Nitrogen removal in a submerged filter with no effluent recirculation

P.C. Chui*, Y. Terashima, J.H. Tay*, H. Ozaki**, and S. Jeyaseelan***

* School of Civil & Structural Engineering, Nanyang Technological University, Nanyang Ave., Singapore 639798

** Department of Environmental Engineering, Kyoto University, Japan

Abstract The performance of a partly aerated submerged filter in treating high nitrogenous wastewaters such as industrial effluents after anaerobic treatment was investigated in this study. The filter was operated without effluent recycling and its response to various test conditions of aeration rates, hydraulic loads, COD/N ratios and hydraulic retention times was evaluated. Results indicated that for an influent concentration of 250 mg N/L and for a loading rate of 0.7 kg N/m^3.d, 60% of the nitrogen can be removed. For a reduced loading rate of 0.25 kg N/m^3.d, nitrogen removal efficiency could reach 86%. Denitrification was hypothesized to have taken place inside the support media in the aerated zone of the filter where oxygen was lacking as well as at locations immediately below the air diffuser where an anoxic zone was formed with the back-flowing nitrified substrate.

Keywords Submerged filters; back mixing; aeration; nitrification; denitrification; nitrogen removal; alkalinity

Introduction

Most biological nitrification-denitrification systems have conventionally been operated as suspended growth systems in activated sludge treatment plants for sewage effluent polishing. More recently, the use of attached growth or biofilm systems has gained much popularity as a pre-treatment process for high nitrogenous industrial wastewaters in addition to nitrogen removal from municipal wastewater.

Single submerged filter systems for carbon oxidation and nitrification of low strength domestic effluents with nitrogen concentrations typically less than 20 mg/L have successfully been applied in laboratory and pilot plants (Haug and McCarty, 1972; McHarness *et al*., 1975; Young *et al*., 1975) and full-scale plants (Paffoni *et al*., 1990; Pujol *et al*., 1992). Denitrification of low and high concentration nitrate wastewaters have also been carried out using submerged filters (Smith *et al*., 1972; Jewell and Cummings, 1975). Complete nitrogen removal using two submerged filters in series for nitrification and denitrification was investigated (Jimenez *et al*., 1987; Cecen and Gonenc, 1992; Ryhiner *et al*., 1994). In these systems, nitrified effluent of the nitrification filter was combined with the feed wastewater and fed to the denitrification filter. The performance of a single filter incorporating both the nitrification and denitrification processes have been reported (Rogalla and Bourbigot, 1990; Tallec *et al*., 1997; Ros and Vrtovsek, 1998).

The performance of a combined filter similar to that of Rogalla and Bourbigot (1990) in the treatment of high nitrogenous synthetic wastewaters was investigated in this study. Effects such as aeration rate, hydraulic loading, organic carbon requirements, retention time, and effluent recycling rates on the performance of the filter system were investigated. This paper presents the part of the results when the filter was operated without effluent recirculation.

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Materials and methods

The filter was fabricated from a 2-m tall acrylic column having an internal diameter of 140 mm. Sampling ports were provided at 400 mm intervals along its height and a distribution plate was placed 75 mm above its base for uniform dispersion of the influent feed. The column was randomly packed with a porous glass rings support medium to a height of 1.8 m. The porosity of the packing was 0.87 and the clean bed liquid volume of the filter was approximately 26 L. Aeration was provided from a location 600 mm from the base of the filter by an air compressor.

The filter was started up by seeding with 2 L of settled activated sludge obtained from a domestic wastewater treatment plant. The filter was operated in a constant temperature room at 35°C and was fed with a synthetic wastewater consisting of tap water and the following chemical constituents : glucose (225 mg/L); peptone (500 mg/L); meat extract (350 mg/L); $NaHCO_3$ (1,500 mg/L); $CaCl_2.2H_2O$ (24 mg/L); $MgSO_4.7H_2O$ (27 mg/L); $FeSO_4.7H_2O$ (20 mg/L); NH_4Cl (400 mg/L); and KH_2PO_4 (100 mg/L). The chemical characteristics of the make-up wastewater were : COD = 1,250 mg/L; TKN = 250 mg/L; NH_3-N = 110 mg/L; total phosphorus = 28 mg/L; alkalinity = 900 mg/L as $CaCO_3$; and pH = 8.0.

