Influence of hydraulic loading and air flowrate on urban wastewater nitrogen removal with a submerged fixed-film reactor

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Abstract

Nutrient disposal to sensitive areas, particularly nitrogen and phosphorus from wastewater treatment plants, provokes eutrophication reducing water quality. Fixed film technology is widely used for the removal of organic matter and nitrogen by the biological process of nitrification–denitrification. This paper studies a nitrification and post-denitrification lab-scale plant with a downflow aerobic submerged filter for removal of organic matter and nitrification, followed by an anoxic upflow biofilter for denitrification. Recycled construction material (clay shists) was employed as support material and methanol was used as carbon source. After 2 weeks of acclimation in which nitrification reached steady-state conditions, different hydraulic loadings (0.35–$1.59\,\mathrm{m^3/m^2\,h}$) and air flowrates ($7.78$–$43.5\,\mathrm{m^3/m^2\,h}$) were applied for 1 year. The highest hydraulic loading which complied with the EU regulation on nitrogen disposal was $0.71\,\mathrm{m^3/m^2\,h}$ ($1.6\,\mathrm{h}$). Hydraulic retention time (HRT), which corresponded to a nitrogen removal of $0.64\,\mathrm{kg\,N/m^3}$ per day operating at an air flowrate of $25.6\,\mathrm{m^3/m^2\,h}$. Concerning to organic matter removal efficiency, the aerobic reactor accepted a maximum chemical oxygen demand (COD) volumetric loading of $16.0\,\mathrm{kg\,COD/m^3}$ per day with a 75% COD removal efficiency. For all the tests carried out, suspended solids (SS) concentration in the outlet water was less than $35\,\mathrm{mg/l}$.
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Keywords: Submerged filter; Post-denitrification configuration; Hydraulic loading; Air flowrate; Wastewater; Nitrification; Denitrification

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1. Introduction

The abusive use of fertilizers and the discharge of incompletely treated industrial and municipal wastewater to rivers and estuaries, results in a high nitrogen concentration of surface water and groundwater. Nitrates and phosphates can stimulate eutrophication where pollution is caused in waterways by heavy algal growth, as they are both rate-limiting nutrients for the process. Nitrate and nitrite contaminated water supplies are also related with several diseases such as methemoglobinemia infants, also called "bluebaby disease" [1]. These two compounds are also capable of inducing mutations of DNA, causing gastric cancer [2].

Average total nitrogen (T-N) concentration in domestic sewage is approximately 45 mg/l [3]. The effect of eutrophication and health consequences in European countries led the European Union (EU) to introduce a more restrictive regulation concerning nitrogen removal in treated wastewater being discharged to sensitive areas. Fixed film technology has raised as an alternative to conventional activated sludge in order to comply with the EU Urban Wastewater Treatment Directive 91/271/EEC.

Most biological treatment of nitrogen involves a combination of two separate reactors under aerobic and anoxic conditions. Nitrification, biological oxidation of ammonia to nitrite and then to nitrate, is achieved by autotrophic aerobic bacteria with a long generation time; while denitrification occurs, the microbiological reduction of nitrate and nitrite to gaseous nitrogen compound, is accomplished by a wide group of mainly heterotrophic bacteria using nitrate as final electron acceptor. The integration of this biological process is possible in two different configurations: pre-denitrification and post-denitrification [4]. Combination of an anoxic process followed by an aerobic process without addition of any external carbon source and with internal recirculation of treated effluent (pre-denitrification) is more frequently applied. However, and alternative aerobic–anoxic process (post-denitrification) which follows the natural sequence of nitrogen removal can be used too.

Denitrification rate is strongly influenced not only by the carbon content but also by its quality [5]. When the pre-denitrification configuration is applied, the carbon content in wastewater (COD) is used, while an external carbon source must be added to the denitrification reactor. Methanol is found to give high nitrogen removal rates and is thus the less expensive choice [6,7].

Among the different fixed film technologies available nowadays, submerged biofilters are applied worldwide, generally for treatment of urban wastewater. The most important features of this process are that it operates as a high rate biological and mechanical filter in the same reactor, eliminating the requirement for separate secondary clarification. Another important characteristic is the achievement of a high biomass concentration, especially of slowly-growing microorganisms such as nitrifiers, immobilizing them in the biofilm matrix.

