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EFFECT OF AERATION AND RECIRCULATION IN THE REMOVAL OF NITROGEN AND CHEMICAL OXYGEN DEMAND FROM SANITARY SEWAGE IN A STRUCTURED BED REACTOR

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ABSTRACT: This study aimed to evaluate a structured bed reactor with intermittent aeration in the simultaneous removal of Chemical Oxygen Demand (COD) and Total Nitrogen (TN) from sanitary sewage. The reactor was operated for 339 days with a Hydraulic Retention Time (HRT) of 12 hours, temperature of 30 ± 1ºC and continuous feeding. Nine trials were carried out in which three different effluent recirculation flow (Qr), 3, 2 and 1 time the incoming flow, were evaluated in three different aeration times, of 1h, 2h, and 3h, in 3 hour cycles. The results showed that different recirculation flows and aeration times did not influence the removal of COD, which reached 89%±12. It was observed that the efficiency of TN removal was higher in the tests with higher COD/TKN ratio, between 7.2 and 7.8, reaching TN effluent values of 17 mg.L^{-1}, being 5 mg.L^{-1} of N-NH_4^+, 2 mg.L^{-1} of N-NO_2 and 10 mg.L^{-1} of N-NO_3^-. It was verified that it is possible to remove the COD and nitrogen from sanitary sewage in a structured bed reactor with intermittent aeration.

KEYWORDS: bioreactor, wastewater, denitrification, nitrification, SND (Simultaneous Nitrification and Denitrification).

INTRODUCTION

The presence of excess nutrients in water bodies can cause eutrophication processes, which leads to the depletion of oxygen concentration, death of aerobic beings and decrease in the water quality used for water supply. In addition, ingestion of excess nitrates can cause serious public health damage (Du et al., 2015; Wan et al., 2014).

The conventional biological processes used for nitrogen removal have a configuration formed by separate reactors for nitrification and denitrification. In this way, an extra source of energy is needed to provide oxygen during the process of ammonia oxidation, nitrification, and an extra source of organic matter for the denitrification step (Ma et al., 2016).

Recent studies have shown that the use of structured bed biological reactors with intermittent aeration is feasible for the joint removal of nitrogen and COD (Santos et al., 2016; Wosiack et al., 2015; Barana et al., 2013). In this type of reactor, the COD removal, the nitrification and denitrification occur simultaneously and without the need for an external carbon source.

Simultaneous nitrification and denitrification (SND) occur within microbial flocs or biofilms, where nitrifying bacteria are easier found in areas where there is a higher concentration of dissolved oxygen, while denitrifying bacteria are active in areas where the final electron acceptor is nitrite and/or nitrate. Thus, autotrophic and heterotrophic microorganisms, nitrifying bacteria and denitrifying bacteria, may exist in a single granule, positioned in different layers, due to the DO gradient in the flocs (Wei et al., 2014). The physical explanation for this phenomenon is that the SND occurs as a consequence of the oxygen concentration gradient in the biofilm due to the diffusion limitations of the gas in this solid medium (Lochmatter et al., 2013; Pradnya, 2013).

The advantages of using SND systems for nitrogen removal when compared to systems where nitrification and denitrification occur in separate environments are: lower construction costs; the

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operating conditions can be kept constant, since there is no need for different combinations between the aerated zone and the anoxic zone; reduction of oxygen demand, because in the reactor there is generation of anoxic zones where nitrate is used as an electron acceptor; lower biomass generation; there is no need for addition of external carbon source; maintenance of pH levels without addition of external agents, since part of the alkalinity returns to the medium in the denitrification process (Zinatizadeh & Ghaytooli, 2015; Canto et al., 2008).

The objective of this study was to evaluate the concomitant removal of COD and total nitrogen from sanitary sewage in a structured bed reactor and intermittent aeration with different aeration times and effluent recirculation ratio.

MATERIAL AND METHODS

The reactor operated for 339 days without interruption. During this period nine operational conditions were evaluated. As detailed below.

Experimental installation

The reactor used was constructed in acrylic with a useful volume of 9.3 L, height of 85 cm, external diameter of 16 cm and internal of 15 cm. In its interior were arranged 13 cylinders of polyurethane foam, with density of 22g.L^{-1}, in longitudinal direction, with 75 cm of height and 2.0 cm of diameter, which allowed the fixation and development of the biomass (Figure 1).