Profile samples collected from the sampling ports along the reactor height were analyzed for dissolved oxygen concentration, pH, oxidation-reduction potential (ORP), alkalinity, COD, volatile suspended solids, and different species of nitrogen. Soluble samples were filtered through 0.45 μm cellulose membrane filters before testing. Ammonia and nitrite nitrogen was determined by the direct nesslerisation and naphthylamine hydrochloride-sulfanilic acid methods, respectively. Nitrate nitrogen was determined either by the brucine colorimetric or the nitrate electrode method. Soluble samples for TKN analysis were first digested and distilled using the modified semi-micro Kjeldahl method, followed by ammonia determination using the direct nesslerisation method. All the test methods were carried out in accordance with the Standard Methods (1995).

The performance of the filter system was investigated through four series of test runs. In the first test series, different aeration rates were applied such that a wide variation in the average DO in the aerated zone was obtained so as to determine its effect on nitrification and denitrification. The reactor was then subjected to different hydraulic loadings of between 0.25 and 1 kg $N/m^3.d$ by increasing the influent flow rate. The requirements on oxidizing substances for denitrification were evaluated using COD to N ratios ranging from 1 to 10. The last test series investigated the nitrogen removal efficiency of the reactor when operated under different influent flowrates but for the same loading.

Results

Start-up of reactor

After seeding, the filter was fed with influent wastewater at a flow rate corresponding to a theoretical hydraulic retention time of 36 hr. The filter was initially operated as a single-pass flow with no effluent recycling. Effluent DO concentration was maintained above 3 mg/L by adjusting the aeration rates. DO were not present below the aeration location in the un-aerated zone where about half of the influent organic nitrogen was converted to ammonia nitrogen. The nitrification process in the aerated zone was slow because of slow growth rate of the nitrifying bacteria. The build up of nitrate in the filter was gradual and eventually reached a value of 42 mg/L only after three months of operation.

The nitrified effluent was then recycled back to the bottom of the filter to facilitate the growth of denitrifiers in the un-aerated zone. A recycle rate similar to the influent rate was selected. After about a month, both nitrite and nitrate nitrogen concentrations in the effluent were reduced to less than 2 mg/L. Total nitrogen removed by the reactor was about 70%

and the remaining nitrogen being excess ammonia was not nitrified. With the denitrifier population established, the recycling was discontinued.

Performance of reactor

The effect of dissolved oxygen was evaluated through a series of test runs with constant inputs of influent flow rate (at 25 mL/min) and concentrations (COD = 1,250 mg/L; TN = 250 mg/L) for airflow rates ranging from 1 to 7.5 L/min. DO concentrations in the aerated zone typically increased with height above the air diffuser and with increasing air flow rates. The bottom un-aerated zone was generally anaerobic/anoxic, with average ORP value at –200 mV compared with 200 to 300 mV in the aerated zone.

The total nitrogen concentrations (sum of TKN plus nitrite and nitrate nitrogen) along the height of the reactor at various airflow rates are shown in Figure 1. A steady decrease in the TN concentration was observed in the aerated zone as airflow rates increased from 1 to 4.4 L/min. Lowest effluent TN concentrations were attained at optimum airflow rates of 4.4 and 5 L/min with average bulk DO in the aerobic zone at between 3 and 4 mg/L. For DO less than 3 mg/L, ammonia was not oxidized as satisfactory nitrification could not be achieved.

At higher airflow rates than 5 L/min, the concentration of TN increased again. Although ammonia was almost completely nitrified, there was a significant build up in the oxidized nitrogen concentrations. This clearly indicates denitrification inhibition, possibly by the effects of oxygen and the depletion of organic matter (Laursen *et al.*, 1994). Furthermore, higher airflow rates could have also thinned out the biofilms and washed out some of the biomass in the aerated zone, resulting in a lower denitrification activity.

