



Ozonation improves physical attributes in domestic sewage effluent

ARTICLES doi:10.4136/ambi-agua.2328

Received: 13 Sep. 2018; Accepted: 24 Jan. 2019

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ABSTRACT

Root Zone Wastewater Treatment (RZWT) is an option of natural wastewater treatment widely studied throughout the world, being a technology based on physical, chemical and biological processes. However, there are some attributes in effluent that occasionally are not eliminated by RZWT. In this context, the use of ozone may contribute to mineralization, disinfection, detoxification, and, especially, color and turbidity removal. Ozone is an excellent oxidant, with great potential to reduce color, turbidity and other attributes in liquids, such as in wastewater treatment. This paper evaluated the effect of different ozone exposure times (0, 30, 60, 90, 120 and 150 minutes) and the application of different dosages of ozone (0, 4, 10, 17, 25 and 35 mg L⁻¹ of O₃) on Hydrogeonic Potential (pH), Electrical Conductivity (EC), Dissolved Oxygen (OD), Total Alkalinity (Alk.), Sodium (Na) and Potassium (K) in domestic sewage effluent treated by the RZWT. Different exposure times and ozone dosages did not influence the attributes Potassium, Sodium, Ammonia and Total Solids. Color and turbidity decreased, while pH, Alkalinity and Dissolved Oxygen increased at all exposure times and O₃ dosages in relation to the non-ozonized effluent.

Keywords: color, ozone, root zone wastewater treatment, turbidity.

Ozonização promove a melhoria de atributos físicos em efluente de esgoto doméstico

RESUMO

O Sistema de Zona de Raízes (SRZ) é uma opção natural de tratamento de efluentes amplamente estudado em todo o mundo, sendo uma tecnologia baseada em processos físicos, químicos e biológicos. No entanto, existem alguns atributos no efluente que ocasionalmente não são eliminados pelo SZR. Nesse contexto, o uso do ozônio pode contribuir para a mineralização, desinfecção, desintoxicação e, principalmente, remoção de cor e turbidez. O ozônio é um excelente oxidante, com grande potencial para reduzir a cor, a turbidez e outros atributos em líquidos, assim como no tratamento de efluentes. O objetivo deste trabalho foi avaliar o efeito de diferentes tempos de exposição ao ozônio (0, 30, 60, 90, 120 e 150 minutos) e a aplicação de diferentes dosagens de ozônio (0, 4, 10, 17, 25 e 35 mg L⁻¹ de O₃) sobre



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Potencial Hidrogeônico (pH), Condutividade Elétrica (CE), Oxigênio Dissolvido (OD), Alcalinidade Total (Alk.), Sódio (Na) e Potássio (K) no efluente doméstico tratado pelo SZR. Diferentes tempos de exposição e dosagens de ozônio não influenciaram os atributos Potássio, Sódio, Amônia e Sólidos Totais. A cor e a turbidez diminuíram, enquanto o pH, a alcalinidade e o oxigênio dissolvido aumentaram em todos os momentos de exposição e as dosagens de O₃ em relação ao efluente não-ozonizado.

Palavras-chave: cor, ozônio, sistema de zona de raízes, turbidez.

1. INTRODUCTION

The use of septic tanks, followed by Root Zone Wastewater Treatment (RZWT), is an option of natural wastewater treatment system widely studied throughout the world, being a technology based on physical, chemical and biological processes which normally occur naturally in ecosystems, such as in natural floodplains (Kadlec and Wallace, 2009).

The main function of a wastewater treatment system is to remove the pollutant load, whether colloidal or soluble compounds. However, some attributes in the effluent occasionally are not eliminated. In this context, the use of ozone may contribute to mineralization, disinfection, detoxification, and, especially, color and turbidity removal (Bhatta *et al.*, 2015).

