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Research Article

Treatability Studies on the Optimization of Ozone and Carbon Dosages for the Effective Removal of Contaminants from Secondary Treated Effluent

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This study investigates the novel and advanced integrated pilot-scale treatment system of removal of contaminants in the secondary effluent from municipal wastewater. The main intent of this work is to assess the combination of pressure sand filter (PSF), ultrafiltration (UF), ozone (O_3), and granular activated carbon (GAC) to treat wastewater and evaluate its suitability for water reuse. The experiments were carried out in a following condition: PSF + UF + O3 + GAC, PSF + O3 + GAC, and PSF + UF + GAC. Configuration 1 was found to be more effective when compared to the other two and almost there occurred complete removal of contaminants. Whereas configuration 2 had the lowest removal efficiency of all, and configuration 3 had quite positive results. The influence of process parameters such as ozone dosage, flow rate, and filtration time was optimized. The optimized filtration time was 20 min with the filtration feed flow rate of 300 LPH. The best configuration of this treatment process produced a removal efficiency of about 80 to 90% with the ozone dosage of 8.33 mg/L with a flow rate of 4 l/min, whereas there occurred complete removal by the subsequent action of GAC. Moreover, the biodegradability of wastewaters as measured by the BOD_5/COD ratio increased from 0.45 to 0.53. The proposed integrated pilot-scale process was effective in removing contaminants to the required level of discharge in the environment or reuse and it will pave the way to provide significant benefits to wastewater treatment.

1. Introduction

The increasing world population, climate crisis, and water scarcity bestowed to the raising demand for managing water resources through sustainable means. Owing to the water demand, industries and municipalities are focused on direct and indirect reuse of water/treatment of effluent discharge through various technologies. The reclaimed water from secondary effluent in municipal wastewater treatment plants are foremost and pave the way for preserving our limited resources of fresh water. However, secondary treated effluent cannot be used directly as it may contain organic and inorganic pollutants that would cause severe impacts on human health and the environment [1–8]. Emerging organic contaminants such as pharmaceuticals, personal care products, endocrine-disrupting chemicals (EDC), and Total Organic Carbon (TOC) can cause hazardous effects even at low concentrations [7, 9]. So, it is important to reduce these contaminants concentrations effectively and reuse them safely to improve environmental and societal sustainability. As we are aware that the conventional treatment process comprises preliminary, primary, and secondary treatment where each process plays its role in removing contaminants but as a higher degree of treatment, it is paramount to have a tertiary or advanced treatment process to produce an effluent with high quality. Nowadays, many advanced treatment processes

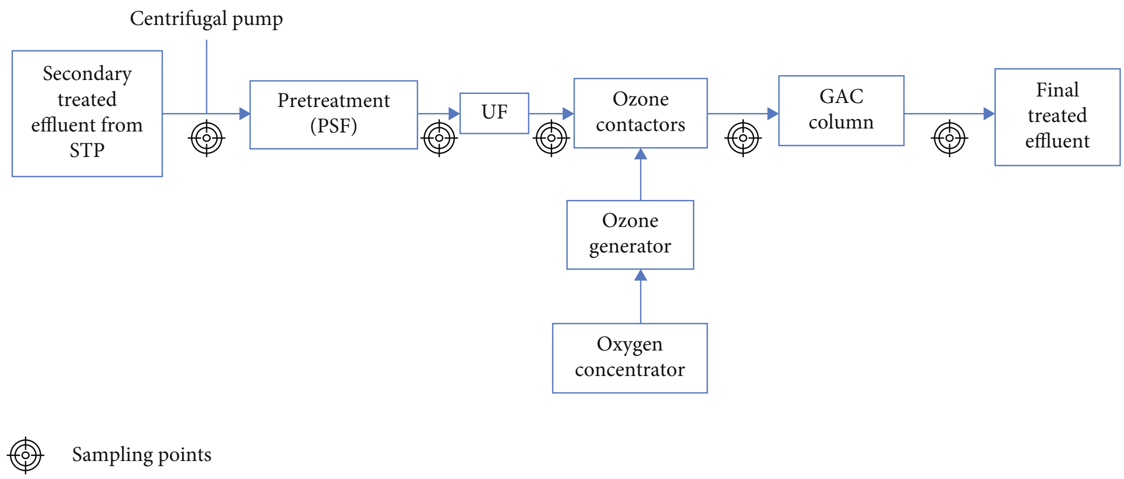

Figure 1: Treatment configuration 1.

have emerged, and the primary objectives of this process lie in the removal of nutrients, pathogens, and turbidity. As a result, coagulation, membrane filtration, chlorination, and ultraviolet radiation are operated. Moreover, there were many other processes available for the treatment of sewage includes membrane distillation, electrodialysis, electrochemical oxidation, photo-electron oxidation, combined coagulation, reverse osmosis membranes, a hybrid reed bed constructed wetland, and microalgae tertiary treatment into activated sludge systems [10–17]. However, the above-mentioned process as an individual is not effective in removing contaminants [18] but an integrated or hybrid approach of treating wastewater is one of the most promising advancements aroused that can be successful and possess more advantages such as reduction of BOD/COD loads, production of favorable effluent quality, eliminates odor, less volume sludge, produce nutrient water for plant irrigation, and increased recycling benefits [19, 20].

