Revista AIDIS de Ingeniería y Ciencias Ambientales:
Investigación, desarrollo y práctica. ISSN 0718-378X

Vol. 8, No. 1, 14 – 25
6 de abril de 2015

REVISTA AIDIS

de Ingeniería y Ciencias Ambientales:
Investigación, desarrollo y práctica.

NITROGEN REMOVAL FROM SEWAGE IN A SERIE OF AEROBIC-ANOXIC SUBMERGED BIOFILTERS

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Recibido el 9 de febrero de 2015; Aceptado el 9 de marzo de 2015

Abstract

This paper presents the results of an alternative technology with combination of aerated and anoxic biofilters, filled with corrugated cut conduit, low cost and simple operation. These filters receiving domestic sewage previously treated in anaerobic reactors. The study was divided in three phases: during the first phase the air flow to the aerated filter was 0.10 m^3/minute, and in phases 2 and 3 air flow increased to 0.15 m^3/minute. In the last phase 1 m^3/day of the anaerobic effluent was used as a supplemental source of carbon to the anoxic filter. The filters were capable of reducing the ammonia concentration to below 10 mg/L. In addition, the system was able to produce the final effluent concentrations of TSS and COD of 5.3 mg/L and 25 mg/L, respectively, without sludge removal, or any other step for phase separation.

Keywords: sewage, denitrification, nitrification, nitrogen removal, submerged biofilter, sludge retention.

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Introdução

The nitrogen present in wastewater is mainly composed of ammonia (gas, NH_3, and salt, NH_4^+) and organic nitrogen (urea, amino acids and other organic substances such as amino group). Occasionally result in traces of oxidized forms of nitrogen such as nitrite (NO^-_2) and nitrate (NO^-_3).

One of the recurrent problems in water bodies, especially in lentic systems, is the cultural eutrophication caused by the increase in nutrient availability. In addition, the ammonia nitrogen in the environment when disposed in inadequate concentrations generates some drawbacks, such as toxicity to living organisms, oxygen consumption to meet the nitrogen demand and also by nitrate contamination of aquifers. For this reason it is often necessary to performed a complementary treatment of wastewater in order to remove or reduce the concentration of nitrogen in final effluents, and one of the most simple and safe procedures is the biological removal.

Generally sewage treatment plants designed to achieve the biological removal of nitrogen include two different reactors: the aerated reactor, where oxidation occurs until ammonia becomes nitrate and the anoxic reactor, where the nitrate will be reduced to molecular nitrogen and thus being eliminated from the system. However, studies have shown that it is possible to nitrify and denitrify using the same environment. The simultaneous nitrification and denitrification (SND) occurs when ammonia is oxidized in an aerobic environment and oxidized compounds are reduced within the same reactor (Meyer et al., 2005, and Chiu, et. al., 2007).

This paper discusses the aerated filter with a support media bed consisting of plastic pieces (high density polyethylene), which provides a high void ratio. In fact is an approach of new treatment technology. This system is in fact an approach because both it contains suspended solids in the interstices, as retains biomass by attaching to the filler material. Although being the same working principle of submerged aerated biofilters, it differs enough of these, because the large amount of sludge retained in the interstices that has an important role in biodegradation, and also from the Moving Bed Biofilm Reactor (MBBR), because the bed is fixed and retains more solids in the interstices of the filler material.

The high void ratio and the accumulation of silt in the interstices may also favor the formation of anoxic zones inside the aerated reactor, providing the occurrence of simultaneous nitrification and denitrification. Aeration through the retained sludge allows respiration of the biomass to enter the endogenous phase, reducing the need for disposal of sludge. Although, eventually a small part of the mineralized sludge escapes through the sludge bed, the final effluent still maintain low concentrations of TSS and turbidity.

Revista AIDIS de Ingeniería y Ciencias Ambientales:
Investigación, desarrollo y práctica. ISSN 0718-378X
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This paper aims to contribute to the development of wastewater treatment technology, using a variation of aerated biofilter, filled with plastic material with high void ratio, capable of removing nitrogen and produce effluent with low concentrations of TSS without generating excess sludge.

Materials and Methods

The studied system is located in the Wastewater Treatment Plant of the Federal University of Rio Grande do Norte (UFRN), in Natal, northeast of Brazil, and treats mainly domestic sewage.