Today, ozone is still used for disinfection, but due to its superior oxidizing ability it is also used for a several other purposes including taste-and-odor control, color removal, iron and manganese oxidation, turbidity removal, algal removal, increasing the biodegradability of natural organic matter (NOM), and oxidation of specific micropollutants (Araújo *et al.*, 2016; Bhatta *et al.*, 2015; Bukhari *et al.*, 2019; Freitas-Silva *et al.*, 2013; Lage Filho, 2010).

Once dissolved in water, the ozone reacts with a large number of organic compounds in two different possible ways: direct oxidation, such as molecular ozone, or indirect reaction through the formation of secondary oxidants, such as free radicals, particularly the hydroxyl radical (Subha and Muthukumar, 2012; Wajahat *et al.*, 2019).

Despite the importance of ozone, it is noteworthy that the use of innovative technologies such as biogenic nanoparticles (BNPs), magnetotactic bacteria (MTB), and Phytogenic Magnetic Nanoparticles (PMNPs) is increasing in wastewater treatment, especially in the removal of heavy metals and specific compounds. In this way, studies related to ozonation should take into account all drawbacks including high cost, complex operation, and toxic and chemical reducing agents required to synthesize and control system operation (Ali *et al.*, 2017; 2018).

Considering the importance of ozonation in wastewater treatment, whether domestic or industrial, the objective of this paper was to evaluate the application of different dosages and exposure times to ozone on physical and chemical attributes in domestic sewage effluent, obtained after Root Zone Wastewater Treatment (RZWT).

2. MATERIAL AND METHODS

The Root Zone Wastewater Treatment (RZWT) is located at the Água Limpa Farm (ALF) of the University of Brasília (UnB) (15°57'16" S, 47°55'89" W and altitude of 1.103 m). The Laboratory of Water Analysis and the Laboratory of Storage and Preprocessing of Agricultural Products of the Faculty of Agronomy and Veterinary Medicine (UnB) performed the analyses of the effluent before and after applying ozonation.

The effluent generated at the Água Limpa Farm is mostly from sanitary discharges and a refectory, which, for its composition, is characterized as domestic effluent. The daily volume

generated is quite variable due to its frequency of use and oscillation of amount of people who attend the ALF, and it is not equal among the days of the week.

The RZWT consists of a set of four equal units built in rectangular fiberglass structures, with dimensions of 2.5 m (width), 6.5 (length) and 0.5 m (height). The first unit was cultivated with taboa (*Typha spp*); the second with Brazilian Papyrus (*Cyperus Giganteus*); the third with umbrella papyrus (*Cyperus alternifolius*), and a fourth system without a plant (witness).

An ozone generator O&LM, Ozone & Life® brand generated the ozone gas. In the process of gas generation, oxygen 90% pure and humidity free was used as input, obtained from the Mark Plus Oxygen Concentrator.

In order to produce the ozone gas (O_3), the oxygen went through a refrigerated reactor, in which the Dielectric Barrier Discharge (DBD) occurs. This type of discharge consists in applying a high voltage between two parallel electrodes, having between them a dielectric (glass) and a free space. In this free space, a discharge is produced in the form of filaments, where electrons are generated with sufficient energy to produce the breakdown of the oxygen molecules, forming the ozone (O_3) (Lage Filho *et al.*, 2008).

To evaluate the effect of exposure times of ozonation (0, 30, 60, 90, 120 and 150 minutes) and to optimize the analyses, two glass containers were used in each stage, where O_3 was equally divided. The containers were hermetically sealed and had 10 cm of internal diameter and 20 cm of useful height, having in its plastic cover, a porous diffuser through which the gas was introduced in a flow of 1.0 L min^{-1} .

The first two treatments were the exposure times of 30 and 90 min (one in each container). As soon as the time of 30 min finished, it was replaced by the 60 min treatment, thus, at the end of the 90 min, four treatments had already been completed (0 - witness, 30, 60 and 90 min). The last two treatments (120 and 150 min) followed the same sequence. At the end of the 120 min treatment, this container was replaced with distilled water until the completion of the experiment, i.e., for another 30 min until the treatment of 150 min was completed.

