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EVALUATION OF COMBINED TREATMENT OF LEACHATE FROM SANITARY LANDFILL AND SANITARY SEWAGE USING OZONE ON UP FLOW OXIDATION REACTOR

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Abstract

This work aimed at monitoring the formation of aerobic granular sludge in a SBR prototype and at evaluating its performance. The SBR was operated in two different phases (A and B), both fed with real domestic sewage and operated with 6-h cycles, but using different types of inocula: biological sludge from an extended aeration activated sludge system (in Phase A) and biological sludge from a conventional activated sludge system (in Phase B). During This study aimed to evaluate the combined treatment of leachate from landfill and sanitary sewage using ozone on upward flow oxidation reactor. The tests were conducted in batches, taking five and two hours of ozonation with 8.9; 9.6 and 10.5 g $O_3.h^{-1}$, applied to the mixture of 2% of leachate from sanitary landfill and 98% of sanitary sewage (v/v). The test was performed in triplicate with monitoring of COD, BOD, color, turbidity and pH. The results showed that ozone is a viable alternative for the pre-treatment of the mixture containing landfill leachate and sewage, facilitating the post-treatment in a biological process. It was observed removals of COD, BOD, apparent and true color and turbidity of 46%, 51%, 86%, 86% and 81% respectively in 5 hours of test, and average removals of 23%, 48%, 78%, 76% and 58% respectively for 2 hours of test.

Keywords: *advanced oxidation, landfill leachate, ozone, oxidation reactor, sewage.*

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Introduction

Landfill leachate (LL) can be characterized as a highly pollutant wastewater, due to its characteristics of composition, containing a large amount of dissolved organic matter (biodegradable and non-biodegradable carbon), ammoniacal nitrogen, potentially toxic metals, chlorinated organic and inorganic salts and xenobiotic substances (Wang *et al.* 2002) and a complex variety of hazardous chemicals (Aziz *et al.* 2004). Although some of these pollutants can be biodegraded, there are limitations in conventional biological processes when the removal of recalcitrant carbonaceous and nitrogenous organic matter, making it difficult to meet the standards for release in hydric bodies (Wang *et al.* 2002; Gao *et al.,* 2015). Facing this situation, a promising alternative to overcome such problems is to evaluate the use of other non-biological processes such as oxidative processes for removal and/or degradation of recalcitrant compounds, facilitating a further biological treatment step. To achieve this effect, a number of methods of treatments can be applied individually or combined, for example: photoreactor using O_3/UVC, O_3/H_2O_2, O_3/H_2O_2/UVC (Gomes *et al.,* 2020), electrocoagulation (Pauli *et al.* 2018), O_3/H_2O_2 (Gomes and Schoenell 2018), flocculation-precipitation, evaporation, adsorption on activated carbon, reverse osmosis and chemical oxidation (Laconi *et al.* 2015). Chemical oxidation using strong oxidizing agents (ozone, hydrogen peroxide, Fenton's reagent), in combination or individually, is a promising alternative due to the efficiency in removing non-biodegradable organic compounds.

The treatment of leachate by ozonation is an important option due to the high oxidation power (2.07 eV) of ozone (O_3), which is very effective in removing color and humic acids (Wang *et al.*, 2017). It is also important because of the oxidation of organic molecules with high molecular weight, commonly found in leachate, generating smaller molecules, with higher oxidation states than the originals, producing water and carbon dioxide (Tizaoui *et al.,* 2007; Cortez *et al.* 2011). Reaction mechanisms are complex and take place by direct or indirect routes from the production of hydroxyl radicals (•OH). These radicals have a high oxidation potential (2.80 eV) not selective. According Bila *et al.* (2005) improving the ratio BOD/COD, facilitating the removal of their products in biological processes. Comparative studies have been conducted using ozone coupled to other technologies, showing different results (Wu *et al.,* 2004; Abu Amr *et al.,* 2013). For example, Oloibiri *et al.* (2015) conducted the study using ozonation and coagulation-flocculation as pre-treatment of sanitary landfill leachate biologically stabilized, preceding a process of adsorption by activated carbon, and verified that the ozonation during 60 min responded by 33% of COD removal, reaching a total removal of 77% after treatment by adsorption.

