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Influence of operational variables on nitrogen removal in two full scale MBR systems

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ABSTRACT

The influence of the operational variables (sludge retention time, temperature, recirculation rate, and organic loading) on nitrogen transformations in two full-scale pre-denitrification submerged membrane bioreactor (MBR) was investigated. The study was carried out in two predenitrification MBR full-scale plants, (ultrafiltration and microfiltration) with different recirculation rates. Both installations were fully automated and recorded continuously all flows, temperature, transmembrane pressure (TMP), and dissolved oxygen concentration (DO). Sludge retention time (SRT), activated sludge temperature and organic loading varied between 20–43 days, 13–30°C, and 0.40–1.1 kg COD/m^3 h, respectively. Biochemical oxygen demand (BOD_5) and chemical oxygen demand (COD) removal yield were over 99.5 and 95%, respectively. Both MBR systems demonstrated excellent N–NH_4^+ removal with yields concerning 99%, and N–NH_4^+ effluent concentrations lower than 1 mg/L independently of operational conditions. In contrast, the total nitrogen (TN) removal was very influenced by operational variables. The most important influence in nitrate removal for MBR systems was the recirculation ratio between MBR and anoxic bioreactor, which determined the presence of DO in anoxic reactors that affect to the denitrification efficiency. These problems were more significant when activated sludge temperature was low.

Keywords: MBR; Denitrification; Temperature; Recirculation; Total nitrigen

1. Introduction

Nitrogen is one of the major nutrients present in urban wastewater, which can cause problems in ecosystems, such as eutrophication, if it is not controlled on dumpling in water bodies.

In biological wastewater treatment plants, nitrogen removal is performed by nitrification–denitrification processes. Nitrogen removal is usually achieved in an MBR by integrating an anoxic bioreactor in the system [1]. For a predenitrification system the nitrogen is converted to nitrates and nitrites in aerobic reactor and then moves through recirculation to anoxic bioreactor, where in the presence of organic matter is used

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as the terminal electron acceptor and transformed to N_2. The main process in nitrogen removal from wastewater may be through denitrification process.

Various advantages make an MBR process a good alternative to a conventional activated sludge system in nitrogen removal. The handle of operational parameters in MBR allows producing changes in nitrogen removal performance.

The nitrifying bacteria are slow-growing microorganism, Grady and Lim [2] reported that heterotrophic bacteria have maximum growth rates of five times and yields of 2–3 times of that of autotrophic nitrifying bacteria, so, the ratio of nitrifying in all bacterial population should therefore be low in urban wastewater treatment plants. This can limit the nitrite production and therefore nitrogen removal, but MBR can work with high sludge retention time (SRT) and total solid retention, retaining slow-growing microorganisms and improving its populations.

Also MBR can operate with hydraulic retention time (HRT) independent from SRT and can work with high HRT, which favors the nitrogen removal due to higher nitrogen assimilation and an increase in denitrification capacity with increased use of slowly biodegradable organic matter of influent [3].

Microbial growth and activity among other physical/chemical properties of organic matter are significantly affected by temperature conditions [4]. An increase in sludge production increases the nutrient removal by assimilation, removing it from the system. Also, nitrificants growth rate and activity is strongly dependent on temperature, in such a way that the fall in temperature causes a decrease in activated sludge nitrifying activity [5].

The organic load to facilities will present influence on nitrogen removal process as denitrifying bacteria that use nitrate as terminal electron acceptor are heterotrophic bacteria. To carry out this process, external carbon source is necessary, usually supplied by organic matter carried by the influent. This organic material will determine the denitrification capacity of the facility.

However, other operational problems appear in MBR operation in terms of nitrogen removal. The internal activated sludge recirculation from the aerobic to the anoxic zones affects the total nitrogen (TN) removal efficiency. A greater recirculation between tanks fed with more nitrates the anoxic zone and prevents these from escaping in the effluent; however, also carries to the anoxic reactor dissolved oxygen (DO). In membrane bioreactors (MBRs), a high input of air is necessary, both for biological activity and for cleaning membranes [6], but different membrane technologies leads to different aeration processes, membrane cleaning, and recirculation ratios.