The pressure sand filter possesses multiple layers of sand with a variety in size and is considered as one of the effective processes in removing suspended, sinkable, and floating particles present in the feed water with a minimum pressure drop. The main advantage of this simple system involves cost-effectiveness and can be adapted to produce desirable yields. Besides, the effective removal of contaminants depends on the thickness of sand, height of the filter, water flow rate, and size of gravels [21–23]. Membrane filtration tends to be one of the most important unit technologies at present. So, this study is aimed at using the ultrafiltration (UF) membrane process for the treatment of secondary effluent. This process yields high separation efficiency on turbidity, total suspended solids (TSS), organic matters, microorganisms, etc [24–29]. According to the World Health Organization (WHO) reuse guidelines, this process can aid in allowing permeate water for reuse [30, 31]. The limitation of this process falls on the membrane fouling that escalates maintenance cost but to overcome this problem, chemical cleaning of the membrane was used [32, 33]. To pursue a reliable and removal of pollutants efficiently, adsorption onto granular activated carbon (GAC) and chemical transformation with ozone appeared to be the best advances worldwide due to its high surface area, economic design, strong oxidizing ability, and easy handling [34–41]. The combination of ozonation and GAC was used in this study as a substantial oxidation power of ozone with the high adsorption capacity of activated carbon can effectively degrade the recalcitrant and toxic organic pollutants [42, 43]. It was reported that the combination of ozone and GAC can enhance the removal of total organic carbon (TOC) [44–46]. It deteriorates spontaneously during the treatment process by a complex mechanism that entails the hydroxyl radical generation. Along these lines, when ozone deteriorates in water, the free radicals that are formed have a great capacity to oxidize and played a predominant role in the disinfection process. Additionally, it assists with controlling the odor and the viability of ozone relies upon contact time [47–50]. But the limitations of this include extreme toxicity, and the high cost of treatment (being both capital and power-serious). This study contributes to the construction of a novel pilot-scale process of treating the secondary effluent with the combination of pressure sand filter (PSF), ultrafiltration (UF), ozonation, and granular activated carbon (GAC) as new trends in wastewater treatments. Aforementioned, the main intent of this paper is to treat the secondary effluent through an integrated system and reuse it effectively for irrigation, household activities, agriculture, and industries; to characterize the secondary effluent for physicochemical properties; and to optimize the parameters such as ozone dosage, adsorbent quantity, and type of suitable carbon for the effective removal of total organic carbon (TOC). Additionally, to understand the mechanism of ozonation to effectively remove the pollutants present in a water matrix. This work can help researchers to get a notion of the effectiveness of integrated treatment systems and also to understand the feasibility of treatment options available for the proper management of wastewater.

2. Material and Methods

2.1. Reagents. Hydrochloric acid, Hypochlorite, and caustic soda were procured from WAP TECH, Chennai. An air-fed ozone generator was used to produce ozone. A pilot

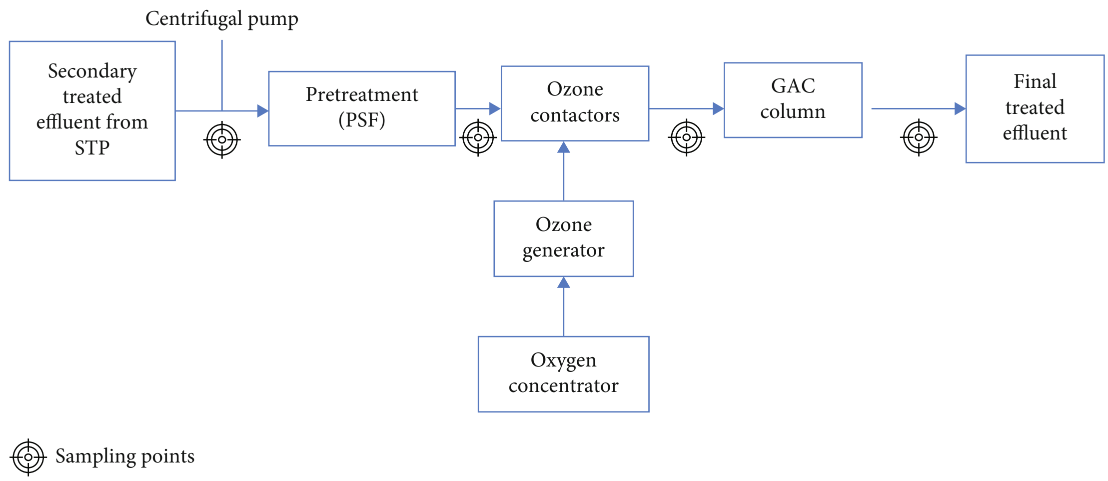

Figure 2: Treatment configuration 2.

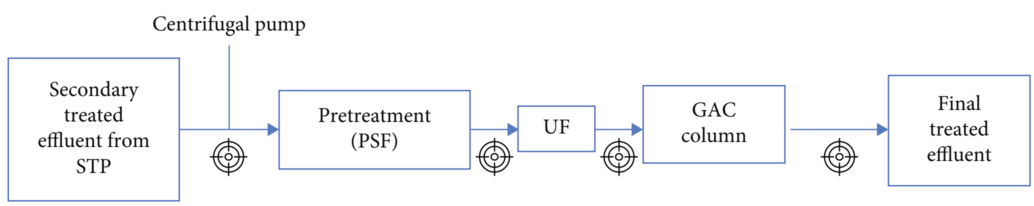

Figure 3: Treatment configuration 3.

Table 1: Design quality parameters of the advanced treatment scheme.

Specification	Pressure sand filter	Ultrafiltration	Granular activated carbon
Diameter	250 mm	—	250 mm
Height	1350 mm	—	1350 mm
Media	Sand & pebbles	Hydrophilic membrane	Sand, activated carbon
Pressure	0.24 MPa	≤0.15 MPa	0.24 MPa
Operating flow	—	250 LPH	—
Permeate flow	—	200 LPH	—
Surface area	—	—	1100 m^2/g
pH	—	1-10	—
Operating temperature (°C)	—	5-45	—

plant was installed by WAP TECH at Sri Sivasubramaniya Nadar College of Engineering, Chennai. The plant consists of a pressure sand filter (PSF), ultrafiltration (UF), ozonation, and activated carbon filter (ACF). Besides, the wastewater was collected from a secondary clarifier in a local sewage treatment plant and is stored in tanks. The treatment was planned to conduct based on three configurations, as shown in Figures. 1–3 ((1) PSF + UF + O_3 + GAC; (2) PSF + O_3 + GAC; (3) PSF + UF + GAC), to predict the best possible contacting patterns for the effective removal of COD, BOD, and TOC from secondary treated effluent.

