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# Removal of 2-Chlorophenol by Ozonation Under Strongly Alkaline Conditions in a Jet Loop Reactor

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**Abstract** This study aimed to investigate the removal of 2-chlorophenol (2CP) using ozonation in a jet loop reactor under high pH (strongly alkaline conditions). The effects of initial 2CP concentration and ozone dosage on the removal of 2CP, chemical oxygen demand (COD), and total organic carbon (TOC) were determined. As the initial 2CP concentration increased, the removal efficiencies of 2CP, COD, and TOC decreased, whereas an increase in ozone dosage resulted in higher removal efficiencies. The initial concentration of 100 mg/L of 2CP was completely removed within approximately 12.5 min at an ozone dosage of 13.1 $gO_3$/h, while after 180 min of ozonation, the COD and TOC removal efficiencies were 97% and 60%, respectively. The removal of 2CP and COD followed pseudo-first-order kinetics in a two-stage process. When examining the kinetics of 2CP removal, the rate constants observed in the second-stage were higher than those observed in the first-stage. In the case of COD removal, unlike in the 2CP, the rate constant values observed in the first-stage were higher than those observed in the second-stage. For both 2CP and COD removal, the rate constant values in both stages exponentially decreased with an increase in the initial 2CP concentration, whereas they linearly increased with an increase in ozone dosage.

**Keywords** Jet loop reactor · Ozonation · 2-chlorophenol · Chemical oxygen demand · Total organic carbon

**Abbreviations**

| | |
|---|---|
| JLR | Jet loop reactor |
| 2CP | 2-Chlorophenol |
| AOSC | Average oxidation state of carbon |
| COD | Chemical oxygen demand |
| PFO | Pseudo-first order |
| TOC | Total organic carbon |

## 1 Introduction

Global water demand is steadily increasing owing to population growth and industrialization. Increasing water demand leads to the depletion and pollution of existing water resources. The discharge

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of industrial wastewater, which has a significantly higher pollutant load than domestic wastewater, into the receiving environment without adequate treatment exacerbates the problem of water pollution (Singh et al., 2022; Xie et al., 2021). Phenols and phenol derivatives are widely used in daily life and are present in significant amounts in industrial wastewater (Igbinosa et al., 2013). Chlorophenols, which are derivatives of phenols, are commonly recognized for their toxicity and persistent structures, and are considered priority pollutants. In the environment, they possess a strong odor and exhibit poor biological degradation (Carucci et al., 2010; Garba et al., 2019; Prashanthakumar et al., 2018). Chlorophenols are a group of organochlorine compounds derived from phenols and characterized by one or more covalently bonded chlorine atoms. They can be divided into five groups: monochlorophenols (2-chlorophenol (2CP), 3-chlorophenol, and 4-chlorophenol), dichlorophenols, trichlorophenols, tetrachlorophenol, and pentachlorophenol (Fan et al., 2015). The most significant sources of wastewater containing chlorophenols are pesticides, paint, solvents, pharmaceuticals, wood-preserving chemicals, and paper and pulp industries (Gao & Wang, 2007). Studies have shown that even at parts per billion levels, chlorophenol compounds can cause odors in water (Lei et al., 2021). Long-term intake of water contaminated with chlorophenols can lead to dizziness, anemia, and other symptoms, significantly affecting the human nervous and respiratory systems and causing liver damage (Garba et al., 2019; Lei et al., 2021). The World Health Organization and International Agency for Research on Cancer have identified many chlorophenols as potential human carcinogens (Igbinosa et al., 2013). Among these compounds, 2CP has been listed as a priority pollutant in the decision 2455/2002/EC of the European Parliament and by the United States Environmental Protection Agency under the Clean Water Act (Lei et al., 2021; Liu et al., 2018). 2CP is an odorous liquid commonly used in the synthesis of pharmaceuticals, disinfectants, pesticides, and dyes (Mohd Zaki et al., 2023; Sharma et al., 2019). It irritates the skin upon contact and has serious health effects when ingested or inhaled. Moreover, high levels of exposure can even lead to fatalities (Elsalamony & Mahmoud, 2017). To reduce the adverse effects of 2CP on human health and the environment, it needs to be removed from wastewater before discharge. Therefore, an environmentally friendly and practical treatment method is required for the efficient removal of 2CP. Various treatment methods such as adsorption (Huong et al., 2016; Koubaissy et al., 2011), aerobic granulation (Khan et al., 2011), electro-catalytic degradation (Liu et al., 2018), catalytic ozonation (Ni & Chen, 2001), photocatalytic degradation (Elsalamony & Mahmoud, 2017), and ozonation (Sung & Huang, 2007) have been applied for the removal of 2CP.

Ozone can be generated from pretreated air or oxygen through UV irradiation or electrical discharge (Eliasson & Kogelschatz, 1991; Pera-Titus et al., 2004). Owing to its high redox potential (2.07 V), the use of ozone in water and wastewater treatment is increasing (Puspita et al., 2015). Since ozone is a powerful oxidizer, it reacts with both organic and inorganic substances, breaking down organic compounds into smaller substances (such as organic acids) or into $CO_2$ and $H_2O$ (Dai et al., 2015; Jin et al., 2012). Ozonation is used for the removal of color, odor, microorganisms, pesticides, and toxic and resistant (refractory) pollutants (Baghirzade et al., 2021; Jakubowski et al., 2003; Kasiri et al., 2013; Lim et al., 2022; Ren et al., 2012; Wei et al., 2017). Moreover, it is a potential pretreatment process to convert refractory compounds present in wastewater into substances that can be more readily removed by conventional methods (such as biological treatment) (Boczkaj & Fernandes, 2017; Pera-Titus et al., 2004). The two ways that ozone and organic materials react are known as direct oxidation with molecular ozone and indirect oxidation with hydroxyl radicals (Canton et al., 2003; Cheng et al., 2018; Wang & Chen, 2020). Hydroxyl radicals are generated through a series of radical chain reactions involving initiation, propagation, and termination (Gottschalk et al., 2010).