For the treatment with different dosages of ozone (0, 4, 10, 17, 25 and 35 mg L^{-1} of O_3), only one container was used. The treatments performed consecutively from the lowest to the highest dosage. The first effluent sample was exposed to a dosage of 4 mg L^{-1} of O_3 for 30 min, the second sample was added and the dosage was increased from 4 to 10 mg L^{-1} which remained for more 30 min. The dosages of 17, 25 and 35 mg L^{-1} of O_3 followed the same sequence.

In order to carry out the laboratory analyses, the methodologies of analysis of the Standard Methods for the Examination of Water and Wastewater (APHA *et al.*, 2005) were used or adapted. APHA Method no. 4500-0 (for Dissolved Oxygen); no. 2320 (for Total Alkalinity); no. 2130 (for Turbidity); no. 2120 (for Color); no. 2540 (for Solids); no. 4500 – NO_3 (for Ammonia); no. 3500 – Na (for Sodium); and no. 3500 – K (for Potassium). To apply different exposure times to O_3 , four collection campaigns were carried out in November 2017, and to evaluate the different dosages of O_3 , three collection campaigns were carried out in December 2017. In each campaign, 6 L of effluent in sterilized containers were collected, always at 1 p.m. due to higher volume of sewage at this time. The containers were packed in ice packs, which were transported to the laboratory and refrigerated at 4°C (APHA *et al.*, 2005).

The results of the experiments were analyzed using the Software Assistant (Silva and Azevedo, 2016), applying the Duncan test at 1 and 5% of probability, considering the collection dates as the replicates.

3. RESULTS AND DISCUSSION

3.1. Potential Hydrogen (pH)

There was an increase in pH values in the effluent samples, either as a function of exposure times due to the variation of the ozone dosages (Figure 1).

The pH values of the effluent exposed to the times of ozonation were higher than the effluent without ozonation, being the largest of 8.38 and the lowest of 7.46 (without ozonation). The time of 30 min differed from the times of 90, 120 and 150 min. A variation of 8.7% might be observed for the time of 30 min, up to 12.3% for the times of 120 and 150 min, demonstrating that the greatest variation occurred up to 30 min, after the beginning of O₃ application (Figure 1).

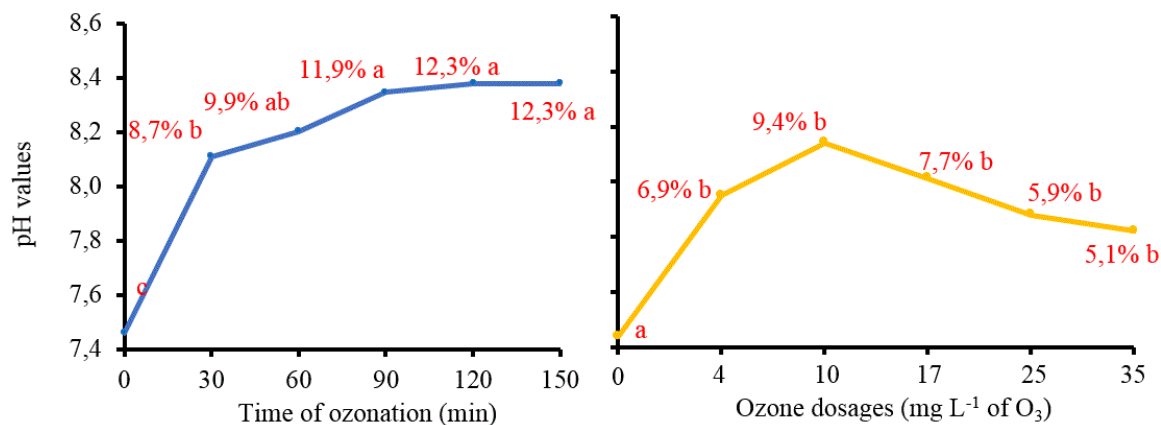


Figure 1. pH values of the domestic sewage effluent as a function of the exposure times 0, 30, 60, 90, 120 and 150 min at the dosage of 12 mg L⁻¹ of O₃ and to the dosages of 0, 4, 10, 17, 25 and 35 mg L⁻¹ of O₃ for 30 min and variation, in %, in relation to the effluent without ozonation. Different letters between the times of ozonation or O₃ dosages differ from each other by the Duncan test at 1% probability.

