

OLAND is feasible to treat sewage-like nitrogen concentrations at low hydraulic residence time

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ABSTRACT

Energy-positive sewage treatment can in principle be obtained by maximizing energy recovery from concentrated organics in the side stream, and by minimizing energy consumption for residual carbon and nitrogen polishing in the main stream with partial nitrification and anammox. To test the feasibility of this concept, oxygen-limited autotrophic nitrification/denitrification (OLAND) was tested on sewage-like nitrogen influent concentrations with a lab-scale rotating biological contactor at 25°C. After a long steady-state operation of the reactor treating 537 mg N L⁻¹, the influent ammonium concentration was stepwise decreased to 278, 146, 66 and 30 mg N L⁻¹ over 41, 48, 52 and 60 days, respectively, maintaining a continuous loading rate by a stepwise decrease in hydraulic residence time (HRT). At the two lowest concentrations and HRT (2 and 1 h), volumetric removal rates were still relatively high with 444 and 383 mg N L⁻¹ d⁻¹, respectively. However, due to the absence of an oxygen control system, more nitrification occurred at lower nitrogen levels, yielding removal efficiencies of 51 and 46%, respectively. At 30 mg N L⁻¹, transiting from continuous to intermittent disc rotation decreased the dissolved oxygen concentration from 1.4 to 1.2 mg O₂ L⁻¹, increasing the removal efficiency with 11%. The latter indicated that desired removal efficiencies could be obtained provided sufficient suppression of nitrification. The results of this study show the OLAND feasibility for treatment of low nitrogen levels and HRT, a prerequisite to energy-positive wastewater treatment.

KEYWORDS

Centralized, footprint, nitrification, nitrite oxidizing bacteria, nutrient, sustainable

INTRODUCTION

For wastewaters with an ammonium level below 5 g N L⁻¹ and a relatively low ratio of biodegradable chemical oxygen demand (bCOD) to nitrogen (typically $\leq 2-3$), autotrophic nitrogen removal through partial nitrification and anammox is economically and ecologically the preferred treatment (Mulder 2003). Oxygen-limited autotrophic nitrification/denitrification (OLAND) is a one-stage realization of this process, in which aerobic ammonium-oxidizing bacteria (AerAOB) oxidize part of the ammonium to nitrite in the outer, aerobic zones of microbial biofilm (partial nitrification), while the anoxic ammonium-oxidizing bacteria (AnAOB) subsequently convert nitrite and ammonium to nitrogen gas in the inner, anoxic zones (anammox) (Kuai & Verstraete 1998, Vlaeminck *et al.* 2010). The dissolved oxygen concentration (DO) plays a key role to balance partial nitrification and anammox and to avoid aerobic conversion of nitrite to nitrate (nitrification) by nitrite-oxidizing bacteria (NOB). The main advantages of this process compared to the conventional nitrification/denitrification, which is currently used in sewage treatment, are the low sludge production, the absence of external organic carbon source addition and the decrease of the aeration costs with almost 63% (Mulder 2003).

Conventional activated sludge (CAS) systems are energy-negative. The aeration required for organic carbon and nitrogen removal constitutes about 60-70% of the total energy consumption of a municipal wastewater treatment plant (WWTP) (Zessner *et al.* 2010). In case primary and secondary sludge are treated through anaerobic digestion, the nitrogen-rich digester liquid is usually brought back into the main wastewater line, increasing the nitrogen inlet by 15-20% and therefore decreasing denitrification capacities due to COD limitation (Siegrist *et al.* 2008). In case OLAND is applied for separate sludge liquid treatment (Fig. 1.A), the aeration requirements can be reduced with 25% without influencing the nitrogen loading rate of the CAS step (Siegrist *et al.* 2008). Over the last five years this concept is applied in a significant number of full-scale OLAND applications (Wett *et al.* 2010).