The conversion of ozone into hydroxyl radicals is catalyzed by hydroxyl ions in an alkaline environment (Qu et al., 2015). Increasing the pH in the ozonation system results in higher production of hydroxyl radicals due to the following reactions (Eq. 1–3) (Alaton et al., 2002; Wu et al., 2008; Yildirim et al. 2011). Indirect reactions with hydroxyl radicals are faster than direct reactions with molecular ozone (Canton et al., 2003). Hydroxyl radicals are powerful oxidizing agents that oxidize organic substances faster than ozone itself (Wu et al., 2008).

$$O_3 + OH^- \rightarrow O_3^{\bullet -} + OH^{\bullet} \quad (1)$$

$$O_3^{\bullet -} \rightarrow O_2 + O^{\bullet -} \quad (2)$$

$$O^{\bullet -} + H^+ \rightarrow OH^{\bullet} \quad (3)$$

In addition to its high production cost, ozone should be used as generated due to its relatively low solubility and low stability in water (Al-Abduly et al., 2014; Boczkaj & Fernandes, 2017; Mehrjouei et al., 2015). Therefore, various types of ozone contactors, such as bubble columns, stirred tanks, and packed columns, have been used to enhance the mass transfer of gaseous ozone into water (Farines et al., 2003; Mizuno & Tsuno, 2010; Tiwari & Bose, 2007). However, these types of reactors have various disadvantages that negatively affect the ozone performance and introduce complexities in their application and maintenance (Mahyar et al., 2017). Jet loop reactors (JLRs) appear to be dependable alternatives because they prevent these issues. It is suitable for ozonation because it provides good interaction between the gas and liquid phases. Compared with conventional reactors, JLRs offer several advantages, low capital and operating costs, including simple construction, increased circulation with the same energy input, high heat and mass transfer coefficients, the lack of moving parts inside the reactor, excellent gas dispersion, the ability to maintain highly homogeneous concentration and temperature profiles, and ease of scaling up from pilot plant to industrial scale (Cengiz et al., 2024; Chu et al., 2017; Değermenci & Yildiz, 2021; Deshpande et al., 2009; Jianping et al., 2002; Maly et al., 2022; Warmeling et al., 2016).

In this study, a jet loop reactor (JLR) was used to explore the elimination of 2CP by ozonation under alkaline conditions. The effects of different ozone dosages and initial 2CP concentrations on 2CP, chemical oxygen demand (COD), and total organic carbon (TOC) removal were examined. The first-order reaction rate constants for 2CP and COD removal were calculated using the experimental data.

## 2 Materials and Methods

### 2.1 Chemicals

All chemicals and reagents used in the analyses were of analytical grade or higher and used without any additional purification. The 2CP required for preparing aqueous solutions containing 2CP (≥99%) was purchased from Sigma, and the NaOH (99%) required to adjust the pH of the solutions was purchased from Merck.

### 2.2 Experimental Setup and Procedure

The JLR consisted of two concentric cylindrical tubes made of stainless steel. The reactor (outer) and draft tube (inner) had inner diameters of 10 cm and 4 cm, respectively. Both the cylindrical tubes had a wall thickness of 2 mm. The gas collection tank (a wider partition located on top of the reactor) had an inner diameter of 20 cm. The total height was 110 cm when the reactor and the gas collection tank were combined. The draft tube, positioned 35 cm below the top of the gas collection tank and 10 cm above the base of the reactor (impact plate), was centered within the reactor using a support. The gas, delivered through a separate line, passes through a nozzle and is sprayed into the draft tube by a circulation-pump-driven liquid. The two-phase flow advancing inside the draft tube moves downward, collides with the impact plate directly below the reactor, and rises between the reactor wall and the draft tube. A portion of the liquid containing gas bubbles, reaching the level of the draft tube, is drawn back into the draft tube and continues to circulate owing to the pressure difference. The nozzle consists of two concentric cylindrical tubes. The outer cylinder, through which the liquid flows, has an inner diameter of 15.7 mm, whereas the inner cylinder, through which the gas flows, has an inner diameter of 4 mm and a wall thickness of 1.2 mm. In the experiments, the liquid circulation flow rate was measured using an analog inline flowmeter and set to 60 L/min using a manually controlled valve. The reactor temperature was maintained at 20 °C using a coil (heat exchanger) placed inside the gas collection chamber through which the tap water passed. The volume of the solution filled into the reactor is 18 L.

With the aid of an ozone gas generator, ozone gas was produced using either dry air or pure oxygen

(COM-AD-08, Anseros). The generated ozone gas was supplied to the reactors at a flow rate of 250 L/h using a flowmeter located in front of the ozone generator. By altering the ozone generator's working capacity, different ozone dosages were adjusted, while maintaining a constant gas flow rate (250 L/h). A schematic view of the experimental system in which the ozonation experiments were conducted is provided in Fig. 1.

## 2.3 Analytical Methods

2CP and COD analyses were performed using a spectrophotometer (SpectroFlex 6600, WTW) according to Standard Methods (APHA 2023). 2CP analysis was conducted using the 4-aminoantipyrine method (5530D) at 500 nm, while COD analysis was performed using the closed reflux method (5220D) at 600 nm. TOC analyses were carried out using the high-temperature combustion method (5310B) in a Tekmar-Dorhman Apollo 9000 instrument according to Standard Methods (APHA 2023). All analyses were performed in triplicate, and the average values are presented in the figures. The 2CP, COD, and TOC removal efficiencies were calculated using the following equation:

$$\text{Removal efficiency } (\%) = \frac{C_0 - C_t}{C_0} \times 100 \quad (4)$$

Non-dispersive ultraviolet absorption technology was used to gauge the amount of ozone in the gas phase (GM-6000-OEM, Anseros). The pH was measured and controlled using a pH controller (Alpha pH500, Eutech Instruments).

