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Research article

Elucidating the synergistic effects of aeration and non-thermal plasma on the degradation pathways of specific pollutants in wastewater

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ABSTRACT

Organic pollutants originating from industrial discharges pose significant threats to human health and ecological balance. Conventional pretreatment methods face challenges due to high costs, limited efficiency, and the generation of residual sludge. Non-thermal plasma (NTP) technology, a promising advanced oxidation process, has attracted substantial research interest for its potential to rapidly and effectively treat industrial wastewater. This study employed a dielectric barrier discharge (DBD) reactor to investigate the feasibility of low-cost, efficient industrial wastewater treatment through NTP-mediated pollutant degradation. NTP generates reactive oxygen and nitrogen species (RONS), capable of complete organic pollutant oxidation. Wastewater samples from Kaveh Industrial City underwent treatment in a DBD reactor to induce the formation of reactive agents. Water quality parameters, including turbidity, total dissolved solids (TDS), total suspended solids (TSS), chemical oxygen demand (COD), biochemical oxygen demand (BOD_5), dissolved oxygen (DO), electrical conductivity (EC), and pH, were measured before and after synergetic plasma treatment. The combination of aeration/filtration and 90 min of plasma treatment significantly reduced turbidity compared to untreated wastewater. A 30-min NTP treatment coupled with aeration/filtration demonstrated superior efficiency in removing TDS and TSS, attributed to NTP-generated active species. Optimal COD and BOD_5 removal was achieved through a 24-h aeration, adsorbent filtration, and 30-min NTP process. While standalone 30-min NTP treatment exhibited lower efficiency, the combined aeration/filtration system reduced EC and increased pH with extended plasma exposure. A comparative study of advanced oxidation processes showed that plasma treatment effectively reduced COD by 65 %. Plasma offers a cost-effective and efficient solution for wastewater treatment, despite slightly higher energy consumption.

These findings underscore the potential of NTP as a viable strategy for industrial wastewater treatment. The integration of NTP with conventional pretreatment methods offers promising prospects for enhancing wastewater quality and environmental protection.

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1. Introduction

Industrial water treatment is paramount in contemporary environmental stewardship, given the escalating global crisis of water pollution induced by rapid industrialization and urbanization [1,2]. The unchecked discharge of untreated wastewater, replete with highly toxic organic and inorganic pollutants from diverse industrial sectors across developing and developed nations, poses a grave threat to primary water resources such as groundwater and surface water bodies, thereby jeopardizing public health [3,4]. Within this complex milieu, metal ions, especially heavy metals originating from metal finishing industries, constitute a significant environmental risk [5,6]. Moreover, the presence of highly carcinogenic substances like polycyclic aromatic hydrocarbons (PAHs) [7], petrochemical byproducts, and other chemical contaminants exacerbates these environmental challenges [8,9]. The introduction of colorants into industrial wastewater further complicates the pollution landscape, indicating the potential presence of dyes and pigments [10,11]. Characterized by high concentrations of inorganic salts, organic matter, and mineral oils, industrial wastewater constitutes a severe environmental hazard. Consequently, the development of innovative industrial wastewater treatment technologies is imperative to safeguard freshwater and marine ecosystems, and ultimately, public health [12].

Industrial wastewater is typically characterized by a suite of physical and chemical parameters, including total dissolved solids (TDS), turbidity (Tur), color, chemical oxygen demand (COD), biological oxygen demand (BOD_5), dissolved oxygen, and pH [13]. Conventional treatment methodologies such as primary sedimentation, coagulation-flocculation, filtration, ion-pair extraction, and disinfection are commonly employed to ameliorate these water quality indicators [8,14,15]. However, the widespread application of these techniques is often hindered by exorbitant costs, limited efficacy, and the generation of problematic sludge residues [16,17]. The recalcitrant nature of many organic and inorganic pollutants in industrial effluents has necessitated the development of advanced treatment strategies. In recent years, advanced oxidation processes (AOPs) utilizing oxidants like hydrogen peroxide (H_2O_2) and ozone, as well as photocatalysis, have emerged as promising alternatives for enhancing wastewater biodegradability [18]. AOPs generate highly reactive hydroxyl radicals (OH^-) capable of mineralizing a wide spectrum of organic contaminants into carbon dioxide and water, including those resistant to conventional treatment methods. While the removal efficiency of AOPs is contingent upon the oxidant employed, the synergistic combination of oxidants with ultraviolet (UV) radiation, as in O_3/UV, H_2O_2/UV, and O_3/H_2O_2/UV systems, has demonstrated potential for augmenting treatment performance [19,20].

Nevertheless, the widespread application of these AOP techniques is encumbered by significant economic costs, elevated energy consumption, and the potential generation of harmful intermediate byproducts arising from incomplete oxidation [21].

Plasma, the fourth fundamental state of matter introduced by Irving Langmuir in 1928, is a quasi-neutral gas comprising electrons, ions, photons, atoms, radicals, and excited molecules within an electromagnetic field [22]. Its versatility has spurred its application across diverse domains, including medicine [23], agriculture [24], industry [25], and environmental science [26]. Non-thermal plasma (NTP) has emerged as a promising technology for the degradation of a wide array of toxic organic micropollutants, such as pharmaceuticals, pesticides, and organic dyes, as well as the removal of heavy metals from industrial wastewater [13,27,28].

NTP generates reactive oxygen and nitrogen species, including HO•, O•, HO_2•, O_3, H_2O_2, ultraviolet light, heat, and shockwaves [18,20,21]. These highly reactive species facilitate the efficient removal of recalcitrant contaminants under ambient conditions. In particular, the hydroxyl radical (HO•) is a potent, non-selective oxidant capable of rapidly degrading contaminants, with its efficacy enhanced when combined with plasma-induced physical processes like ultraviolet radiation and shockwaves [29,30]. Given its potential for treating industrial wastewater, NTP technology has garnered significant attention within the scientific community in recent years [31]. Typically, a plasma discharge system comprises a power source, an electrode, and a working gas. The system's energy output is influenced by both power input and gas composition. Plasma discharges for wastewater treatment can be categorized into four primary types based on plasma distribution, reactor configuration, and operational parameters. Recent research has increasingly focused on the correlation between operational conditions and the efficacy of plasma systems, as well as the reactive species generated during discharge. Hameedl and Kadhem (2020) constructed a gliding arc reactor to treat bacteria-contaminated water in a Petri dish using magnetic stirring. Their findings indicated a correlation between increased treatment duration and decreased bacterial survival rate [32]. Alternatively, Jose et al. (2019) developed a pulsed corona discharge reactor equipped with a circular metallic ground electrode and seven tungsten needles connected to a high-voltage power supply. Plasma was generated through pulsed voltage-induced air ionization. The study demonstrated complete dichlorination within 10 min and a 91 % reduction in total organic carbon (TOC) after 12 min of treatment [33].