A better energy balance can in principle be obtained by maximizing energy recovery from concentrated organics, and by minimizing energy consumption for residual nitrogen and carbon polishing. In this way, energy neutrality was achieved at an Austrian WWTP, by digesting the sludge from two subsequent activated sludge steps with a high and low loading rate, respectively, and removing nitrogen in the side stream with an OLAND system (Wett *et al.* 2007). Given the high energetic content of the sewage COD however, energy-positive sewage treatment should be possible (Verstraete *et al.* 2009, Kartal *et al.* 2010). This requires an advanced biological or physicochemical COD concentration step to increase energy recovery from anaerobic digestion (Fig. 1.B). Sewage has a typical nitrogen content of about 30-100 mg N L⁻¹ and a typical COD/N ratio between 10 and 15 (Henze *et al.* 2008, Metcalf & Eddy 2003). The advanced concentration step is expected to decrease the COD/N ratio of the main stream to around 2. Indeed, the COD concentration in the main stream will decrease with at least 75% while most of the nitrogen is left in the main stream (Verstraete *et al.* 2009, Kartal *et al.* 2010, Salomé 1990) and a part of the co-captured nitrogen is returned to the main stream through the digestate (Fig. 1.B). Consequently, the resultant stream will have a low COD/N ratio, enabling OLAND treatment without the risk that AnAOB are overgrown by heterotrophic biomass (Lackner *et al.* 2008).

Until now, the OLAND process has been applied for high-strength nitrogen wastewaters (> 0.2 g N L⁻¹) such as landfill leachate and digestate from sewage sludge, specific industrial streams and concentrated black water (Table 2). However, AerAOB and AnAOB have a relatively high affinity for their nitrogen substrates (K_s : 0.05-2.4 mg N L⁻¹; Lackner *et al.* 2008), enabling in principle to treat low-strength wastewaters with a sufficiently high removal efficiency. To obtain reasonably high nitrogen removal rates (400 mg N L⁻¹ d⁻¹), the treatment of low nitrogen levels (< 80 mg N L⁻¹) has to occur at low hydraulic residence times (HRT), in the order of some hours, rendering biomass retention an important requirement. In this study, sewage-like nitrogen influent concentrations were tested in a lab-scale rotating biological contactor (RBC) at 25°C. This is one of the first reports on the OLAND treatment of such low nitrogen concentrations and HRT, a prerequisite to energy-positive sewage treatment.

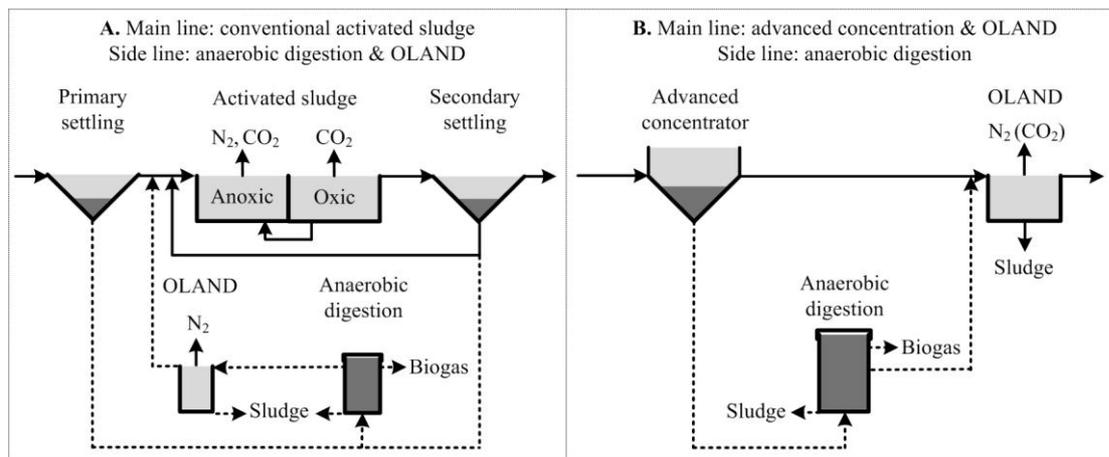


Figure 1. Simplified layout of WWTP with OLAND implementation in the side line (A) and new innovative scheme which includes a first physicochemical or biological concentration of the COD sewage and implementation of the OLAND process in the main line. Full vs. dashed lines display the main vs. side stream, the latter representing around 5-10% of the overall flowrate

METHODOLOGY

OLAND Rotating biological contactor (RBC)

The lab-scale RBC was based on an airwasher LW14 (Venta, Weingarten, Germany) with a rotor consisting of 40 discs interspaced at 3 mm, resulting in a disk contact surface of 1.32 m². The reactor had a liquid volume of 3.6 L, immersing the discs for 64%, and the rotor speed was around 3 rpm. The reactor temperature was set at 25°C and the pH was adjusted to be higher than 7.3 by the addition of NaHCO₃. The dissolved oxygen concentration was not directly controlled.