This aeration condition affects the DO concentration in the activated sludge, and as a result, the DO concentration in the MBR can easily fluctuate above 4 mg O_2/L [7,8] and part of this oxygen is dragged along to the anoxic zone. In addition, the presence of DO has negative aspects in other parts and processes of the facility, since its presence in the anoxic reactor creates problems in the mechanisms of elimination of nitrogen compounds, particularly in the step of denitrification, as oxygen affects denitrification process by three factors: competitive effect, being more cost-effective energy use as the electron acceptor O_2 [9], for enzyme inhibition [10], which reduce their activity, or genetic [11], avoiding the generation of enzymes. This is even more crucial in situations of low load of organic matter in the influent.

The influence over the nitrogen removal of the operational parameters such as sludge retention time (SRT), temperature, organic loading, and recirculation flow were analyzed in order to determine its influence over biomass assimilation and denitrification process in MBR systems.

2. Materials and methods

2.1. Experimental installation and operating conditions

The experimental installation used in this study were two full-scale MBR installations configured in pre-denitrification mode (Fig. 1), installed at the WWTP of Granada (Spain). Both installations were fed with pretreated real urban wastewater. The first installation was equipped with ultrafiltration membranes (0.034 microns mean pore size) made in polyvinylidenefluoride (PVDF) with a maximum treatment capacity of 120 m^3/day with a recirculation ratio between tanks seven times the permeate flow. The other installation was equipped with microfiltration membranes (0.4 μm medium pore size) made in chlorine polyethylene (PE). Its maximum treatment capacity is 36 m^3/day and recirculation rate reaches five times the effluent flow.

Both installations were controlled by SCADA and fully automated. The plants were equipped with sensors and flow meters for total control and measurement in the installations and recorded continuously all flows, temperature, transmembrane pressure (TMP), and DO. SRT and hydraulic retention time (HRT) were fixed by permeate flow and sludge purge management. The anoxic fraction in tanks was 25% of total volume.

The experiment was carried out during 900 days, where SRT, activated sludge temperature and organic loading vary between 20–43 days, 13–30°C, and 1.1–0.45 kg COD/m^3 d, respectively. A similar HRT of 38 h was kept during the period of study and DO was maintained in the range 0.5–1.6 mg O_2/L in the aerobic and MBRs.