This paper presents the experimental results of a new fixed-film reactor response operating downflow for nitrification and upflow for denitrification (post-denitrification configuration), treating urban wastewater and packed with clay schists from recycled construction material. Our main objective was to study the influence of the hydraulic loading and the air flowrate, on the performance of the system in order to meet the terms of the European Directive 271/91 with regard to the removal of organic matter (75%), nitrogen (70–80%) and suspended solids (SS, 90%).

2. Experimental section

2.1. Experimental set-up

A schematic diagram of the experimental set-up is depicted in Fig. 1 and Table 1. The lab scale plant consisted of two identical Plexiglas cylindrical columns in series (2 m high, 6 cm i.d. each). The first biofilter was aerobic and operated downflow. It removed organic matter and oxidized all nitrogen forms to nitrate (nitrifying column). This filter was connected by means of a valve, which allows the separate cleaning of the system, to an upflow anoxic biofilter where methanol was added to achieve denitrification. The system operated first downflow and secondly upflow, working as communicating vessels, needing only one pump to treat the sewage. In addition, organic and suspended solids loading of the influent developed filter clogging in the upper part of the first column, making its backwashing easier than if it were an upflow filter. This also allowed a filtered influent to enter the denitrifying column, making its cleaning cycle less frequent.

Both columns were packed with clay schists from recycled construction material. Process liquid was employed to start-up the system according to Mann et al. [8]. Water and methanol flowrates (a flowrate of 56 m^3/m^2 h was necessary to provide 1 kg dissolved oxygen per hour)
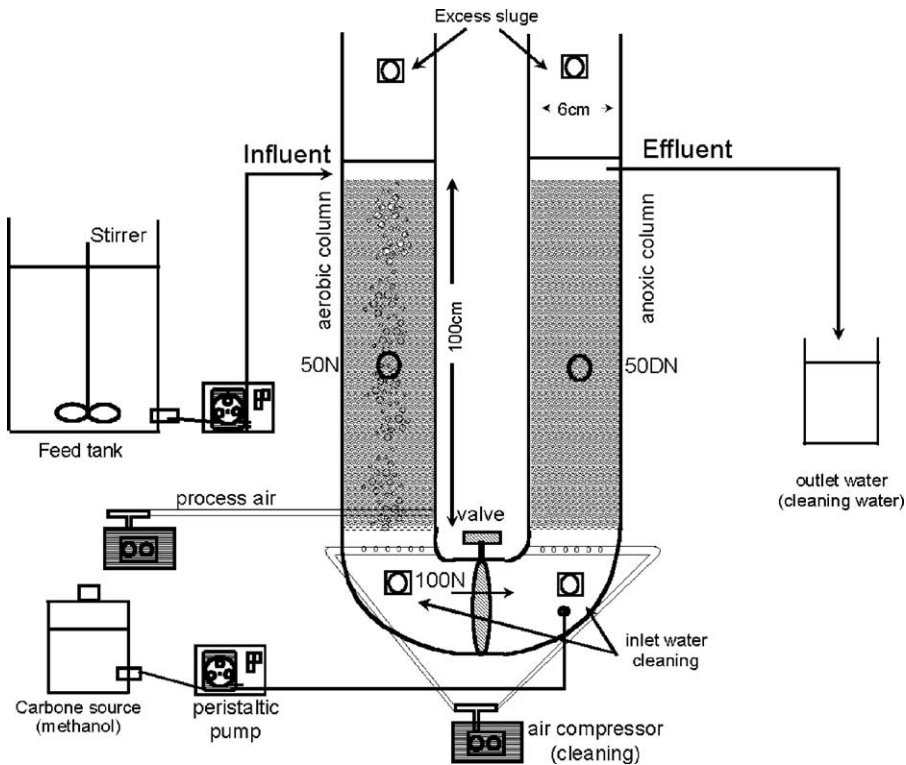

Fig. 1. Lab scale plant process.

Table 1
Operational parameters and capacity of the tested units

Hydraulic loading	0.35–1.59 m^3/m^2 h
Air flowrate	7.8–43.5 m^3/m^2 h
Hydraulic retention time	3.2–0.7 h
Bed height (aerobic–anoxic)	1.0 m
Surface	2.82×10^{-3} m^2
Single bed volume	2.82×10^{-3} m^3
Two bed volume	5.65×10^{-3} m^3
Porosity support material	0.56
Mean diameter support material	5–7 mm
Density	1.75 g/cm^3
Media type	Clay schists
Temperature	22–25 °C

were regulated by peristaltic pumps, Watson Marlow 505S and Ismatec REGLO Analog MS-4/6, respectively. In order to avoid filter clogging of the aerobic bed, backwashing was carried out on a daily basis to remove excess biomass, while the anoxic biofilter was cleaned every 7 days.