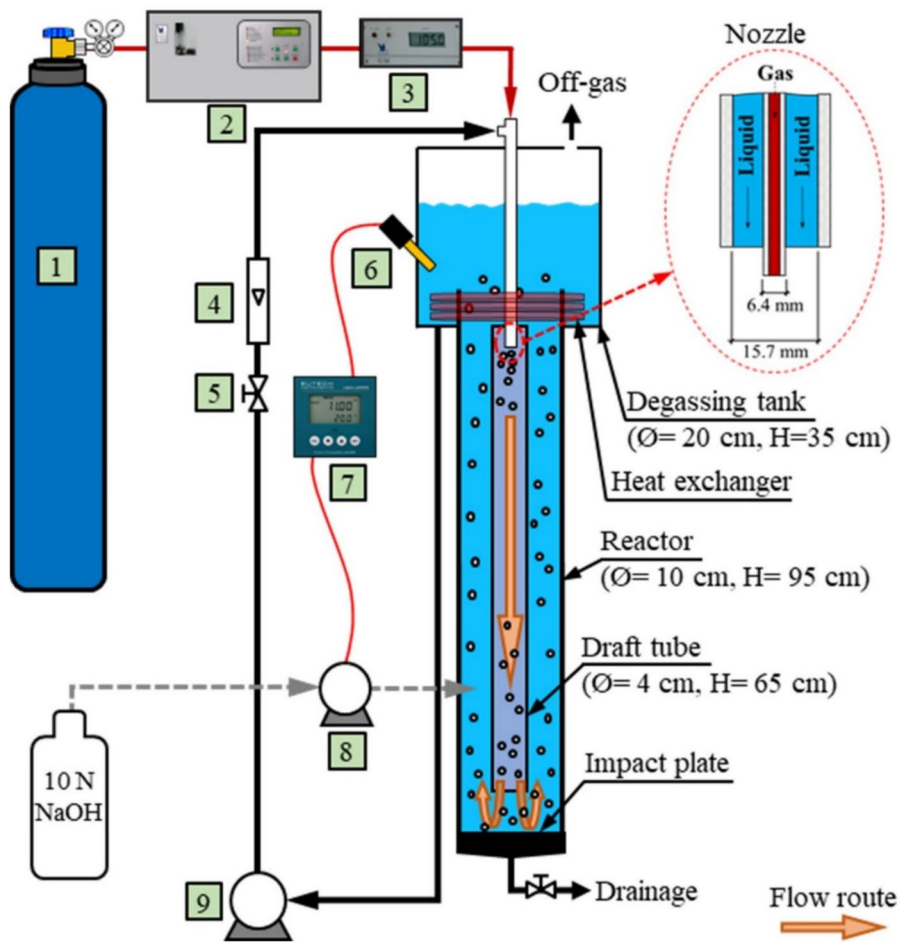


**Fig. 1** The JLR system in which the experiments were conducted: (1) oxygen/dry air cylinder, (2) ozone generator, (3) ozone analyzer, (4) flowmeter, (5) valve, (6) pH probe, (7) pH controller, (8) peristaltic pump, and (9) centrifuge pump

## 3 Results and Discussion

### 3.1 Effect of Initial 2CP Concentration

In the literature, the effects of different 2CP concentrations on adsorption, biodegradation, and other advanced oxidation processes have been investigated (Moradnia et al., 2022; Moussavi et al., 2014; Poulopoulos et al., 2008; Sharma et al., 2023). When evaluating the effect of the initial 2CP concentration, the concentration ranges reported in the literature were taken into consideration. To investigate this effect, ozonation experiments were conducted in the JLR using solution with different initial 2CP concentrations (100, 200, 300, and 500 mg/L), (Fig. 2). At high pH levels, the formation of hydroxyl radicals increases, leading to a more rapid and efficient degradation of organic pollutants (Barlak et al., 2020). Therefore, a pH of 11 was selected to ensure optimal oxidation conditions. During the experiments, the temperature, the pH value of the solution, and the ozone dosage were kept constant at 20 °C, 11, and 4.38 $gO_3$/h respectively. The changes in the removal of 2CP, COD, and TOC were monitored over time (Fig. 2).

As shown in Fig. 2(a), after a 30-min ozone treatment, the complete removal of 2CP was achieved at an initial concentration of 100 mg/L, whereas the removal percentages at initial concentrations of 200, 300, and 500 mg/L were 81%, 62%, and 36%, respectively. Owing to the constant amount of oxidant available for increasing the pollutant concentrations, the