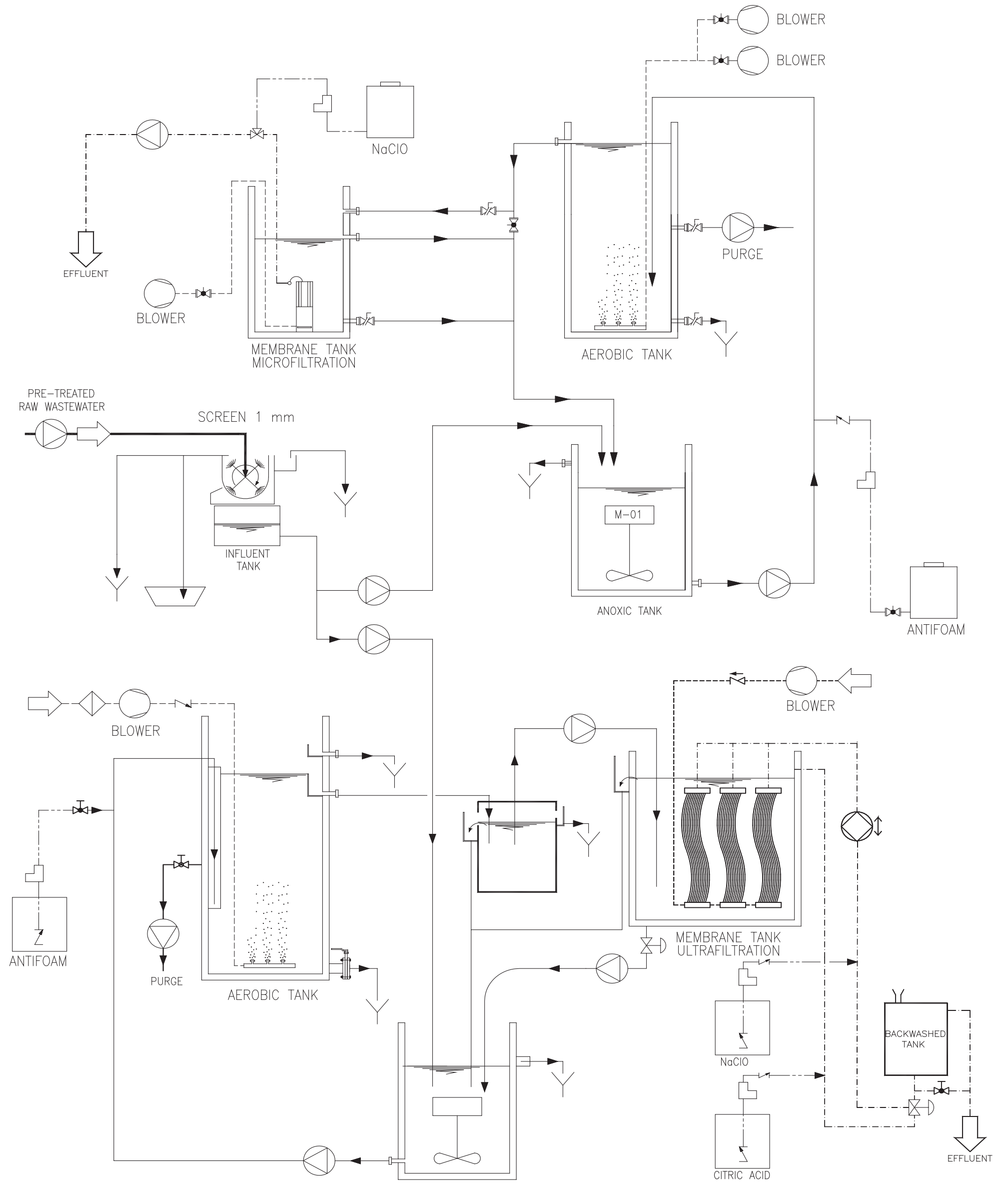

Fig. 1. Schematic flow of the experimental facilities.

2.2. Physical and chemical analysis

Twenty four hours composite samples were collected daily from both the influent and the effluent. All samples were analyzed for total (TSS) and volatile (VSS) suspended solids, total and filterable (0.45 μm) biochemical oxygen demand (BOD_5), chemical oxygen demand (COD), TN and NH_4^+, NO_3^-, NO_2^-. Activated sludge samples were collected daily from each bioreactor to determine TSS and VSS.

TSS were calculated with a gravimetric method using 0.45-μm filters, dried at 105°C and the fixed and VSS solids ignited at 550°C. COD was measured using the COD closed reflux micro method with potassium dichromate and by measuring the absorbance of the digestate colorimetrically at 600 nm. BOD_5 was determined by the manometric method, incubating the sample in darkness at 20°C for five days. Allylthiourea was added to inhibit nitrification. All measures were carried out in accordance with *Standard Methods for the examination of water and wastewater* [12].

NH_4^+, NO_3^-, NO_2^- were measured using ion selective electrodes (Orion 9307BNWP, 9512BNWP and Crisson 96-64 nitrite Electrode). Electrode slopes were automatically determined by using a standard of known concentration. To determine TN, 50 mL of unfiltered diluted sample (1/10) was oxidized at 120°C for 30 min in the presence of boric acid, sodium hydroxide, and potassium peroxodisulphate. The result of the oxidation was analyzed by Merck-Spectroquant analytical kits for NO_3^- (Kit No: 1.14773.0001).