2.2. Process Description. The filtration time for the treatment process took 20 min for each trial. Raw water flows through the filter bed and the suspended matter gets retained between sand surface and the sand grains. There is a steady rise in the loss of head over a while and the stream get decreased once the pressure drop across the filter is excessive. To assist in cleaning the bed for PSF and ACF, the backwash operation cum rinse leaded via air scouring by a way of agitation through the underdrain system. This is being performed once after each trial. The air scouring agitates the sand with a scrubbing action, which tends to loosen up the interrupted particles and then the filter is ready to be put back into service. Further, the wastewater passes through the pore size of 100 μm bag filter and is collected in the feed tank upstream of the UF system. During the backwash period of 10 s, 6% of hydrochloric acid, 33% of hypochlorite, and 99% of caustic soda were pumped into the permeate

Table 2: Physico-chemical characterization of feed water, treated water quality, and National standards for reuse and discharge of wastewater.

Parameter	Feed water (secondary effluent)	PSF	UF	O_3	GAC	National standards
pH	7.47–7.65	7.30–7.37	7.11–7.12	6.43–6.52	6.29–6.33	5.5 to 9.0
TOC (mg/L)	80.2–137.9	47.5 ± 1.8	41.2 ± 0.62	2.2 ± 0.16	0	—
COD (mg/L)	200–400	188.5 ± 5.6	172.4 ± 1.9	55 ± 0.5	0	200
BOD_5 (mg/L)	120.1–153.3	101.5 ± 1.04	92.5 ± 0.30	27.1 ± 0.14	0	100
Turbidity (NTU)	3–4	2.6–2.7	2.9–1.1	0.5	0	10
TSS (mg/L)	12	9	5	2	0	200
TP (mg/L)	29	22	19	14	0	—

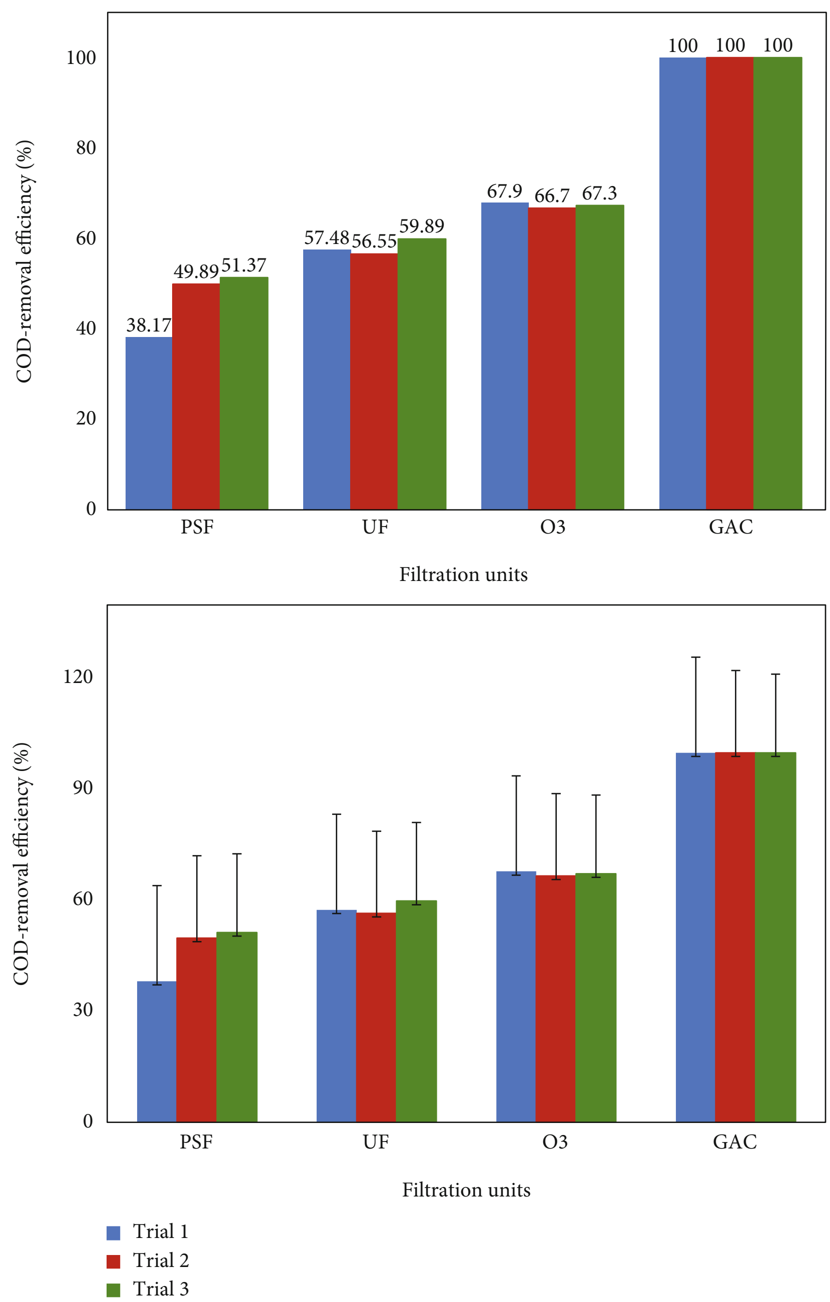

Figure 4: Bar chart representing the removal efficiency of pollutants in PSF, UF, O_3, and GAC.