As the O₃ dosages varied, the pH values increased, reaching 8.14 (10 mg L⁻¹ of O₃), differing from the effluent without ozonation, but equal to the other dosages. After the dosage of 10 mg L⁻¹ of O₃, a reduction in the pH values was observed until the highest dosage evaluated (35 mg L⁻¹ of O₃), but not significantly (Figure 1). The pH drop from the 25 mg L⁻¹ O₃ dosage can be attributed to the formation of short chain acid compounds (acetic and oxalic acid, for example), resulting from the partial oxidation of more complex substances promoted by ozone as observed by Aquino and Pires (2016).

As ozone acts on the effluent, there is the release of hydroxide ions (OH⁻) making these predominant over hydrogen H⁺ ions. When the pH reaches 8.0, practically half of the O₃ introduced is decomposed into several intermediate forms of oxygen, over a period of 10 min (Lage Filho *et al.*, 2008; Coelho *et al.*, 2015).

This fact may explain the increase in pH values in the effluent samples with variation of the exposure time. When the pH value of the effluent is above 8.0, most of the ozone is decomposed into OH⁻, which induces an increasingly alkaline condition, interfering with the attributes of the effluent sample.

Bhatta *et al.* (2015) observed a slight increase in pH values in their study on the treatment of wastewater by ozone produced in dielectric barrier discharge, in agreement with the results found here. Likewise, Silva and Daniel (2015) observed an increase in pH values ($p = 0.05$) for the ozonized effluents compared to the UASB reactor effluent.

3.2. Electrical Conductivity (EC) and Total Alkalinity

Ozonation promoted a significant reduction in EC values as a function of time of ozonation and a reduction of Alkalinity as a function of O₃ dosages (Figure 2).

As the time of exposure of the effluent to the O₃ increased, there was a reduction of 7.5% in the EC values at 120 min compared to the effluent without exposure to O₃. The grand mean

of the treatment for different ozone dosages was $1281 \mu\text{S cm}^{-1}$, with values ranging from 1295 (0 mg L^{-1} of O_3) to 1272 (17 mg L^{-1} of O_3), but not differing between dosages.

Regarding total alkalinity, the grand mean obtained by the different exposure times was 56 mg L^{-1} of CaCO_3 , with values ranging from 53 to 59 mg L^{-1} of CaCO_3 , showing no significant difference by the 1% Duncan test.