2.3. Statistical analysis

Data obtained in the study were analyzed using the statistical program STATGRAPHICS Plus 3.0 for Windows. An ANOVA test was used to assess homogeneity of variance with a significance level of 5% ($p < 0.05$) of the samples between comparative periods. The least significant differences test (LSD-Test) was used to measure the homogeneity of data between comparative periods.

3. Results and discussion

Nitrogen removal in those MBR systems was carried out in different steps and under different mechanisms. Nitrogen compounds present in the influent can be either assimilated in sludge growth or converted to gaseous nitrogen through nitrification–denitrification process. The cell-assimilated nitrogen can be removed by sludge wasting and the gaseous nitrogen will escape from the MBR to the atmosphere. The remaining nitrogen compounds will remain in the wastewater and leave the MBR system carried out in the effluents.

The values of TN detected were changing during the investigation, as originally detected high values above 80 mg N/L, while subsequently decreased to values around 70 mg N/L and then rise again. The average values were 81.07 mg N/L, with minimum values of 20.88 mg N/L and maximum up to 158.99 mg N/L.

The mean percentage distribution of TN in its components was observed as most of the nitrogen, 95.7%, is in the forms of organic N and NH_4^+ the shapes that make total Kjeldahl nitrogen (TKN), with 3.3% in form of NO_3^- and a 1% of NO_2^-. Comparing these data with NT values detected in urban wastewater characterizations was observed that the influent has medium-high concentrations of TN [13].

It was observed a heavy influent organic loading input to the experimental facilities, with average COD values of 965 mgO_2/L, obtaining a maximum in 2,806 mgO_2/L and minimum of 204 mgO_2/L.

One of the mechanisms for nitrogen removal is the assimilation, in biomass generation. The presence of nitrogen in the biomass is considered a 10% of the total weight [13]. The concentration of influent N required for incorporation into sludge mass was considered equal to the N content of the mass of sludge (VSS) purged daily, according to the next equation.

$$N_{\text{assimilated}} = i_{\text{xvss}} \times X_{\text{vss}} \times Q_{\text{w}} \quad (1)$$

The average values of nitrogen assimilation in both plants were 36.85 mg N/L in the ultrafiltration plant and 22.23 mg N/L in the microfiltration plant, representing a nitrogen removal of 43.92 and 32.7%, respectively. Fig. 2 shows the nitrogen assimilation in biomass during the investigation in both plants.

It was observed that both plants obtain similar nitrogen assimilation, when it works at the same time, and the difference in the average values was based mainly in the first phase when only ultrafiltration plant operated. When both plants worked with the same operational conditions, no statistically significant differences where noticed (p-value = 0.8020).

In the statistical study, the influence of operational parameters in nitrogen assimilation through the comparison of homogeneous groups with the same characteristics shows that the temperature presented a clear influence on nitrogen assimilation. In the comparable groups, statistical differences appear (p-value = 0.00001) with more assimilation in situations with average temperature (15–20°C), being more limited in situations with high or low temperatures (<15°C or >25°C).

The SRT did not present statistically significant differences between comparable groups (p-values of 0.2907 and 0.0691), but shows a tendency to increase assimilation at lower SRT. The organic loading presented influence (p-value = 0.00001) and with higher organic loadings were obtained better results in nitrogen assimilation.

All those effects were related with biomass generation. The increase in biomass generation produces more nitrogen assimilation to the new biomass generated and the effects of the operational parameters on nitrogen removal were related to its effects in biomass generation.