The system consists of two in series submerged biofilters being the first aerated (AER) and the second non-aerated (ANX). The initial idea was that the second filter had performed as an anoxic reactor, but the remainder oxygen from the first aerated filter still provided dissolved oxygen to the second, although at low concentrations. However, for the purpose of reporting the results, it was adopted the terminology anoxic for the second filter (ANX). The filters were 4.00 m long and 0.70 m wide and with total and working height of 1.22 m and 1.10 m, respectively, resulting in total and working volumes of 3.42 m^3 and 3.08 m^3, respectively. The filters were filled with 2 cm in diameter corrugated cut conduit, with an average length of 2.90 cm, a specific surface area of 277 m^2/m^3 and a void ratio of 90% (see Figure 1).

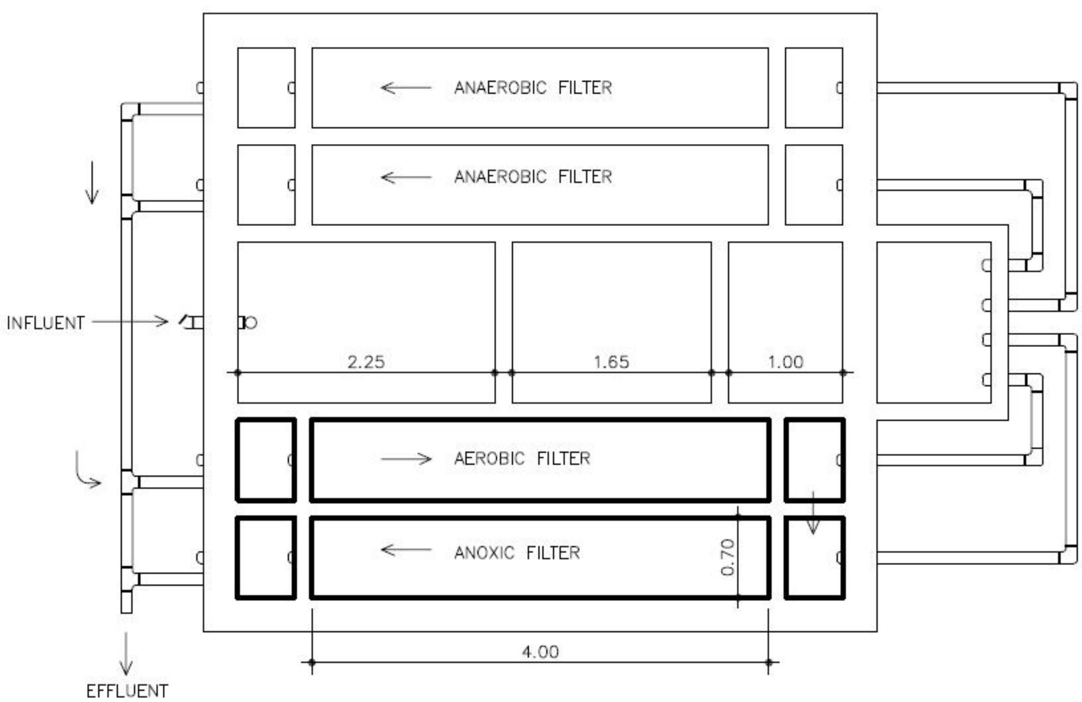

Figure 1: Schematic details of the system.

The biofilter startup was made under anaerobic conditions to avoid the agitation caused by aeration on biofilm adherence and development on support plastic media. After one week, aeration was initiated at a flow rate of 0.05 m^3/min and, after another week, the aeration flow rate was increased to the value adopted in first phase of the research (0.10 m^3/min). Then, for one month, bacterial community was allowed to growth and stabilize. During this period of bacterial acclimatization, samples were taken twice a week and analyzed for the same parameters presented in Table 2.

The air was supplied by means of two parallel working compressors with the flow rates calibrated by two rotameters. Inside the filter the air was distributed through three ½ inch hoses, with 1/32 inch holes each 5 cm; for protection each hose was placed inside a 3 inch PVC pipe with ¼ inch holes each 5 cm, located at the bottom, which were used to distribute the influent. The aerated biofilter was fed with the effluent from an anaerobic system consisting of septic tank followed by two parallel anaerobic filters.

