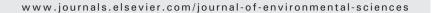


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## Denitrification and biofilm growth in a pilot-scale biofilter packed with suspended carriers for biological nitrogen removal

### from secondary effluent

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#### ABSTRACT

Tertiary denitrification is an effective method for nitrogen removal from wastewater. A pilot-scale biofilter packed with suspended carriers was operated for tertiary denitrification with ethanol as the organic carbon source. Long-term performance, biokinetics of denitrification and biofilm growth were evaluated under filtration velocities of 6, 10 and 14 m/hr. The pilot-scale biofilter removed nitrate from the secondary effluent effectively, and the nitrate nitrogen (NO<sub>3</sub>-N) removal percentage was 82%, 78% and 55% at the filtration velocities of 6, 10 and 14 m/hr, respectively. At the filtration velocities of 6 and 10 m/hr, the nitrate removal loading rate increased with increasing influent nitrate loading rates, while at the filtration velocity of 14 m/hr, the removal loading rate and the influent loading rate were uncorrelated. During denitrification, the ratio of consumed chemical oxygen demand to removed NO<sub>3</sub>-N was 3.99-4.52 mg/mg. Under the filtration velocities of 6, 10 and 14 m/hr, the maximum denitrification rate was 3.12, 4.86 and 4.42 g N/(m<sup>2</sup>·day), the half-saturation constant was 2.61, 1.05 and 1.17 mg/L, and the half-order coefficient was 0.22, 0.32 and 0.24 (mg/L)<sup>1/2</sup>/min, respectively. The biofilm biomass increased with increasing filtration velocity and was 2845, 5124 and 7324 mg VSS/m<sup>2</sup> at filtration velocities of 6, 10 and 14 m/hr, respectively. The highest biofilm density was 44 mg/cm<sup>3</sup> at the filtration velocity of 14 m/hr. Due to the low influent loading rate, biofilm biomass and thickness were lowest at the filtration velocity of 6 m/hr. © 2015 The Research Center for Eco-Environmental Sciences, Chinese Academy of Sciences. Published by Elsevier B.V.

#### Introduction

Conventional secondary effluent in wastewater treatment plants containing high concentrations of nitrate and nitrogen is one of the limiting factors inducing eutrophication in receiving water bodies. Tertiary denitrification of the secondary effluent is an effective method for nitrate removal to control the eutrophication of receiving water bodies (Boltz et al., 2012). During denitrification, oxidized nitrogen is denitrified to nitrogen gas under anoxic conditions, with organic carbon as the electron donor. However,

the amount of biodegradable organic carbon in the secondary effluent is limited. Therefore, external organic carbon should be supplied for tertiary denitrification, and commonly used sources include methanol, ethanol and glucose (Park et al., 2009). Compared with other external carbon sources, denitrification with ethanol has several advantages as follows: (1) short acclimation duration (Nyberg et al., 1996), (2) high denitrification rate (Taljemark et al., 2004; Welander and Mattiasson, 2003), (3) less affected by temperature (Mokhayeri et al., 2006), and (4) less harmful to the environment.

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High-density carriers such as quartz and ceramic sands have been often used as the media for tertiary denitrification (de Barbadillo et al., 2006), while few studies have focused on biofilters with suspended carriers. The main drawback of biofilters with high-density carriers includes high head loss and high energy requirement for backwashing, while the effluent from biofilters with suspended carriers often contains a high concentration of suspended solids (SS), and additional processes are required to remove SS. For example, in the South Caboolture Water Reclamation Plant, a moving bed biofilm reactor (MBBR) with suspended carriers was shown to be able to reduce nitrate to below 1 mg/L, and filtration was subsequently carried out to remove SS (Wilson et al., 2008). In order to remove oxidized nitrogen and SS simultaneously, a new type of biofilter packed with composite suspended and sand carriers was designed for post-denitrification of secondary effluent, and the lab-scale system could remove nitrate and SS efficiently (Shi et al., 2014).