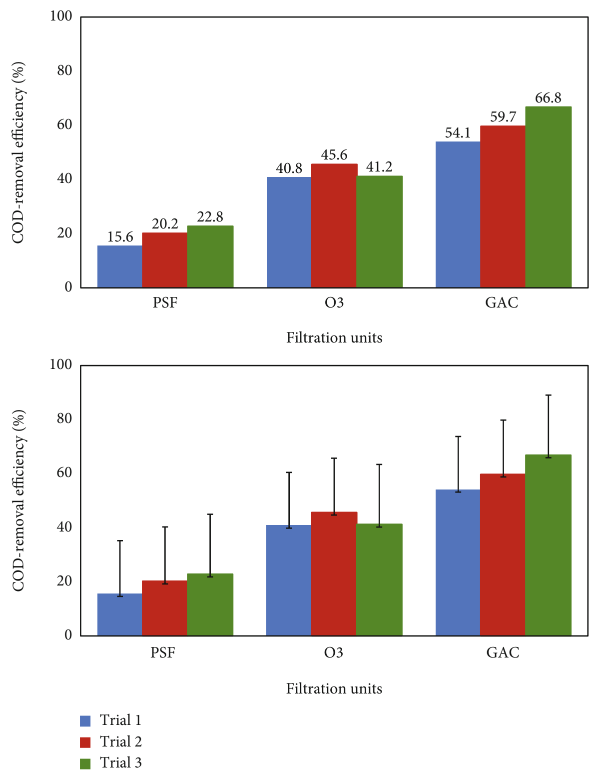

Figure 5: Bar chart representing the removal efficiency of pollutants in PSF, O_3, and GAC.

port at a certain period of interval. The design quality parameters of PSF, UF, and GAC are reported in Table 1.

Ozonation contact tank consists of a 100 L capacity to hold wastewater with a height of 62 cm, an outer diameter of 50 cm, and an inner diameter of 48 cm. The gas mixture containing ozone was sent to the tank through the porous ceramic diffuser built at bottom of the tank with the concentration of 8.33 mg/L (nominal ozone pump capacity of 5 gm/hr) and the required amount of oxygen was 4 L/min with the ozonation contact time of 10 min. The ozonated water was fed into the granular activated carbon (GAC) filter vessel which had a packing size of 2.5 kg/cm^2 and the type of carbon used here is Activated Carbon IV 900. There were 3 trials in each scheme that was being performed. Backwashing is done once after trial, before heading, and operating the treatment unit. After, performing the trials, the samples were stored at 4°C until they were processed for analysis within a day. The pilot plant can be operated in manual mode, partially automated, and fully automated mode, and they were controlled by a control panel. Importantly, the treated effluent was collected in a storage tank and used for gardening.

2.3. Effluent Quality Parameters and Analysis. The samples were characterized in terms of COD, BOD, pH, turbidity, total organic carbon (TOC), and total suspended solids (TSS). A DRB 200 reactor was used to measure COD and TOC, whereas BOD was evaluated using the respirometric method. Turbidity was measured using a turbidity meter and the pH was determined using a portable pH meter procured from Hach. Whereas TSS was analyzed using DR 9000 from Hach.

3. Results and Discussion

3.1. Characterization of Source Water. The foremost physical-chemical parameters of the feed wastewater and the effluent of the full-scale treatment (PSF, UF, O_3, and GAC) relevant to this study are summarized in Table 2.

3.2. Effect of pH. Significant pH control is foremost for ozonation. Besides, when it comes into contact with PSF, UF the pH was about at 7.44 to 8.14. Carboxylic acids are formed due to the response of molecular ozone with organic compounds [51], which barely respond with ozone, and bringing about decrease in pH values in the effluent during the underlying phase of ozonation. At the point when ozonation is done in acidic conditions, the solubility of carbon dioxide and the generation of carboxylic acids are repressed because of the increased presence of hydrogen ions in the system. Thus, the variations of pH in the effluent are dejected. The solubility of carbon dioxide and the generation of carboxylic acids are advanced within the presence of hydroxyl ions. At the point, when ozonation is carried out in basic conditions, consequently, fast reduction of pH in the effluent occurs during the underlying phase of ozonation. In any case, in basic conditions, hydroxyl radicals gradually mineralize the carboxylic acids, which prompt an increase in pH during the subsequent stages of ozonation, and the effective inhibitors (CO_3^{2-} and HCO_3^-) of hydroxyl radicals, react with hydroxyl radicals and hydroxide ions. At pH above 8, ozone decomposition was observed due to a strong effect of hydroxide ions. In an alkaline medium the formation of free • OH radicals create

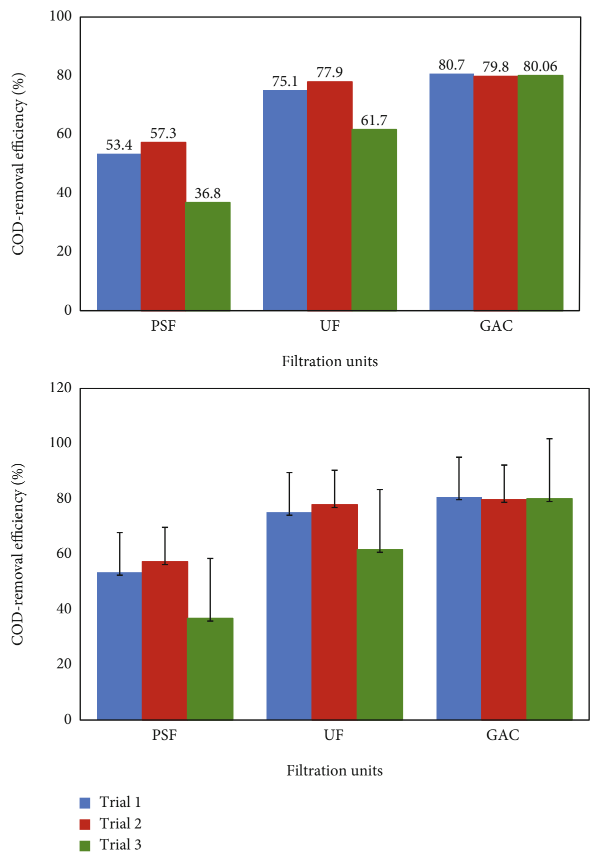

Figure 6: Bar chart representing the removal efficiency of pollutants in PSF, UF, and GAC.

chain reactions by increasing the speed of destruction of ozone [52]. Since the formation of free radicals occurred at faster rate above 8 pH, it can be inferred that the best ozonation performance occurred at alkaline pH using less ozone and thereby increasing the ozone transfer efficiency [53].