Operating conditions

The reactor was maintained at 30 ± 1°C with the aid of an aquarium heater connected to a thermostat. The 12-hour HRT was maintained constant with the aid of a Masterflex peristaltic pump model 77800-60 (A1). The recirculation was done by a diaphragm pump; model GmbH-69123, of the brand Prominent Dosier Technik (A2/A4). Aeration was obtained with the use of two aquarium aerators connected to a timer. The reactor effluent was collected by the top of the reactor (A3).

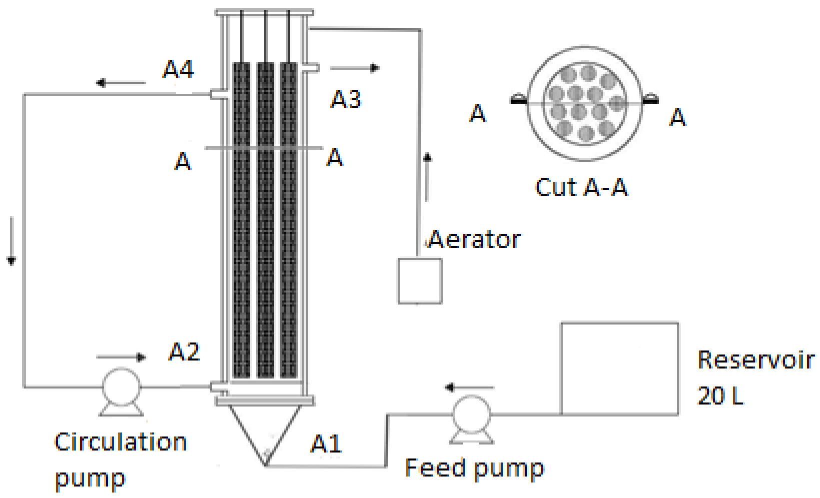

FIGURE 1. Scheme system.

Inoculum

The inoculum used was from an activated sludge reactor of a sewage treatment unit of Sanepar of the city of Curitiba-PR. Their choice was due to their high nitrifying activity. The inoculation was done according to the methodology proposed by Zaiat et al. (1994) apud Moura et al. (2012).

The inoculated cylinders were inserted into the reactor, which operated with 24 hours HRT and constant aeration, until the presence of nitrite and nitrate in the effluent. This phase lasted 13 days, and after that, the reactor started to operate under pre-established normal conditions.

Substrate

To perform the experiment was used as substrate sanitary sewage, collected after the steps of grid and sand removal at a Sewage Treatment Plant of SANEPAR of the city of Ponta Grossa-PR. The collection was done weekly in gallons of 5 L polypropylene. The sewage was kept under refrigeration at 8ºC until the use.

Before being used the influent had its alkalinity corrected with $CaCO_3$, to obtain the ratio of 7.14 mg $CaCO_3$/mg of TKN. The maintenance of this relationship is necessary for the occurrence of the nitrification process.

Physical and chemical analyzes

For monitoring the samples were collected daily and analyzed in triplicate. The following variables were analyzed: pH, alkalinity, chemical oxygen demand (COD), total Kjeldahl nitrogen (TKN), ammoniacal nitrogen ($N\text{-}NH_4^+$), nitrite (NO_2^-) e nitrate (NO_3^-). The alkalinity determination analyzes were performed according to the methodology described by Ripley et al. (1986), the nitrate ones according to Cataldo et al. (1975) and the others were performed according to APHA; AWWA; WEF (1998).

Calculation of nitrification efficiency, denitrification and TN removal

The evaluation of nitrogen removal was done using Equations I to III, which it was possible to calculate the efficiency of nitrification, denitrification and TN removal. In all the equations it was considered that in the influent there was only nitrogen in the organic and ammoniacal forms, since their oxidized forms were not detected in the analyzes performed.