Filtration velocity affects the kinetics of denitrification and biofilm growth, and consequently affects the system performance of the biofilter. At low filtration velocity, a long reaction duration will be required to metabolize pollutants thoroughly, and in addition, due to the low influent loading rate, a biomass-limited condition often exists. At high filtration velocity, the reaction duration is short and denitrification may be inadequate, and the biofilm will be thin with high shear stress. Wei et al. (2014) found that the denitrification rate increased with increasing filtration velocity. In a denitrifying biofilm system, biomass, density and thickness of biofilm affect the substrate conversion rate, thus affecting the denitrification efficiency. For example, in an aerobic biofilm reactor, the penetration depth of oxygen in the biofilm is in general 100-150 μm, therefore, for maximizing aerobically biological processes, the biofilm thickness should not exceed about 150 μm (Tijhuis et al., 1994). The thickness of the biofilm is determined by balancing growth and detachment of biofilm biomass. The substrate loading rate affects the growth of the biofilm directly, and a high substrate loading rate leads to a high amount of biomass and a thick biofilm (Tijhuis et al., 1994). The detachment of biofilm is affected by hydrodynamic conditions, backwashing frequency and the filling ratio of carriers. Neethling et al. (2010) found that a balance existed between the backwashing frequency and the appropriate thickness of biofilm. Melo and Vieira (1999) found that the biofilm density increased with increasing flow rate, and the biofilm density ranged between 14 and 28 mg/cm<sup>3</sup>. Wäsche et al. (2002) showed that the biofilm density increased with increasing sheer stress and substrate load rates, and the biofilm density ranged between 10 and 65 mg/cm<sup>3</sup>. To date, studies on the biokinetics of denitrification and biofilm growth for tertiary biofilm systems have been relatively few, and further investigations are required.

A pilot-scale biofilter packed with composite suspended and sand carriers was operated under filtration velocities of 6, 10 and 14 m/hr (corresponding to empty bed retention times (EBRTs) of 17.8, 10.7 and 7.6 min, respectively) with ethanol as the organic carbon source. Long-term performance of the system and denitrifying kinetics were investigated so as to clarify the denitrification performance for the removal of nitrogen. In addition, biofilm growth was measured so as to correlate it with denitrifying performance.

#### 1. Materials and methods

#### 1.1. Pilot-scale biofilter and its operation

The schematic diagram of the experimental biofilter is shown in Fig. 1. The pilot-scale biofilter was made from a plexiglass column with a diameter of 20 cm and a height of 300 cm, and its effective volume was 91 L. The packed height of suspended carriers (specific surface area of 500 m²/m³, SPR-1 type, Spring, Qingdao, China) was 178 cm. Sampling ports were provided at different heights along the biofilter. There was a pre-mixing zone with a height of 12 cm on the top of the reactor to facilitate the mixing of the secondary effluent and the organic carbon. At the bottom of the biofilter, there was a support gravel stone layer of 10 cm and a quartz sand layer of 20 cm with sizes between 2 and 4 mm for the removal of SS.

The secondary effluent in the 7th Wastewater Treatment Plant, Kunming, China, was used as the feed. During the study, the influent chemical oxygen demand (COD) concentration was 20 mg/L, nitrate nitrogen (NO<sub>3</sub>-N) was 11.2 mg/L, ortho-phosphate (PO<sub>4</sub>-P) was 0.45 mg/L, nitrite nitrogen (NO<sub>2</sub>-N) was 0.25 mg/L, pH was 6.9 and dissolved oxygen (DO) was 2.4 mg/L.

During operation, the external carbon dosage was 5.4 g COD/g  $NO_3$ -N. The treated wastewater and ethanol were fed via peristaltic pumps from the top of the biofilter. The filtration velocity was controlled by the speed of the peristaltic pump. The biofilter was backwashed every 24 hr for 15 min with combined air and water. During backwashing, the water flow rate was 10 L/( $m^2$ -sec) and the air flow rate was 5 L/( $m^2$ -sec). During the start-up period, the filtration velocity was 6 m/hr, and after the system reached steady state and adequate data were collected, it was then increased to 10 and 14 m/hr sequentially.

#### 1.2. Batch experiments

Long-term performance, denitrifying biokinetics of backwashed biofilm biomass and biofilm biomass on suspended carriers, and characteristics of biofilm were tested under the filtration velocities of 6, 10 and 14 m/hr, respectively.

During the long-term operation, parameters such as COD,  $NO_3$ -N,  $NO_2$ -N, ammonium nitrogen ( $NH_4$ -N), pH and DO were tested daily to examine the dynamics of nutrient removal in the biofilter.

Under steady state at each filtration velocity, samples were taken at 0, 23, 53, 88, 118, 148, 178, 208, 238 and 278 cm along the biofilter depth, and concentrations of typical parameters (NO<sub>3</sub>-N, NO<sub>2</sub>-N, COD, DO and pH) were tested so as to investigate the denitrifying biokinetics of the biofilter. At different stages of the experiment, 2 L of backwashed biofilm biomass was taken for batch experiments. Potassium nitrate and ethanol were dosed to achieve the initial NO<sub>3</sub>-N and COD of 30 mg/L and 200 mg/L, respectively. After the beginning of the batch experiment, samples were taken at 5 min intervals. The samples were centrifuged at 12,000 r/min for 2 min, and then the supernatant was stored at 4°C for further analysis of NO<sub>3</sub>-N, NO<sub>2</sub>-N and COD.

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