Wang *et al.* (2005), observed COD removals of 41.7% using only ozone in the treatment of leachate, and when combined with other oxidizing agents such as O_3/H_2O_2 and O_3/UV they verified an increase in removal in the order of 15%. Koslowzka (2018), was found that the most effective process in landfill leachate treatment is the combination of ozonation with hydrogen peroxide addition (COD:H_2O_2=1:10). The COD, TOC and BOD values were 65%, 62% and 36%

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lower, respectively, in comparison to ozonation process conducted alone. According to these authors, high COD removals in the treatment of leachate of sanitary landfills are possible to be obtained, when oxidizing agents such as those used are employed in the pretreatment combined with biological processes.

The ozonation has also been used in the treatment of leachate of composting (Mokhtarani *et al.*, 2014) and replacing the chlorine to treat water for public supply (Silva and Daniel, 2015). Treatment in consortium of leachate of sanitary landfills and sanitary sewage (SS) is an alternative that has been employed in Brazil, and other countries, as a way of harnessing the idle capacity of some sanitary sewage treatment plants, generating income for the system operator, as it is charged by treated organic load. However, there is no common ground on the impact that the leachate can lead in biological treatment of sanitary sewage, given that they weren't designed for this purpose. This is a way to reduce costs of deployment and operation of sanitary landfills.

In view of this reality, this study aimed to evaluate the efficiency of the treatment of the leachate whih recalcitrant characteristics from sanitary landfill of São Leopoldo, (RS) combined with sanitary sewage generated at the University of the Sinos Valley (UNISINOS), using O_3 in lab-scale semi-batch an up flow oxidation reactor.

Experimental

Characterization of landfill leachate (LL) and sanitary sewage (SS)

Table 1. Physico-chemical characteristics of the landfill leachate and sanitary sewage.

Landfill leachate	
Parameter	Value
Apparent color (mg.L^{-1}Co/Pt)	5575.2 ± 2260.8
True color (mg.L^{-1}Co/Pt)	4447.9 ± 1935.8
Turbidity (NTU)	151.8 ± 213.7
COD (mg O_2. L^{-1})	1634.6 ± 352.8
BOD_5 (mg O_2.L^{-1})	164.2 ± 64.7
pH	8.1 ± 1.15
N-NH_3	998.2 ± 503.2
P_{total}	18.5 ± 9.2
BOD_5/COD	0.1 ± 0.02
Sanitary sewage	
Apparent color (mg.L^{-1} Co/Pt)	948.2 ± 421.4
True color (mg.L^{-1} Co/Pt)	275.7 ± 27.4
Turbidity (NTU)	124.3 ± 34.8
COD (mg O_2.L^{-1})	438.6 ± 34.3
BOD_5 (mg O_2.L^{-1})	213.8 ± 19.1
pH	7.5 ± 0.54
N-NH_3	107.3 ± 34.7
P_{total}	3,9 ± 2.5
BOD_5/COD	0.5 ± 0.07

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The characteristics of LL and SS can be observed in table 1. The landfill leachate was collected in the equalization tank that feeds the accumulation ponds of the sanitary landfill of São Leopoldo/RS. Collected LL was stored at 4ºC for later physical-chemical characterization and use. The samples were brought to room temperature before use. All the tests were performed with the same leachate. The SS was collected after the preliminary treatment (hand-cleaned bar rack followed by a grit chamber) of the sewage treatment plant before each test.

Experimental procedure

Six tests with ozonation were carried out, divided into two steps. First step took 300 min (Experiment A - tests 1, 2 and 3) and the second step took 120 min (Experiment B - tests 4, 5 and 6) of ozonation. It was applied 8.9, 9.6 and 10.5g $O_3.h^{-1}$ for each test, generated from an airflow of 3, 4 and 5 $L.min^{-1}$ O_2, respectively. The table 2 presents a summary of the characteristics of each test.

Table 2. Summary of the characteristics of each test.

