Combined partial nitritation/Anammox system for treatment of digester supernatant

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Abstract One-year (2004) comprehensive investigations in a semi-industrial pilot plant (5 m^3) were carried out with the aim of assessing the influence of operational parameters on the partial nitritation/Anammox system performance. In the system designed as a moving-bed biofilm reactor, the influent nitrogen load to the Anammox reactor was progressively increased and a stable Anammox bacterial culture was obtained. Interaction between subsequent aerobic and anaerobic conditions in the partial nitritation and Anammox reactors, respectively, granted conditions to remove nitrogen through the nitrite route. It implies that the oxygen supply can be limited to a high extent. A control strategy for the partial nitritation step relied on concomitant adjustment of the air supply with a variable influent nitrogen load, which can be monitored by both pH and conductivity measurements. In the Anammox reactor, an influent nitrite-to-ammonium ratio plays a vital role in obtaining efficient nitrogen removal. During the 1-year experimental period, the Anammox reactor was operated steadily and average nitrogen removal efficiency was 84% with 97% as the maximum value.

Keywords Anammox; biofilm; moving-bed reactor; nitrogen removal; partial nitritation; supernatant

Introduction

The deammonification process technology started in the 1990s when many research groups conducted experiments with the new technology for nitrogen removal based on an anaerobic ammonium oxidation (Anammox) (van Loosdrecht and Jetten, 1998; Hippen, 2001; Hippen *et al.*, 2001; Fux, 2003; Szatkowska *et al.*, 2004; Trela *et al.*, 2004; Gut, 2006). In contrast to traditional nitrification/denitrification, the deammonification process results in savings amounting to 60% of the oxygen supply, no additional chemicals are needed and sludge production is very low. Biological systems for nitrogen removal can be improved by separate treatment of highly concentrated waters (up to 2.3 g NH_4-N L^{-1}), such as supernatant produced during dewatering of digested sludge, or effluents from the fertiliser industry, fish canning industry, manure treatment and landfill leachates. In wastewater treatment plants (WWTP) with anaerobic sludge digestion, a recirculated supernatant contributes to 15–20% of the influent nitrogen load. Therefore, it is proposed to separately treat the supernatant rather than return it to the WWTP inlet for treatment as a part of the main flow.

The deammonification process may be executed in two steps as a combination of the aerobic partial nitritation process and the Anammox process. According to van Dongen *et al.* (2001), in the first step, around 55–60% of the ammonium needs to be converted to nitrite nitrogen by nitrifying aerobic microorganisms. In the second, it is possible to remove nitrogen (ammonium and nitrite are converted to nitrogen gas under anaerobic or oxygen-limited conditions) by Anammox bacteria (Egli, 2003). The reactors that are used to develop and maintain the Anammox culture must be chosen with regard to the slow growth rate of the bacteria. Biofilm systems (moving-bed reactor, fixed bed reactor,

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fluidised bed reactor, gas-lift reactor) or sequential batch reactors (SBR) are especially suitable for cultivation of Anammox bacteria (Strous *et al.*, 1997; Siegrist *et al.*, 1998; Dapena-Mora *et al.*, 2004).

In Sweden, the partial nitritation/Anammox moving-bed biofilm system for nitrogen removal is investigated and evaluated at two pilot plants: laboratory-scale pilot plant at the Royal Institute of Technology, Stockholm (supplied with supernatant from Bromma WWTP) and a semi-industrial scale pilot plant at Himmerfjärden WWTP, supplied directly with the supernatant from dewatering of digested sludge (Płaza *et al.*, 2002; Szatkowska, 2004; Trela *et al.*, 2004; Gut, 2006). The pilot plants were started up with the goal of developing the Anammox bacterial culture as biofilm on Kaldnes rings. The seeding material was obtained from the full-scale nitrification basin at Himmerfjärden WWTP in the year 2000 and, after more than 1 year, an Anammox culture was established. In this paper, the results from 1-year operation (2004) of the semi-industrial scale pilot plant are presented and discussed.