2.2. Wastewater composition

The feed water was domestic wastewater from primary treatment of the wastewater treatment plant "Los Vados" EMASAGRA (Granada, Spain). The water to be treated was collected daily at the same time and stored at 4 °C. Its mean composition was: biological oxygen demand (BOD$_5$) 135 ± 13 mgO$_2$/l; chemical oxygen demand (COD) 450 ± 37 mgO$_2$/l; total nitrogen 80 ± 11 mgN/l, suspended solids 120 ± 15 mg/l, total phosphorous 12 ± 2 mgP/l and pH 8 ± 0.3. Water samples were taken from three different locations of the aerobic column: influent (I), 50 cm (50N) and outlet water (100N); and two locations of the anoxic reactor: 50 cm (50DN) and effluent water (E). A methanol solution (1.5%) was applied to the denitrification column, at different flowrates, to achieve a range of concentrations from 0 to 3.5 g methanol/g NO$_3^-$-N.

2.3. Analytical methods

Samples were analysed for SS, COD, BOD$_5$ and T-N in accordance with the Standard Methods for the Examination of Water and Wastewater [9]. Dissolved oxygen (DO) was measured using a Crison Oxi320 oxymeter, temperature and pH were measured with a Crison GLP 22 pH-meter. Merck-Spectroquant analytical kits were used to analyse ammonium (Kit No: 1.14559.0001), nitrate (Kit No: 1.14773.0001), and nitrite (Kit No: 14776). Measurements were carried out by using a Hitachi V-2000 spectrometer. The following physical parameters were recorded periodically: temperature, headloss, pH, DO, water and air flowrates. All measurements were taken three times daily. Differences among the three measurements under the same experimental conditions were negligible, showing less than 5% deviation from the average.

3. Results and discussion

3.1. System start-up and methanol concentration

T-N, COD and SS concentrations in the outlet water were analysed to study the time needed to reach steady-state conditions (Fig. 2). In addition, NH_4^+-N, NO_2^--N and NO_3^--N concentration of the aerobic filter were obtained to determine the period of time needed to establish a biofilm capable of nitrifying inlet nitrogen to nitrate (Fig. 3). The start-up operating conditions were: hydraulic loading $0.35 \, m^3/m^2$ h, air flowrate $29.9 \, m^3/m^2$ h using methanol as carbon source. The reactor was run for 13 days until nitrogen removal reached steady-state conditions; in the case of carbonaceous and solids removal the process took 3 days.

In order to determine the concentration of methanol needed for denitrification, 0–3.5 g methanol/g NO_3^--N ratios were tested (Fig. 4). Different denitrification efficiencies were observed when methanol concentration was increased ($P < 0.001$), removing 98% of nitrate when 2.37 methanol/g NO_3^--N was applied. When methanol concentration was decreased,
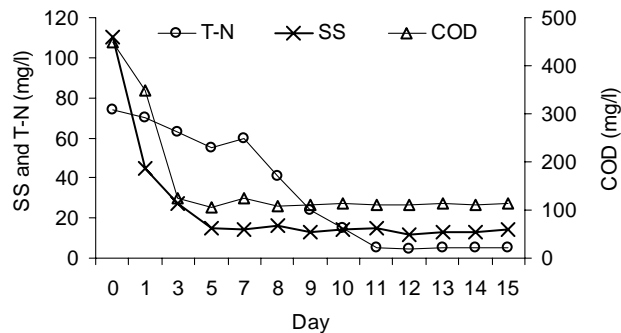