The temperature strongly affects the biomass generation. The increase in temperature present an affection in biomass production and biomass degradation,

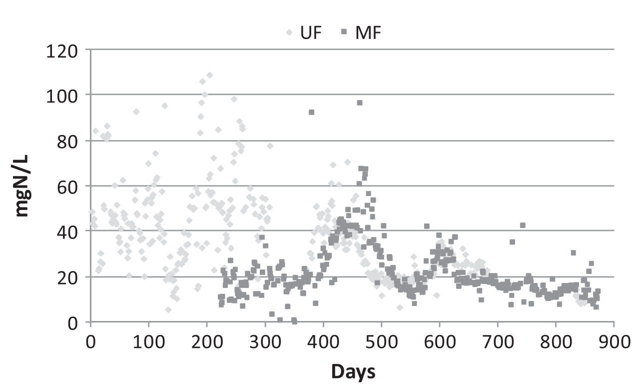

Fig. 2. Nitrogen assimilation.

when the temperature was high (>25°C) the biomass generation was lower and biological activity was high so its produce endogeneous conditions and biomass degradation, and in lower temperatures (<15°C) the biological activity was lower and there were less biomass generation, and less nitrogen assimilation [14,15].

The effect of SRT on biomass generation was limited in these conditions due to the high values of SRT, always over 20 days. Low SRT means greater volumes purged from the system and more biomass generation. The SRT have different effects over b_h and Y_h, both parameters decrease with low SRT, but the relation with b_h was exponential and with Y_h linear, so at low SRT the sludge production was greater [16]. A bigger organic load in the influent increases the sludge generation and nitrogen assimilation due to a bigger substrate load [17].

Of the TN in the effluent, only a small part corresponds to ammonium, with average concentrations of 2.23 mg N–NH_4^+/L for ultrafiltration plant with maximum up 35.98 mg N–NH_4^+/L and minimum of 0 mg N–NH_4^+/L. On the other hand, the microfiltration installation presented ammonium values in the effluent of 0.91 mg N–NH_4^+/L with maximum up to 26.81 mg N–NH_4^+/L and minimum of 0 mg N–NH_4^+/L (Fig. 3), without significant statistical differences between plants (p-value = 0.9935).

The ammonium present in the effluent corresponding to soluble non nitrifiable ammonium fraction present in the wastewater [18], showing a total nitrification of nitrifiable nitrogen.

Under our conditions, statistically significant differences between the values during the different study periods were obtained (p-value = 0.0001) due to operational variables, presenting differenced a statistically significant groups on the days around 450 of operation, which generated different homogeneous groups on the LSD test.

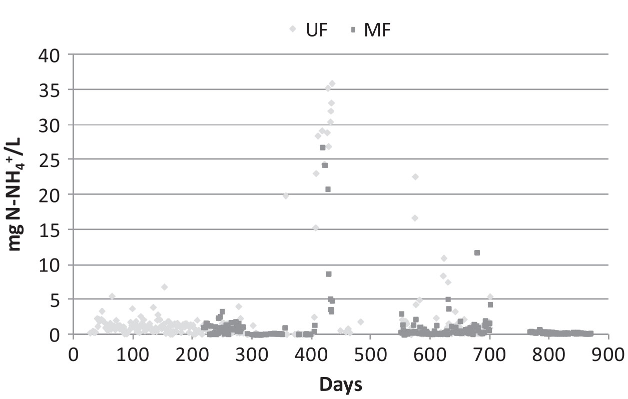

Fig. 3. NH_4^+, concentration in effluents.

Nitrification processes are influenced by the temperature, SRT and DO concentration under which this process are limited. Ekama and Wentzel [18] proposed a simplified expression from a nitrification balance in activated sludge systems to determine the minimum SRT under which nitrification would not occur.

$$\mathrm{SRT}_m = \frac{1}{\mu_{\mathrm{AmT}}(1 - f_x) - b_{\mathrm{AT}}} \tag{2}$$

These components are temperature dependent, which will determine the days required for the nitrification can occur based on the process temperature. To avoid nitrification problems in low temperature periods, it is necessary to keep SRT over the growth rate of the nitrifying bacteria at this specific activated sludge temperature [19]. During the investigation it had worked with a minimum of 20 days SRT and minimum punctual temperatures of 10°C. In the worst temperature conditions, the minimum SRT for nitrification was around 21 days, so we can expect a complete nitrification for both facilities.