In the calculations of nitrification efficiency (Equation II), the assimilation of nitrogen for the cellular synthesis was not considered, which, according to some authors, can vary from 10 to 20% (Foco et al., 2015). It was also considered the occurrence of complete nitrification, where all nitrite is converted to nitrate. In general, at low nitrogen concentrations, such as those found in sanitary sewage, there is no accumulation of nitrite, except under special conditions, such as low concentrations of DO, high pH and high concentrations of ammonia. However, even in these cases, the microorganisms that oxidize nitrite to nitrate tend to adapt to the environment (Yoo et al., 1999).

$$TNremoval(\%) = \frac{TKNi - TKNe - Nnitrite - Nnitrate}{TKNi} X100 \quad (1)$$

$$Nitrification(\%) = \frac{TKNi - TKNe}{TKNi} X100 \quad (2)$$

$$Denitrification(\%) = \frac{TKNi - TKNe - Nnitrite - Nnitrate}{TKNi - TKNe} X100 \quad (3)$$

where,

TKNi = total Kjeldahl nitrogen influent;

TKNe = total Kjeldahl nitrogen effluent;

N.nitrite = nitrogen in nitrite form,

N.nitrate = nitrogen in nitrate form.

Experimental procedures

The experimental conditions used were defined based on the study developed by Moura et al. (2012) and Barana et al. (2013) (Table 1). Nine trials were evaluated that differed by the aeration time in each cycle of 180 minutes and the recirculation ratio (Qr).

TABLE 1. Operating conditions of the reactor.

Trials	Time of trails (days)	Aeration /No aeration (min)	Qr*
1	52	180/0	
2	46	90/90	3
3	27	60/120	
4	43	180/0	
5	42	90/90	2
6	35	60/120	
7	24	180/0	
8	30	90/90	1
9	40	60/120	

* Qr = Recirculation flow / Input flow

Statistical analysis

In all the results obtained, the Shapiro-Wilk tests were performed to verify if the data presented normal distribution, using the R program. The results were then analyzed statistically by the analysis of variance (ANOVA), where the confidence level of 95% was considered. When there was a significant difference among the treatments, they were compared by Tukey's Test. In order to evaluate the influence that the independent variables, time of aeration and recirculation had on the dependents, after the statistical analysis of variance, the existence of a correlation coefficient was evaluated through the program Statistica 7.0, with confidence level of 95%.

RESULTS AND DISCUSSION

The results presented refer to the averages obtained during the stationary period of the tests performed.

Removal of nitrogen and COD

Table 2 shows the mean values of influent and effluent of the concentration of TKN, $N\text{-}NH_4^+$, COD and the effluent concentration averages of $N\text{-}NO_2^-$ and $N\text{-}NO_3^-$obtained in each test.

The COD influent concentration ranged from 224 ± 21 $mg.L^{-1}$ to 308 ± 156 $mg.L^{-1}$. The results of COD removal indicated that there was no statistical difference between the nine trials at 95% confidence level and presented values from 69 ± 4 to 89 ± 12% (Table 3).

The local legislation determines as release standard of COD values for 125 $mg.L^{-1}$ (Ordinance 1304/2007 - SUDERHSA). Thus, it was verified that the reactor was efficient in the removal of this parameter, which presented maximum concentration in the effluent of 74 ±81$mg.L^{-1}$.

The high efficiency of COD removal in this type of reactor is explained by the fact that organic matter can be removed in both aerobic and anoxic phases. In the aerobic phases, in presence of DO, aerobic heterotrophic bacteria acts, that use DO as final acceptor of electrons. In the anoxic phases, heterotrophic denitrifying bacteria uses $N\text{-}NO_3^-$ as final acceptor of electrons and organic matter as a source of energy and carbon.

It has been observed in studies carried out with this type of reactor, that the efficiency obtained in the removal of COD was always greater than 70%. Barana et al. (2013) used the same reactor design to treat poultry slaughterhouse effluent with COD of 418 $mg.L^{-1}$ and observed COD removal above 88%. Moura et al. (2012), who evaluated the treatment of synthetic sewage with COD of 353 $mg.L^{-1}$ in the same reactor, using HRT of 8 to 12 hours, Qr of 5 and aeration of 120 aerobic minutes followed by 60 without aeration, did not notice difference in the efficiency of COD removal, which varied between 85 and 89%.

Pradnya (2013) verified that the COD removal did not vary with different values of COD/N influent.

The Shapiro-Wilk test showed that the results of COD removal efficiency and TN presented a normal distribution. The Anova test indicated that there was no significant difference between the efficiency results obtained, for a 95% confidence interval, regardless of the conditions employed. Thus, if there was a need to choose one of the tests based on COD removal efficiency, it would be interesting to choose the number 9. The test 9, compared to the others, is the one with the lowest energy consumption, because it has the lowest recirculation rate, Q_r equal to 1, and the shortest aeration time of 60 minutes.