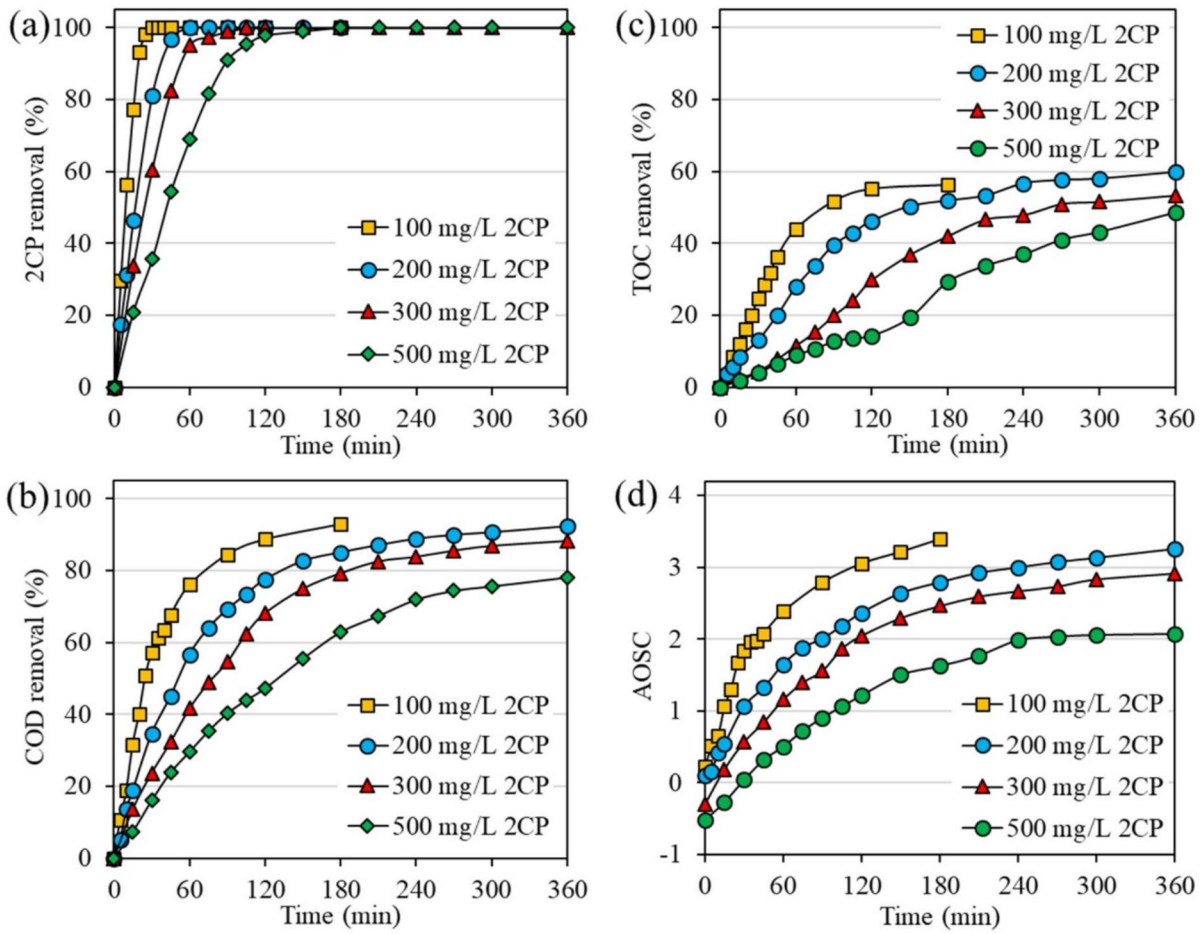


**Fig. 2** 2CP removal (**a**), COD removal (**b**), TOC removal (**c**), and AOSC factor evolution (**d**) at different initial 2CP concentrations (Ozone dosage = 4.38 $gO_3$/h, pH = 11, T = 20 °C)

ozonation time required for the complete degradation of 2CP increased as the initial 2CP concentration increased. Complete degradation (mineralization and conversion to other intermediate products) of 2CP required approximately 30 min at 100 mg/L, 60 min at 200 mg/L, 105 min at 300 mg/L, and 180 min at 500 mg/L. It can be stated that there is a linear relationship between the increasing 2CP concentration and the time required for its complete degradation.

Figure 2(b) shows the COD removal at different initial 2CP concentrations. The measured COD values for the initial 2CP concentrations yielded a ratio of $1.56 \pm 0.04$ mg COD/mg 2CP. This value was close to the theoretically calculated value (1.62 mg COD/mg 2CP). Although 2CP was completely consumed by ozonation at high pH in the JLR, the breakdown of COD and TOC continued. The oxidation products causing COD and TOC are intermediate products formed from the degradation of 2CP, which can only originate from 2CP. Some of these intermediate products formed from the oxidation of 2CP mentioned in the literature include chlorohydroquinone, 3-chlorocatechol, muconic acid, maleic acid, tartaric acid, dihydroxymaleic acid, and oxalic acid (Hsu et al., 2007). These intermediate products that are created during the breakdown of 2CP have a lesser degradability than 2CP itself (Hsu et al., 2007). Therefore, the removal of 2CP occurred at a faster rate than the removal of COD. After 180 min of ozone treatment, the COD removal percentages were measured as 93%, 85%, 79%, and 63% for 100, 200, 300, and 500 mg/L of 2CP, respectively.

Figure 2(c) shows the TOC removal efficiencies at different initial 2CP concentrations. The measured TOC values for the initial 2CP concentrations yielded ratio of $0.57 \pm 0.03$ mg TOC/mg 2CP. This value was close to the theoretically calculated value (0.56 mg TOC/mg 2CP). Similar to COD removal, although 2CP is completely consumed, it causes TOC reduction owing to the oxidation of intermediate products formed. After 180 min, the TOC removal percentages were 56, 52, 42, and 29% for 100, 200, 300, and 500 mg/L of 2CP, respectively. The ozonation of oxalic acid, an intermediate product, was found to have an inclination to accumulate in the solution at high pH levels, according to a prior study (Hsu et al., 2007), and it is not easily mineralized (Hu et al., 2016). From the results obtained, an increase in the initial 2CP concentration led to a decrease in the rates of 2CP, COD, and TOC removal. For higher initial 2CP concentrations, the desired efficiencies of 2CP, COD, and TOC removal could be achieved by increasing the ozonation time while keeping other parameters influencing the ozone gas concentration. The removal rate hierarchy for different initial 2CP concentrations was determined to be 2CP > COD > TOC.

The changes in the average oxidation state of carbon (AOSC) for different initial 2CP concentrations are shown in Fig. 2(d). The AOSC value of the solution was calculated using Eq. 5 (Espejo et al., 2014; Garg & Mishra, 2010).

$$AOSC = 4 - 1.5(COD/TOC) \tag{5}$$

The AOSC values ranged from −4 (representing methane) to +4 (representing carbon dioxide). High AOSC values reflect a high oxidation state of the organic molecules, whereas low AOSC values indicate incomplete oxidation (Asgari et al., 2020). However, it should be noted that in the TOC analysis, the $CO_2$ present in the solution was removed and not measured, and AOSC does not fully account for mineralized compounds (Pocostales et al., 2011). The calculated AOSC values for initial 2CP concentrations of 100, 200, 300, and 500 mg/L are 0.22, 0.10, −0.30, and −0.52, respectively. It has been observed that the AOSC values increase over time as a result of the oxidation of 2CP. After 180 min of ozonation, the AOSC values increased to 3.39, 2.61, 2.55, and 1.63 for 100, 200, 300, and 500 mg/L, respectively. Thus, a higher oxidation state was observed at the lowest 2CP concentration. This can be attributed to the fact that the given ozone dosage was approximately the same for all 2CP concentrations, resulting in a higher ozone dose per unit of pollutant for the lower 2CP concentration.