Shen et al. (2016) employed a glow discharge reactor to investigate phthalate degradation in aqueous solution. Their findings demonstrated highly efficient removal of phthalate derivatives, exceeding 99 % within 60 min of treatment using the glow discharge plasma process [34]. Shang et al. (2021) adopted a different approach, constructing a falling film dielectric barrier discharge (DBD) reactor to characterize the generation of long-lived in-situ reactive species. The reactor exhibited exceptional performance in producing and transferring active species from the gas to the liquid phase, thereby facilitating pollutant degradation [35,36].

NTP offers an environmentally benign alternative to chlorine-based wastewater treatment by generating highly reactive species without the introduction of additional chemicals, thereby mitigating secondary pollution [37,38]. While effective in pollutant removal, careful consideration must be given to the potential formation of persistent byproducts that may pose risks to aquatic ecosystems. Due to its versatility, cold plasma can be employed independently or integrated with conventional treatment processes. NTP's advantages, including efficient heavy metal removal, low energy consumption, and cost-effectiveness, position it as a promising technology for water reuse applications [29,30].

While both NTP and other advanced oxidation processes, such as photocatalysis, have demonstrated efficacy in wastewater treatment, NTP offers several distinct advantages. Unlike photocatalysis, which primarily relies on hydroxyl radicals generated through photoexcitation, NTP produces a broader spectrum of reactive species, enabling the degradation of a wider range of pollutants.

Additionally, NTP exhibits greater robustness, as it is less susceptible to operational parameters like pH, temperature, and turbidity. Furthermore, NTP's faster reaction kinetics and potential for complete mineralization of organic pollutants distinguish it as a promising alternative for treating complex wastewater matrices, including landfill leachate [39,40].

A 300-liter wastewater sample, subjected to only preliminary physical and biological (aeration) treatment prior to undergoing the final stages of wastewater treatment, was collected from the Kaveh Industrial City and Special Economic Zone in Arak, Markazi Province, Iran, and transported to the laboratory for analysis. Characterized by a chemical oxygen demand (COD) of approximately 150 mg/L and total dissolved solids (TDS) of around 3856 mg/L, the wastewater sample significantly exceeded environmental discharge standards for absorption wells. As previously discussed [3,4], industrial effluents are notorious for their high organic load and recalcitrant pollutants, necessitating advanced treatment strategies beyond conventional methods.To address these challenges, this study implemented an integrated approach combining aeration, adsorption, and non-thermal plasma (NTP)-based dielectric barrier discharge (DBD). The primary objective was to substantially reduce COD, biological oxygen demand (BOD_5), and total TDS to meet stringent discharge regulations. To comprehensively assess treatment efficacy, dissolved oxygen (DO), electrical conductivity (EC), turbidity, and pH were monitored. Furthermore, the study determined the specific energy consumption (SEC) and total cost per cubic meter of wastewater treated, facilitating comparison with other advanced oxidation processes. Ultimately, this research aimed to develop a sustainable and environmentally sound wastewater treatment process.

2. Materials and experimental methods

2.1. Materials s and instruments

pH measurements were conducted using a standard F20-std-Kit pH meter manufactured by Mettler Toledo (Switzerland). BOD_5 was determined using a BODTrakTM system. COD was calculated by preparing treatment samples in a reactor and employing a HACH model DRB200 (HACH Company, United States) for digestion at 150 °C for 2 h. Subsequent COD analysis was performed using a HACH DR6000 spectrophotometer (HACH Company, United States). Turbidity measurements were obtained using an AQUALYTIC HACH turbidity meter (HACH Company, United States). Total suspended solids (TSS) were determined according to standard method number 2540. A HACH-based HQ440d multiparameter probe (HACH Company, United States) was utilized for the analysis of total dissolved solids (TDS), pH, and electrical conductivity (EC). Color was measured at a wavelength of 450 nm using a HACH DR6000 spectrophotometer (HACH Company, United States) according to ASTM D2120 and expressed as platinum-cobalt units. Phenol concentration was determined according to ASTM D5530-D using a Hach DR6000 spectrophotometer at 510 nm. The pyridine colorimetric method was employed for the analysis.

2.2. Source of industrial wastewater collection

The wastewater employed in this study underwent only preliminary physical and biological (aeration) treatment within 300-liter polyethylene containers located at the Kaveh Industrial City and Special Economic Zone in Arak, Markazi Province, Iran (S0). This industrial park is situated approximately 120 km southwest of Tehran and 160 km north of Arak (Fig. 1). The Kaveh Industrial City boasts a total wastewater treatment capacity of 5000 cubic meters per day. The collected wastewater samples were stored at 4 °C without dilution prior to immediate analysis. The initial characterization of the raw and pre-treated wastewater is presented in

Fig. 1. Aerial map of the sampling location for industrial wastewater (Kaveh Industrial City wastewater treatment plant).

Tables 1 and 2.

As indicated in Table 2, the wastewater sample exhibited COD, BOD_5, EC, and TDS values of 142, 86, 6479, and 3856 mg/L, respectively, which significantly exceeded environmental discharge standards for absorption wells. As previously discussed, traditional treatment methods are often associated with substantial drawbacks, including high operational costs, generation of toxic byproducts, incomplete pollutant removal, and the management of hazardous chemical sludge [41].

To address these limitations, this study incorporated additional aeration to effectively eliminate malodorous compounds and enhance the biodegradability of organic matter [42,43]. Subsequently, a combination of coarse and fine silica sand filters and activated carbon filters was employed to reduce TDS, remove residual particulates and colloids, and adsorb recalcitrant organic pollutants, thereby improving overall effluent quality [44]. Finally, to eliminate persistent organic compounds and other recalcitrant micropollants resistant to conventional treatment, plasma-based technology was integrated into the treatment process [45].

2.3. Experiment design

Experiments were conducted utilizing a pilot-scale treatment system with a capacity of 100 L per hour, equivalent to 2.4 cubic meters per day. A schematic representation of the treatment process is provided in Fig. 2.

2.3.1. Physical pre-treatment

Following the aeration durations of 24, 48, and 72 h investigated by Doan and Lohi [46], an aeration period of 48 h was adopted for this study, aligning with standard practices at the wastewater treatment plant. The pretreatment stage of the S0 wastewater treatment system encompassed aeration and filtration. Initially, wastewater underwent a 24-h aeration process within an aeration tank (S_1). Subsequently, the treated effluent was transferred to the filtration phase, consisting of three distinct PVC pipes filled with coarse and fine silica sand, a carbon-silica mixture, and activated carbon, respectively (S_2).