Related to DO concentration, the activity of nitrifying bacteria substantially decreases at lower DO levels. Campos et al. [20] claimed that in a nitrifying activated sludge, ammonia was completely oxidized to nitrates at DO levels higher than 1 mg O_2/L, whereas at DO concentrations of 0.4 and 0.6 mg O_2/L ammonia and nitrite accumulation were observed. In these experimental facilities, the DO concentration was kept in the range 0.5–1.6 mg O_2/L in the aerobic and MBRs. This produced a complete nitrification in most of the situations developed. But it can be seen a peak in the concentration of ammonia in the effluent around day 450. This was due to a sudden increase in suspended solids in the activated sludge that produced insufficient aeration and therefore low concentrations of DO, leading to incomplete nitrification [21].

As for denitrification capacity appears starting differences between the two facilities, since not all the nitrate produced in the aerated tank reaches anoxic tanks, but leaves a portion of the effluent defined by recirculation ratios. It was possible to establish a balance to determine the amount of nitrate in the effluent of each installation driven by recirculation.

$$N_{\mathrm{red}} = N_{\mathrm{ox}} \times \frac{R}{1 + R} \tag{3}$$

Ultrafiltration membrane installation had a recirculation ratio of seven times the permeate flow rate (R = 7), so 87.5% of nitrate was susceptible to oxidation in the installation and 12.5% came out in the nitrate form. For microfiltration installation with a recirculation of five times the permeate flow rate ($R = 5$), 83% of the nitrate was susceptible to oxidation in the installation, so 17% left in nitrate form. Fig. 4 shows the measured nitrate concentration in the effluent from both facilities.

In order to know the maximum denitrification capacity of experimental installations, kinetic equation was applied [18] through which we can determine the concentration of $N–NO_3^-$ disposable.

$$D_{p1} = S_{bi}\left[\frac{f_{Sbs}(1 - f_{cv}Y_h)}{2,86} + \frac{K_2 f_x Y_h \mathrm{SRT}}{(1 + b_H \mathrm{SRT})}\right] \quad (4)$$

The application of this equation indicates that the average concentration of $N–NO_3^-$ to reduce by systems were 58.62 and 60.48 mg N/L from ultrafiltration and microfiltration respectively. The nitrogen remaining after assimilation and nitrification were 62.24 and 59.86 mg N/L for ultrafiltration and microfiltration, being capable of total denitrification.

The effluent obtained for microfiltration facility, presented an average concentration of 14.4 mg N/L. This assumes an average yield of 71.16%, although the final concentrations were elevated, being composed primarily of NO_3^-. For ultrafiltration, the effluent obtained had an average concentration of 25.6 mg N/L. This represents an average yield of 52.5%, wherein the effluent comprising mainly NO_3^-. There were clearly significant differences (p-value = 0.00001) for the nitrogen content of the effluent from each of the facilities to compare their lower performance highlighting the ultrafiltration plant.

There was no appreciable influence of the process parameters to study in the case of denitrification (SRT, HRT, temperature, organic load) even if it theoretically should exhibit an influence on the process. This was because its effect was masked by the effect of other variables in the denitrification, as the presence of DO in the sludge.

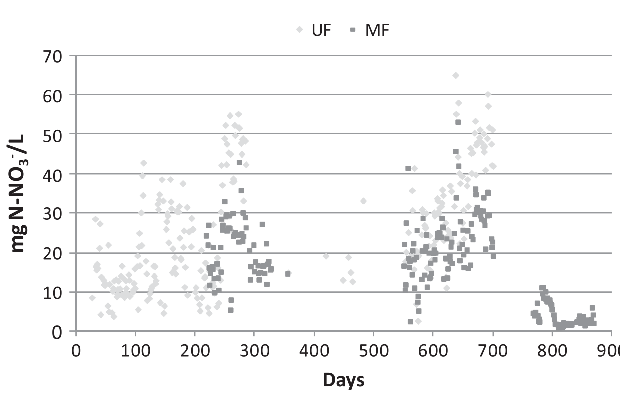

Fig. 4. $N–NO_3^-$ concentration in both effluents.