To determine the reaction rate constants for the removal of 2CP through ozone treatment in the JLR, kinetic calculations were performed assuming a pseudo-first-order (PFO) rate of decomposition. Similarly, PFO kinetic model has been used for the removal of phenol, 2-nitrophenol, and parabens from aqueous solutions through ozone treatment (Barlak et al., 2020; Chen et al., 2003; Tay et al., 2010). Therefore, time versus ln(2CP) and ln(COD) graphs were plotted. Figure 3(a) and Fig. 3(b) demonstrate that the degradation of both 2CP and COD

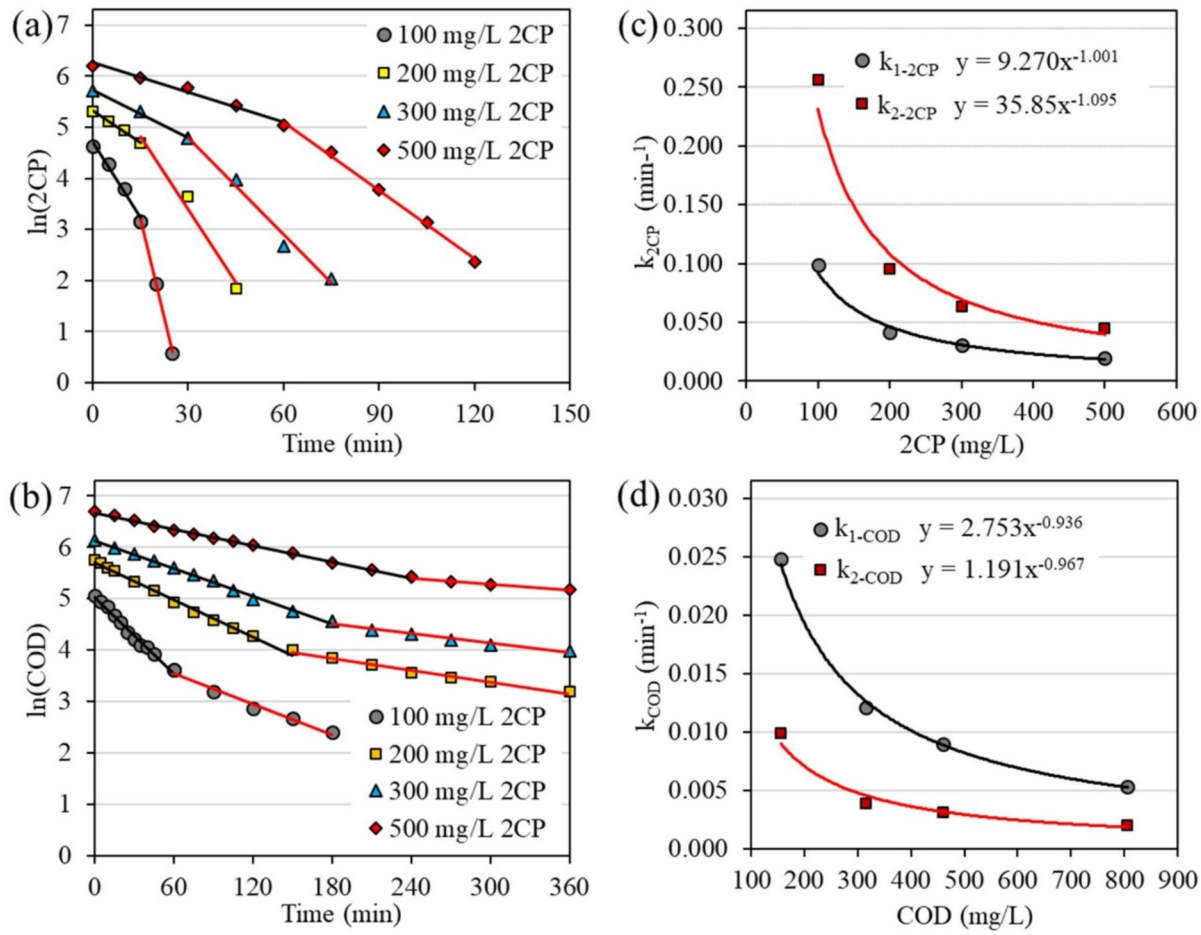


**Fig. 3** Under different initial 2CP concentrations, the variations of ln(2CP) (**a**), ln(COD) (**b**), the change in PFO constant for 2CP (**c**), and PFO constant for COD (**d**) (Ozon dosage = 4.38 $gO_3$/h, pH = 11, T = 20 °C)

occurred in two-stage. The results indicated that the rate constant observed in the first-stage of 2CP degradation ($k_{1\text{-}2CP}$) was lower than the rate constant ($k_{2\text{-}2CP}$) observed in the second-stage (Fig. 3(c)). For a 2CP concentration of 100 mg/L, $k_{1\text{-}2CP}$ is 0.0984 min$^{-1}$ while $k_{2\text{-}2CP}$ is 0.2565 min$^{-1}$. In the literature review, no studies were found that demonstrate the degradation of 2CP through a two-stage PFO kinetic model. However, in a study focusing on the ozonation of 2-nitrophenol, the first-stage and second-stage rate constants were reported as 0.4246 and 0.7512 min$^{-1}$, respectively, under conditions of solution pH 9 and ozone dosage of 6 $gO_3$/h. Similarly, in a study on the removal of parabens, two-stage reaction kinetics were reported and the second-stage rate constant was higher than the first-stage rate constant (Tay et al., 2010). When the results obtained in our study are compared with these studies in the literature, they show a similar two-stage degradation kinetics (Table 1). In addition, it was observed that the ratio of the second-stage rate constant to the first-stage rate constant

**Table 1** Comparison of first- and second-stage rate constants for the ozonation of different pollutant

| Pollutant | $k_{1\text{-stage}}$ (min$^{-1}$) | $k_{2\text{-stage}}$ (min$^{-1}$) | References |
|---|---|---|---|
| Parabens | 0.1030–0.4400 | 0.2610–1.1700 | (Tay et al., 2010) |
| 2-Nitrophenol | 0.0449–0.4844 | 0.0819–0.8970 | (Chen et al., 2003) |
| 2CP | 0.0193–0.2617 | 0.0447–0.5474 | Present work |

was approximately 2, which is consistent with other studies (Chen et al., 2003; Tay et al., 2010).