The research was conducted in three phases, with the influent flow rate of 10 m^3/day. The air flow to the aerated filter was 0.10 m^3/minute, during the first phase, and 0.15 m^3/minute in following phases corresponding to oxygen application rates of 51 and 77 m^3/m^2.day, respectively. During the third phase 1 m^3/day of the anaerobic effluent with mean concentration of 56 mg/L of ammonia, 167 mg/ L of COD, 352 mg$CaCO_3$/L of alkalinity, and 57 mg/L of TSS, was diverted directly to the ANX filter as a supplemental source of carbon. In the first two phases the hydraulic retention times (HRT) were 7 h and 20 min in two filters. In phase three, with the diversion of part of the anaerobic effluent to the ANX filter the HRT rose to 8 h and 10 min in the AER filter and was not changed in the ANX filter. (Table 1).

A total of 35 samples from effluents of anaerobic treatment (septic tank + anaerobic filter - ANA), aerated filter (AER) and anoxic filter (ANX) were collected from August/2010 to April/2012, being 14 in phase 1, 11 in phase 2 and 10 in phase 3. During this period there was no sludge removal. Table 1 summarized the operational variables.

Table 1. Operational variables

	Sewage flow (m^3/d)		Air flow (m^3/min)	source of carbon (m3/d)	HRT (hh:mm)		Phase duration (Days)	samples
	AER	ANX			EAR	ANX		
Phase 1	10	10	0.10	-	07:20	07:20	92	14
Phase 2	10	10	0.15	-	07:20	07:20	77	11
Phase 3	9	10	0.15	1	08:10	07:20	60	10

Results and discussion

Table 2 shows the averages (in bold) and standard deviations (in parentheses) of the parameters analyzed in the three phases of the research and Table 3 presents the loadings of COD and total nitrogen. As a general view was observed that the system have a great capacity to retain the solids within the interstices favoring the increase on cell retention time which may leads to biomass respiration through endogenous phase and the consumption of retained sludge and, thus eliminating the secondary sedimentation unit.

Table 2. Means (bold) and standard deviations (in brackets) during the three research phases

	Phase 01			Phase 02			Phase 03		
	ANA	AER	ANX	ANA	AER	ANX	ANA	AER	ANX
N- Ammonia (mg/L)	**59.6** (9.3)	**27.7** (12.0)	**28.3** (10.0)	**53.3** (9.8)	**9.4** (3.1)	**8.9** (4.8)	**51.9** (11.3)	**11.2** (5.5)	**9.6** (5.8)
N-Organic (mg/L)	**2.4** (0.8)	**0.9** (1.1)	**0.6** (0.7)	**2.1** (0.9)	**1.6** (0.8)	**0.8** (0.3)	**2.5** (0.8)	**1.7** (0.8)	**1.5** (0.8)
N-NO_2 (mg/L)	**0.0** (0.0)	**2.3** (1.5)	**1.3** (1.6)	**0.0** (0.0)	**0.1** (0.1)	**0.0** (0.0)	**0.0** (0.0)	**0.1** (0.1)	**0.0** (0.0)
N-NO_3 (mg/L)	**0.0** (0.0)	**7.3** (5.2)	**5.9** (4.5)	**0.0** (0.0)	**18.4** (4.0)	**16.0** (4.9)	**0.1** (0.0)	**22.5** (3.7)	**17.5** (3.8)
DO (mg/L)		**2.6** (1.1)	**0.7** (1.1)		**2.9** (1.5)	**1.0** (0.7)		**0.3** (0.3)	**1.1** (1.1)
pH	**7.6** (0.2)	**7.3** (0.3)	**7.4** (0.3)	**7.2** (0.0)	**6.5** (0.3)	**6.6** (0.3)	**7.3** (0.3)	**6.0** (0.6)	**6.7** (0.5)
Alkalinity (mg$CaCO_3$/L)	**381** (54.4)	**217** (69.5)	**244** (72.1)	**357** (33.8)	**95** (23.3)	**106** (42.0)	**365** (43.0)	**79** (30.3)	**99** (46.8)
T (º C)	**25.6** (1.8)	**25.8** (1.9)	**25.7** (1.9)	**27.0** (0.6)	**27.1** (0.6)	**27.3** (0.7)	**27.3** (1.2)	**27.8** (1.5)	**27.7** (1.3)
COD (mg/L)	**140** (31.1)	**59** (21.6)	**52** (24.5)	**146** (40.3)	**63** (54.8)	**50** (30.8)	**104** (23.8)	**55** (31.1)	**25** (21.5)
Turbidity (UT)	**39.2** (8.1)	**7.6** (7.9)	**2.0** (1.3)	**34.7** (5.0)	**12.2** (9.7)	**5.0** (8.3)	**34.9** (7.0)	**12.1** (16.1)	**4.1** (3.5)
TSS (mg/L)	**32.5** (15.1)	**8.7** (8.7)	**4.5** (7.2)				**27.6** (8.0)		**5.3** (4.2)