2.3.2. Non thermal plasma-based treatment

Pre-treated wastewater was pumped into a plasma reactor equipped with multiple quartz-based dielectric barrier discharge (DBD) tubes generating cold atmospheric plasma from ambient air at a voltage of 15 kV and a frequency of 8 kHz. Emission spectra of the air plasma within the 500–700 nm wavelength range were analyzed using optical emission spectroscopy (OES) conducted in Iran. Optical radiation was captured via an optical fiber positioned 4 cm from the plasma. The wastewater stream was subjected to the reactor's electric discharge as a thin film and circulated multiple times according to desired treatment durations ranging from 5 to 90 min (S_3-S_{10}). Samples (S_0-S_{10}) were collected at the conclusion of each treatment stage (Table 3) to assess wastewater characteristics and the impact of varying plasma treatment durations. Subsequently, turbidity, TDS, TSS, COD, BOD_5, DO, and EC parameters were measured.

2.4. Statistical analysis

Data were collected in triplicate for each treatment group and subjected to normality testing prior to statistical analysis. Statistical analyses were performed using Statistica (27) and SPSS software packages. To assess differences among multiple time points, a one-way repeated measures analysis of variance (ANOVA) was conducted, followed by Duncan's multiple comparison post hoc test.

3. Results

Industrial wastewater, subjected to preliminary physical and biological pretreatment, underwent a final stage of physical treatment. A non-thermal plasma (NTP) technology-based dielectric barrier discharge (DBD) reactor was employed as an innovative approach to remove hazardous contaminants from the industrial wastewater. The effectiveness of the pollutant removal process was evaluated by monitoring COD, BOD_5, TDS, EC, TSS, turbidity, and DO.

Table 1
Characterization of the raw wastewater from kaveh industrial city.

EC (μs/cm)	7911
COD (ppm)	4019
BOD_5 (ppm)	2892
BOD_5/COD	0.719
TDS (ppm)	4020
TSS (ppm)	736
Turbidity NTU	432
pH	7.3
Color (Co-Pt)	675
Phenolic compound (ppm)	3.7
DO	1.11

COD and BOD_5: Chemical and biological oxygen demand, TDS: Total dissolved solids DO: Dissolved oxygen, EC: Electrical conductivity.

Table 2
Characterization of the pre-treated wastewater from Kaveh Industrial City wastewater treatment plant.

EC (μs/cm)	6479
COD (ppm)	142
BOD_5 (ppm)	86
BOD_5/COD	0.719
TDS (ppm)	3856
TSS (ppm)	25
Turbidity (NTU)	21
pH	7.5
Color (Co-Pt)	41
Phenolic compound (ppm)	3.11
DO	6.2

COD and BOD_5: Chemical and biological oxygen demand, TDS: Total dissolved solids DO: Dissolved oxygen, EC: Electrical conductivity.

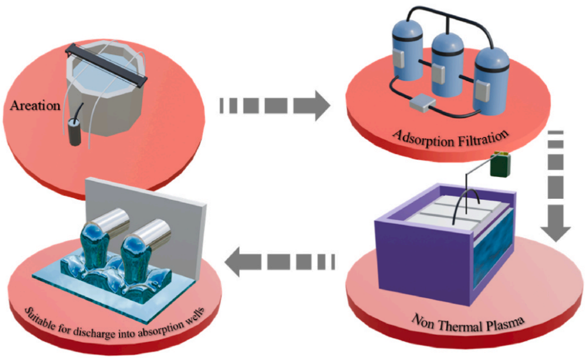

Fig. 2. Schematic of the treatment process.

Table 3
The purification stages of wastewater.

Purification stages	
S_0	The pre-treated wastewater from Kaveh Industrial City wastewater treatment plant
S_1	24 h aeration
S_2	S_1/Adsorbent filtration step
S_3	S_2/Plasma for 5 min
S_4	S_2/Plasma for 10 min
S_5	S_2/Plasma for 15 min
S_6	S_2/Plasma for 20 min
S_7	S_2/Plasma for 25 min
S_8	S_2/Plasma for 30 min
S_9	S_2/Plasma for 60 min
S_{10}	S_2/Plasma for 90 min

3.1. Plasma characteristics

To identify reactive species generated during plasma discharge in contact with industrial wastewater, optical emission spectroscopy (OES) was employed. Fig. 3 presents the OES results obtained from the plasma generated in our reactor operating at 15 kV and 8 kHz. As can be seen in the figure, the peak related to nitrogen oxide (NO) compounds is formed at around 290 nm. After that, the peak corresponding to hydroxyl radicals (OH) has been appeared at around 310 nm. The nitrogen second positive system (SPS) peaks, which correspond to the transitions between the $C^3\Pi_g \rightarrow B^3\Pi_g$ levels, can be observed at wavelengths of 337 nm, 357 nm, and 380 nm. Also, the nitrogen first positive system (FPS) peaks, which correspond to the transitions between the $B^3\Pi_g \rightarrow A^3\Pi_g$ levels, are present in the wavelength range of 650–750 nm. In addition, peaks related to oxygen species were created in the spectrum diagram at wavelengths of 610 nm for excited oxygen and 777 nm for singlet oxygen. All these results indicate that due to the application of an electrical field, the air molecules are excited and ionized, and then reactive oxygen and nitrogen species (RONS) are created in the plasma.

The injected energy induces the dissociation of nitrogen and oxygen molecules by energetic electrons within the discharge. This process leads to the formation of hydroxyl radicals (OH), ozone (O_3), nitrogen oxides (NO and NO_2), and excited N_2 through various

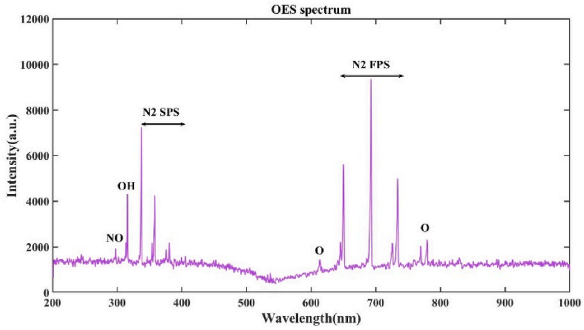

Fig. 3. Optical Emission Spectra of non-thermal plasma in ambient air.

reaction pathways. In the liquid phase exposed to non-thermal plasma, nitrate (NO^{3-}), nitrite (NO^{2-}), and hydrogen peroxide (H_2O_2) are commonly detected. These species can subsequently generate NO, NO_2, hydroxyl radicals (OH), and nitric acid (HNO_3) within the liquid phase.

3.2. Turbidity

Turbidity, a measure of suspended impurities within a fluid [47], exhibited a linear decrease in the industrial wastewater subjected to the combined treatment process, as illustrated in Fig. 4. Initial turbidity of 21 NTU was significantly reduced to 2.8 NTU following aeration, filtration, and NTP treatment. This substantial reduction is attributed to the synergistic effects of these processes. Aeration facilitated the biological degradation of organic matter [48], while filtration effectively removed suspended solids and adsorbable organic compounds [49].