Reactor operation

The influent of an OLAND lab-scale rotating biological contactor (RBC), as used by Vlaeminck *et al.* (2009) to treat digested black water, was switched to synthetic wastewater consisting of (NH₄)₂SO₄, NaHCO₃, KH₂PO₄ (10 mg P/L) and 2 mL L⁻¹ of a trace element solution (Kuai & Verstraete 1998). After a longer term steady-state operation of the reactor treating 537 mg N L⁻¹, the influent ammonium concentration was stepwise decreased to 278, 146, 66 and 31 mg N L⁻¹ over 41, 48, 52 and 60 days, respectively, maintaining a continuous loading rate (about 840 mg N L⁻¹ d⁻¹) by a stepwise decrease in hydraulic residence time (HRT) (Table 1). Reactor pH, DO and temperature were daily monitored and influent and effluent samples were taken at least thrice a week for ammonium, nitrite and nitrate analyses.

Chemical analyses

Ammonium (Nessler method) and volatile suspended solids (weighing and drying) were determined according to standard methods (Greenberg *et al.* 1992). Nitrite and nitrate were determined on a 761 compact ion chromatograph equipped with a conductivity detector. DO and pH were measured with respectively, an electrode installed on a C833 meter (Consort, Turnhout, Belgium) and a HQ30d DO meter (Hach Lange, Düsseldorf, Germany).

RESULTS

The reactor was operated at an influent concentration of 537 mg N L^{-1} for almost 2 months until stable operation was obtained. The reactor performance after this stabilization period is shown in Figure 2. The influent nitrogen concentration was gradually decreased from 537 mg N L^{-1} to finally 31 mg N L^{-1} . During this experiment the hydraulic residence time gradually decreased from 16 to 1 hours while the total nitrogen loading rate was kept constant due to changes in the influent flow rate (Table 1). The other operational parameters such as pH and DO (Table 1) did not change much during the experiment. However, a significant decrease ($p < 0.05$) of the total nitrogen removal rate was observed over time. Therefore the total nitrogen removal efficiency decreased from $79 \pm 9\%$ in the beginning of the experiment to $35 \pm 7\%$ at the end of phase V^a. Due to a stable ammonium removal rate and subsequent ammonium removal efficiency (Fig 2; Table 1), this decrease in total nitrogen removal was caused by an imbalance in the nitrifying microbial community.