Fig. 2. T-N, SS and COD concentrations in the outlet water during start-up (mean inlet water concentrations for T-N, SS and COD were 80 ± 11, 120 ± 15 and 450 ± 37 mg/l, respectively).
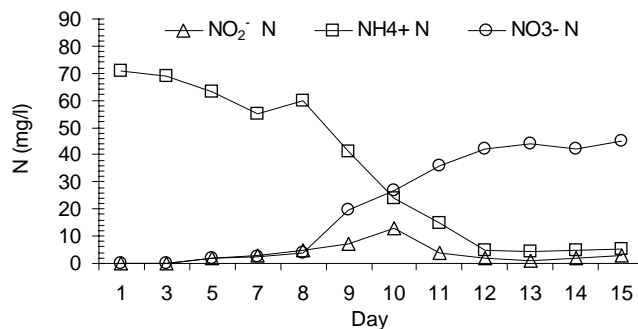

Fig. 3. NH_4^+-N, NO_2^--N and NO_3^--N concentration in the outlet water of the aerobic reactor during start-up (mean inlet water concentrations for NH_4^+, N-NO_2^- and N-NO_3^- inlet water were 75 ± 9, 0 and 0 mg/l, respectively).
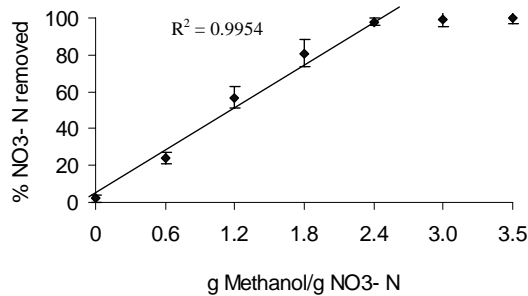
Fig. 4. Effect of methanol/NO_3^--N ratio on denitrification.

nitrate removal efficiency steadily decreased down to 2% in total absence of methanol. This low efficiency reflects the denitrification capacity available from residual COD in the aerobic filter. No linear relation between nitrate removal and methanol/NO_3^--N ration was observed for methanol/NO_3^--N ratio over 2.37, needing 3.0 g methanol/g NO_3^--N to reach 100% nitrate removal. A methanol excess is required to achieve complete denitrification. These results showed similar response as those obtained by other authors [10–12]. Once the system reached stationary conditions, 2.37 g methanol/g NO_3^--N was fixed in order to meet the demand of the system without any methanol leakage in the effluent water. This aspect was important owing to the operational and material costs incurred by this leakage and the increments in COD disposal in the effluent. A high correlation coefficient was found between nitrogen removal and methanol concentrations. Fig. 4 illustrates the linear regressions established for methanol ratio between 2.37 and 0, obtaining a positive slope of 41.4.

The time needed to produce nitrates (Fig. 3) was consistent with that described by [13] who indicated that nitrifying bacteria, need 9 days at 25 °C to attain chemolithoautotrophic growth and manifest its oxidizing activity. Once the nitrate concentration reached stationary conditions, nitrite concentration nearly disappeared; this result is in accordance with Metcalf and Eddy [3], who described that generation time needed for ammonia oxidizers was larger than for nitrite oxidizers; the ammonia oxidation step becomes the rate-limiting step in nitrification.

On the other hand, nitrogen removal obtained under our experimental conditions was not only due to the nitrification–denitrification process. Obviously, there was also an overall loss of nitrogen, which would logically be assumed to be nitrogen assimilation [14], ammonia volatilization [15], adsorption on the biofilm due to different electric charges [16] or through simultaneous nitrification–denitrification in the aerobic column [17]. Neither assimilation, volatilisation, adsorption nor aerobic denitrification were directly measured or individually calculated for this study.

3.2. Nitrogen removal

After start-up, the reactor was operated at different hydraulic loadings starting from 0.35 to 1.59 m³/m² h. Each hydraulic loading was maintained constant as air flowrate was
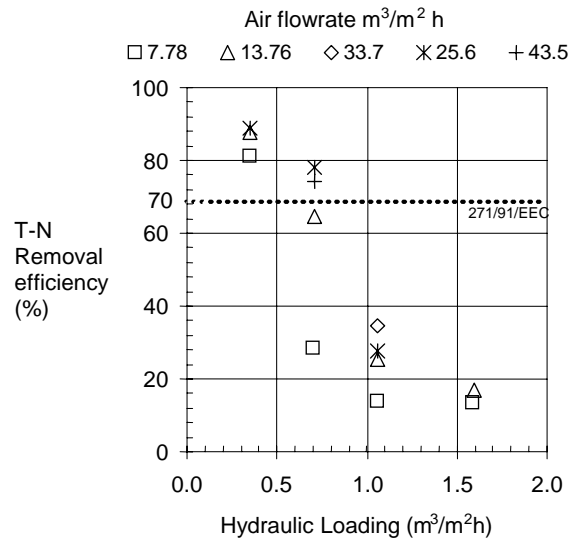
Air flowrate m^3/m^2 h