Methods

Pilot plant description

The used pilot plant (Figure 1) is located at Himmerfjärden WWTP (258,000 p.e., flow 101,000 $m^3 \cdot d^{-1}$), serving the south-west Stockholm area and consists of two separate reactors in which different biological processes are established. The influent digester supernatant is continuously pumped to a primary settling tank (equalisation tank). Afterwards, the supernatant is directed to the partial nitritation reactor (R1). Next, after the settling tank, the Anammox reactor (R2) is placed. Each reactor has a working volume of 2.1 m^3 and consists of three zones equally sized. In the second reactor, zones 1 and 2 are inter-connected. The biomass of nitrifying and Anammox bacteria has been developed as biofilms on Kaldnes rings – a plastic ring-shaped support. Both reactors are filled up with Kaldnes carriers to obtain an overall volumetric filling of approximately 45–50%. The pH correction was carried out in the first zone of the Anammox reactor during four initial months of the described period.

Operational and analytical procedures

The experiment on increase of the influent nitrogen load was planned with the goal to not overload the system and damage the Anammox bacterial culture. The decision of the increase in the influent nitrogen load to the Anammox reactor was based on the precise analyses of the concentrations of inorganic nitrogen forms in the effluent and in the reactor. The effluent from the nitritation reactor was diluted with tap water according to a schedule. Samples to analyse for ammonium (NH_4-N), nitrite (NO_2-N) and nitrate (NO_3-N) nitrogen were taken once a week. Additional analyses of soluble chemical oxygen demand (COD), total soluble phosphate phosphorus (PO_4-P) and alkalinity were

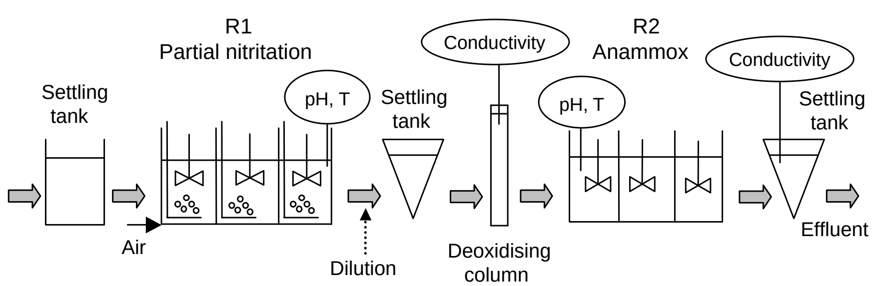

Figure 1 Scheme of the semi-industrial scale pilot plant at Himmerfjärden WWTP

performed as a routine. The samples were directly filtrated and analysed by drLange spectrophotometer. On working days, manual measurements were carried out: pH values, dissolved oxygen (DO) concentrations, temperature, conductivity, inflow rates and $Na_2CO_3/NaHCO_3$ solution dosage. Moreover, on-line pH and conductivity measurements were collected.

Results and discussion

Influent characteristics

Table 1 shows statistical evaluation of the parameters describing the characteristics of the influent supernatant. The system must have been prepared to operate with the influent ammonium nitrogen concentration varying between 532 and 917 g NH_4-N·m^{-3}. An influent buffering capacity expressed by the quotient of alkalinity/NH_4-N (average value 1.6) indicated an excess of alkalinity to partly oxidise ammonia to nitrite. The alkalinity measurements may, to some extent, have been influenced by the presence of organic acids.

Partial nitritation reactor

Table 2 demonstrates experimental results for the partial nitritation process over the period of 1 year. The pH value was gradually decreasing along the zones, which is in agreement with the reaction of the biological oxidation of ammonia by nitrifying bacteria. The conductivity decreases to the highest extent in zones 1 and 2, where most of the oxidation of ammonia to nitrite takes place. The DO concentration is difficult to maintain below 1.5 mg O_2 L^{-1} due to manual regulation and the variable characteristics of supernatant. The temperature was in an optimal range by the use of heat supply.

