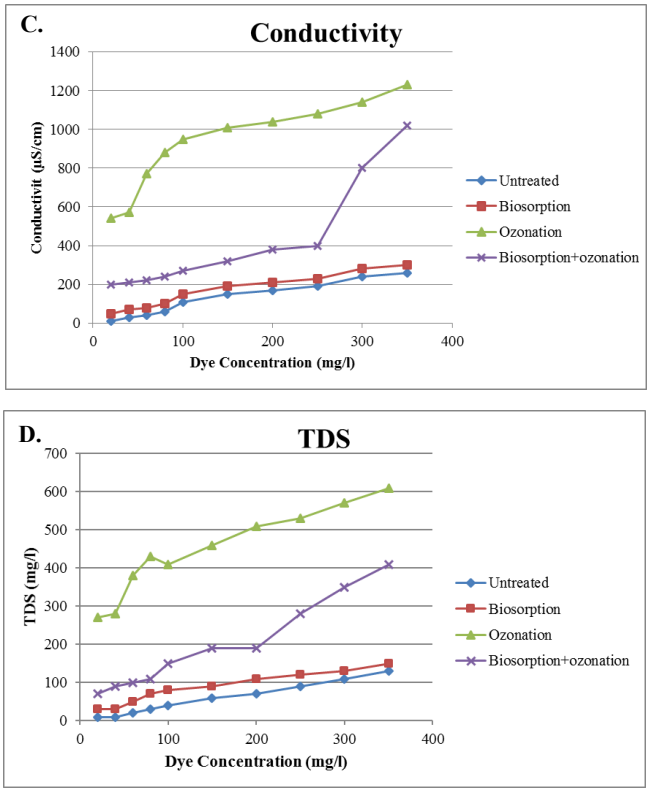




**Figure 1:** Comparison of pH (a), Comparison of turbidity (b) Comparison of conductivity (c), Comparison of Total Dissolved Sol-

### Authors Contribution

Experimental design: Anam Khalid, Azhar Ali; Experimental work: Iqra Sardar and Gulfam Abid; Analyzed data and paper write up: Anam Khalid, Azhar Ali, Sana Noor; Iqra Sardar

### Compliance with ethical standards

### Conflict of Interest

The authors declare that they have no conflict of interests.

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