Plasma treatment significantly enhances contaminant removal through the generation of highly reactive species that oxidize and mineralize refractory organic compounds. Moreover, it effectively inactivates microbes, pathogens, and other harmful agents present in wastewater [50,51].

3.3. Total dissolved solids (TDS)

As depicted in Fig. 5, the adsorbent filtration step (S2) exhibited approximately half the solid removal efficiency of S0 and S1, suggesting the significant removal capacity of coarse- and fine-grained silica and activated carbon within the filtration process, likely due to substantial organic compound sedimentation. Pushpalatha et al. [44] emphasized the pivotal role of adsorption technology in treating various wastewater types, primarily targeting the removal of organic and inorganic particles. Adsorption, a critical component of wastewater treatment, involves the transfer of substances from aqueous phases to solid surfaces.

Adsorbent filtration is widely recognized as a straightforward, cost-effective, and efficient purification method applicable across a broad pH range and characterized by ease of operation. Consequently, it is extensively employed in purification processes.

TDS levels exhibited a decreasing trend with increasing treatment time (5–30 min) when employing NTP. The dominant oxidizing species generated by NTP, namely OH^- and O_3, effectively degraded organic compounds. However, a subsequent increase in TDS levels was observed following treatment durations of 60 and 90 min, suggesting a potential negative impact of NTP on this parameter at extended exposure times. Prolonged treatment may induce over-oxidation, leading to incomplete pollutant fragmentation and

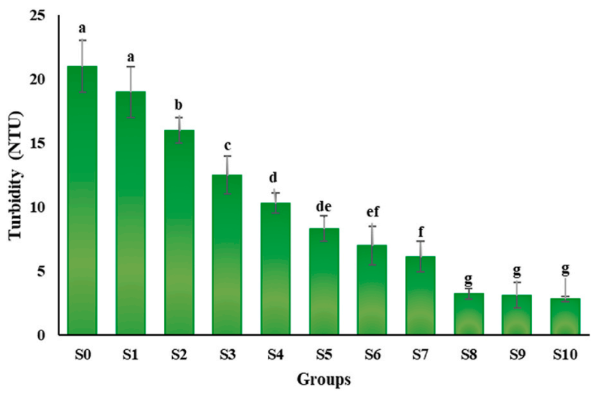

Fig. 4. Duncan tests of means of turbidity parameter with respect to the treatment condition (untreated ($\mathbf{S_0}$) and treated wastewater ($\mathbf{S_1}$-$\mathbf{S_{10}}$)).
* Means followed by the same letter are not significantly different ($p < 0.05$).

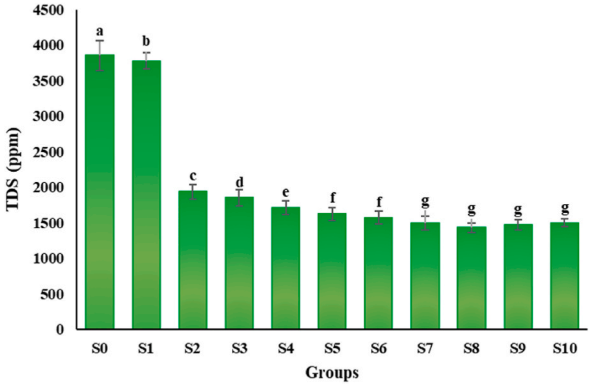

Fig. 5. Duncan tests of means of TDS parameter for untreated (**S_0**) and treated wastewater (**S_1-S_{10}**).
* Means followed by the same letter are not significantly different ($p < 0.05$).

elevated TDS levels [52–54]. Consequently, the results underscore the synergistic efficacy of NTP treatment (30 min) in pollutant removal through the generation of reductive species.

3.4. Total suspended solids (TSS)

This study investigated the removal efficiency of total suspended solids (TSS). As illustrated in Fig. 6, increasing the contact time of NTP in combination with aeration and filtration resulted in a significant reduction of TSS. The aeration group (S_1) exhibited a 16.0 % (21.0 ppm) TSS removal rate, while the maximum removal rates achieved after aeration, filtration, and NTP treatment for 30 min (S_8), 60 min (S_9), and 90 min (S_{10}) were 96.0 % (1.0 ppm), 99.7 % (0.08 ppm), and 99.9 % (0.03 ppm), respectively. The enhanced removal efficiency of NTP can be attributed to the scavenging activity of hydroxyl radicals (OH^-) through their reaction with organic compounds [55]. The experimental TSS results demonstrate that the overall wastewater quality was significantly improved by all plasma treatment conditions [56].

3.5. COD and BOD_5 of wastewater

COD removal efficiencies for S_0 to S_8 were determined to be 13.30 %, 30.98 %, 34.5 %, 42.95 %, 46.47 %, 51.40 %, 56.33 %, 64.08 %, 53.52 %, and 50.00 %, respectively. Similarly, BOD_5 removal efficiencies for the same series were 19.76 %, 39.53 %, 45.34 %, 52.32 %, 54.65 %, 56.97 %, 60.46 %, 67.44 %, 59.30 %, and 51.16 %, respectively. COD and BOD_5 concentrations demonstrated a decreasing trend with increasing plasma treatment time up to 30 min when combined with aeration and filtration, followed by an increase in both parameters after 60 and 90 min of NTP exposure (Fig. 7a and b). The optimal COD and BOD_5 removal efficiencies of 64.08 % and 67.44 %, respectively, were achieved under conditions of 24 h of aeration, adsorbent filtration, and 30 min of NTP treatment (S_8). The substantial reduction in COD (Fig. 7a) indicates efficient decomposition of biodegradable organic substances, suggesting a decrease in oxidized organic compounds. The lower BOD_5 concentration in the S_8 group reflects improved wastewater quality attributed to the removal of organic and inorganic residues through 30 min of NTP treatment compared to aeration alone (S_1) and aeration/filtration treatment (S_2) [57].

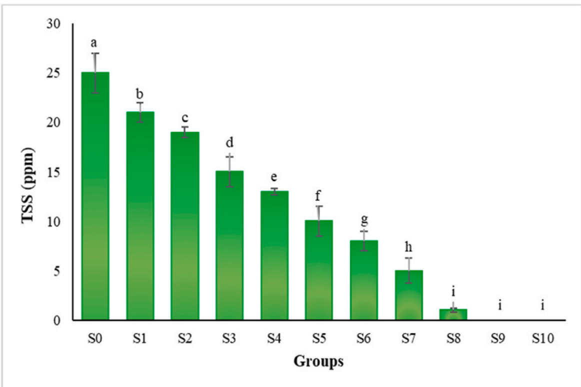

Fig. 6. Duncan tests of means TSS parameter for untreated (**S_0**) and treated wastewater (**S_1-S_{10}**).
* Means followed by the same letter are not significantly different ($p < 0.05$).