Time series data of the inorganic nitrogen forms with compensation for hydraulic retention time (HRT) are shown in Figure 2. Stable oxidation of ammonia to nitrite with only minor formation of nitrate nitrogen (an average value of 8.3 g NO_3-N·m^{-3}) was attained. A favourable effluent nitrite-to-ammonium ratio (NAR) at the value around 1.2 was predominantly maintained, with the changes (0.8–2.0) caused by variability of the influent ammonium concentration and process conditions. The ammonium nitrogen conversion rate amounted to 5.8 g NH_4-N·m^{-3}·d^{-1} and somewhat exceeded nitrite nitrogen production rate that was equal to 5.1 g NO_2-N·m^{-3}·d^{-1} (1-year mean values).

The pH value, depending highly on the alkalinity of the wastewater, is an excellent parameter for monitoring the nitritation process in order to keep the effluent NAR close to the theoretical value of 1.3. Figure 3 shows correlation between removed pH (between the inlet and zone 3 of R1) and the NAR in the effluent. The NAR is reaching the value of 1.3 when the average pH value in the influent (7.8, Table 1) is decreased by the amount of 1.5. The relationship between the pH parameter and the average value of DO in reactor 1 was also found (Figure 4). The relationship indicates that for the required pH drop by 1.5, the average value of DO in the bulk liquid should be kept around 1.0 mg O_2 L^{-1}. Furthermore, the conductivity parameter shows a high correlation with the influent ammonium nitrogen concentration (Figure 5). The relationship between converted ammonium nitrogen and removed amount of conductivity (Figure 6) demonstrated that a suitable drop of conductivity in the partial nitritation reactor monitors the conversion of ammonium to nitrite nitrogen. It is recommended to use on-line DO measurements in combination with an on-line conductivity control device to optimise the air supply for a specific influent nitrogen load.

Table 1 Statistical evaluation of the influent digester supernatant

	NH_4-N ($g \cdot m^{-3}$)	pH (−)	Alkalinity ($mmol \cdot L^{-1}$)	Conductivity ($mS \cdot cm^{-1}$)	Alkalinity/ conductivity ($mmol \cdot L^{-1}/mS \cdot cm^{-1}$)	Alkalinity/NH_4-N ($mmol \cdot L^{-1}/mmol \cdot L^{-1}$)	COD ($g\ O_2 \cdot m^{-3}$)	Organic acids ($g \cdot m^{-3}$)	PO_4-P ($g \cdot m^{-3}$)
Average	717.0	7.8	86.1	4.94	16.9	1.6	369	101	19.1
Minimum	532.5	7.4	22.8	2.37	4.8	0.5	203	24	2.3
Maximum	917.5	8.1	189.3	6.47	36.3	3.2	691	259	46.4
S.D.*	96.3	0.1	40.2	0.80	7.1	0.6	136	79	17.4
n	55	241	46	241	46	46	24	8	18

*S.D. - Standard deviation

Table 2 Experimental results for the partial nitritation reactor (R1)

Parameter	Unit	Average	Range
HRT*	d	2.0	1.8–2.3
Influent inorganic N	$g{\cdot}m^{-3}$	717.0	532.5–917.5
Effluent inorganic N	$g{\cdot}m^{-3}$	674.3	398.2–887.2
pH value – influent (median)	–	7.8	–
pH value – effluent (median)	–	6.5	–
DO concentration	$g\ O_2{\cdot}m^{-3}$	Z*1 1.01	Z1 0.6–3.5
		Z2 1.20	Z2 0.4–5.3
		Z3 1.10	Z3 0.3–4.9
Conductivity	$mS{\cdot}cm^{-1}$	In 4.90	In 2.37–6.47
		Z1 3.87	Z1 2.25–5.85
		Z2 3.50	Z2 2.29–4.81
		Z3 3.45	Z3 2.19–4.72
Temperature	°C	31.2	24.2–39.5

*HRT, hydraulic retention time; Z = zone

Anammox reactor

The statistical evaluation of parameters measured and calculated during the operation of the Anammox reactor is demonstrated in Table 3. The oxygen-limited conditions were assured as well as proper temperature. Deviations from the average in case of pH and conductivity values depend on the influent characteristics and the performance of the preceding nitritation process. Additionally, in the Anammox reactor, an increase in the pH value is obtained. The increase of the pH value by a unit of 1 is an indication of the stability of the process whereas any values below 1 could be an early sign of a disturbance in the process performance. It must also be stressed that the increase of the pH value in the Anammox reactor implies that no addition of chemicals for pH correction is necessary.