As shown in Figure 1, nitrogen concentrations were relatively constant throughout the aerated zone for all cases, indicative of a well-mixed zone. TN concentrations in the effluent ranged from 54 mg/L under an airflow rate of 5 L/min to 161 mg/L at the lowest airflow rate. The corresponding nitrogen removal efficiencies were from 78% to 35% for a loading rate of 0.33 kg N/m^3.d. These results show that nitrogen removal is possible even without effluent recirculation in the reactor. Denitrification could have occurred in areas below the air diffuser brought about by backflow of nitrified substrate, which was observed throughout the study. Simultaneous nitrification and denitrification could also have taken place in the aerobic zone, inside the support media where oxygen was lacking.

The effect of hydraulic loading on nitrogen removal was investigated by varying the retention times from 6 to 24 hr while keeping substrate concentrations at 1,250 mg COD/L

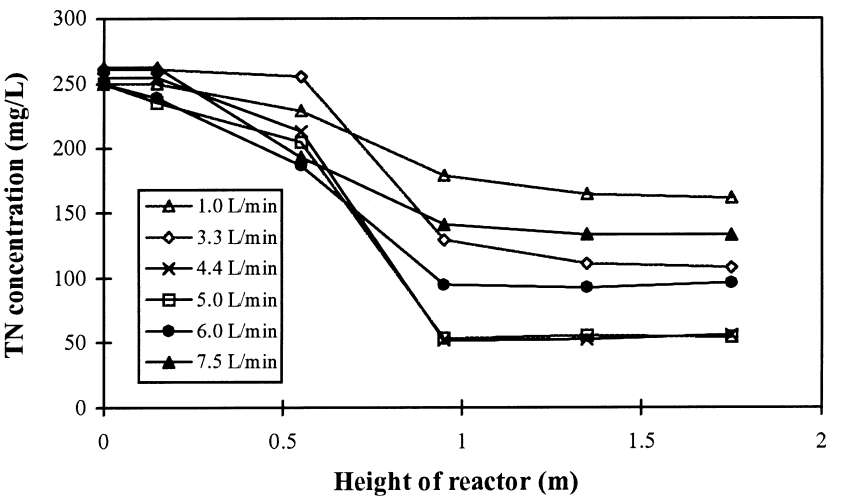

Figure 1 Total nitrogen profiles under various aeration rates

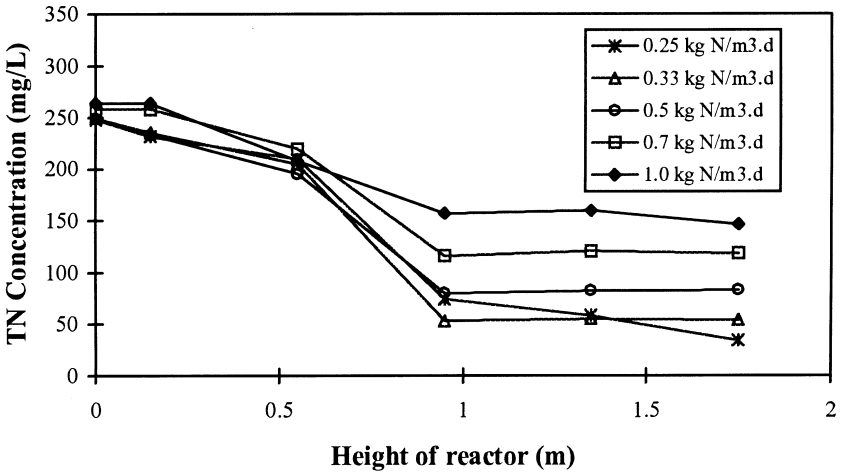

Figure 2 Total nitrogen profiles under various hydraulic loadings

and 250 mg N/L. The resulting nitrogen loading rates applied were between 0.25 and 1.0 kg/m^3.d. The average DO in the aerobic zone for HRTs of 18 and 24 hr was able to maintain at above 3 mg/L. However, for shorter HRTs of 6, 9, and 12 hr, the maximum average bulk DO attainable was only 2.5 mg/L because of the higher organic loadings and inefficiency in the air diffuser system. The variations of TN concentrations with filter depth for the various hydraulic loading rates are shown in Figure 2. The results show that apart from the initial loss of about 15% of nitrogen to ammonia assimilation by heterotrophic bacteria in the anoxic zone, TN concentrations did not change appreciably over the range of hydraulic loading rates. It did, however, increase steadily from 34 to 146 mg/L in the aerobic zone with increasing loading rates. Maximum nitrogen removal of 86% was achieved at the lowest loading rate of 0.25 kg/m^3.d; while at 1.0 kg/m^3.d, only 42% of the influent nitrogen concentration were eliminated.