TABLE 2. Average values of COD, TKN, $N\text{-}NH_4^+$, $N\text{-}NO_2^-$, $N\text{-}NO_3^-$ and number of samples analyzed at each stage.

Trails (N)	Influent ($mg.L^{-1}$)			Effluent ($mg.L^{-1}$)				
	COD	TKN	$N\text{-}NH_4^+$	COD	TKN	$N\text{-}NH_4^+$	$N\text{-}NO_2^-$	$N\text{-}NO_3^-$
1 (18)	261±193	61±23	42±2	74±81	21±14	14±19	2±4	19±10
2 (18)	264±28	52±3	39±5	62±23	8±3	3±4	3±3	21±4
3(10)	302±216	49±2	41±6	38±5	5±3	4±3	4±4	27±2
4 (18)	308±156	42±6	29±1	40±36	5±4	3±4	1±1	18±5
5 (18)	298±103	40±5	26±7	30±11	5±3	3±5	1±1	15±3
6 (13)	276±70	39±3	30±6	32±4	5±1	4±1	2±1	10±1
7 (14)	225±40	39±2	30±2	61±19	6±2	5±1	3±1	17±1
8 (17)	232±30	38±2	31±4	70±19	8±2	4±1	2±2	15±2
9 (19)	224±21	39±4	26±3	53±9	8±2	4±1	5±2	16±3

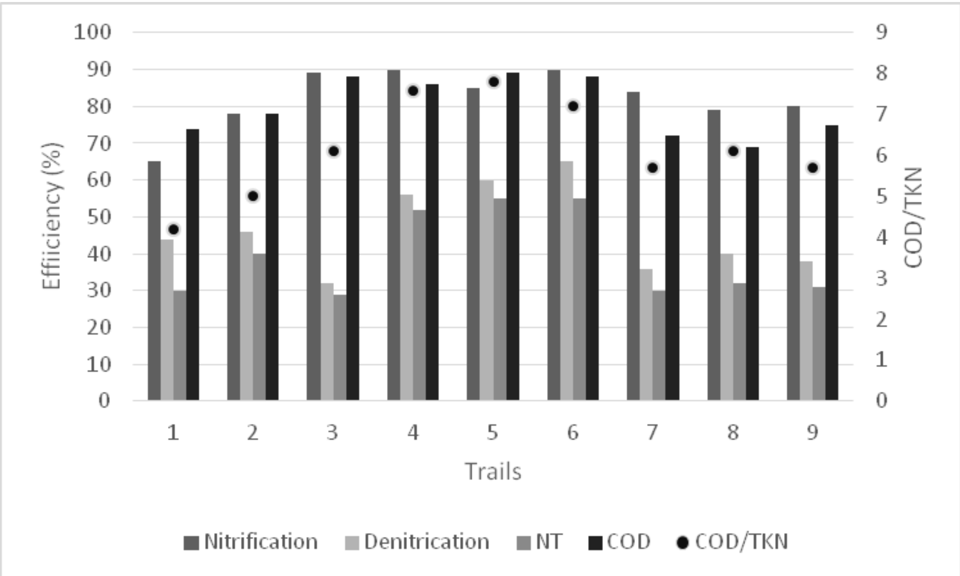

FIGURE 2. Efficiency of nitrification, denitrification, NT removal, COD removal and COD / TKN influent ratio.

The Tukey's test ($p \leq 0.05$) did not show statistical difference between TN removal efficiency results of trials 1, 2, 3, 7, 8 and 9. The TN removal efficiency results of trials 4, 5 and 6 also showed no statistical difference between themselves.

The tests that presented the best TN removal efficiencies were those with the highest C/N ratios of 7.6; 7.8 and 7.2, in trials 4, 5 and 6, respectively. The removal of TN was higher the higher the COD/TKN ratio, indicating that probably the denitrification rates were lower in the tests where organic matter was lacking for the heterotrophic denitrifying bacteria (Figure 2).

Wei et al. (2014) evaluated TN removal by SND with COD/N ratios varying from 3 to 10. They found that TN removal ranged from 48 to 88% for COD/N ratios of 3 to 9. With a COD/N ratio of 10 the efficiency of TN removal decreased to 83%.

Asadi et al. (2016) also observed that TN removal in effluent from dairy and soft drink plants ranged from 39 to 72% when COD/N ratio ranged from 3 to 6.2.