	Experiment A			Experiment B		
Test	1	2	3	4	5	6
$gO_3.h^{-1}$	8.9	9.6	10.5	8.9	9.6	10.5
Time (min)	0 - 300			0 - 120		

The airflow rate was set with the flowmeter coupled to a regulating valve (UNITEC FX010). All tests of each experiment (A and B) and analytical parameters were analyzed in triplicate. The total volume of the mixture LL + SS in each test was 160 L, 2% (3.2 L) being composed of the LL.

Up flow oxidation reactor (UFOR)

The oxidation reactor was composed of a 250 mm diameter PVC pipe with 2220 mm of length (useful volume was 108L). UFOR had 2/3 of the volume filled with 1.5" Pall Rings® in order to optimize the internal hydraulic conditions by preventing the emergence of preferential paths. The UFOR was placed inside a 500L fiberglass tank, which received the mixture (LL + SS) for treatment.

The mixture was pumped from the tank to the UFOR using a helical pump (Netzsch (BN) model NM031BY01L06B) with a constant flow rate of 2.0 $m^3.h^{-1}$. A centrifugal pumb was used for recirculation at a flow rate of 7 $m^3.h^{-1}$ with a constant flow. A fraction of mixture being treated was collected at a point just above the section bundled with Pall Rings®, i.e., 1330 mm from the base to the top of the reactor and recirculated to a Venturi tube, in order to generate a negative pressure, to suction the ozone to the base of the reactor, as show in figure 1.

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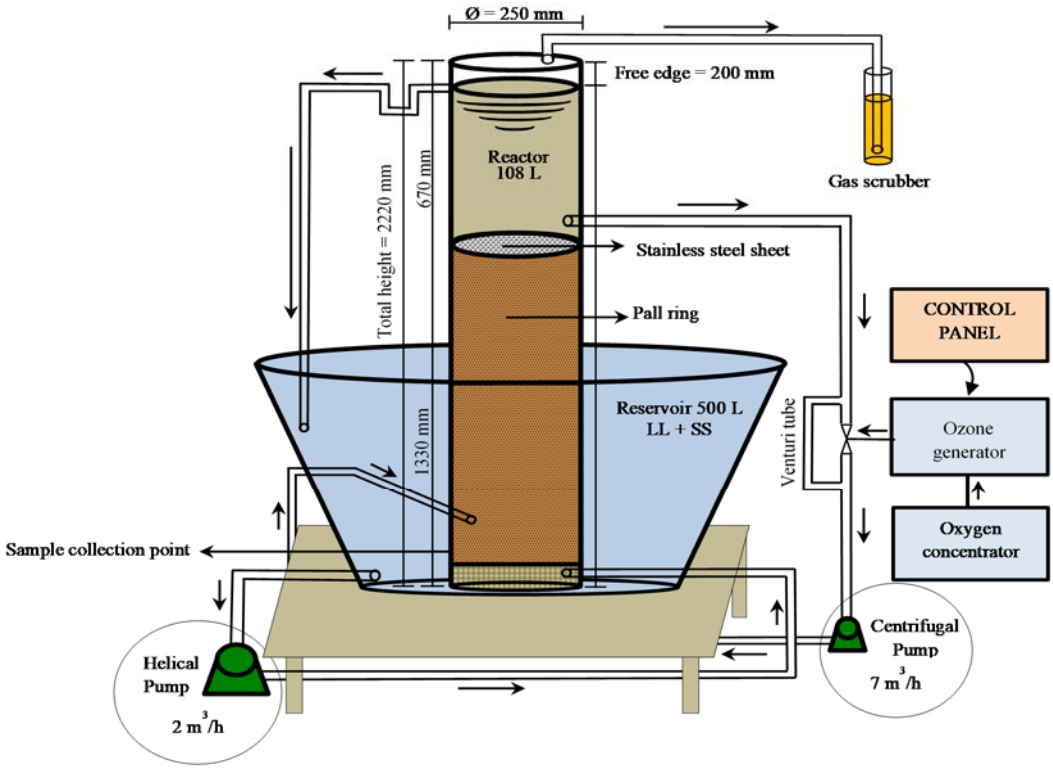

Figure 1. Schematic diagram of up flow oxidation reactor.