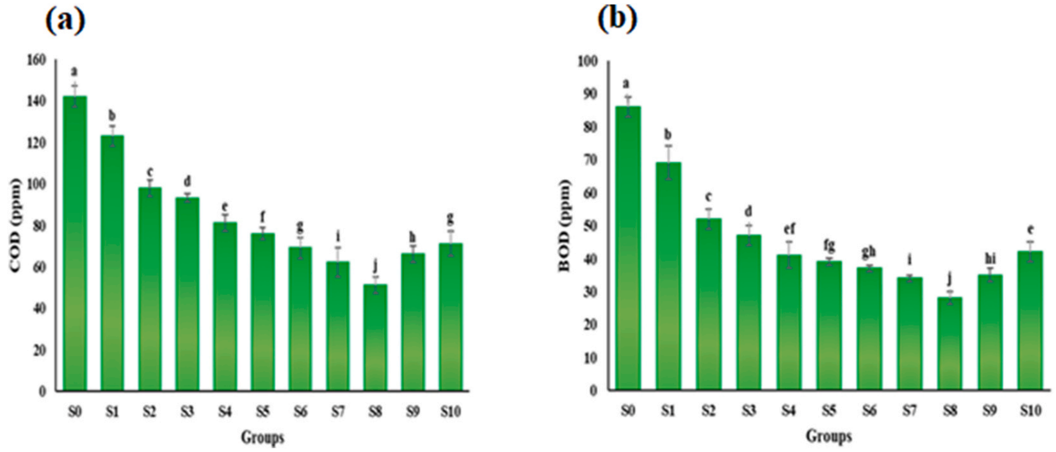

Fig. 7. Duncan tests of means of COD and BOD_5 for all stages of aeration (**S_0**), filtration (**S_1**) and the combination with NTP (**S_2-S_{10}**) with the treatment various times.
* Means followed by the same letter are not significantly different ($p < 0.05$).

3.6. Dissolved oxygen (DO)

DO levels in wastewater can fluctuate due to factors such as ionic activity, atmospheric pressure, aeration, and various chemical and biological reactions [58]. This study investigated the impact of aeration, filtration, and NTP on DO in wastewater samples. As shown in Fig. 8, DO levels exhibited a slight decrease from S_1 to S_8, followed by an increase in S9 and S10. The S8 group, subjected to 30 min of combined NTP treatment, displayed the lowest DO levels compared to S_0 and other treatment groups. This phenomenon can be attributed to oxygen consumption during chemical reactions leading to the formation of ozone (O_3), hydrogen peroxide (H_2O_2), and organic peroxyl radicals (ROO-) in the water, which contribute to pollutant degradation [59,60].

3.7. Electrical conductivity (EC)

A significant reduction in electrical conductivity (EC) was observed in the wastewater sample following aeration (S_1; 6398 ± 192 μS/cm) compared to untreated wastewater (6479 ± 172 μS/cm). Subsequently, the EC decreased by approximately 49.9 % in the S_2 group (3245 ± 71 μS/cm) relative to the untreated sample. A continued decrease in EC was observed from S_3 to S_8 with increasing NTP treatment time. The optimal EC reduction of 61.8 % was achieved at 30 min of plasma treatment (S_8), resulting in a value of 2485 ± 55 μS/cm. However, a slight increase in EC to 2573 ± 57 μS/cm and 2602 ± 49 μS/cm was observed after 60 and 90 min of NTP treatment, respectively (Fig. 9). Wastewater conductivity is primarily influenced by conductive ions, including inorganic materials and dissolved salts such as alkali, chlorides, and carbonates [61]. The observed EC reduction following cold plasma DBD treatment is attributed to the degradation of organic and inorganic contaminants by reactive species, predominantly hydroxyl radicals and ozone. These potent oxidants effectively mineralize contaminants, leading to a decrease in ion concentration. The strong correlation between total dissolved solids (TDS) (Fig. 5) and EC supports the hypothesis that the reduction in TDS is the primary factor driving the observed EC decrease [19].

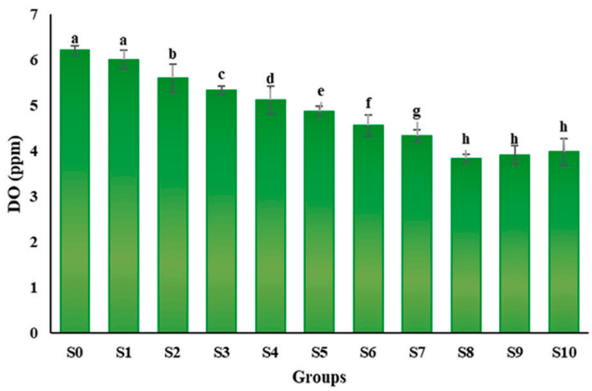

Fig. 8. Duncan tests of means DO parameter with respect to the treatment condition (untreated (**S_0**) and treated wastewater (**S_1-S_{10}**)).
* Means followed by the same letter are not significantly different ($p < 0.05$).

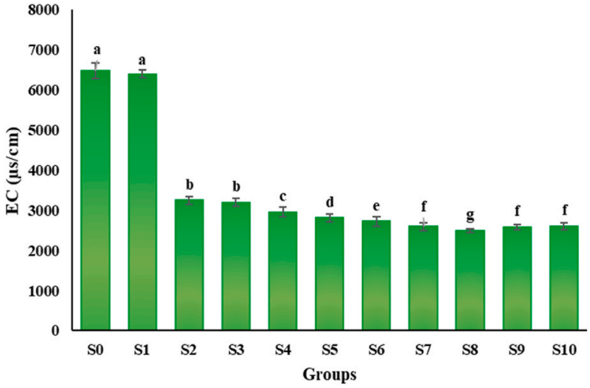

Fig. 9. Duncan tests of means of conductivity for all stages of aeration (**S_0**), filtration (**S_1**) and the combination with NTP (**S_2-S_{10}**) with the treatment various times.
* Means followed by the same letter are not significantly different ($p < 0.05$).

Conversely, at treatment times of 60 and 90 min, an increase in reactive species led to a corresponding increase in wastewater EC [62]. These ions have been reported to penetrate cell wall structures, resulting in microbial inactivation [63].

3.8. pH

pH, a measure of solution acidity or alkalinity, was assessed to characterize the solution's chemical environment. Due to the presence of bicarbonates and carbonates, most wastewaters exhibit alkaline properties (Fig. 10). pH reflects the concentration of hydrogen ions (H^+) in a solution. The untreated aqueous solution (S_0) displayed a neutral pH of 7.3. Subsequent aeration (S_1) and filtration (S_2) treatments shifted the solution towards a basic medium. While NTP treatment combined with aeration and filtration for 5 and 10 min resulted in a slight pH increase to 8.0 and 8.4, respectively, the pH stabilized thereafter. This elevation is attributed to increased ammonia concentrations resulting from the NTP-mediated oxidation of nitrogen-containing organic compounds in the wastewater matrix [64]. The final pH range of 8.3–8.4 aligns with the acceptable pH range for domestic water use (6.5–8.5) [65].

