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# Optimized aeration strategies for nitrogen and phosphorus removal with aerobic granular sludge

*Samuel Lochmatter, Graciela Gonzalez-Gil, Christof Holliger**

*Ecole Polytechnique Fédérale de Lausanne (EPFL), School of Architecture, Civil and Environmental Engineering, Laboratory for Environmental Biotechnology, Lausanne, Switzerland*

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ABSTRACT

Biological wastewater treatment by aerobic granular sludge biofilms offers the possibility to combine carbon (COD), nitrogen (N) and phosphorus (P) removal in a single reactor. Since denitrification can be affected by suboptimal dissolved oxygen concentrations (DO) and limited availability of COD, different aeration strategies and COD loads were tested to improve N- and P-removal in granular sludge systems. Aeration strategies promoting alternating nitrification and denitrification (AND) were studied to improve reactor efficiencies in comparison with more classical simultaneous nitrification–denitrification (SND) strategies. With nutrient loading rates of 1.6 gCOD $L^{-1}$ $d^{-1}$, 0.2 gN $L^{-1}$ $d^{-1}$, and 0.08 gP $L^{-1}$ $d^{-1}$, and SND aeration strategies, N-removal was limited to 62.3 ± 3.4%. Higher COD loads markedly improved N-removal showing that denitrification was limited by COD. AND strategies were more efficient than SND strategies. Alternating high and low DO phases during the aeration phase increased N-removal to 71.2 ± 5.6% with a COD loading rate of 1.6 gCOD $L^{-1}$ $d^{-1}$. Periods of low DO were presumably favorable to denitrifying P-removal saving COD necessary for heterotrophic N-removal. Intermittent aeration with anoxic periods without mixing between the aeration pulses was even more favorable to N-removal, resulting in 78.3 ± 2.9% N-removal with the lowest COD loading rate tested. P-removal was under all tested conditions between 88 and 98%, and was negatively correlated with the concentration of nitrite and nitrate in the effluent ($r = -0.74$, $p < 0.01$). With low COD loading rates, important emissions of undesired $N_2O$ gas were observed and a total of 7–9% of N left the reactor as $N_2O$. However, $N_2O$ emissions significantly decreased with higher COD loads under AND conditions.



## 1. Introduction

The aerobic granular sludge process is a promising technology for biological wastewater treatment. It offers the possibility to remove organic carbon (hereafter referred to as chemical oxygen demand, COD), nitrogen (N) and phosphorus (P) in a single reactor (Zeng et al., 2003; de Kreuk et al., 2005). One of the major challenges of this technology is the efficient N-removal, a two-step process with first, nitrification under aerobic conditions followed by denitrification under anoxic conditions and requiring COD. The association of these two processes in a single stage system is delicate and can result in considerable accumulation of nitrate ($NO_3^-$) or nitrite ($NO_2^-$) in the effluent. Residual effluent $NO_x^-$ concentrations should be

* *Corresponding author*. EPFL ENAC IIE LBE, Station 6, 1015 Lausanne, Switzerland. Tel.: +41 216934724; fax: +41 6934722.
E-mail address: christof.holliger@epfl.ch (C. Holliger).
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minimized to protect the aquatic ecosystem, and to prevent deterioration of the P-removal in reactors operated in sequencing batch mode (Furumai et al., 1999; Peng et al., 2010).

In biofilms of a certain thickness, nitrification and denitrification can occur simultaneously (SND) during aeration (Münch et al., 1996; Keller et al., 1997) since the penetration depth of oxygen is limited due to microbial activity. Hence, the outer layer of aerobic granules is aerobic allowing nitrification and the inner layer anoxic allowing denitrification. The main parameters governing SND in granular sludge are the concentration of dissolved oxygen (DO) in the bulk liquid, the granule size, and the microbial activity. Several studies have shown that a higher DO favors nitrification but limits denitrification, whereas a lower DO enhances denitrification but limits nitrification (Beun et al., 2001; de Kreuk et al., 2005; Mosquera-Corral et al., 2005). For the granule size, a maximal N removal was observed with average diameters of 1.3 mm (de Kreuk et al., 2005). However, no strategy to control the granule size is yet known. Parameters influencing the granule size are the food-to-microorganism ratio (Li et al., 2011) and the hydrodynamics during the mixing phase (Tay et al., 2004).

Besides the spatial dimension, there is also a temporal dimension influencing the oxygen penetration depth in granular biofilm. Modeling studies and micro-sensor measurements have shown that oxygen penetrated much deeper into the granules at the end of the aeration phase than at the beginning because of decreased aerobic microbial activity toward the end of aeration (de Kreuk et al., 2007a,b; Yilmaz et al., 2008). Once COD oxidized and ammonium completely nitrified, oxygen fully penetrates the biofilm (Meyer et al., 2005) which also stops denitrification because of inhibition by oxygen.