The ozone generator uses the Corona effect, being composed of an O_2 concentrator using the atmospheric air to generate O_3 (Brasil Ozônio – BRO3-PLUS2). Ozone was injected into the recirculating stream through the Venturi tube, allowing the mixture O_3 + (LL + SS) for each test with a constant flow of an ozone. The effluent from the UFOR was transferred from the top of the reactor to the 500 L tank, being recirculated through the helical pump with flow rate of 2 m^3.h-1. In this way, the liquid level inside the reactor was kept 690 mm above the collection point of the Venturi tube. This cycle was maintained by the established times in each test. Every new batch the entire contents of the reactor was removed and a new mixture (LL + SS) was prepared. The ozone generating equipment was calibrated hourly during each test according to 2350-E (Ozone Demand/Requirement- Semi-Batch Method) (APHA, 2012).

Analytical methods

To assess the efficiency of the treatment system, the following physical and chemical parameters were analyzed: pH, apparent color (AC), true color (TC), turbidity, BOD_5^{20} and COD. The

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determination of the true and apparent color was proposed to rapidly evaluate the degree of oxidation of the soluble organic matter. Also to verify possible changes in suspended solids concentration during ozonization. All parameters were analyzed according to APHA (2012).

Statistical analysis

The data was compared by using the t-Student test for means. The test performed is bilateral with a significance of 1% and 4 degrees of freedom. This work performed two types of tests for equality of means. In the first test (Equation 1 and 2), the equality of the means between the BOD (COD) tests was compared at the same time; in the second test (Equation 3 amd 4), the equality of the means between the times in the same BOD (COD) test was compared.

For the first case:

$$\begin{cases} H_0: \bar{x}_{i,s} = \bar{x}_{r,s} \\ H_1: \bar{x}_{i,s} \neq \bar{x}_{r,s} \end{cases}$$ **Equation (1)** **Equation (2)**

were s is the time $(0, 30, 60, 120, 180, 300)$, i and r are the test (1,2,3,4,5,6) with $i \neq r$.

For the second case:

$$\begin{cases} H_0: \bar{x}_{i,j} = \bar{x}_{i,s} \\ H_1: \bar{x}_{i,j} \neq \bar{x}_{i,s} \end{cases}$$ **Equation (3)** **Equation (4)**

were i is the test (1,2,3,4,5,6), j and s are the time $(0, 30, 60, 120, 180, 300)$ with $j \neq s$.

Results and discussion

The leachate collected presented physico-chemical characteristics with BOD/COD recalcitrant = 0.1 (see Table 1) indicating the impossibility of treatment by biological processes. The ozonation is an alternative pretreatment to improve the ratio BOD/COD enabling biological treatment.

The LL treatment with SS is a low-cost option that deserves attention, and has been used in Brazil as an alternative to the LL treatment, avoiding the need to invest in new WWTP solely for this purpose. There is no consensus as to the percentage of LL to be added to the SS because it depends on the physicochemical characteristics of the LL. According to Çeçen and Aktas, 2004; Renou *et al.* 2008; Yu *et al.* 2010; Fudala-Ksiazek *et al.* 2011, the percentage of LL added to SS should not overpass 10%, under risk of inhibition of the microbiota of the WWTP due to high concentrations of ammonia nitrogen normally present in the LL.

Removal of color and turbidity

The removal of apparent and true color and turbidity was intense in the first 120 minutes of treatment in UFOR for different rates of O_3 used, for 120 and 300 minutes of contact, as shown in Figure 2.