3.3. Effect of Removing COD and BOD_5. The influent COD before heading it into the filtration units were different for each scheme such that the influent COD for configurations 1, 2, and 3 was about 290 mg/L, 287 mg/L, and 463 mg/L, respectively. All the trials for each configuration were performed and analyzed in a single day to deduct the functioning and to understand the capability of filtration units for better removal. Based on three trials performed figures 4–6 represents the effect of COD concerning configurations 1,2, and, 3, respectively. While, the error bars represent the standard deviation. COD concentration averaged 121.8 mg/L (SD = 5.006) in the ozone influent and 94.6 mg/L (SD = 1.650) in the ozone effluent for configuration 1 and there occurred complete removal in granular activated carbon. All the configurations were operated under the flow rate of 300 l/h and contact time of 20 min. For second configuration, the concentration averaged 221.4 mg/L (SD = 10.479) in the influent of ozone and 168.6 mg/L at ozone effluent (7.644), whereas the COD concentration was 95.1 mg/L (SD = 18.340) in granular activated carbon. As configuration 3 does not contain ozonation, the inlet concentration of COD in granular activated carbon was 177.3 mg/L with an outlet concentration of 92.3 mg/L. When overall efficiencies of all the configurations are taken into an account, the removal efficiency was effective in configuration 1, which was about 100% as it accompanied with all the units such as pressure sand filter (PSF), ultrafiltration (UF), ozonation, and granular activated carbon (GAC). The removal efficiency of configuration 2 was about 60.24% and 40.21% in configuration 3 and it was shown in Figures 4–6. The concentration of COD was low in configuration 2 due to the UF performance which was mislaid as it is well-known for its removal of particulate and macromolecules from the wastewater, but it was not present in configuration 2. On the other hand, there occurred a poor reduction of COD in configuration 3 due to the absence of ozone demand. In this regard, it can be inferred that the first configuration performed well as PSF, UF, ozonation, and GAC played a major role in reducing COD. Ozonation had a great effect on the operational performance thus resulting in a better reduction. Furthermore, GAC (with an average removal of 96%) performed better than sand filter (with the average removal of 47%) [54]. As predicted, it had a great influence on the reduction of contaminants from the wastewater. A set of oxygen flow rates including 1 l/min, 2 l/min, 3 l/min,

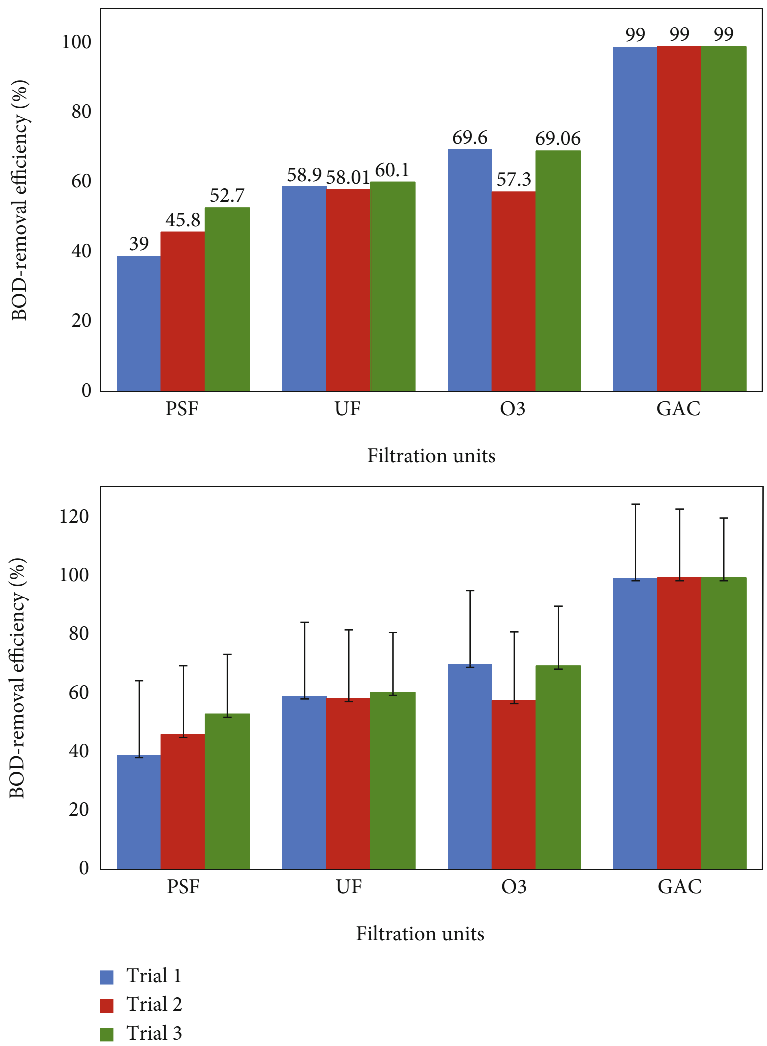

Figure 7: Bar chart representing the removal efficiency of pollutants in PSF, UF, O_3, and GAC.

and 4 l/min were scrutinized to investigate its effect on degradation performance and the optimized flow rate was at 4 l/min which had a greater impact on the removal efficiency of about 99%. Besides, the removal efficiency of 1 l/min, 2 l/min, and 3 l/min were 33%, 47%, and 60%, respectively.