The reaction rate constant is influenced by various experimental parameters such as the solution pH, 2CP concentration, and ozone gas concentration. Under these experimental conditions, the solution pH and ozone gas concentration were maintained constant throughout the experiment. Therefore, the significant increase in the rate constant observed in the second-stage can be attributed to a decrease in the 2CP concentration. The formation of intermediate products was initiated by the degradation of 2CP by hydroxyl radicals. Owing to the non-selective behavior of hydroxyl radicals, there is expected to be competition between the intermediate products and 2CP for reaction with hydroxyl radicals. Consequently, the reaction of hydroxyl radicals with 2CP, rather than intermediate products, can occur more rapidly with decreasing 2CP concentration. This explains the increase in the rate constant observed in the second-stage. As the initial 2CP concentration increased, both first-stage and second-stage reaction rate constants decreased because the input gas concentration remained constant throughout the experiments. In studies investigating the removal of 2-nitrophenol and parabens by ozonation, it was found that the degradation kinetics occur in two-stage and the reaction rate constants in both stages decrease as the concentration of the target pollutant increases (Chen et al., 2003; Tay et al., 2010).

The measured COD values from the beginning of ozonation include both the unreacted 2CP and its intermediate products. When 2CP is completely oxidized through ozonation, the reaction proceeds with intermediate products. Therefore, the reaction rate constants related to the oxidation of 2CP were calculated based on COD removal. The first-stage rate constant ($k_{1\text{-COD}}$) observed for COD removal was higher than the second-stage rate constant ($k_{2\text{-COD}}$) (Fig. 3(d)). For an initial 2CP concentration of 100 mg/L, $k_{1\text{-COD}}$ is 0.0248 $min^{-1}$ while $k_{2\text{-COD}}$ is 0.0099 $min^{-1}$. Similar results have also been reported for COD removal via ozonation in different types of wastewaters (Table 2). The higher rate constant in the first-stage compared to the second-stage indicates that the reaction is faster initially and slows down due to the formation of intermediate products. In a study, it has been reported that the intermediate products formed during the ozonation of 2CP include chlorohydroquinone, 3-chlorocatechol, muconic acid, maleic acid, tartaric acid, dihydroxymaleic acid, and oxalic acid (Sung & Huang, 2007). It has been stated that among these intermediate products, oxalic acid tends to accumulate in the solution upon ozone treatment at a high pH (Hsu et al., 2007) and is not easily mineralized (Hu et al., 2016). Therefore, it can be said that increasing the initial 2CP concentration leads to an increase in the amount of intermediates product formed and a decrease in COD removal rates.

### 3.2 Effect of Ozone Dosage

Studies conducted to determine the effect of ozone dosage on 2CP removal are shown in Fig. 4. Ozone dosage is an important operational parameter, particularly for determining hydroxyl radical formation at high pH levels (Barlak et al., 2020). The removal of 2CP, COD, and TOC over time was investigated at a temperature of 20 °C, pH of 11, and an initial 2CP concentration of 100 mg/L. The efficiencies of 2CP, COD, and TOC removal were significantly influenced by inlet ozone dosage (Fig. 4). Figure 4(a) shows the percentage of 2CP removal over time at different ozone dosages. Increasing the ozone dosage led to an increase in the 2CP removal rate. As the ozone dosage increased, it is observed that the time required for the complete degradation of 2CP decreased. At an ozone dosage of 2.55 $gO_3/h$, 2CP was completely eliminated after approximately 60 min of ozone treatment. When the ozone dosages were 4.38, 8.25, and 13.1 $gO_3/h$, the corresponding times required for the complete depletion of 2CP were 30 min, 20 min, and 12.5 min, respectively.

Figure 4(b) illustrates the effect of various ozone dosages on COD removal. Although 2CP was completely consumed, the degradation of COD continued

**Table 2** Comparison of first- and second-stage rate constants for COD removal in different wastewater samples

| Wastewater type | $k_{1\text{-COD}}$ | $k_{2\text{-COD}}$ | References |
|---|---|---|---|
| Coking wastewater | 0.0045–0.0080 | 0.0010–0.0013 | (Wang et al., 2022) |
| Biotreated landfill leachate | 0.0082 | 0.0028 | (Wang et al., 2023) |
| 2CP—synthetic wastewater | 0.0053–0.0786 | 0.0020–0.0129 | Present work |