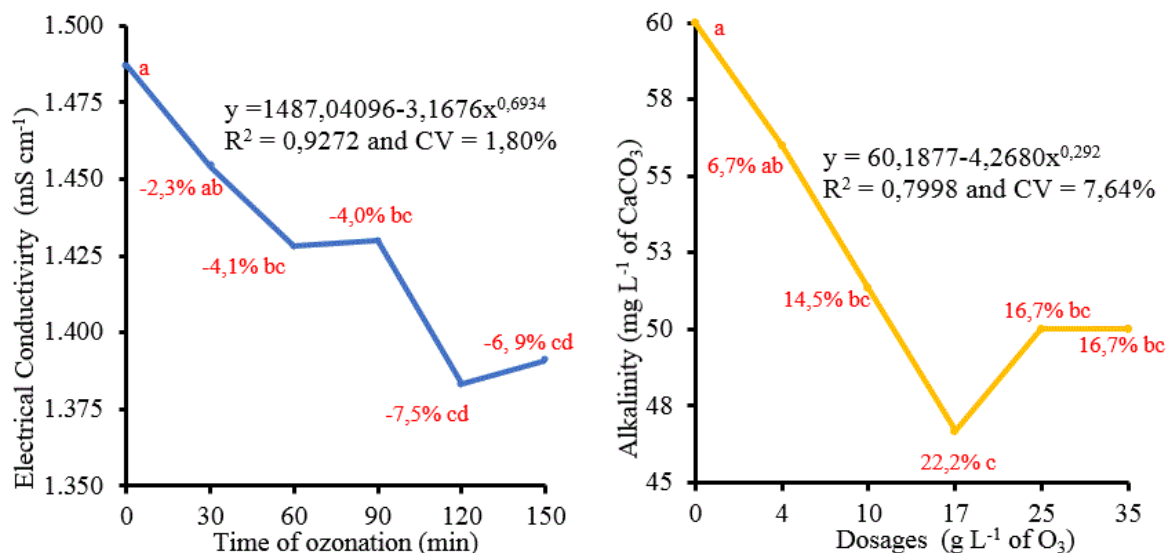


Figure 2. Electrical Conductivity of the domestic sewage effluent as a function of the exposure times of 0, 30, 60, 90, 120 and 150 min at the dosage of 12 mg L^{-1} of O_3 , and total alkalinity as a function of the time of exposure of the effluent at the dosages of 0, 4, 10, 17, 25 and 35 mg L^{-1} of O_3 for 30 min and variation, in %, in relation to the effluent without ozonation. Different letters between times of ozonation or O_3 dosages differ from each other by the Duncan test at 1% probability.

In the treatment as a function of the O_3 dosages, there was a reduction in the alkalinity values with the increase in the ozone dosages up to 17 mg L^{-1} , tending to stabilize from this dosage. The grand mean in the treatment was 52.33 mg L^{-1} of CaCO_3 , with values ranging from 46.67 mg L^{-1} (17 mg L^{-1} of O_3 dosage) to 60 mg L^{-1} (without ozonation), an average reduction of 22.2% (Figure 2).

Considering the basic pH values in both treatments, the buffering power (alkalinity) of the effluent treated with O_3 is adequate. It is worth highlighting that alkalinity is not the standard classification for natural waters or emission of sewage, showing the importance of this parameter in the control of certain unit processes used in water treatment plants (Von Sperling, 2014).

3.3. Dissolved Oxygen (DO)

The Dissolved Oxygen (DO) values were higher at all times of ozonation and at all O_3 dosages compared with the effluent without ozonation (Figure 3).

The grand mean in the treatment as a function of time of exposure to O_3 was $15.16 \text{ mg O}_2 \text{ L}^{-1}$, with values ranging from 5.5 to $19.3 \text{ mg O}_2 \text{ L}^{-1}$ for 0 and 90 min, respectively, representing elevation of 351%. While at the exposure times of 30, 60, 120 and 150 min the increases were 253, 282, 349 and 318%, respectively (Figure 3).

The grand mean when varying the O_3 dosages was $21.28 \text{ mg O}_2 \text{ L}^{-1}$, with values ranging from 6.2 to $26.2 \text{ mg O}_2 \text{ L}^{-1}$. There was an increase in DO values of 424% (17 mg L^{-1} O_3) and 413% (25 mg L^{-1} O_3) in relation to the effluent without ozonation.