The effect of removing BOD_5 was relatively good in configuration 1 with the rate of removing that keeps in between 39 to 99%. It was observed that the BOD_5 increased when the concentration of ozone was supplied in a small quantity, and there was a decline in the BOD_5 level when the contact time and the concentration of ozone was increased about 4 l/min with 8.33 mg/L. Thus, stating that ozonation had a great impact on the reduction of BOD_5. Furthermore, the BOD_5/COD ratio is one of the paramount indicators to check the effluent biodegradability. Predominantly, if the BOD_5/COD falls greater than 0.3, represents a readily biodegradable effluent, it was calculated from treated effluent and there was a steady increase in the ratio of about 0.45 to 0.53, indicating that the treated wastewater has a great potential for biodegradation [55]. Based on three trials performed figures 7–9 represents the effect of BOD concerning configurations 1,2, 3, respectively, and the error bars represent the standard deviation.

3.4. Solids and Turbidity. In configuration 1, TSS in the ozone effluent was greatly reduced and it averaged 2 mg/L. Besides, there occurred complete removal in GAC (about 100% efficiency). Pressure sand filter and granular activated carbon filter performed similarly concerning turbidity removal. PSF contributes as a sustainable and effective treatment option for suspended solids removal, whereas heterogeneous and biological oxidation occurs on the surfaces of granular material since contaminants either get adsorb or are oxidised by microbes attached to the granular filter media. When GAC is used as a postfiltration process, it receives a high-quality effluent to adsorb organic compounds which were not been adsorbed in previous stages. As per the WHO standards, ultrafiltration can effectively remove organic materials from wastewater [56–58]. In configuration 2 and 3, the rate of removal was less where TSS in the ozone effluent averaged 8 to

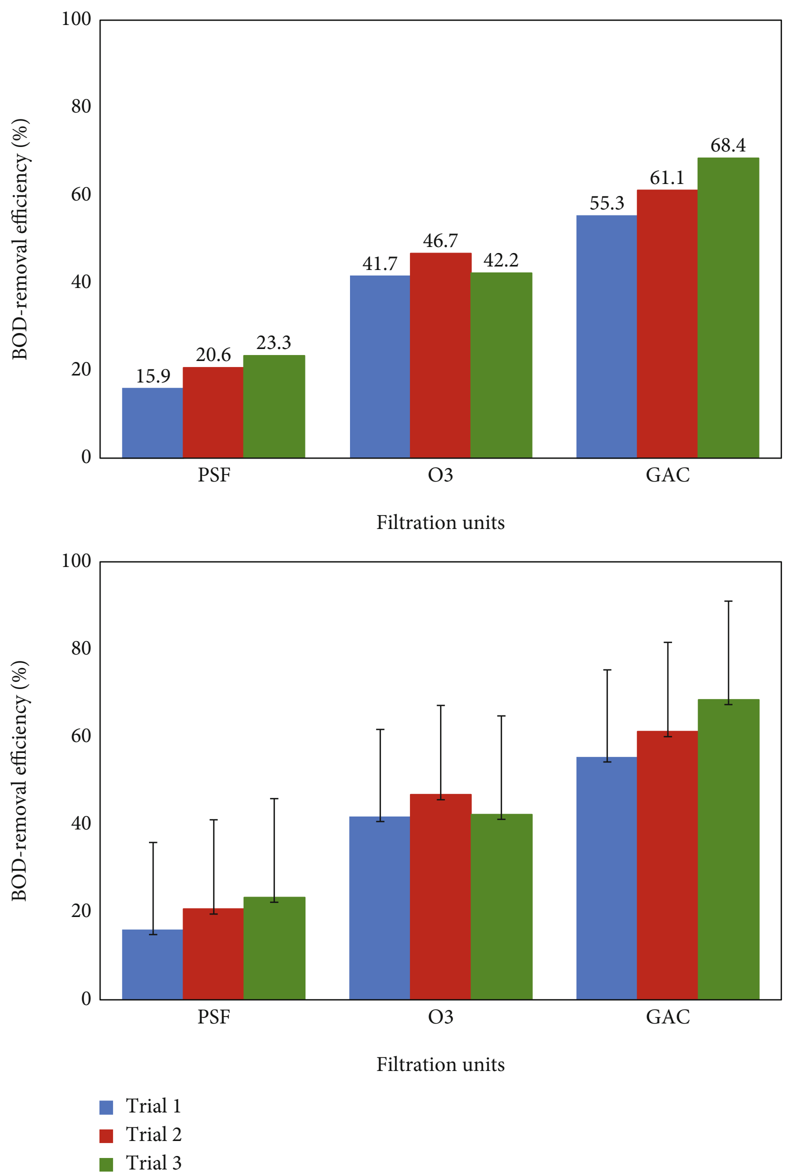

Figure 8: Bar chart representing the removal efficiency of pollutants in PSF, O_3, and GAC.

10 mg/L. For turbidity removal to be considered, in configuration 1, both the filters performed very similar, and the removal averaged 3-4 NTU for PSF, whereas, complete removal took place in GAC.