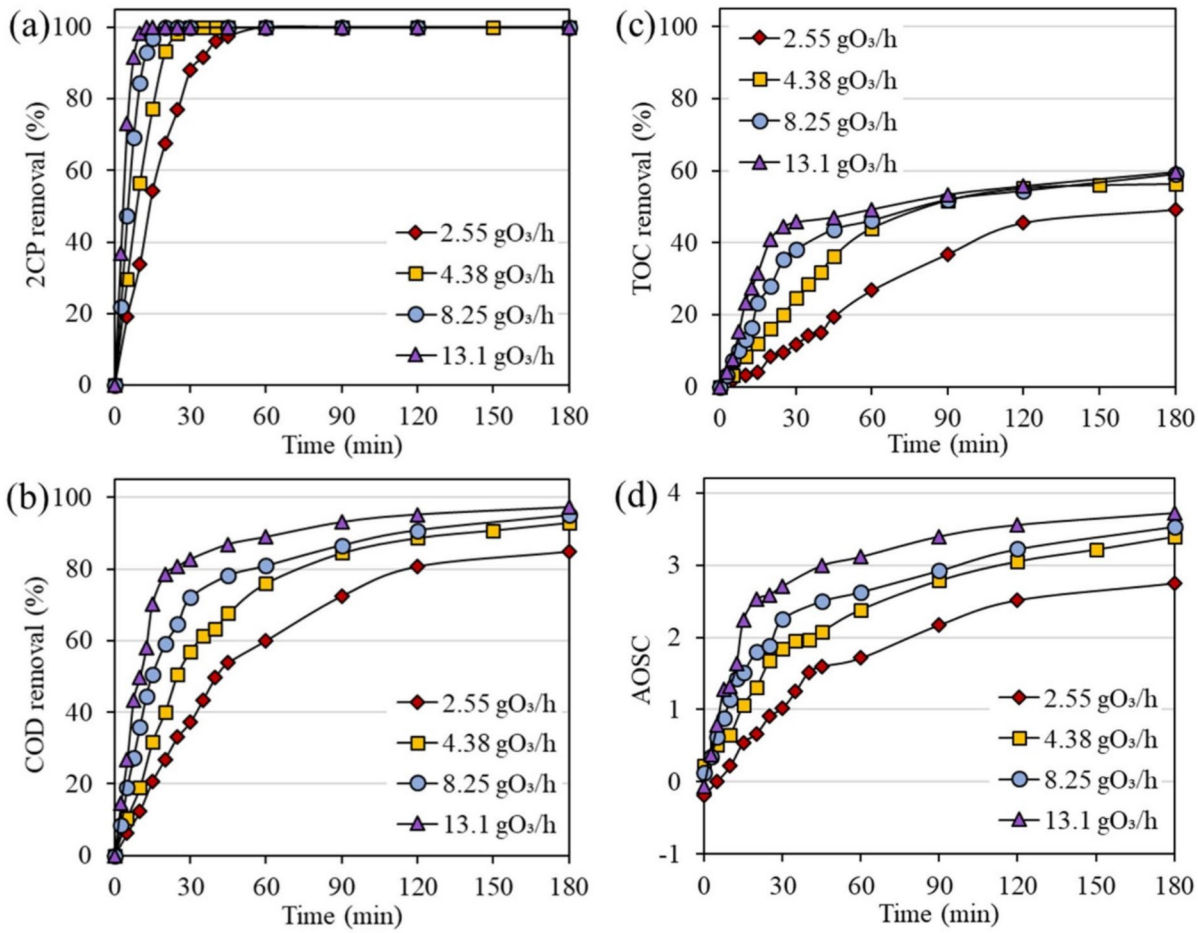


**Fig. 4** Removal efficiencies of 2CP (**a**), COD (**b**), TOC (**c**), and AOSC (**d**) for different ozone dosages in JLR (2CP = 100 mg/L, pH = 11, T = 20 °C)

because of the formation of intermediate products. Increasing ozone dosage increased the COD removal rate. After 20-min ozonation, COD removal efficiencies were 27%, 40%, 59%, and 79% for ozone dosages of 2.55, 4.38, 8.25, and 13.1 $gO_3$/h, respectively. After 180-min ozonation, the COD removal efficiencies were 91%, 93%, 95%, and 97% for ozone dosages of 2.55, 4.38, 8.25, and 13.1 $gO_3$/h, respectively. The reaction times for the complete depletion of 2CP (60, 30, 20, and 12.5 min) were associated with COD removal efficiencies of approximately 60%, 57%, 59%, and 58%, respectively. Even if 2CP has been totally consumed, COD continues to degrade because intermediate products are produced.

Figure 4(c) illustrates the effect of different ozone dosages on the TOC removal. Similar to COD removal, although 2CP was completely consumed, the degradation of TOC continued because of the formation of intermediate products. After 180 min, the TOC removal efficiencies were 49%, 56%, 59%, and 60% for ozone dosages of 2.55, 4.38, 8.25, and 13.1 $gO_3$/h, respectively. At ozone dosages of 2.55, 4.38, 8.25, and 13.1 $gO_3$/h, 2CP was completely depleted in 60, 30, 20, and 12.5 min, respectively. The corresponding TOC removal efficiencies at these reaction times (60, 30, 20, and 12.5 min) were approximately 27%, 25%, 28%, and 27%, respectively.

Figure 4(d) displays the variation in AOSC values at different ozone dosages. In all experiments in which the effect of ozone dosage was tested, it was observed that the AOSC value, as a result of 2CP oxidation, increased over time from an initial

value close to zero. For instance, at an ozone dosage of 2.55 $gO_3$/h, the initial AOSC value of −0.18 gradually increased during ozonation and reached a value of 2.75 after 180 min. At ozone dosages of 4.38, 8.25, and 13.1 $gO_3$/h, the AOSC values after 180 min were 3.39, 3.53, and 3.72, respectively. Based on these results, it can be concluded that the intermediate products formed during ozonation of 2CP were not completely mineralized. One of these intermediate products, oxalic acid, exhibits high resistance to ozonation and tends to accumulate in the solution (Hsu et al., 2007; Hu et al., 2016). Therefore, complete mineralization was not observed. This is also evident from the gradual decrease in COD and TOC removal efficiencies over time.

Figure 5(a) and Fig. 5(b) illustrate the two-stage degradation of 2CP and COD, respectively, with varying ozone dosages. In the degradation of 2CP, the reaction rate constants in both stages increased linearly with increasing ozone dosage, and it was determined that the rate constant observed in the first-stage ($k_{1\text{-}2CP}$) was lower than that observed in the second-stage ($k_{2\text{-}2CP}$) (Fig. 5(c)). For example, for an ozone dosage of 13.1 $gO_3$/h, $k_{1\text{-}2CP}$ is 0.2617 $min^{-1}$ while $k_{2\text{-}2CP}$ is 0.5774 $min^{-1}$. Because the solution pH and ozone dosage were kept constant during the experiment, the increase in the rate constant in the second-stage was attributed to the decrease in the 2CP concentration. The formation of intermediate products begins as 2CP starts to break down by hydroxyl radicals. Due to the non-selective nature of hydroxyl