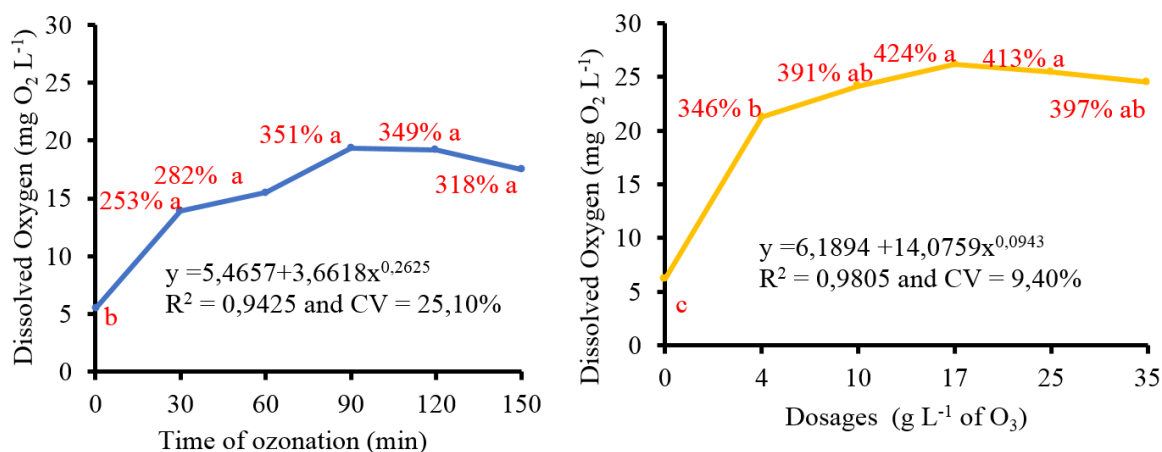


Figure 3. Dissolved Oxygen (mg O₂ L⁻¹) of the domestic sewage effluent as a function of times of the exposure of 0, 30, 60, 90, 120 and 150 min at the dosage of 12 mg L⁻¹ of O₃ and as a function of the dosages of 0, 4, 10, 17, 25 and 35 mg L⁻¹ of O₃ for 30 min and variation, in %, in relation to the effluent without ozonation. Different letters between times of ozonation and O₃ dosage differ from each other by the Duncan test at 1% probability.

The significant increase in DO as a function of the time of exposure and to the O₃ dosages is explained by the fact that the O₃ applied to the effluent is rapidly converted into O₂, raising its concentration in the environment. This phenomenon was also observed by Bhatta *et al.* (2015) where the DO values of the effluent, after the treatment with ozone, increased significantly, reaching the value of 9.55 mg O₂ L⁻¹ in that study.

3.4. Turbidity

There was a decrease in the turbidity values in the effluent samples both in relation to the exposure times and in relation to O₃ dosages (Figure 4).