3.5. Total Organic Carbon (TOC). TOC is a one of the paramount quantitative measures of the total amount of organic constituents present in the wastewater. Figures 10–12 represent the effect of TOC concerning configurations 1,2, and, 3, respectively, whereas, the error bars represent the standard deviation. The influent TOC before heading it into the filtration units were different for each scheme such that the influent TOC for configurations 1, 2, and 3 were 80.2 mg/L, 79.2 mg/L, and 139.2 mg/L, respectively. All the trials for each scheme were performed and analyzed in a single day to deduct the functioning and understand the capability of filtration units for better removal. According to the theory of fixed-bed adsorption, at a steady-state, the effluent concentration ought to equal the influent concentration. However, under wastewater treatment conditions the effluent concentration of TOC seldom reaches the influent concentration; but in this study the effluent concentration raised to a steady-state value that was lower than the influent concentration. In effect, the GAC column continues to remove the influent TOC indefinitely, and more or less the constant steady-state removal is usually attributed to biodegradation, thus, resulting in better separation efficiency. Since GAC has a finite adsorption capacity, effluent concentration increases with run time. The combination of ozone and GAC

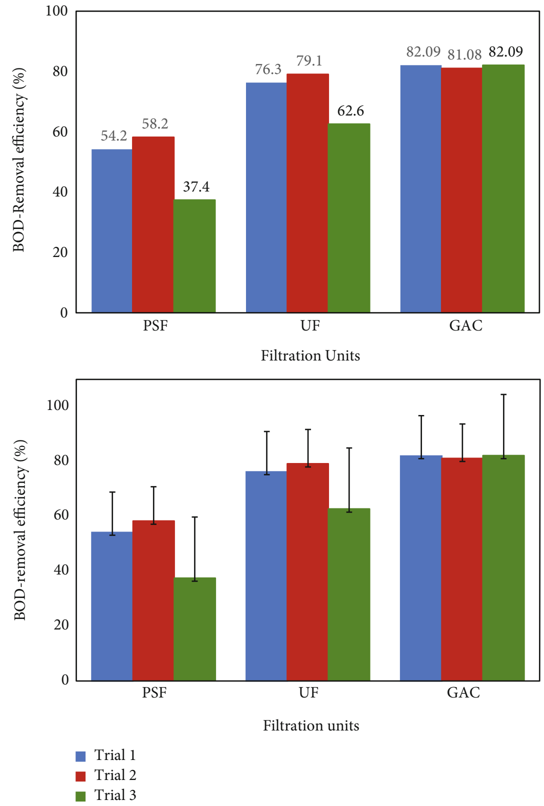

Figure 9: Bar chart representing the removal efficiency of pollutants in PSF, UF, and GAC.

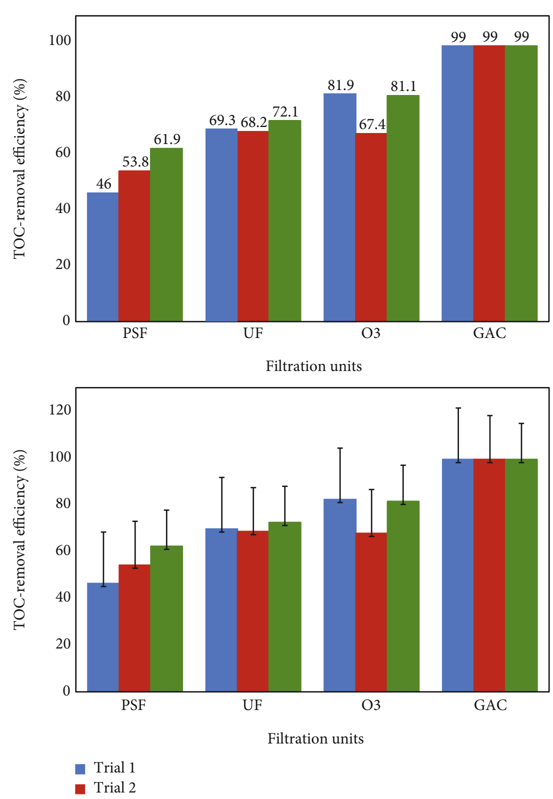

Figure 10: Bar chart representing the removal efficiency of pollutants in PSF, UF, O3, and GAC.

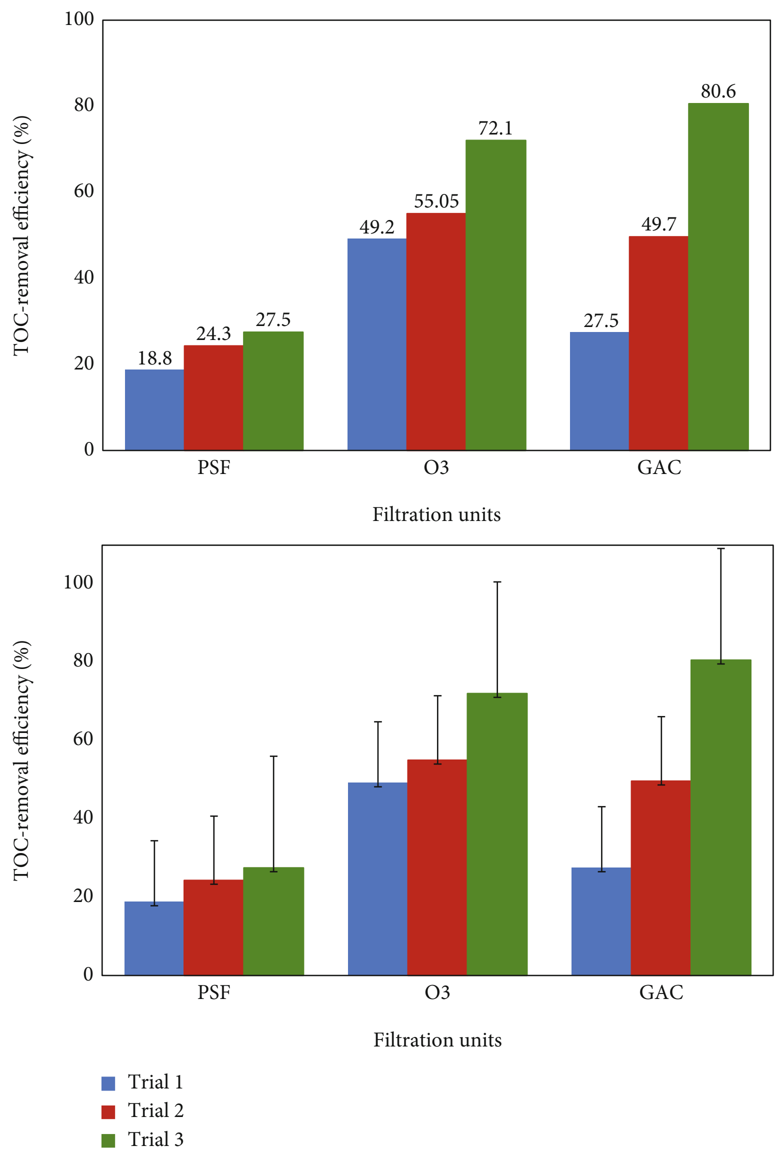

Figure 11: Bar chart representing the removal efficiency of pollutants in PSF, O3, and GAC.

decreased the TOC loading from 137.9 mg/L to complete removal with the increase in removal efficiency from 30% to 100%.