□ 7.78   △ 13.76   ◇ 33.7   ✳ 25.6   + 43.5

Fig. 5. Steady-state T-N removal efficiency at different hydraulic loadings and air flowrates (mean inlet water concentration for T-N was 80 ± 11 mg/l).

increased from 7.78 to 43.5 m^3/m^2 h, remaining steady-state conditions for 3 weeks. Upon completion of data collection, at each flowrate, hydraulic loading was increased. Due to the similar response of the nitrification at high hydraulic loading, not all the air flowrates were tested for each hydraulic loading. Fig. 5 shows the performance of the system regarding nitrogen removal at different operational conditions. It was clearly manifested that for a fixed air flowrate an increase in the hydraulic loading represented a decrease in T-N removal. Fig. 6 illustrates the progression in the concentration of NH$_4{}^+$-N, NO$_2{}^-$-N and NO$_3{}^-$-N at the sampling points.

Our experimental results indicated that for a hydraulic loading of 0.354 m^3/m^2 h (hydraulic retention time (HRT) aerobic reactor = 1.6 h) and every air flowrate tested (7.8, 13.8 and 25.6 m^3/m^2 h) the reactor achieved complete nitrogen removal. In the case of 25.6 m^3/m^2 h air flowrate, only 50 cm of the aerobic bed was needed for the complete nitrification. For a hydraulic loading of 0.71 m^3/m^2 h (HRT aerobic reactor = 0.8 h), a larger air flowrate was needed to achieve nitrification; for 7.8 m^3/m^2 h (air) no nitrification was achieved; to start producing nitrate 13.8 m^3/m^2 h (air) was needed, whereas to remove 79% of the existing nitrogen 25.6 m^3/m^2 h (air) was required. A higher air flowrate (43.53 m^3/m^2 h, data not shown) was applied to determine if total oxidation of ammonia was achieved in the first 50 cm of the bed. Results confirmed that the whole aerobic filter bed was needed. In addition, when 43.53 m^3/m^2 h (air) was applied, a negative effect was observed due to an occasional biofilm detachment. Furthermore, oxygen concentration at the bottom of the anoxic column reduced denitrification and increased methanol consumption.

With a hydraulic loading rate of 1.1 and 1.59 m^3/m^2 h no nitrification was reported for any of the air flowrates tested. Therefore, HRT of the aerobic column of 0.55 or 0.35 h