In addition to too high DO, the COD to N and P ratio in the influent wastewater can be a reason for impaired denitrification. In combined N- and enhanced biological P-removal (EBPR) systems, the COD concentration in the influent is often the limiting parameter (Kuba et al., 1996; Keller et al., 1997). During anaerobic feeding, polyphosphate-accumulating organisms (PAO) and glycogen-accumulating organisms (GAO) store carbon as polyhydroxyalkanoates. These intracellular stocks of COD allow denitrification during the aeration phase in the anoxic parts of the biofilm. Denitrification by PAO with concurrent ortho-phosphate uptake is called denitrifying P-removal and allows saving COD.

From an operating perspective, lowering the DO has been proposed as solution to improve SND in aerobic granular sludge (Beun et al., 2001). This has been confirmed by lab-scale tests (de Kreuk et al., 2005), however it has also been reported that a lower DO can lead to granule disintegration (Mosquera-Corral et al., 2005). Besides a lower DO, the addition of an anoxic phase after aeration has been tested to remove accumulated $NO_x^-$ (Kishida et al., 2006; Yilmaz et al., 2008; Adav et al., 2009). The introduction of an anoxic phase leads to a temporal separation of nitrification and denitrification. Instead of SND, the main N-removal occurs via alternating nitrification and denitrification (AND). A modeling study has come to the conclusion that such AND strategies would have a higher N-removal potential than SND (Xavier et al., 2007). Drawbacks of a post-anoxic phase can be that no more COD is available for denitrification, or that P is re-released. Hence, another AND strategy was proposed, combining alternating anoxic/oxic conditions with step-feeding (Chen et al., 2011). The step-feeding provided COD for denitrification. With this strategy denitrification could be markedly improved, however the study focused on COD and N-removal only without including biological P-removal.

The present study investigated the potential of different aeration strategies to reach maximum efficiency of both N- and P-removal. We tested two strategies for optimized SND and two for AND. The AND strategies were designed to have an alternation of aerobic/anoxic phases during the famine phase and not only an additional post-anoxic phase. In addition, we also varied the influent COD concentrations to detect limitations due to lack of organic substrate.

## 2. Materials and methods

### 2.1. Reactor setup

Two similar bubble-column SBRs were used, with designs described in Ebrahimi et al. (2010) and Weissbrodt et al. (2012), respectively. The first had an internal diameter of 5.2 cm and a working volume of 2.4 L, whereas the second had an internal diameter of 6 cm and a working volume of 3.5 L. The effluent withdrawal point was positioned at a height of 54 and 61 cm, respectively, resulting in equal volumetric exchange ratios per cycle of 50%. The SBR cycle length was 3 h with 60 min of anaerobic plug-flow feeding, 112 min of aeration, 3 min of settling and 5 min of withdrawal. The hydraulic retention time was for both reactors 6 h. The temperature was controlled at 20 °C and the pH was regulated between 7.0 and 7.3 by adding 1 mol $L^{-1}$ NaOH and HCl solutions.

The up-flow gas flow rate was maintained constant at 3.6 L $min^{-1}$ with diaphragm gas pumps (KNF Laboport). DO setpoints were varied according to the defined oxygenation strategies. To control DO, an off-gas recirculation system was installed with the possibility to add air or nitrogen gas, as described by Mosquera-Corral et al. (2005). If the measured DO value was outside the range of ±3% of the DO setpoint, air or nitrogen gas was added in order to increase or decrease, respectively, the oxygen content in the recirculated off-gas. The quantity of air or nitrogen gas added was defined by PID and mass flow controllers (Brooks).

### 2.2. Synthetic wastewater composition

The influent consisted of a mixture of two synthetic media and tap water adapted from de Kreuk et al. (2005). For the smaller reactor 130 mL of each medium was diluted with 940 mL of tap water and for the bigger reactor 190 mL of each medium with 1370 mL of tap water, resulting in the same influent concentrations. Medium A was composed of 3.51 g $L^{-1}$ of sodium propionate, 0.89 g $L^{-1}$ of $MgSO_4 \cdot 7H_2O$, and 0.36 g $L^{-1}$ of KCl, and medium B of 1.89 g $L^{-1}$ of $NH_4Cl$, 0.73 g $L^{-1}$ of $K_2HPO_4$, 0.23 g $L^{-1}$ of $KH_2PO_4$, and 5 mL $L^{-1}$ of a trace element solution as described by Vishniac and Santer (1957). For the tests conducted with higher COD loading rates, the concentration of sodium propionate was increased