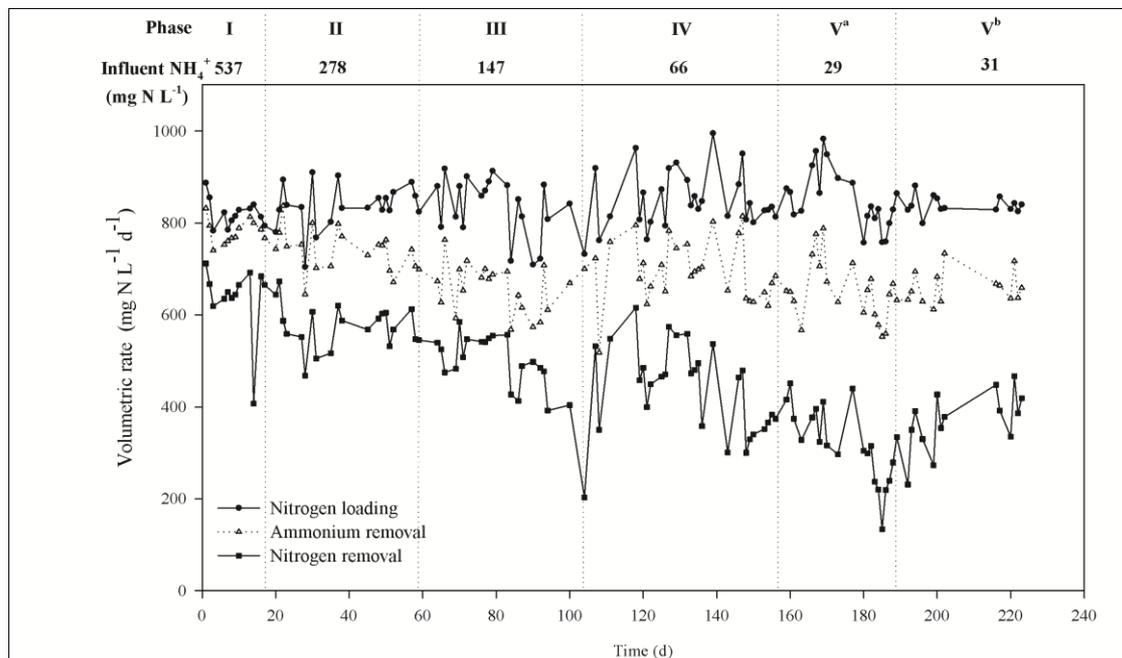


Figure 2. Volumetric loading and removal rates of the rotating biological contactor under a stepwise decrease of the influent ammonium levels from 537 mg N L^{-1} (I) to 278 (II), 146 (III), 66 (IV) and 29 mg N L^{-1} (V^a) under continuous rotation. At phase V^b (31 mg N L^{-1}) intermittent rotation was applied.

Table 1. OLAND rotating biological contactor conditions and performance (average \pm standard deviation) over the periods with stepwise decreases of the ammonium influent concentration and hydraulic residence time (HRT). In periods I-V^a, rotation was continuous, whereas this was intermittent in period V^b. For the nine bottom rows statistical analysis were performed and the phases that were NOT significantly different ($p>0.05$) are indicated with the number of the similar phase. d: days; h: hours; DO: dissolved oxygen level; prod.: production; cons.: consumption

Period	I	II	III	IV	V ^a	V ^b
Duration (d)	21	41	48	52	31	29
Number of samples (-)	14	18	29	36	23	12
Influent NH ₄ ⁺ level (mg N L ⁻¹)	537 \pm 13	278 \pm 11	146 \pm 21	66 \pm 5	29 \pm 8	31 \pm 1
Influent flow rate (L d ⁻¹)	5.4 \pm 0.2	10.5 \pm 0.3	20.5 \pm 1.5	42.9 \pm 2.3	82.6 \pm 2.0	83.6 \pm 0.7
HRT (h)	16.0 \pm 0.5	8.3 \pm 0.3	4.2 \pm 0.4	2.0 \pm 0.1	1.0 \pm 0.0	1.0 \pm 0.0
N loading rate (mg N L ⁻¹ d ⁻¹)	819 \pm 30	840 \pm 49	832 \pm 68	855 \pm 56	851 \pm 66	840 \pm 20
DO level (mg O ₂ L ⁻¹)	1.4 \pm 0.2 ^{III,Va}	1.2 \pm 0.2 ^{IV,Vb}	1.4 \pm 0.2 ^{I,Va}	1.2 \pm 0.1 ^{II,Va}	1.4 \pm 0.4 ^{I,III}	1.2 \pm 0.1 ^{II,IV}
pH (-)	7.6 \pm 0.1	7.5 \pm 0.1	7.3 \pm	7.4 \pm 0.1 ^{III}	7.3 \pm 0.2 ^{III,Vb}	7.3 \pm 0.0 ^{III,Va}
Free ammonia (mg N L ⁻¹)	0.91 \pm 1.58 ^{II,III}	0.40 \pm 0.15 ^{III,I}	0.40 \pm 0.17 ^{I,II}	0.10 \pm 0.03	0.04 \pm 0.02 ^{Vb}	0.04 \pm 0.01 ^{Va}
N removal rate (mg N L ⁻¹ d ⁻¹)	642 \pm 72	565 \pm 42	471 \pm 88 ^{IV}	444 \pm 84 ^{III,Vb}	303 \pm 75	383 \pm 52 ^{IV}
NH ₄ ⁺ removal rate (mg N L ⁻¹ d ⁻¹)	757 \pm 79	737 \pm 48	650 \pm	693 \pm 60 ^{III,Va}	652 \pm 67 ^{III,IV,Vb}	664 \pm 36 ^{III,Va}
N removal efficiency (%)	79 \pm 9	67 \pm 3	58 \pm 9	51 \pm 8 ^{Vb}	35 \pm 7	46 \pm 6 ^{IV}
NH ₄ ⁺ removal efficiency (%)	94 \pm 10	91 \pm 3 ^{IV,Va,Vb}	72 \pm 26	89 \pm 4 ^{II,Va,Vb}	77 \pm 31 ^{II,IV,Vb}	91 \pm 5 ^{II,IV,Va}
NO ₃ ⁻ prod./NH ₄ ⁺ cons. (%)	12 \pm 2	22 \pm 2 ^{IV}	18 \pm 8 ^{IV}	21 \pm 6 ^{II,III}	45 \pm 11	32 \pm 6
Total N effluent (mg N L ⁻¹)	105 \pm 15 ^{II}	93 \pm 8 ^I	66 \pm 24	32 \pm 5	24 \pm 2	20 \pm 2