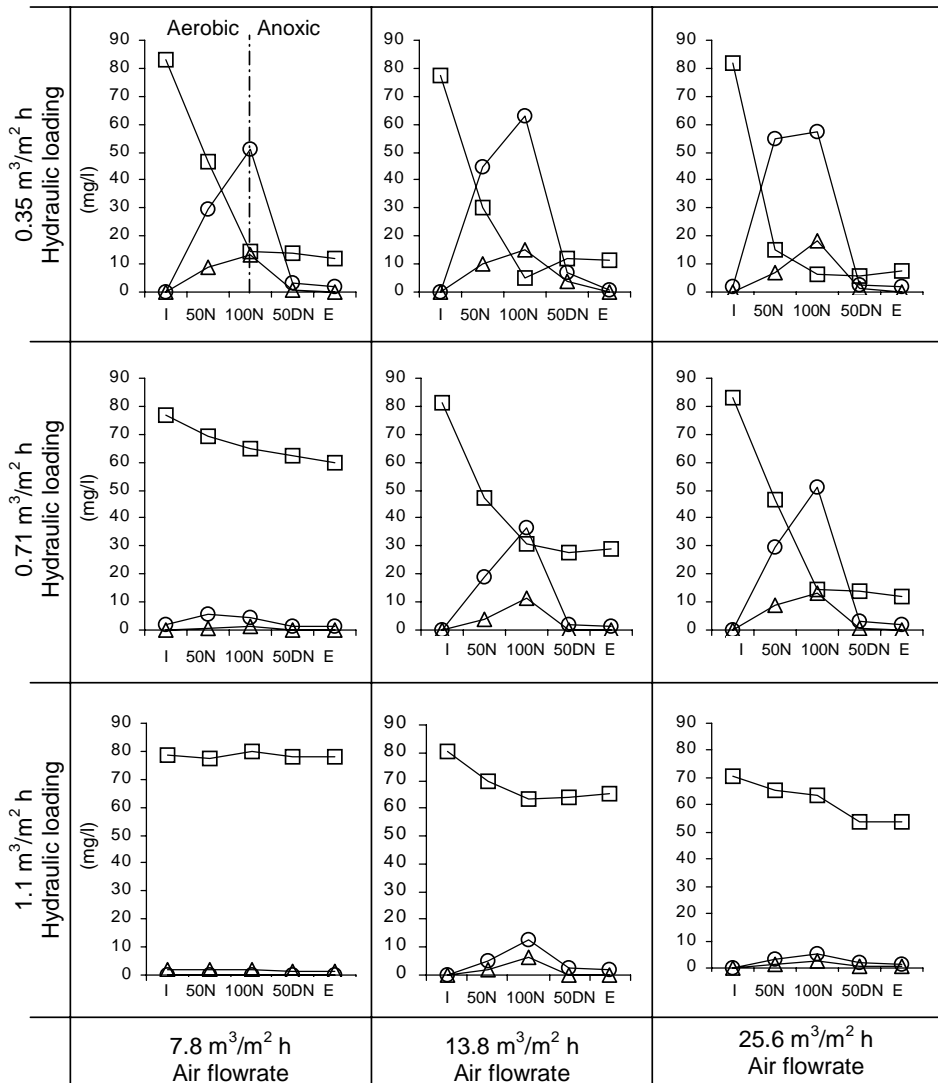Fig. 6. Influence of air flowrate and hydraulic loading on the concentration of NH_4^+-N($\square$), NO_2^--N ($\triangle$), NO_3^--N ($\bigcirc$) at 50 cm of each bed (50N, 50DN), influent (I), outlet of the nitrifying column (100N) and effluent water (E).

were insufficient to nitrify; in this case, nitrogen elimination was only due to ammonia volatilisation, adsorption or microbial assimilation.

Although dissolved oxygen concentration was one of the most important factors affecting nitrification [18], increments in oxygenation of the aerobic reactor could not fulfil the European Directive with hydraulic loadings higher than $0.71\,\mathrm{m^3/m^2}$ h. With aeration below $25.6\,\mathrm{m^3/m^2}$ h not all the inlet nitrogen was oxidized to nitrate, making denitrification inadequate to comply with the regulation.
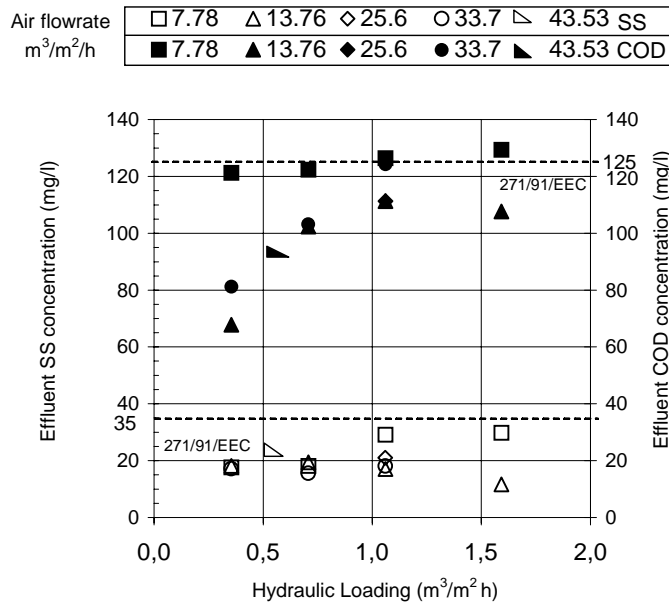
Fig. 7. Steady-state SS and COD concentration (mg/l) in the effluent water at different operation conditions (mean inlet water concentration for SS and COD was 120 ± 15 mg/l and 450 ± 37 mg/l, respectively).