4. Conclusion

The present study focused on the novel and advanced integrated pilot-scale treatment system (PSF, UF, O_3, and GAC) for treating municipal wastewater from secondary effluent. The potential of ozonation and carbon dosages had been scrutinized for the reduction of COD, BOD, and TOC that were detected and quantified in wastewater. In the light of the analysis obtained and discussions that had presented in the preceding sections, concluding remarks may be summarized as follows:

(i) The feasibility of applying ozonation, the biodegradation process, and the optimization of the reaction process were scrutinized. The selection, exploration of reactions, and water chemistry conditions such as ozone flow rate, pH, and filtration time were optimized and falls as a deciding factor for the treatment efficiency. Thus, it can be concluded that the ozonation contributed to the higher COD, BOD, and TOC removal efficiencies and better biodegradability

(ii) No detection of dissolved ozone was observed during the first 2 to 4 min. Chemical reactions took place at a faster rate, and due to the high enhancement factor of ozone at the initial stage ozone mass transfer appeared as a limiting step. By the

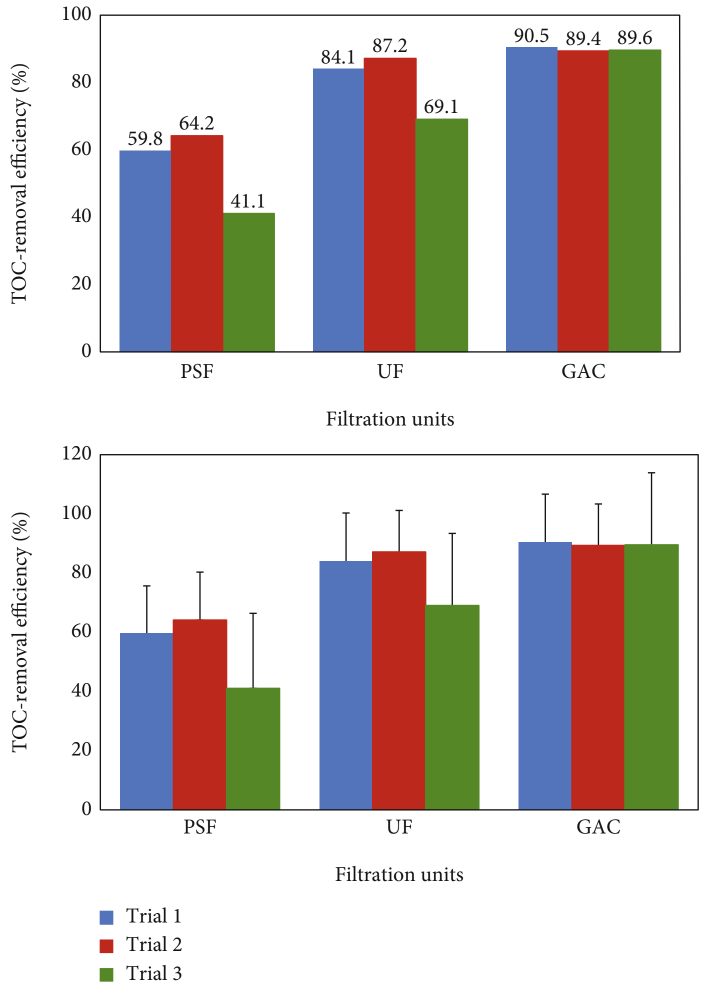

Figure 12: Bar chart representing the removal efficiency of pollutants in PSF, UF, and GAC.

continuous ozonation, with the contact time of about 10 min, was found to be good and the fast kinetic pattern transitioned to a moderate pattern. There occurred a decrease in the removal efficiency in the lower flow rate of about 1 l/min and also in the high flow rate of above 5 l/min. So, the optimized ozone flow rate detecting better-removing efficiency of recalcitrant was about 70 to 80% was observed at 4 l/min with the concentration of 8.33 mg/L

(iii) From the study, ozone degradation has a great potential to produce high quality treated water with reduced toxicity. It cannot be used as a stand-alone treatment process, which also requires a posttreatment process for removing the oxidation by-products before reusing the water. Therefore, the GAC process was performed, and it acted as an additional polishing effect and thus helped in better reduction of targeted COD, BOD, and TOC concentrations. By combining GAC as a posttreatment process, the complex odor issues were well controlled. The high level of TOC using GAC was greatly reduced and attained a removal efficiency of about 99%.

(iv) Thus, the study exerts an effective removal of contaminants from municipal wastewater using an advanced integrated treatment system. As the results of the pilot system were quite positive and the effluent quality meets the National Standards for discharge to the sewer network as well as for reuse the treated water can be used it further or safely discharged to the environment. The proposed pilot-scale integrated system presents a successful process for obtaining high-quality water with low operational and running costs. Besides, PSF and UF can effectively help in removing suspended solids and bacteria, also, act as a pre-treatment, whereas GAC acts as a post-treatment process and adsorbs organic compounds that were not filtered in previous stages. Ultimately, the application of this treatment process with the combination of pressure sand filter, ultrafiltration, ozonation, and granular activated carbon will pave the way to provide significant benefits for treating wastewater

Data Availability

Data are available on request.

Conflicts of Interest

The authors declare that they have no conflicts of interest.

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