It is observed in Table 2 that the system was capable of producing final effluents with low mean values of turbidity (2 – 5 UT) and TSS (4.5 – 5.3 mg/L). The system was also effective in the removal of COD (63 to 76%) reaching final concentrations around 50 mg/L, in phases 1 and 2, and 25 mg/L in phase 3. The increase of the HRT and oxygen flow rate in the AER filter, changes in the ANA effluent quality, and the best performance of ANX filter may be influenced for best efficiency on COD removal during phase 3.

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Table 3. Volumetric loadings of COD and total nitrogen

	COD (kg/m³.d)		Total Nitrogen (kg/m³.d)	
	AER	ANX	AER	ANX
Phase 1	0.51	0.21	0.22	0.11
Phase 2	0.53	0.23	0.20	0.11
Phase 3	0.34	0.24	0.18	0.14

However, the good efficiency displayed by the system in the three phases could have contributed to the inefficiency of denitrification due to the low input concentrations of COD to the anoxic filter which may have been insufficient to meet the demand of carbonaceous denitrifying bacteria. This ability of COD removal is presented by more complex systems than the ones commonly used in the treatment of sewage in Brazil. As an example is the combination of UASB-MBBR presented by Tawfik *et al.* (2010), which reached the same efficiency with a HRT of 13.5 h.

The low rate of denitrification may also be explained by the high concentration of DO. This element is priority in the respiratory chain and may have inhibited the respiration of nitrate.
Analyzing the concentration of dissolved oxygen it was observed average values of 2.6 and 2.9 mg/L in AER filter effluent (phases 1 and 2, respectively) being in the range (1 to 4 mg/L) which Yuan e Gao (2010) observed the nitrification in domestic wastewater, and within the range of 0.8 and 4.0 mg/L stated by Jun et al. (2007) that it is possible to occur simultaneous nitrification and denitrification. The dissolved oxygen values were sufficient to oxidize up to 82% of the ammonia. However, the accumulation of sludge in the interstices, which on one hand may favors the respiration of biomass to get into the endogenous phase, on the other hand may hinders the contact of oxygen with the biomass located in the center of the sludge mass, not allowing a higher ammonia oxidation.

In ANX the average values of dissolved oxygen between 0.7 and 1.1 mg/L were above the 0.3 – 0.5 mg/L recommended by Hocaoglu et al. (2011) as the maximum concentration to achieve denitrification, and well above the value reported by Hu et al. (2010), which observed denitrification of synthetic wastewater in anoxic environment with the absence of dissolved oxygen. Besides, the mean dissolved oxygen in the ANX filter was 1.0 mg/L which Ferreira (2000) indicates as the maximum level prior to the inhibition of denitrification to occurs. Thus the dissolved oxygen concentrations in the ANX filter, combined with low concentrations of COD, may have contributed to the inefficiency of denitrification.

Figure 2 presents the ANOVA factor analysis for the ammonia nitrogen comparing the points of analysis and research phases. It is possible to observe a significant difference ($p < 0.05$) between the collection points, anaerobic effluent (ANA) and aerated effluent (AER), and also among the phases, suggesting that aeration causes a significant impact on the concentration of the $N\text{-}NH_3$. As expected there was no significant difference between the concentrations of ammonia nitrogen from AER and ANX filters ($p > 0.05$). The ANX filter N-NH3 remained almost unchanged, however, it is normal to have a slight decrease as occurred in phases 2 and 3, due to ammonia assimilation by the retained biomass and other complex processes and phenomena.