3.9. Color

As illustrated in Fig. 11, the combined application of aeration, adsorbent filtration, and non-thermal plasma significantly reduced color intensity in the wastewater. Aeration initially decreased color from 41 to 38 Pt-Co units, primarily through the biodegradation of chromogenic organic matter by microorganisms [66]. Subsequent treatment with coarse and fine silica-based adsorbents and activated carbon achieved an additional 34.2 % reduction, attributed to the adsorbents' high surface area and affinity for colored compounds [67,68]. The final application of non-thermal plasma resulted in a remarkable reduction to 9 Pt-Co units, likely due to the generation of highly reactive species that effectively mineralized chromophores [69,70].

3.10. Phenolic compounds

As illustrated in Fig. 12, the combined application of aeration, adsorbent filtration, and non-thermal plasma significantly reduced

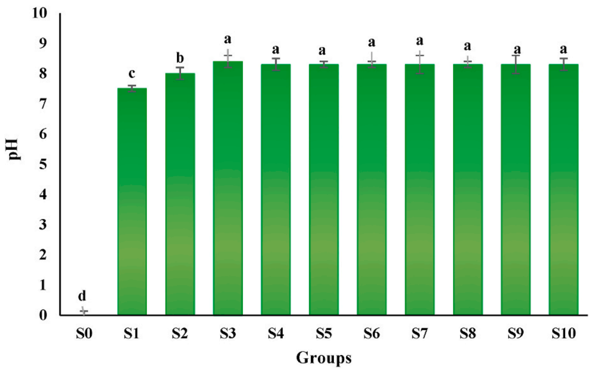

Fig. 10. Duncan tests of means of pH of wastewater with respect to the treatment condition (untreated (**S_0**) and treated wastewater (**S_1-S_{10}**)).
* Means followed by the same letter are not significantly different ($p < 0.05$).

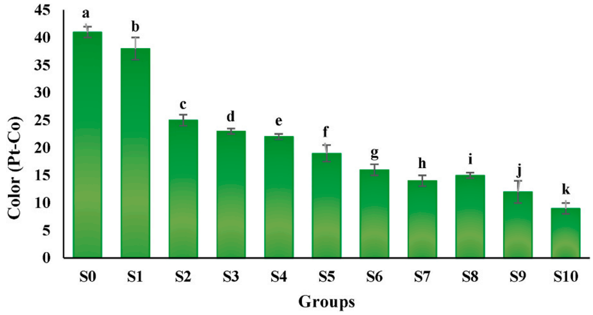

Fig. 11. Duncan tests of means of color for all stages of aeration (S_0), filtration (S_1) and the combination with NTP (S_2-S_{10}) with the treatment various times.
* Means followed by the same letter are not significantly different ($p < 0.05$).

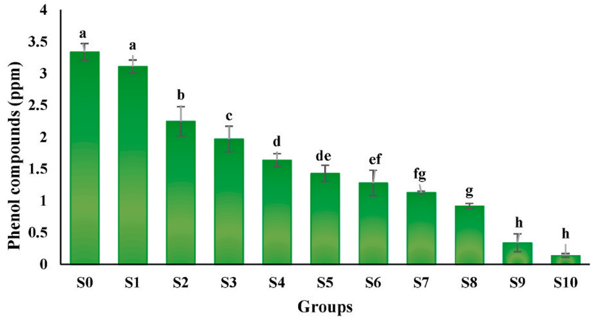

Fig. 12. Duncan tests of means of phenolic compounds for all stages of aeration (S_0), filtration (S_1) and the combination with NTP (S_2-S_{10}) with the treatment various times.
* Means followed by the same letter are not significantly different ($p < 0.05$).

phenol concentrations in the wastewater. Aeration initially decreased phenol levels from 3.34 ppm to 3.11 ppm, primarily attributed to enhanced oxygen transfer and microbial activity [71]. Subsequent treatment with coarse and fine silica-based adsorbents and activated carbon achieved an additional 27.6 % reduction, owing to the adsorbents' high surface area and affinity for phenolic compounds [72,73]. The final application of non-thermal plasma resulted in a remarkable reduction to 0.14 ppm, likely due to the generation of highly reactive species that effectively mineralized residual phenolic compounds [74,75].

To optimize pollutant removal efficiency, Fig. 13 presents a comparative analysis of untreated, pre-treated, and treated solutions. The most significant improvements in removal efficiency or reduction percentage were observed in the 30-min plasma treatment (S_8). This treatment demonstrated the highest efficacy in reducing COD by 64.1 %, BOD_5 by 67.4 %, and TDS by 62.9 %. For a more detailed analysis, Table 4 provides a comparative assessment of all parameters in S_8 relative to the control sample (S_0) to evaluate overall pollutant reduction efficiency and to S_2 to assess the specific impact of plasma treatment on parameter reduction.

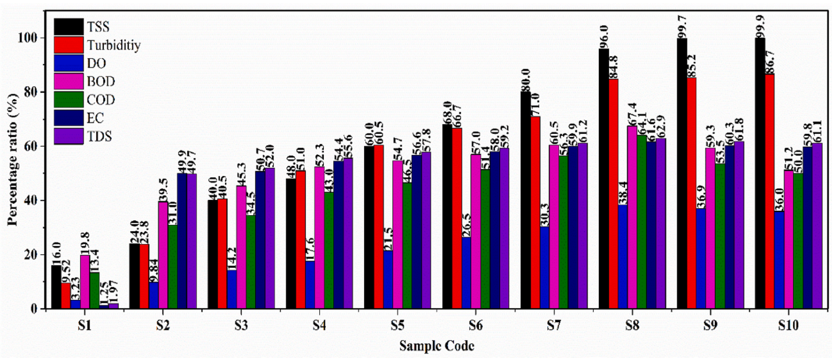

Fig. 13. The percentage of removal efficiency to remove pollutants.

Table 4
The effectiveness of plasma utilization in treating the S_8 group as opposed to the S_0 and S_2.

Parameter	Absolute percentage S8 compared to S0	Absolute percentage S8 compared to S2	Reduction percentage compared to S0 (Efficiency)	Reduction percentage compared to S2 (Efficiency)
TDS	37.14	73.85	62.86	26.15
EC	38.35	76.58	61.65	23.43
COD	35.91	52.04	64.08	47.96
BOD_5	32.56	53.85	67.446	46.15
DO	61.62	68.34	38.39	31.66
Turbidity	15.24	20	84.769	80
TSS	4	5.27	96	94.74
pH	113.70	103.75	−13.70	−4
color	36.58	60	63.41	40
phenolic compounds	27.54	40.89	72.45	59.11

3.11. Comparison of specific energy consumption (SEC), operational costs and COD removal efficiency in some advanced oxidation methods

A 2017 study by Rodrigo et al. compared two advanced oxidation processes (AOPs) - ultrasound combined with ozone ($US + O_3$) and solar/ozone (with and without hydrogen peroxide) - in terms of specific energy consumption (SEC) and operational costs. Table 5 presents the SEC and operational costs for each treatment evaluated. The results indicate that solar energy significantly reduces energy consumption, and the lowest operational cost per cubic meter is achieved through the simultaneous application of solar energy and ozone. However, the COD removal efficiency in this configuration was deemed unsatisfactory.