Both plants had been working in parallel treating the same water and under similar conditions, the main difference between them was the presence of residual DO in the anoxic tank driven from the membrane tank through recirculation. These concentrations were low, between 0.1 mgO_2/L in the best conditions and 0.6 mgO_2/L in the worst case scenario, although slightly higher in ultrafiltration installation due to the higher recirculation ratio. MBR systems presented high aeration rates due to higher sludge concentration and the need of membrane aeration, which leads to high DO concentrations.

The denitrification was supported by two kinetic processes, one on the rapidly biodegradable COD and second kinetic on slowly biodegradable COD. The presence of DO prevents the denitrification because denitrifying bacteria are facultative bacteria that prefer using O_2 as final electron acceptor over nitrate as it is a more effective energetically [9], and also, presenting problems such as inhibition assay [10] or genetic problems in bacteria [11]. Therefore, the oxygen leads competition over nitrate in consumption biodegradable organic matter. To avoid this, it was recommended to increase the available organic matter, which can be done by increasing the second kinetics of denitrification. This should be helped by the increase in the temperature of the facilities, the increase of HRT or increases in the percentage of sludge anoxic fraction in installations [18]. It was also interesting to reduce the amount of DO that reaches the anoxic reactors by reducing recirculation and using more exhaustive control of sludge DO values and membranes aeration.

4. Conclusions

Results show that the operational parameters present a significant effect on nitrogen removal. Temperature presents a clear effect on nitrogen assimilation and nitrification process, being the process more favored at medium temperatures. The assimilation was limited at maximum and minimum temperatures due to its effects on biomass productions, but nitrification was improved at high temperatures.

High SRT had not a significant effect in nitrogen removal. SRT over 20 retention days conditions the effects of this variable, not being a factor in none of the removal mechanisms.

Organic load determine the biomass generation due to the input substrate. In periods with higher loads the nitrogen assimilation was improved and the denitrification potential was bigger, being capable of a greater denitrification.

For nitrification process, it was necessary a correct evaluation of oxygen demands taking into account the sudden increase in sludge concentration. In this case, the operational parameters tested did not show clear effects over denitrification, because it was masked by the effects of another variable, the DO concentration in anoxic tanks. This DO was dragged to the anoxic fraction due to recirculation ratio, and this ratio determines the nitrogen removal. Under the same conditions, the plant with higher recirculation ratio presented more DO in the anoxic reactor and significant lower denitrification rates, with more nitrates in effluent. The DO competes versus nitrate in advantage in organic matter consumption, being necessary actions such as reducing the drag of oxygen.

The oxygen supply in MBR systems requires a precise control of DO supply with sufficient concentrations for complete nitrification and prevent membrane fouling, but trying to minimize their impact on the denitrification process.

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Abbreviations

MBR — membrane bioreactor
SRT — sludge retention time
TMP — transmembrane pressure
DO — dissolved oxygen
HRT — hydraulic retention time
WWTP — wastewater treatment plant
TN — total nitrogen
i_{xvss} — nitrogen content by weight of the MLVSS concentration
X_{vss} — MLVSS concentration
Q_w — sludge purge flow
μ_{Am} — maximum specific growth rate for autotrophic bacteria
b_A — endogenesis constant for autotrophic bacteria
f_x — anoxic fraction of the reactor
N_{red} — denitrifiable nitrogen
N_{ox} — oxidized nitrogen
R — recirculation ratio
D_{p1} — primary denitrification potential
S_{bi} — Influent biodegradable COD
$f_{Sb's}$ — rapidly biodegradable COD fraction related to influent biodegradable COD
f_{cv} — relation VSS/COD
K_2 — second specific denitrification ratio
b_h — heterotrophic endogenous constant
Y_h — Heterotrophic biomass generation rate

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