A hydraulic loading of $0.71\,\mathrm{m}^3/\mathrm{m}^2\,\mathrm{h}$, corresponded to a nitrogen removal $0.64\,\mathrm{kg}\,\mathrm{N}/\mathrm{m}^3$ per day with a HRT of 1.6 h. Higher water flowrates coincided with higher organic loadings, which may have favoured the development of heterotrophic bacteria due to their short generation time. According to Grady et al. [19] nitrifying bacteria compete with heterotrophs in the use of oxygen, nutrients and location in the biofilm. Slow growing nitrifiers remain in the inner part of the biofilm making nitrification difficult [20]; this has been corroborated by respirometric tests carried out by Lazarova and Manem [21]. Direct competition between aerobic heterotrophs and autotrophic bacteria can be one of the main causes of the decrease in nitrifying activity [22]. The influent organic loading strongly affected ammonia oxidation, and consequently nitrification should be preceded by organic matter elimination. According to Cheremisinoff [23] and Canziani et al. [24], in order to achieve a high nitrification rate, organic matter concentration should be low to avoid oxygen and nutrients limitation.

For all tests, at 50 cm from the denitrifying column, more than 95% of denitrification was achieved. This was an expected result due to methanol selection of denitrifying rapid kinetics microbiota, which catalysed the reaction faster than nitrification [25].

3.3. COD and SS removal

Carbonaceous removal efficiency, represented as COD removal, is depicted in Fig. 7. These results were obtained working only with the aerobic column, at 100N sampling port.

For every operating condition tested, except for $7.78\,m^3/m^2\,h$ air flowrate and hydraulic loading over $1.1\,m^3/m^2\,h$, COD removal was higher than 75%. A lower carbon removal yield corresponded with reduced aeration, as previously described [26]. According to these results the aerobic reactor working at a hydraulic loading of $1.59\,m^3/m^2\,h$ easily complied with the EU regulation, achieving a maximum volumetric loading of approximately $16.029\,kg\,COD/m^3$ per day removing $12.862\,kg\,COD/m^3$ per day with a HRT of 0.35 h. When nitrification and denitrification was not fully achieved and not all the methanol added was utilized for denitrification, a COD concentration near 175 mg/l was found in the effluent water. Another casual source of increments in COD in the outlet water might be a deficient filter backwashing, due to a biofilm detachment.

Fig. 7 illustrates the relationship between the SS concentrations in the effluent water at different hydraulic loadings. Even with low aeration conditions, all the analysed samples showed less SS concentration than 35 mg/l. Hydraulic loading of $1.59\,m^3/m^2\,h$ did not diminished filter efficiency tolerating a maximum volumetric loading of $2.0\,kg\,SS/m^3$ per day removing $1.6\,kg\,SS/m^3$ per day with a HRT of 0.7 h. These results can be explained by the packed structure of the support material, the low flow velocity of water and the roughness of the biofilm surface which facilitates SS attachment [27]. Other dominant forces that can control the attachment of particles are electrical interactions between charged particles and the charged media surface [28].

4. Conclusions

Influence of hydraulic loading and air flowrate on the removal of organic matter, nitrogen and SS was studied in a post-denitrification system with a downflow–upflow biofilter. Both columns were packed with clay schists from recycled construction material. The following conclusions were obtained.

1. Thirteen days were required to establish a biofilm capable of nitrifying inlet nitrogen to nitrate and denitrify the nitrate, whereas 3 days were enough to achieve carbon and solids removal.
2. An amount of 2.37 g methanol/g $NO_3{}^-$-N was needed in order to meet denitrification demand. When nitrification was not fully achieved, methanol concentration had to be reduced.
3. Operating with an air flowrate range of $7.8\text{–}43.5\,m^3/m^2\,h$, COD removal efficiency complied with the EU Urban Wastewater Treatment Directive 91/271/EEC, removing a maximum of $12.9\,kg\,COD/m^3$ per day with a HRT of 0.8 h at the outlet of the aerobic filter.
4. Hydraulic loading of $0.71\,m^3/m^2\,h$ corresponded to a nitrogen removal $0.64\,kg\,N/m^3$ per day with a HRT of 1.6 h. When hydraulic loading was higher, nitrification was strongly affected in the first step, oxidation of ammonia to nitrite.
5. Denitrification occurred in the first half of the denitrifying column needing only 50 cm of the bed to achieve more than 95% of nitrate removal.
6. Even with low aeration conditions all the samples analysed showed less SS concentration than 35 mg/l in the outlet water.

Acknowledgements

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