Figure 3 shows the proportion of the different nitrogen species contributing to the removal of ammonium in the RBC. The contribution of the different processes was calculated assuming that the OLAND process was responsible for the total nitrogen removal and no denitrification occurred. Indeed, there was no organic carbon present in the influent and a relatively high DO concentration was measured in the reactor. The difference between the total nitrogen and ammonium removal rate was caused by an excess AerAOB activity causing nitrite accumulation, nitrate production by AnAOB (11% of the ammonium converted by OLAND) and nitrate production by NOB activity. In the beginning of the experiment 95% of the ammonium removed was converted by the OLAND process and excess AerAOB or NOB activity was negligible (Fig 3). However, during the experiment excess nitrification activity was observed resulting in nitrate production by NOB (nitrification) or nitrite accumulation. Nitrification increased significantly when decreasing the influent ammonium concentration from 537 ± 13 to 278 ± 11 mg N L⁻¹, resulting in an increase of the relative nitrate production (Fig. 3, Table 1). During the following 2 phases, the NOB activity was slightly lower, but due to a high AerAOB activity, a higher nitrite accumulation was observed. Consequently the contribution of the full OLAND process started to decrease over time. In the fifth phase, again an increase in NOB activity was observed and this resulted in a further decrease of the contribution of OLAND and therefore a decrease in total nitrogen removal rate. This resulted in a total nitrogen effluent concentration of 24 mg N L⁻¹ (Table 1), which mainly consisted of nitrate (15 mg N L⁻¹).

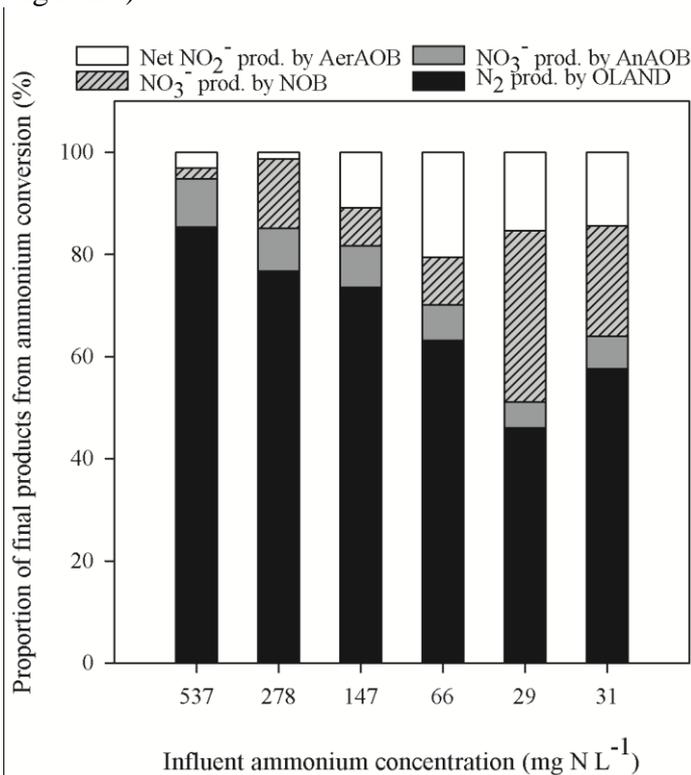


Figure 3. Proportions of the nitrogen species produced from the influent ammonium over the different operational periods: nitrogen gas production through OLAND (nitrogen in – nitrogen out), nitrate production through anammox (11% of ammonium converted by OLAND), nitrite accumulation due to additional nitrification (nitrite out – nitrite in), nitrate accumulation due to additional nitrification (nitrate out – nitrate in – anammox nitrate production)