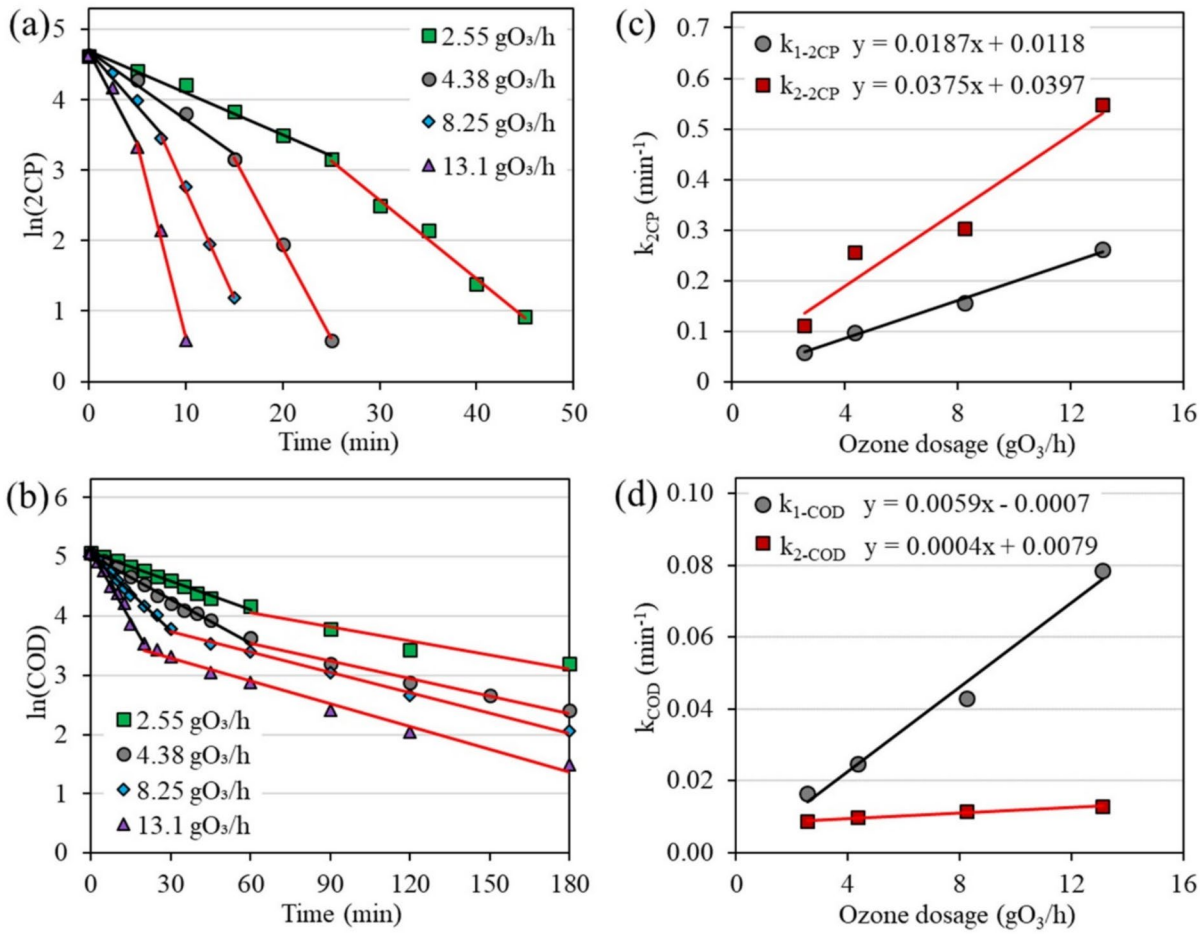


**Fig. 5** Under different ozone dosages, the variations of ln(2CP) (**a**), ln(COD) (**b**), the change in the PFO constant for 2CP (**c**), and PFO constant for COD (**d**) (2CP = 100 mg/L, pH = 11, T = 20 °C)

radicals, competition between intermediate products and 2CP for reaction with hydroxyl radicals is expected. Consequently, the reaction of hydroxyl radicals with 2CP can occur more rapidly with decreasing 2CP concentration. This explains the increase in the rate constant observed in the second-stage.

In the kinetics of COD removal, unlike 2CP, the rate constant observed in the first-stage ($k_{1\text{-COD}}$) was higher than that observed in the second-stage ($k_{2\text{-COD}}$) (Fig. 5(d)). For an ozone dosage of 13.1 $gO_3/h$, $k_{1\text{-COD}}$ is 0.0786 $min^{-1}$, while $k_{2\text{-COD}}$ is 0.0129 $min^{-1}$. The reaction rate constants in both stages increased linearly with ozone dosage. When examining the kinetics of COD removal, it was observed that the process progressed rapidly in the first-stage (during the degradation of 2CP) but slowed down owing to the formation of intermediate products (chlorohydroquinone, 3-chlorocatechol, muconic acid, maleic acid, tartaric acid, dihydroxy maleic acid, and oxalic acid) resulting from the oxidation of 2CP (Hsu et al., 2007; Hu et al., 2016; Sung & Huang, 2007).

## 4 Conclusion

In this study, we investigated the removal of 2CP via ozonation at high pH values. The removal of 2CP was accompanied by monitoring of COD and TOC removal efficiencies. The effect of initial 2CP concentration and ozone dosage from operating parameters were investigated. An increase in the initial concentration of 2CP resulted in longer durations required for the complete depletion of 2CP. The concentration of 2CP in the solution containing 100 mg/L was completely removed in approximately 12.5 min at an ozone dosage of 13.1 $gO_3/h$, while after 3-h ozonation, the COD and TOC removal efficiencies were 97% and 60%, respectively. The kinetic study revealed that the degradation of 2CP and COD followed two-stage PFO kinetics. In the case of 2CP removal kinetics, the observed rate constants in the second-stage were higher than those in the first-stage. However, for COD removal, the rate constants in the first-stage were higher than those in the second-stage. The rate constant values in both stages decreased exponentially with an increase in the initial concentration of 2CP, whereas they increased linearly with increasing ozone dosage. It was found that the rate constant values in both stages for 2CP and COD removal exhibited an exponential decrease with an increase in the initial concentration of 2CP, and a linear increase with an increase in ozone dosage.

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**Data Availability** The authors confirm that the data supporting the finding of this study is available on request.

**Declarations**

**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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