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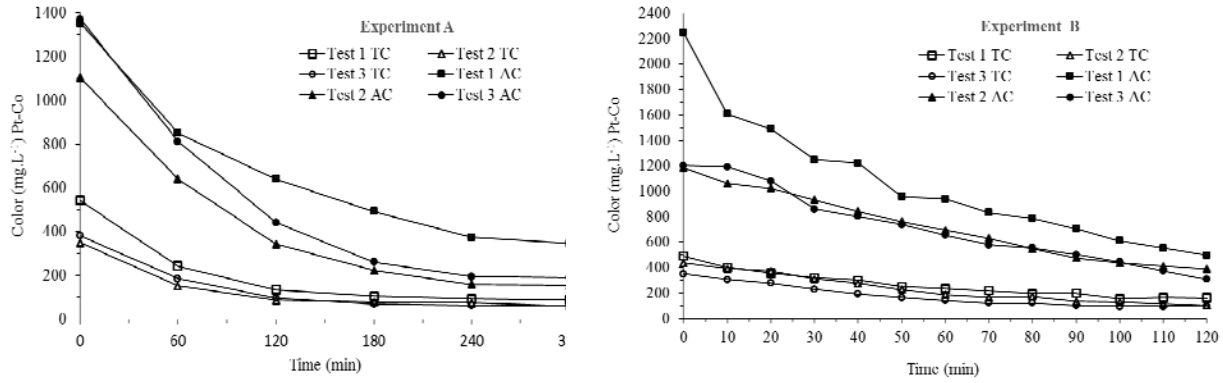

Figure 2. Removal of apparent color and true color for the tests with 120 and 300 minutes of treatment in UFOR in the different ozonation rates.

All tests were performed with the same rates of application of ozone and the collection of samples was performed at intervals of 10 minutes to identify the contact time in which maximum removal of color and turbidity was observed. The increase in the ozone application rate from 8.9 to 9.6 and 10.5g of $O_3.h^{-1}$ not proved to be decisive in reducing turbidity and color. However, for 120 minutes of treatment, a decrease of true color of 14.3%, 11.6% and 19.3% in tests 1, 2 and 3 respectively was observed, compared to tests with 300 minutes of ozonation. The same behaviour was observed for removal of apparent color, with 13%, 14% and 9.3% respectively. With respect to turbidity, a minor removal was observed in tests with 120 min of ozonation. This effect was attributed to the longer treatment, therefore a greater time of effluent recirculation on treatment, causing the destruction of existing particles in mixture due to turbulence caused by hydraulic fluid passage through the section bundled with Pall ring.

The increase in ozone concentration also resulted in an increase in the removal of BOD and COD on tests 1, 2 and 3 with 300 minutes long, while the same was not observed when the ozonation time was reduced to 120 minutes, with only a increase in BOD removal when applied 10.5 g of $O_3.h^{-1}$. It should be noted the difficulty of obtaining results in a definite pattern, allowing the establishment of relations of efficiency versus operating conditions of the pilot system, when the substrate presents great variability in their physical-chemical characteristics, which is natural for this type of effluent. These results show the conversion of recalcitrant compounds to components with higher biodegradability, and that conversion depends on the ozonation time. These effects were also observed by other authors (Poblete *et al.* 2019; Cortez *et al.* 2010; Chaturapruek *et al.* 2005; Marttinen *et al.* 2002), but in different experimental conditions. The convergent point among the other researches is the possibility of using ozone as an oxidizing agent. However, it is necessary to develop reactors for such possibilities and to establish efficient operational methods.

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Removals of AC and TC of 86% were obtained, and the largest removal of color was observed during the first two hours of treatment, with 76% for TC and 69% for AC, as show in Fig. 2. After 2h of treatment, color removal has decreased. This rapid decrease in color can be assigned, according to Tizaoui *et al.* (2007), to the occurrence of reactions between ozone and color-causing substances present in the mixture in treatment, leading to formation of by-products of lower intensity of color. Studies by Wu *et al.* (2004), highlight that the hydroxyl generated in the process of ozonation reacts specifically with the conjugated chains of organic molecules that give color to the leachate.

Similarly, Cheng *et al.* (2011), notes that the color in effluents is caused by the existence of organic compounds whose chemical structure features unsaturated bonds such as C=C, C=O, C=N and N=N. Ozone is a strong oxidizer and can act breaking double or triple bonds in the organic molecules decreasing the effluent color. The decrease of color in the mixture in treatment occurred in function of time and ozone concentration applied. However, it was not possible to determine a default color removal in line with the rate of application of ozone.