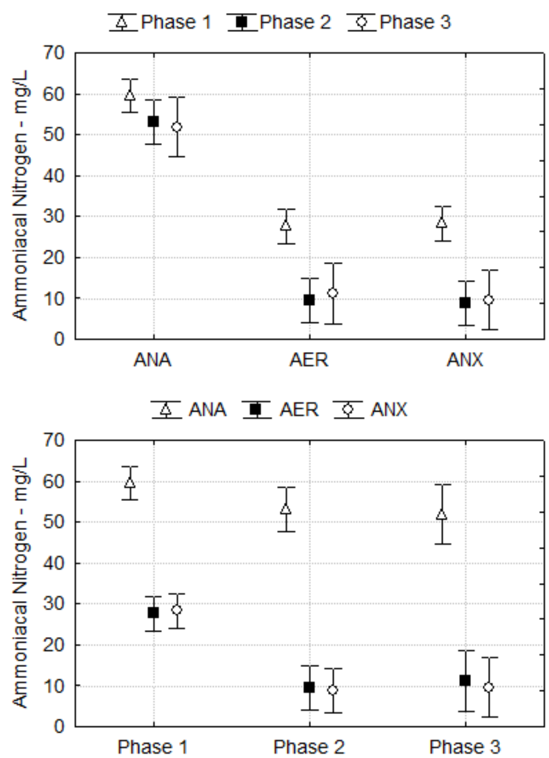

Figure 2. ANOVA factor analysis comparing Ammonia Nitrogen in sampling points and research phases

The increase in 50% in aeration flow rate from phase 1 to phase 2 leads to a significant decrease in the concentration of ammonia nitrogen in AER and ANX filters (Figure 2), to 27.7 mg/L and 28.3 mg/L, respectively (first phase), to 9.4 mg/L and 8.9 mg/L, respectively (second phase), resulting in overall removal of 83%. The increase in HRT of AER filter in phase 03 appears to have caused no impact on final $N\text{-}NH_3$ concentration and, as well as adding 1 m^3/day from the ANA effluent directly to ANX filter, as an additional carbon source, caused no considerable change in the final concentration of $N\text{-}NH_3$.

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The concentration of nitrite was found to be statistically different in AER and ANX filters from phase 01 in relation to other phases. This was due to the untypical accumulation of nitrite in the filters during the first phase, probably caused by the large period of nitrating biomass acclimation. It is noteworthy, however, that there was a downward trend in nitrite concentration at the end of phase 1. Low oxygen concentrations can inhibit nitration resulting in nitrite accumulation in the media. Ruiz et al. (2006) observed that the maximum nitrite accumulation occurred when the dissolved oxygen concentration was between 0.7 and 1.4 mg/L. Ge et al. (2012) argue that the accumulation of nitrite can also occurs during the adaptation of microbial community to substrate type, pH and temperature.

As expected the aeration in all phases was sufficient to produce a nitrate concentration statistically higher than its concentration in the anaerobic step. However, during phase 1, the average nitrate concentration was only 7.3 mg/L and, although nitrification obviously occurred, the results were below expectations and the main factor may have been insufficient aeration to meet the nitrogen demand. Then, an increase in the aeration for the following phases was performed.

Referring to Figure 3, it is noted that denitrification, although occurring, especially during phase 3, the nitrate concentration did not differ statistically from AER and ANX filter effluents in all phases ($p > 0.05$). The most likely cause is the absence of carbonaceous material to efficiently meet the need of responsible bacteria for the conversion of nitrate into molecular nitrogen. In addition, the dissolved oxygen in the ANX filter may have presented peak concentrations that inhibited denitrification. Taking phase 3 as example, it is observed that the $C/N\text{-}NO_3$ relation was 2.45, below the ideal relation of 4.50 proposed by Ivanovic and Leiknes (2011) on works with denitrification of wastewater using MBBR.

Also in phase 3, although it has been observed an increase in denitrifying activity in the ANX filter is possible to say that the prevailing environmental conditions were not efficient to impose a significant denitrification. In this case the problem may have been primarily the failure of supplementary carbon source.