The free ammonia concentrations decreased sharply over time due to the low nitrogen concentrations and the low pH (Table 1). However, levels were always too low to avoid NOB activity. The very low free ammonia concentration ($< 0.1 \text{ mg N L}^{-1}$), which occurred in phases V^a and V^b allowed for relative nitrate productions above 30% (Table 1). Due to the stable oxygen concentration (always above $1 \text{ mg O}_2 \text{ L}^{-1}$) during the experimental period, no relation between oxygen concentration and nitrate production could be found.

In an attempt to increase the total nitrogen removal efficiency, a discontinuous movement of the rotor was introduced (phase V^b). It was rotated only 1/3 of the time, equally spread over time (1 min on, 2 min off). This intermittent rotation resulted in a significant decrease of the oxygen concentration from 1.4 ± 0.4 to $1.2 \pm 0.1 \text{ mg O}_2 \text{ L}^{-1}$. The total nitrogen removal efficiency increased to the same level as in phase IV ($p > 0.05$) from 35 ± 7 to $46 \pm 6\%$ due to a lower relative nitrate production (Table 1). Due to the lower oxygen concentration it was possible for the anammox bacteria to use a larger part of the produced nitrite (Fig. 3). The excess AerAOB activity decreased while the net nitrite accumulation remained constant. Consequently, the contribution of the NOB decreased. This intermittent aeration caused a decrease in the total nitrogen effluent concentration (20 mg N L^{-1}) mainly due to a decrease in nitrate effluent concentration (11 mg N L^{-1}).

DISCUSSION

Operation of the OLAND RBC on sewage-like nitrogen concentrations (66 and 31 mg N L^{-1}) at low HRT (2 and 1 h), resulted in considerably high nitrogen removal rates ($383\text{--}444 \text{ mg N L}^{-1} \text{ d}^{-1}$). The removal percentages were around 50% and lower than previously reported for this type of reactors (Schmid *et al.* 2003, Pynaert *et al.* 2003), mainly due to the additional nitrification activity, given the lack of DO control in our set-up. According to Flemish standards, the effluent nitrogen concentrations (around 20 mg N L^{-1}) were still slightly above the discharge requirements ($< 15 \text{ mg N L}^{-1}$; Vlaamse regering 2006), and it is therefore needed to aim for a nitrogen removal efficiency above 75 and 50% for influent concentrations of 66 and 30 mg N L^{-1} , respectively.

The DO level in the RBC was always above $1 \text{ mg O}_2 \text{ L}^{-1}$ (Table 1) and therefore not low enough to suppress the growth of NOB, resulting in significant nitrate production by NOB (Fig. 3). A decrease of the DO with $0.2 \text{ mg O}_2 \text{ L}^{-1}$ during phase V^b could already decrease the NOB activity with 10% (Fig. 3), demonstrating the link between DO and nitrification. In some applications, the free ammonia concentration is used as NOB inhibitor (Anthonisen *et al.* 1976, Vlaeminck *et al.* 2009). However, due to the low nitrogen concentrations and relatively low pH in this study, free ammonia levels were always too low to efficiently suppress NOB. Therefore, DO control is the predominant control parameter for nitrification suppression treating low-strength wastewaters. It is anticipated that the use of a DO-controlled set-up with a low DO setpoint (e.g. $0.3 \text{ mg O}_2 \text{ L}^{-1}$) will more effectively suppress the NOB at long term, allowing for nitrogen removal efficiencies exceeding 80% (Joss *et al.* 2009). Using the RBC configuration, it is suggested to control DO by a variation of the rotation speed of the rotor (Meulman *et al.* 2010).