3.2. Removal of organic matter

The data summarized in Table 3 show that the largest COD and BOD removal was obtained in experiment “A” with 46.2% and 51.0%, after 300 minutes of ozonation, while in experiment “B”, with ozonation of 120 minutes, the removal of COD and BOD was 20.3% and 27.3%, respectively.

The results allows us to suggest that the use of ozonation can be an alternative to an integrated process of leachate treatment, for example, preceding a biological treatment step. In this way the recalcitrant organic substances normally present in the LL can be partially or fully oxidized by ozone, improving the biodegradability of the mixture affluent to the biological step. Laconi (2012) studied the use of ozone after biological treatment of different effluents, among them LL, and noted that biological treatment followed by ozonation does not guarantee depurative levels sufficient for discharge for LL. On the contrary, thanks to the synergy between biological degradation and ozonation, integrated treatment significantly improves the process performance, thus allowing the discharge limits to be met

BOD/COD ratio showed an increase of 13.5% in test 5 and 20.7% in test 6. In other tests the increase in the ratio was below 5%. The increase of the biodegradability coefficient can be attributed to the ozone action. According to Qin, *et al.* (2013) and Yva *et al.* (2019) the ozone can be increase in the biodegradability coefficient, because it reacts with humic acids in waters and effluents. Humic acids in water may react with ozone via direct or indirect pathway, resulting in a decrease in molecular weight and an enhancement of biodegradability. In this way the increase of biodegradability can be attributed to a sum of factors such as the decrease in color, turbidity and ammoniacal nitrogen removal by stripping, which may have been potentiated by ozone

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bubbling in the inflow current to the reactor. The presence of high color, turbidity and ammoniacal nitrogen can inhibit the proliferation of microorganisms, resulting in a decrease in the biodegradability coefficient. Although the ozone treatment has not achieved high results in the removal of organic matter, it is suggested the use as pre-treatment, as have been observed an increase of biodegradability and color removal of the mixture.

Table 3. Results of COD, BOD520 and pH after treatment with ozone.

		Experimental conditions A				Experimental conditions B		
Parameter	RTa	(1) 8.9 gO3.h-1	(2) 9.6 gO3.h-1	(3) 10.5 gO3.h-1	RTa	(4) 8.9 gO3.h-1	(5) 9.6 gO3.h-1	(6) 10.5 gO3.h-1
COD ($mg\ O_2.L^{-1}$)	0	518	318	364	0	339	332	459
	30	490	286	309	30	310	300	401
	60	477	253	275	60	290	297	389
	180	470	221	215	120	261(19.4)	270(48.0)	370(50.5)
	300	417(19.5)	215(32.4)	198(45.6)				
BOD_5^{20} ($mg\ O_2.L^{-1}$)	0	227	187	269	0	184	210	209
	30	210	156	200	30	165	158	187
	60	195	131	177	60	154	150	108
	180	193	123	146	120	129(29.9)	141(32.8)	90(56.9)
	300	183(38.0)	97(18.7)	133(19.4)				
pH	0	7.2	7.1	6.9	0	7.2	7.1	7.2
	30	7.1	7.3	7.2	30	7.1	7.3	7.1
	60	7.3	7.4	7.3	60	7.3	7.4	7.3
	180	7.4	7.5	7.5	120	7.4	7.5	7.4
	300	7.5	7.7	7.7				

With the tests 4, 5 and 6, it was found that the removal of organic matter occurred mainly in the first two hours of ozonation, that being the time in which it was observed the higher removal. After 2 hours of treatment to remove organic matter decreased probably due to the formation of by-products of ozonation, and because of the difficulty of degradation of recalcitrant compounds.

Results of statistical analysis

Tables 4 and 5 show, respectively, the results of the statistical tests carried out according to section 2.5 for the two cases addressed. In Tables 4 and 5 the gray cells with the x are where the test means are statistically equal at 1% of significance level.

In Table 4 the blank cells indicate that there is no evidence to accept that the means of the tests ate the same time are statistically equal to the 1% significance level.

In Table 5 the blank cells indicate that there is no evidence to accept that the means of the same test at different times are statistically equal to the level of significance of 1%. In both Tables (4 and 5) the crossed cells are where the tests do not apply.