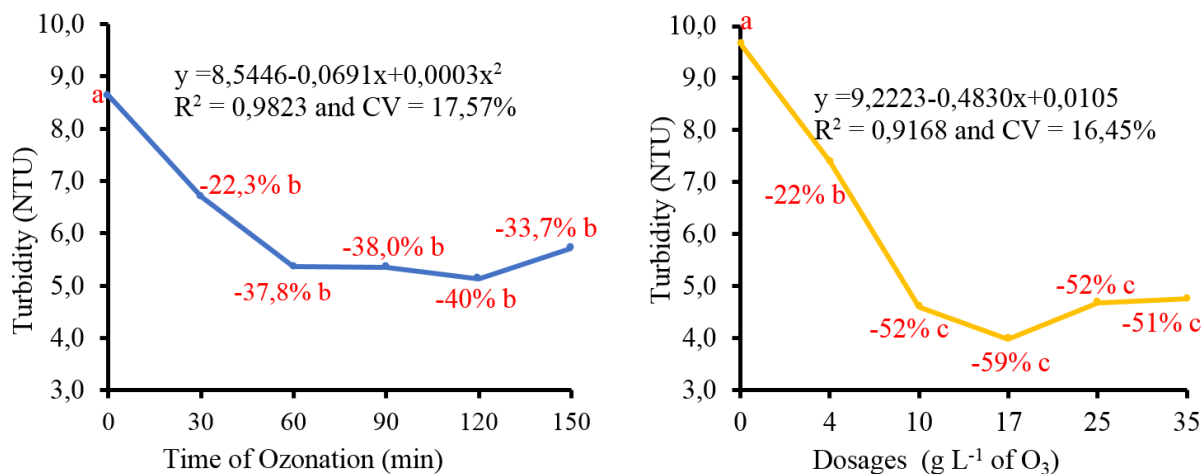


Figure 4. Turbidity in NTU of domestic sewage effluent treated as a function of the exposure times of 0, 30, 60, 90, 120 and 150 min at the dosage of 12 mg L⁻¹ of O₃ and as a function of the dosages of 0, 4, 4, 10, 17, 25 and 35 mg L⁻¹ of O₃ for 30 min and variation, in %, in relation to the effluent without ozonation. Different letters between times of ozonation and O₃ dosages differ from each other by the Duncan test at 1% probability.

The grand mean in treatment as a function of time of exposure was 6.2 NTU, being the highest value 8.63 and the lowest 5.1 NTU (0 and 120 min of exposure), representing a reduction of 40.0%.

As a function of the O_3 dosages, the grand mean of turbidity was 5.8 NTU, representing a reduction of 23.42% in the dosage of $mg L^{-1}$ of O_3 , and of 52.43% and 58.86% in the dosages 25 and 35 $mg L^{-1}$ of O_3 , respectively (Figure 4).

In this study, the greatest reductions in turbidity values were 40.0% and 58.9% at 120 min of exposure and 17 $mg L^{-1}$ of O_3 , respectively. Cabrera-Díaz *et al.* (2016) observed higher turbidity removal values, reaching 99.2% in combined treatment of vinasse (anaerobic filter-reactor and ozonation process); however, using a concentration of 100 $mg L^{-1}$ of O_3 and a contact period of 3 hours.

The reason for the decreasing values of turbidity while increasing ozone dosages and exposures times are that some particles and high weight organic compounds would be destroyed by ozone and converted into dissolved compounds like observed by Xu *et al.* (2002).

The reduction in turbidity values were positive, considering there was a reduction close to 60.0% in the dosage of 17 $mg L^{-1}$ of O_3 for 30 min. On the other hand, Li *et al.* (2015), in a study on biofiltration and ozonation for effluent treatment, observed a turbidity reduction of 90.0% using a dosage of 5 $mg L^{-1}$ of O_3 and a flow of $1.5m^3 h^{-1}$, respecting local limits for the discharge of urban effluents into water bodies.

3.5. Color

There was a reduction in color values at all exposure times and at all ozone dosages (Figure 5).

The grand mean of the color as a function of exposure times to O_3 was 96.0 of standard APHA color with progressive reduction, reaching 49.7 at 120 min of exposure. In percentage, the reduction was 47.0% at 30 min and 76.2% at 120 min

Regarding the treatment with different dosages of ozone, there was a reduction of 67.7% at the dosage of 4 $mg L^{-1}$ of O_3 and of 81.5% at the dosage of 17 $mg L^{-1}$ of O_3 (Figure 5).