Kummer et al. (2011) evaluated the denitrification of tilapia slaughterhouse effluent, and obtained 100% nitrate removal with a COD/N ratio between 3.2 and 5.4. With a C/N ratio greater than 5.4 and less than 3.2 denitrification efficiency decreased. The reduction of the efficiency in the C/N tests below 3.2 was attributed to the lack of organic matter for the heterotrophic denitrifying bacteria. On the other hand, the reduction of efficiency in values of C/N ratio greater than 5.4 was attributed to the excessive development of other microorganisms, which may have contributed to the organic substrate with the denitrifying bacteria.

It can be verified that the maximum effluent concentration of ammoniacal nitrogen obtained, considering the 9 tests, was 14 $mg.L^{-1}$, a value that would be within the release standards defined by the Brazilian federal legislation for industrial effluents, which is a maximum of 20 $mg.L^{-1}$. Thus, this system can be considered efficient for the removal of ammoniacal nitrogen.

When the correlation coefficient analysis was used for the recirculation, aeration, nitrification, denitrification, TN removal and COD removal, a positive and strong correlation of 0.9691 between denitrification and TN removal, indicating that denitrification was the limiting phase of the process. The correlation between COD removal and denitrification was also strong, 0.6475, indicating that denitrification was in fact influenced by the COD content of the medium.

The influent pH values ranged from 7.30 ± 1.7 to 8.08 ± 0.3. The effluent presented pH values between 6.80 ± 1.8 and 8.00 ± 0.2. These pH values are within the ideal range for the development of nitrifying and denitrifying and heterotrophic aerobic microorganisms (Mayer et al., 2009; Won & Ra, 2011).

Guadie et al. (2013) did not perceive a significant difference in the COD removal and $N\text{-}NH_4^+$ from sanitary sewage in a membrane reactor when the pH ranged from 7.5 to 9.5. He et al. (2014) evaluated the influence of pH values of 7.5 to 9.0 on SND and observed better reactor performance with pH values lower than 8.0.

The influent alkalinity varied between 211 ± 51 and 308 ± 33 mg $CaCO_3.L^{-1}$ and the effluent between 67 ± 12 and 112 ± 81 mg $CaCO_3.L^{-1}$. The influent alkalinity was measured before mixing it with the recirculated.

During the nitrification process, the oxidation of $N\text{-}NH_4^+$ releases H^+ ions to the medium, causing the pH to drop. In this step, every 1 mg of NH_4^+ oxidized in the reactor requires 7.14 mg alkalinity in the form of $CaCO_3$. In the denitrification process there is alkalinity production, unlike nitrification, where there is alkalinity consumption. About 50% of the alkalinity is recovered in the denitrification, where for every milligram of NO_3^- reduced to nitrogen gas (N_2), is generated approximately 3.57 mg of $CaCO_3$ (Won & Ra, 2011; METCALF & EDDY, 2003). During the whole studied period the value of effluent alkalinity indicated that there had been denitrification, since there was a balance of alkalinity. In this way, it is proven once again the occurrence of SND. It can be considered that effluent recirculation helped to maintain buffer capacity of the reactor, since it promoted the dilution of NH_4^+ with NO_3^-, helping in the medium alkalinization.

CONCLUSIONS

The structured bed reactor allowed the removal of up to 55% of TN and 89% of COD, generating effluent with 3 mg.L^{-1} of N-NH_4^+ and 30 mg.L^{-1} of COD.

There was no correlation between the aeration time and COD removal efficiency. In this way, it can be suggested for future applications that the aeration time employed is the lowest studied, so that there is energy savings with aerators.

The different recirculation rates (Qr) studied did not interfere in the COD removal. However, with Qr equal to 2 it was possible to obtain the highest rates of TN removal, from 52 to 55%. With Qr equal to 3 TN removal rates varied from 29 to 40% and with Qr equal to 1 these rates ranged from 30 to 32%. In all the studied trials it was possible to obtain effluent with concentrations of N-NH_4^+ of 3 to 14mg.L^{-1}, indicating the potential of this reactor for nitrogen removal.

It can be concluded that the structured bed reactor with intermittent aeration and recirculation of the effluent was efficient in the joint removal of COD and TN, proving itself as a technically feasible alternative for the removal of nutrients and organic matter in a single compartment.

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