Plasma technology offers a distinct advantage over other advanced oxidation methods due to its simultaneous generation of ozone, hydrogen peroxide, and UV radiation, significantly enhancing hydroxyl radical production in treated water, wastewater, and leachate. Consequently, plasma treatment exhibits superior pollutant removal rates compared to alternative methods. Table 6 presents the specific energy consumption (SEC) and operating costs associated with the plasma treatment process over a 30-min duration. The results indicate that plasma can achieve a 64.08 % reduction in COD following pretreatment, with an estimated operating cost of $1.2/$m^3$, significantly lower than those reported in Table 5. While the plasma-based process demonstrates slightly higher SEC compared to Solar/O_3 and Solar/O_3/H_2O_2 processes, it surpasses $US + O_3$ and US + O3/H_2O_2 processes in terms of energy efficiency. A key strength of plasma treatment is its exceptional COD reduction performance, approximately double that of other methods, highlighting its superior ability to remove organic pollutants. Furthermore, the plasma-based approach offers a significant cost advantage over all other processes evaluated.

4. Discussion

Rapid industrialization and urbanization in developing countries have exacerbated water pollution, leading to increased disease prevalence and decreased life expectancy compared to developed nations [77]. The World Health Organization (WHO) reports that contaminated drinking water is responsible for 80 % of diseases and 50 % of child deaths [78]. While various physical, chemical, and biological treatments have been proposed for water decontamination, advanced oxidation processes (AOPs) have emerged as effective methods for degrading organic pollutants. AOPs generate reactive oxygen species (ROS), including hydroxyl radicals (OH^-), singlet oxygen ($^1O_2$), superoxide radicals (O_2^-), and molecular oxidants such as ozone (O_3) and hydrogen peroxide (H_2O_2), through interactions with water and oxygen [79]. The highly reactive hydroxyl radical (•OH) is particularly effective in oxidizing organic compounds, breaking them down into smaller intermediates that are ultimately mineralized to carbon dioxide (CO_2) and water, without generating solid secondary pollutants. Several AOPs, including Fenton, photo-Fenton, photocatalysis, ozonation, electrochemical oxidation, and sonolysis, have been investigated for pollutant degradation [80].

Non-thermal plasma (NTP), an innovative AOPs-based technology, has been widely employed to degrade harmful organic compounds in aqueous solutions through high-voltage discharge without generating secondary pollution [81]. A key advantage of NTP is its ability to operate without external additives or adjustments to temperature and pH, requiring minimal treatment time. As previously discussed, NTP, as an effective wastewater treatment alternative, generates neutrons, photons, UV–visible light, positive and negative ions, high-energy electrons, and electric fields [82]. The initial formation of radicals such as O_3 and O• occurs in the gas phase, followed by rapid transfer to the liquid phase [83]. While free electron discharge in the aqueous phase contributes to plasma formation alongside other complex chemical processes, it is crucial to recognize that this is not the sole outcome. Free electron discharge can also induce the formation of a diverse range of free radicals and molecular oxidants through water decomposition, including H^+, OH^-, O^-, O_2^-, O_3, and H_2O_2 [84]. Notably, hydroxyl radicals possess the ability to decompose target contaminants into less toxic or non-toxic compounds [85]. The generation pathways of short-lived radicals produced by NTP are outlined in Equations (1)–(6) [86,87].

$$H_2O + e^- \rightarrow OH^\cdot + H^\cdot \text{ and } H_2 + O^\cdot \tag{1}$$

$$H^\cdot + H^\cdot \rightarrow H_2 \tag{2}$$

Table 5
Specific energy consumption (SEC) and operational costs for each treatment studied [76].

	SEC(Wh/mgCOD/L)	Operational cost			Pollutants elimination (%)	
		Reagent or oxidant	Energy	Total (USD/m^3)	Color	COD
US + O_3	0.127	0.0	16.9	16.9	26.4	14.7
US + O_3/H_2O_2	0.048	20.2	16.9	37.1	41.5	33.8
Solar/O_3	0.013	0.0	3.5	3.5	45.1	27.9
Solar/O_3/H_2O_2	0.011	26.9	3.5	30.5	59.7	34.4

Table 6
Specific energy consumption (SEC) and operating costs for the plasma treatment method over a 30-min period.

Treatment	SEC (Wh/mgCOD/L)	Operational cost			Pollutants elimination (%)	
		Reagent or oxidant	Energy	Total (USD/m^3)	Color	COD
After 30 min Plasma treatment (UV + O_3+H_2O_2)	0.043	–	4	1.2	63.41	64.08

$$OH^{\cdot} + O^{\cdot} \rightarrow O_2 + H^{\cdot} \tag{3}$$

$$OH^{\cdot} + OH^{\cdot} \rightarrow H_2O + O^{\cdot} \tag{4}$$

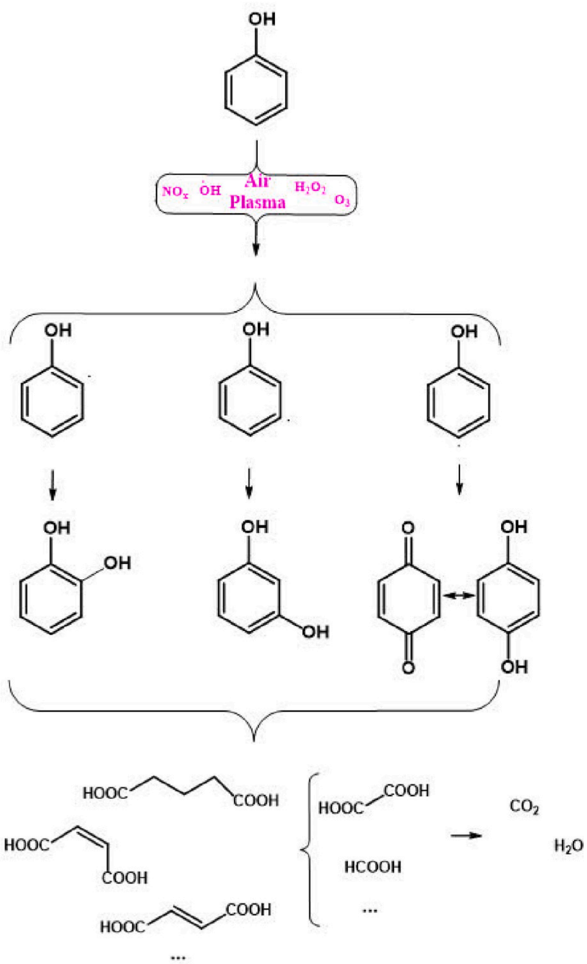

Fig. 14. Mechanism for phenol degradation in a non-thermal plasma system.