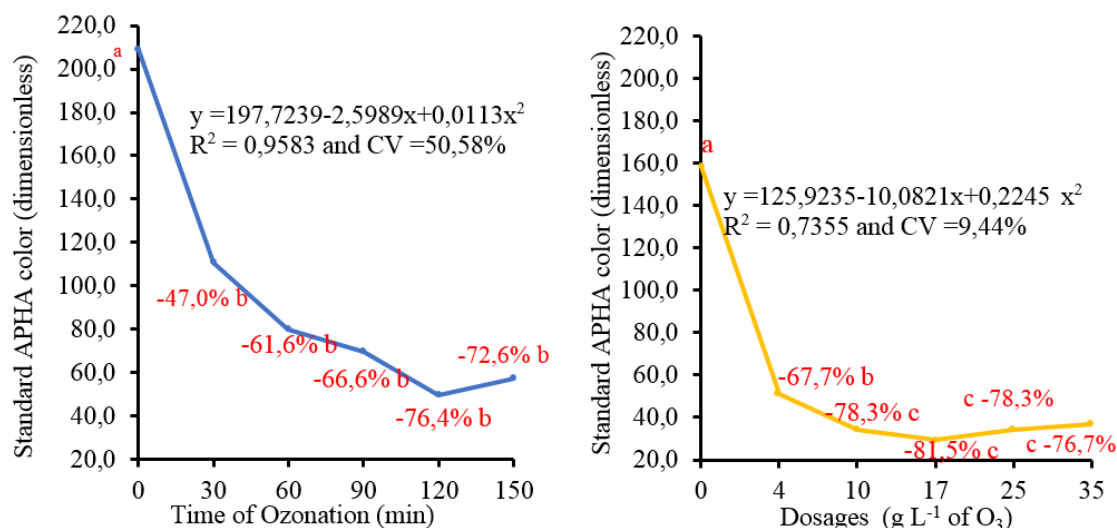


Figure 5. Standard APHA color (dimensionless) values of treated domestic sewage effluent as a function of the exposure times of 0, 30, 60, 90, 120 and 150 min at the dosage of 12 $mg L^{-1}$ of O_3 and as a function of the dosages of 0, 4, 10, 17, 25 and 35 $mg L^{-1}$ of O_3 for 30 min and variation, in %, in relation to the effluent without ozonation. Different letters between times of ozonation and dosages of O_3 differ from each other by the Duncan test at 1% probability.

The greatest color reductions (76.2% and 81.5%), are corroborated by Cabrera-Díaz *et al.* (2016) and Li *et al.* (2015), in which Cabrera-Díaz *et al.* (2016) observed color removals of 93.7% (UPt-Co) in a combined treatment of vinasse (anaerobic filter-reactor and ozonation process) emphasizing the effectiveness of the ozonation process in the color and turbidity

removal of this residue. Li *et al.* (2015), on the other hand, using biofiltration and ozonation for effluent treatment, observed a general efficiency for color removal of 70.0%, similar to the one obtained in the present study. Bhatta *et al.* (2015) observed visually, in their study on the treatment of wastewater by ozone, that the ozonized samples went from a dark gray color to a light color, close to the standard color of the water.

3.6. Ammonia, Sodium, Potassium and Total Solids

The average values of ammonia, sodium, potassium and total solids were not influenced as a function of exposure times and O₃ dosages.

Considering the average values, there was a tendency of ammonia elevation in absolute values, and there was no statistical difference, attributed, in part, to the great variability of the results, i.e., there was no consistent evidence that the time of ozonation and the O₃ dosages have influence on the ammonia contents.

Regarding sodium and potassium, there was great homogeneity of the values, both as a function of time and of O₃ dosages, i.e., parameters that were not influenced by ozonation.

The total solids, although showing some variations between the exposure times and O₃ dosages, did not present significant differences, leading to the conclusion, as well as in the parameters ammonia, sodium and potassium, that ozonation does not influence their values.

4. CONCLUSIONS

Ozonation reduced color and turbidity values both as a function of exposure times and as a function of O₃ dosages. There was a maximum reduction of 76.2% in color at 120 min of exposure and of 81.5% in turbidity at the concentration of 17 mg L⁻¹ of O₃.

There was an increase in pH, Total Alkalinity and Dissolved Oxygen (OD) values. At 120 min of exposure, there was a 12.30% increase in pH and at the dose of 4 mg L⁻¹ of O₃ the increase was 9.4%. Alkalinity increased 22.2% at the dose of 17 mg L⁻¹ of O₃, whereas OD values increased 351% at 90 min of exposure and 424% at the dose of 17 mg L⁻¹ of O₃.

Potassium, sodium, ammonia and total solids were not influenced by exposure time and ozone dosages.

5. ACKNOWLEDGMENTS

To CNPq for the financial assistance. Universal Call - MCTI / CNPq N° 14/2013. Process: 480332 / 2013-4.

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