Test runs in the third series were conducted for COD to N ratios of 1, 2.5, 5, 7.5, and 10, by varying the influent COD concentrations from 250 to 2,500 mg/L while maintaining TN concentration at 250 mg/L. For a HRT of 9 hr, the constant nitrogen loading rate applied to

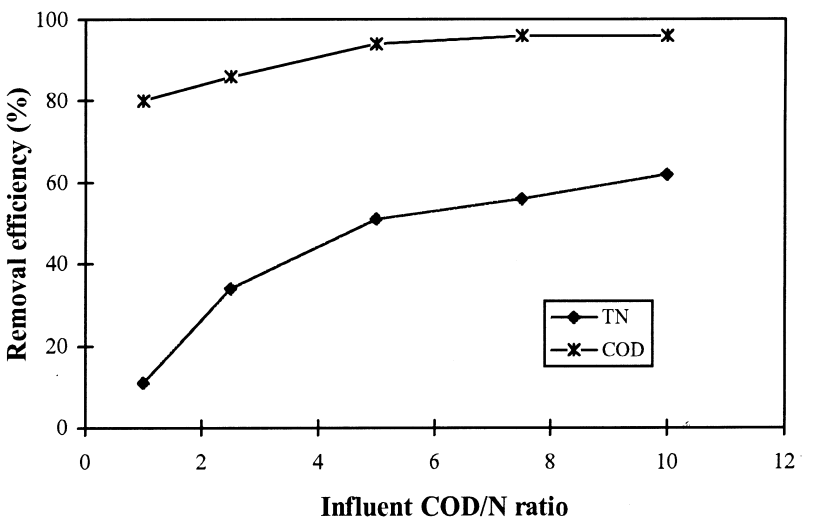

Figure 3 Removal efficiencies at various influent COD to N ratios

the filter was 0.7 kg N/m^3.d. Average DO in the aerobic zone was again kept within the optimum range to minimize the effects of DO.

Summary curves of TN and COD removal efficiencies as a function of the influent COD/N ratios are shown in Figure 3. The results indicate that TN and COD removals increased with increasing COD/N ratios. However, the increase is more appreciable for nitrogen, with removal efficiency increased from 10% to 60% when the COD/N ratio was increased from 1 to 10. On the other hand, COD was almost completely eliminated for all the influent COD/N ratios as a significant amount of the organic matter was removed in the anaerobic zone of the filter.

At the two lowest COD/N ratios, ammonia was completely removed but a significant build up in the oxidized nitrogen concentration of up to 80% of the effluent nitrogen concentration occurred. As expected, denitrification activity was limited due to insufficient COD. The results indicate that the minimum influent COD/N ratio required is about 5. This is higher than the theoretical value of 2.85 g methanol-COD/g nitrate-nitrogen for the denitrification reaction (St. Amant and McCarty, 1969) because of organics removal in the lower anaerobic zone. Higher DO present in the reactor and growth of the bacterial population would also create additional carbon requirement (Smith *et al.*, 1972). However, higher COD/N ratios than necessary are undesirable for the reactor as aeration has to be raised substantially because of the higher COD loadings. This is detrimental to denitrification in the aerated zone for reasons given earlier. A higher organic loading also results in a greater growth of heterotrophic bacteria over denitrifiers in the un-aerated zone, which will lower the denitrification rate.

Finally, four test runs were conducted with HRT at 4, 9, 12, and 18 hr. Loading rates were maintained at 2.5 kg COD/m^3.d and 0.5 kg N/m^3.d by varying the substrate concentration. The other operating conditions remained the same as in the previous tests. The removal efficiencies of nitrogen and COD for the four different HRT investigated are shown in Figure 4. COD removal was not affected by low HRTs and efficiencies were above 90%. The highest nitrogen removal efficiency of 67% was achieved at 12 hr HRT.