Low HRT were applied in order to obtain reasonably high nitrogen loading and removal rates. At a nitrogen influent concentration of 66 and 29 mg N L⁻¹, a HRT of 2h and 1h was applied, respectively. Compared to described OLAND systems, the applied HRT in this study was very low (Table 2). For anammox systems it was suggested that a decrease in hydraulic residence time could have a positive effect on the removal rate due to the wash out of possibly toxic byproducts (Tsushima *et al.* 2007). However, in the latter study the loading rate was not constant while decreasing the HRT, so it is not clear whether the increase in AnAOB activity was due to a higher loading rate or lower HRT. Nitrogen removal efficiencies for AnAOB activity decreased slightly when a lower HRT until 1h was applied (Tsushima *et al.* 2007). In this study the low HRT seems not to affect the nitrogen removal rate. Due to suppression of NOB activity in phase V^b, the removal rate increased again to a similar level ($p > 0.05$) as during phase IV, where a higher HRT was applied. However, these low HRT implies very good biomass retention in the reactor to avoid wash out of AerAOB and AnAOB. So, it is anticipated that biofilm-based systems, such as RBC and fixed/moving bed reactors, or suspended-based system with very good settling sludge, such as granular sequencing batch reactors (SBR) and gas lift reactors, will be used under the described conditions.

Table 2. Influent concentration, nitrogen loading, nitrogen removal and hydraulic residence time for one step nitrification/anammox processes

Wastewater	Influent concentration (mg N L ⁻¹)	N-loading rate (g N L ⁻¹ d ⁻¹)	N-removal rate (g N L ⁻¹ d ⁻¹)	HRT (d)	Reference
Digested black water	1023	0.94	0.71	1.33	(Vlaeminck <i>et al.</i> 2009)
Sludge digestate	800	0.74	0.67	0.93	(Jeanningros <i>et al.</i> 2010)
Sludge digestate	650	0.54	0.51	1.20	(Joss <i>et al.</i> 2009)
Industrial digestate	250-350	2.0	1.17	0.18	(Abma <i>et al.</i> 2010)
Landfill leachate	209	0.38	0.38	0.55	(Hippen <i>et al.</i> 2001)
Landfill leachate	100-400	0.45-0.90	0.25-0.57	0.19-0.83	(Siegrist <i>et al.</i> 1998)
Sewage-like	66	0.86	0.44	0.08	This study
Sewage-like	30	0.84	0.38*	0.04	This study

*Intermittent rotation of the discs

The prerequisites for the OLAND process to fit in the new concepts are a low energy consumption and a dischargeable effluent quality, which will be obtained by a good biomass retention and low nitrification rate. The choice of type of reactor will have an influence on both requirements. A RBC reactor, as used in this study, is known for its low energy requirements (0.36 - 0.9 kWh kg⁻¹ N), simplicity and good biomass retention (Mathure & Patwardhan 2005). However, good oxygen control is more complicated in this reactor type and nitrification is therefore more difficult to avoid. In reactor types such as a SBR, oxygen control is more efficient, but good biomass retention relies on well settling morphology of the sludge (Vlaeminck *et al.* 2010, De Clippeleir *et al.* 2009) and energy requirements are higher due to active aeration (1.2 kWh kg⁻¹ N) (Wett *et al.* 2010). The balance between energy requirements and removal efficiency will determine the choice of reactor type for the application of OLAND in the main stream of the sewage treatment system.

The interest to implement the OLAND process in the main stream of the WWTP is currently rising (Kartal *et al.* 2010). Next research challenges relate to a decrease of the process temperatures from 25°C to 10-15°C, as typical for a moderate climate in

the cold season (Wanner *et al.* 2005) and to the operation of the OLAND process in the presence of moderate levels (30-100 mg L⁻¹) of biodegradable COD. The tested low nitrogen levels in this study are expected if the energy-positive sewage treatment concept is applied on the current sewage composition. However, in the future dilution prevention could increase the pollutant concentrations with a factor 5, through combining separation between wastewater and stormwater, a decrease in sewer infiltration with 50%, water conservation with 25% and the addition of grinded kitchen waste (Brombach *et al.* 2005, Henze 1997).

CONCLUSIONS

This study shows the feasibility of the OLAND process to treat low strength wastewaters. At the two lowest concentrations (66 and 29 mg N L⁻¹) and HRT (2 and 1 h), volumetric removal rates were still relatively high: 444 and 303 mg N L⁻¹ d⁻¹, respectively. More efficient suppression of the nitrification in the reactor should lead to higher removal efficiencies and a dischargeable nitrogen effluent quality.

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