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Table 4. Test for equality of the means of COD (BOD) at the same time.

Time	Test 6/5	Test 6/4	Test 6/1	Test 6/2	Test 6/3	Test 5/1	Test 5/2	Test 5/3	Test 1/2	Test 1/3	Test 2/3
Comparison between the means of the COD tests											
0	SD	x	x	SD	SD	SD	x	x	SD	SD	x
30	SD	x	SD	SD	x	SD	x	x	SD	SD	x
60	x	x	SD	SD	x	SD	x	x	SD	SD	x
120	x	x	x	NA	NA	NA	NA	NA	NA	NA	NA
180	NA	NA	NA	NA	NA	NA	NA	NA	SD	SD	x
300	NA	NA	NA	NA	NA	NA	NA	NA	SD	SD	x
Comparison between the means of the BOD tests											
0	x	x	x	x	x	x	x	SD	x	x	SD
30	SD	x	x	x	x	x	x	x	x	x	x
60	SD	x	x	x	x	x	x	x	x	x	x
120	SD	x	x	NA	NA	NA	NA	NA	NA	NA	NA
180	NA	NA	NA	NA	NA	NA	NA	NA	x	x	x
300	NA	NA	NA	NA	NA	NA	NA	NA	SD	x	SD

Note: (x) Statististical Equal; (SD) Statististical Different; (NA) Not Applicable

Table 5. Test for equality of the means of COD (BOD) for different times (min) in the same test.

Time	COD/Test						BOD/Test					
	1	2	3	4	5	6	1	2	3	4	5	6
0-30	x	x	x	x	x	SD	x	SD	SD	x	x	x
0-60	x	x	x	x	x	SD	x	SD	SD	x	SD	SD
0-120	NA	NA	NA	x	x	SD	NA	NA	NA	x	SD	SD
0-180	x	SD	SD	NA	NA	NA	x	SD	SD	NA	NA	NA
0-300	x	SD	SD	NA	NA	NA	x	SD	SD	NA	NA	NA
30-60	x	x	x	x	x	x	x	x	x	x	x	SD
30-120	NA	NA	NA	x	x	SD	NA	NA	NA	x	x	SD
30-180	x	x	x	NA	NA	NA	x	x	x	NA	NA	NA
30-300	x	x	x	NA	NA	NA	x	SD	SD	NA	NA	NA
60-120	NA	NA	NA	x	x	x	NA	NA	NA	x	x	x
60-180	x	x	x	NA	NA	NA	x	x	x	NA	NA	NA
60-300	x	x	x	NA	NA	NA	x	x	x	NA	NA	NA
180-300	x	x	x	NA	NA	NA	x	x	x	NA	NA	NA

Note: (x) Statististical Equal; (SD) Statististical Different; (NA) Not Applicable

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Conclusions

The use of ozone in the conditions of this experiment proved to be effective in the treatment of the mixture LL + SS. Color removal of 86% was observed in the first two hours of treatment. The increase in ozone concentration has not presented an increase in efficiency of color and turbidity removal, evidencing the existence of a threshold from which O_3 ceases to be effective in the treatment.

The decrease in ozonation time from 300 to 120 minutes promoted a decrease in removal efficiency of true color and a similar effect was observed for turbidity, which is assigned to the form associated with the destruction of particles by turbulence brought in by the recirculation through reactor bundled session.

The increase in ozone concentration has enabled an increase in the removal of BOD_5 and COD. Removal of BOD_5 and COD was 51% and 44%, respectively. Removal of AC and TC was 86% and occurred mainly in the first two hours of ozonation. BOD/COD ratio showed an increase of 13.5% in the test 5 and 20.7% in the test 6, when the ozonation lasted 300 minutes. In the other tests, the increase was below 5%. In this way the increase of biodegradability can be attributed to a sum of factors that result in the decrease in color, turbidity and ammoniacal nitrogen.
The use of the up-flow oxidation reactor proved to be efficient to promote oxidation by ozone and to allow the ideal mixing conditions for the removal of compounds present in the mixture between LL and SS.

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