Comparing nitrate concentrations during the three phases of the study (Figure 3), it is noted that the increase in aeration resulted in a substantial increase in nitrate concentration from phases 1 to 2 due to a most efficient ammonia oxidation. However, a detail should be noted, analyzing phase 1, the ANA total nitrogen (62.0 mg/L) was reduced in 42% in the ANX final effluent (36.1 mg/L). A concentration of 1.4 mg/L (Concentration of NO_3 in AER minus concentration of NO_3 in ANX) was removed by denitrification in the ANX filter; another part may have been removed by ammonia volatilization, mainly as a result of the agitation caused by aeration. Another small part of nitrogen may have been assimilated by the biomass retained within the reactor. So, it is likely to have occurred simultaneous nitrification and denitrification in the aerobic filter. Making the same calculation for the phases 2 and 3, there have been removal efficiencies of 54% and 48% nitrogen, respectively. Therefore, even with low capacity of denitrification in the ANX filter, the system still was significantly capable of removing nitrogen compounds being the occurrence of simultaneous nitrification and denitrification the most likely hypothesis for the elimination of much of the nitrogen.

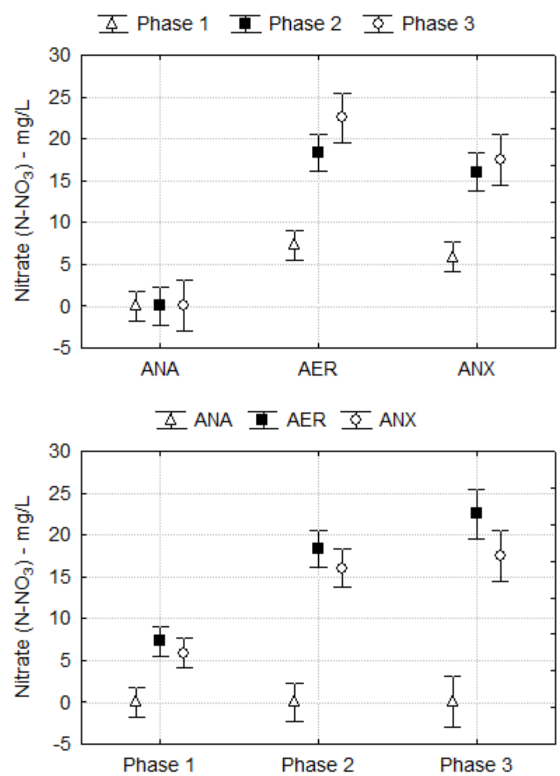

Figure 3. ANOVA Factorial Analysis comparing the Nitrate at sampling points and research phases

The simultaneous nitrification and denitrification is justified because the difficulty of penetration of oxygen inside the concentrated sludge in the interstices and/or its unequal distribution within the reactor, as well as low concentrations of dissolved oxygen, occurrence of anaerobic zones. The processes of nitrification and denitrification may occur in the same reactor as studied by Jun et al. (2007), Zhang and Qi (2007) and Meyer et al. (2005).

The mean concentration of total nitrogen in phases 1, 2 and 3 decreased, respectively, from 62 mg/L (influent) to 36.1 mg/L (effluent); 55.4 mg/L (influent) to 25.7 mg/L (effluent) and 54.4 mg/L (influent) to 28.6 mg/L (effluent), corresponding to removals of 40%, 54% and 47%, respectively, in phases 1, 2 and 3.

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Alkalinity concentrations were significantly higher in ANA when compared with the AER effluent ($p < 0.05$) which is clearly explained by its consumption during nitrification (Figure 4), but because the low denitrifying activity, a proportional generation of alkalinity was not observed and, as a result, the alkalinity concentration were not statistically different in AER and ANX effluents ($p > 0.05$) during all phases (Figure 4). From Figure 4 is can observed that the consumption of alkalinity in phases 2 and 3 was significantly higher than that of phase 1. During phase 1, the average influent alkalinity was 381 mg $CaCO_3$/L, decreased to 217 mg $CaCO_3$/L through the nitrification process in AER, increased slightly during the low activity of the denitrifying bacteria in the ANX, reaching in the final effluent a mean concentration of 244 mg $CaCO_3$/L.