In order to determine the alkalinity requirements, alkalinity destroyed (as $CaCO_3$) during the removal of nitrogen for all the test runs was plotted against the ratio of oxidized nitrogen to TN concentration in the effluent as shown in Figure 5. Alkalinity destroyed was obtained by subtracting the effluent alkalinity from the influent alkalinity and total nitrogen

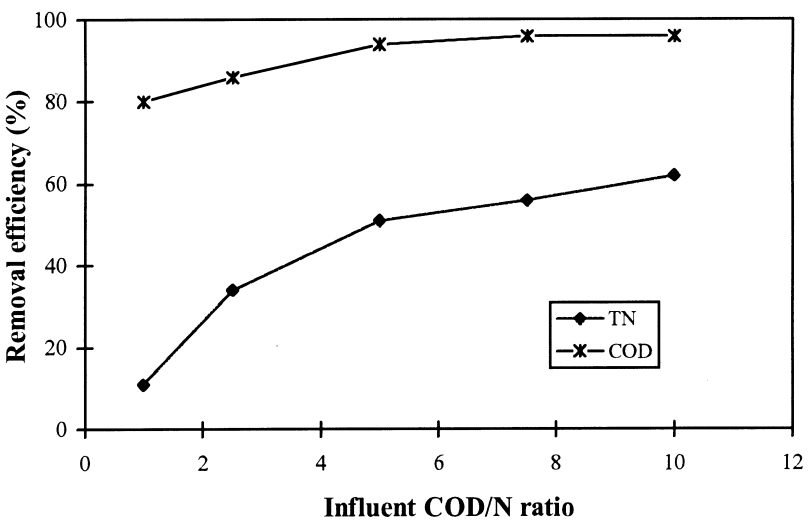

Figure 4 Removal efficiencies at various HRT.

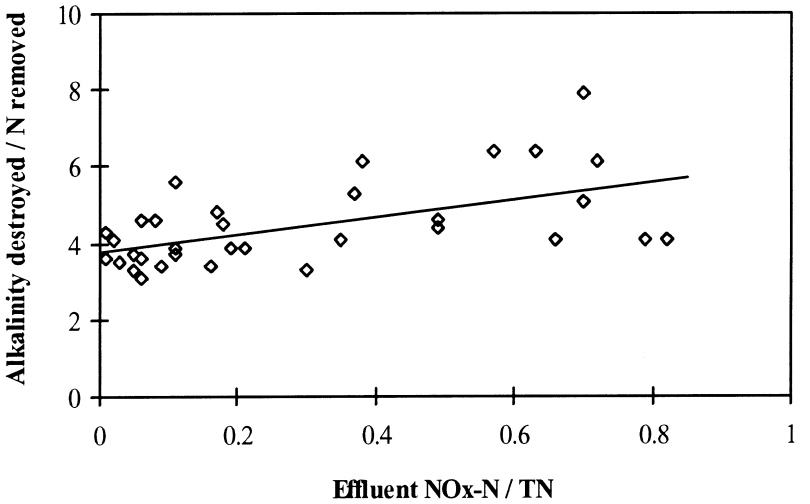

Figure 5 Correlation of alkalinity destroyed/TN removed ratio

removed was similarly computed. At low NO_x-N/TN ratios where denitrification was complete, the alkalinity consumption rate was close to the stoichiometric value of 3.57 g $CaCO_3$ per g of nitrogen removed. At high NO_x-N/TN ratios, denitrification was incomplete and the alkalinity consumption rate was greater than the theoretical value.

Conclusions

The performance of an upflow submerged filter in treating a nitrogenous synthetic wastewater was investigated. The partly aerated biofilter was operated without effluent recycling but denitrification was shown to have taken place in locations where oxygen was absent. Two such locations have been identified in the reactor: inside the support media in the aerobic zone and that immediately below the aeration point. The results of the study indicated that the filter was capable of removing between 60% and 86% of the influent nitrogen concentration of 250 mg/L when operated at the theoretical HRTs of 9 and 24 hr respectively. The corresponding nitrogen loads removed by the reactor were 0.4 and 0.22 kg/m^3.d. COD removals were consistently above 90% for loading rates up to 6.7 kg/m^3.d. However, at COD loads greater than 2.5 kg/m^3.d, nitrogen removal efficiency of the filter was affected as the air diffuser system was unable to provide the optimum average bulk DO of between 3 and 4 mg/L in order to achieve simultaneous nitrification and denitrification in the filter.

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