$$OH^{\cdot} + OH^{\cdot} \rightarrow H_2O_2 \tag{5}$$

$$OH^{\cdot} + H_2O_2 \rightarrow HO_2^{\cdot} + H_2O \tag{6}$$

Previous experimental studies have demonstrated that the hydroxyl and hydroperoxyl radicals generated during water dissociation by NTP can initiate the oxidation of linear hydrocarbons, producing alkyl and alkoxy radicals, which subsequently form gaseous products and carboxylic acids [88]. Aromatic hydrocarbons and phenol have been shown to degrade into formic acid, maleic acid, and oxalic acid, ultimately yielding CO_2 and H_2O [88]. The removal mechanisms for As^{3+}, Cr^{6+}, and organic pollutants involve the electrophilic attack of plasma-generated active species, such as ozone, on electron-rich sites within aromatic and olefinic structures [59, 89]. Additionally, the longer-lived aqueous oxidants, O_3 and H_2O_2, can undergo photolytic decomposition to generate OH^- radicals, further contributing to pollutant degradation [90,91], as outlined in Equations (7)–(9).

$$O_3 + h\nu \rightarrow O + O_2 \tag{7}$$

$$O + H_2O \rightarrow 2OH^{\cdot} \tag{8}$$

$$H_2O_2 + h\nu \rightarrow OH^{\cdot} + OH^{\cdot} \tag{9}$$

Furthermore, electrons generated by NTP can induce water decomposition, contributing to pollutant removal, as depicted in Equation (8) [91].

$$H_2O + e^- \rightarrow OH^{\cdot} + H^{\cdot} + e^- \tag{8}$$

Collectively, OH^-, O_3, H_2O_2, H_2O^+, and $HO_2^{\cdot}$ radicals contribute to pollutant removal within the gas-liquid interface [92,93]. The observed decrease in TDS levels following NTP treatment corresponded to a reduction in EC values, indicating a decline in cationic and anionic species within the wastewater. This phenomenon can be attributed to the interaction between these ions and the dominant oxidizing species, OH^- and O_3, generated by NTP [94]. The combined application of aeration, filtration, and 30 min of plasma treatment effectively decomposed micropollutants within a shorter timeframe, resulting in significant reductions in COD (64.1 %), BOD_5 (67.4 %), TDS (62.9 %), and color (63.4 %) compared to the control sample (S_0).

Consistent with the findings of Du et al. [95], our investigation into phenol compound reduction suggests that active species generated within the plasma, including hydroxyl radicals, hydrogen peroxide, and ozone, play a pivotal role in organic pollutant degradation. However, while Du and colleagues focused on soil-based systems, our study explored the application of cold plasma within the context of industrial wastewater treatment. Hydroxyl radicals, characterized by their strong electrophilic properties, primarily target pollutant active sites, initiating deep oxidation processes. Conversely, hydrogen peroxide and ozone degrade phenol through direct and indirect oxidation mechanisms. Fig. 14 illustrates the proposed mechanism for phenol degradation within a non-thermal plasma system. Active species preferentially attack the aromatic ring of phenol at the ortho, para, and meta positions, triggering a series of reactions leading to the formation of hydroxylated intermediates and quinones. Subsequently, ring opening and deep oxidation processes occur, culminating in the formation of carbon dioxide and water as final mineralization products.

5. Conclusion

Plasma technology has emerged as a promising approach for wastewater treatment due to its capacity to generate high densities of reactive oxygen and nitrogen species (ROS and RNS) via high-energy electron interactions, enabling the removal of a wide range of contaminants. This study investigated the combined effects of aeration, filtration, and non-thermal plasma (NTP) treatment over varying time intervals on turbidity, TDS, TSS, COD, BOD_5, DO, EC, and pH. Results demonstrated that the integrated aeration/filtration/NTP system significantly reduced TDS by 62.86 %, EC by 61.65 %, turbidity by 84.76 %, TSS by 96 %, COD by 64.1 %, BOD_5 by 67.4 %, color by 63.4 %, and phenol by 72.4 % (S_0-S_8) compared to the untreated control (S0). These findings underscore the substantial contribution of NTP to pollutant degradation through the generation of reactive species. The optimal treatment configuration was achieved with 30 min of plasma treatment combined with aeration and filtration (S_8). This study concludes that non-thermal plasma technology, when integrated with aerobic, adsorbent, and filtration processes, presents a highly competitive and effective approach for wastewater treatment.

A comparison of specific energy consumption, operational costs, and COD removal efficiency among various advanced oxidation processes and plasma revealed that plasma treatment effectively reduced COD by nearly 65 %, outperforming other methods. Despite slightly higher energy consumption, plasma offers significantly lower operating costs and superior COD removal efficiency, making it a promising technology for wastewater treatment.

Non-thermal plasma (NTP) technology has emerged as a promising approach for wastewater remediation due to its capacity to generate high concentrations of reactive species and its potential for minimal secondary contamination. NTP's versatility, which allows for configuration adjustments to inhibit microbial activity, remove color and odor, and degrade harmful pollutants more effectively than conventional or advanced oxidation processes (AOPs), further underscores its potential. Additionally, NTP can be seamlessly integrated with other remediation techniques, serving as either a preliminary or final treatment for persistent wastewater contaminants.

While the potential benefits of integrating NTP technology into wastewater treatment are substantial, several challenges must be addressed to optimize its effectiveness. The complex interplay of reactive species generated within the air plasma requires comprehensive characterization to elucidate their precise roles in pollutant degradation. Moreover, the inefficient energy utilization of NTP reactors, where a significant portion of energy is dissipated rather than contributing to reactive species generation, necessitates optimization of reactor configurations to improve energy efficiency and reduce costs. Although extensively studied in laboratory settings, the transition of NTP technology from bench-scale research to commercial-scale applications remains limited by the scarcity of pilot and industrial-scale implementations.

CRediT authorship contribution statement

Mahdiyeh Bakhtiyari-Ramezani: Supervision, Resources, Project administration, Methodology, Investigation, Funding acquisition, Conceptualization. **Narges Ziveh:** Writing – original draft, Visualization, Formal analysis, Data curation. **Navid Ghaemi:** Writing – review & editing, Validation.

Ethics approval statement

The research presented does not involve any animal or human study.

Data availability statement

Data are available within the article. The data associated with this study have not been deposited into a publicly available repository, it will be made available upon request.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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