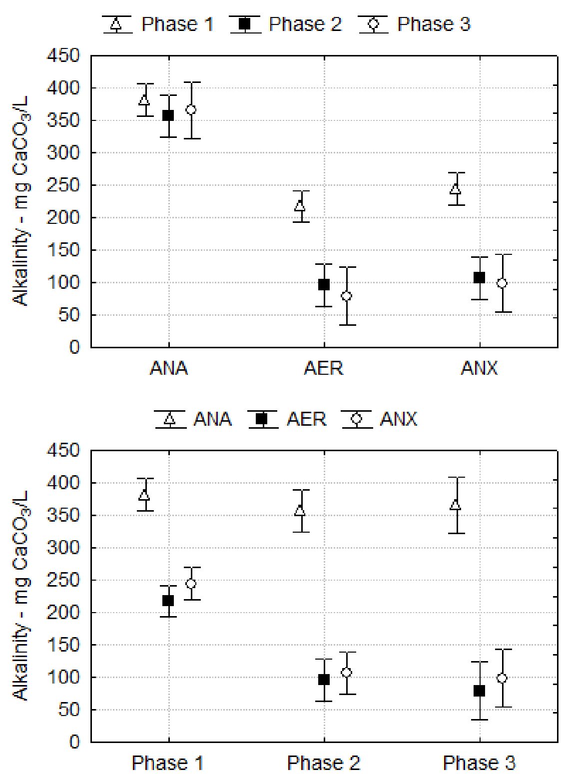

Figure 4. ANOVA factorial analysis comparing the alkalinity at collection points and the phases of the research

Therefore, it can be inferred that 164 mg $CaCO_3$, were consumed to the production of 7.3 mg of N-NO_3 or 22.5 mg $CaCO_3$ were consumed/mg of N-NO_3 produced. On the other hand, for the denitrification to 1.4 mg of N-NO_3 were supplied 27 mg of $CaCO_3$, which means that it was produced 19.3 mg of $CaCO_3$ per mg of N-NO_3 converted into N_2. In phase 2, with the most pronounced nitrifying activity, the consumption of alkalinity was therefore more intense: 262

mg $CaCO_3$/L were consumed to produce 18.4 mg/L of N-NO_3, or 14.24 mg $CaCO_3$ for each mg of nitrate formed. In the ANX filter a reduction of 2.4 mg/L of N-NO_3 was observed, caused by denitrification, and an increase of 10 mg of $CaCO_3$/L, which means that for each mg of N-NO_3 reduced to N_2 were provided 4.2 mg of $CaCO_3$. In phase 3, the relation of consumed alkalinity and nitrate produced was 12.7 $CaCO_3$. The effluent from AER filter showed a mean alkalinity of 79.0 mg $CaCO_3$/L, and thus theoretically still have sufficient alkalinity to oxidize about 6 mg/L ammonia to nitrate. Denitrification provided 19.7 mg $CaCO_3$/L for removal of 5mg/L of nitrate and, therefore, it has been produced 3.94 mg $CaCO_3$/mg of N-NO_3 removed.

Phase 3 showed the highest production of N-NO_3 among the three phases and consequently the greater consumption of alkalinity. However, it had the lowest consumption of $CaCO_3$ per mg of N-NO_3 produced. The consumption of alkalinity in phase 3 is still greater than the theoretical consumption of 7.07 mg $CaCO_3$/mg N proposed by Metcalf & Eddy (2004), but it is similar to 11 mg $CaCO_3$/mg N found by Araújo et al (2009). Among the three studied phases of the average alkalinity supply to the environment in relation to the amount of reduced nitrogen of 3.94 mg $CaCO_3$/mg in phase 3, was the lowest. This value is similar to the theoretical value of 3.57 mg $CaCO_3$/mg N proposed by Metcalf & Eddy (2009).

Conclusions

The aeration flow rate of 0.10 m^3/minute was insufficient to efficiently meet the nitrogen demand under the studied conditions. With the aeration rate 50% higher (0.15 m^3/minute) the ammonia oxidation was more efficient, producing effluent with a final concentration of ammonia around 9 mg/L. Therefore, it was concluded that it was possible to promote the nitrification in submerged biofilter filled with cut conduit, using a simple aeration system, and without sludge removal.

For the operating conditions applied, it was possible to promote significant denitrification without the addition of an external source of substrate for the denitrifying bacteria. The use of effluent from the anaerobic treatment unit as a carbon source provided an increase in denitrification but, according to the statistical tests, the increase was not significant. This result may have been caused by insufficient amount of carbon to supply the nitrogen demand.

A large influence of simultaneous nitrification and denitrification in aerobic environment was observed. This ensured that even with denitrification not significantly occurring in the anoxic filter, the system was able to remove up to 57% of the influent nitrogen. The consumption of alkalinity performed within the considered normal in the literature. The alkalinity in the influent was sufficient to meet the demand of the nitrifying bacteria.

Revista AIDIS de Ingeniería y Ciencias Ambientales:
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