Figure 7 shows concentrations of inorganic nitrogen in the influent and effluent analysed over the period of 1 year. In the year 2004, a slow build-up of the system was initiated. Activity of the Anammox culture was considerably enhanced during a period of five initial months. The total inorganic nitrogen load was enlarged step-by-step to obtain a four-fold increase in the nitrogen load. In August 2004, nitrite nitrogen concentration in the reactor was analysed above $50\,g{\cdot}m^{-3}$ and therefore the nitrogen load was decreased. The peak removal of total inorganic nitrogen amounted to as much as $367\,g\ N{\cdot}m^{-3}$. The influent load enlargement from 0.032 to $0.133\,kg\ N{\cdot}m^{-3}{\cdot}d^{-1}$ (calculated for reactor's volume decreased by the volume of Kaldnes rings) resulted in concomitant removal of ammonium and nitrite nitrogen with small nitrate nitrogen production, which proved that the Anammox process was established successfully. The maximum removal of the influent nitrogen load was calculated as $0.122\,kg\ N{\cdot}m^{-3}{\cdot}d^{-1}$.

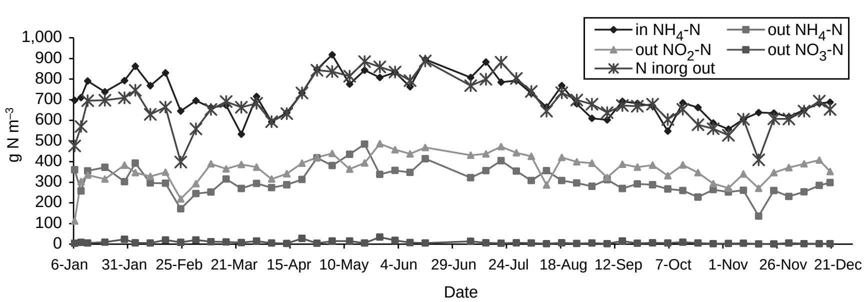

Figure 2 Variations of inorganic nitrogen forms in the partial nitritation reactor (R1)

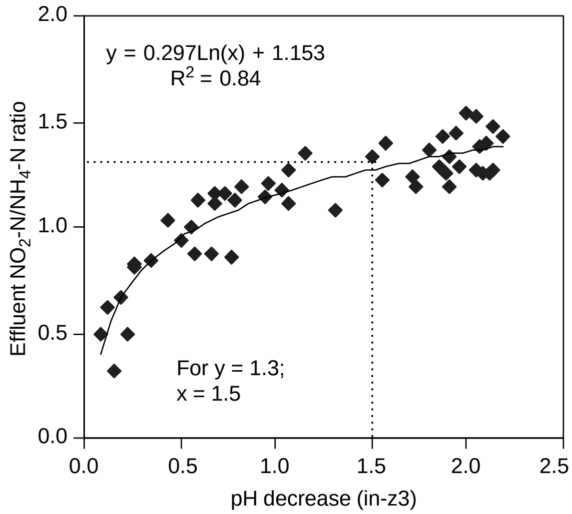

Figure 3 The pH decrease and the effluent NO_2-N/NH_4-N ratio in R1 (based on data from 2003 and 2004)

The maximum values of the removed nitrite nitrogen surpassed 200 g NO_2-N·m^{-3}, which was parallel to a high removal of ammonium nitrogen (exceeding values of 170 g NH_4-N·m^{-3} and corresponding to almost complete removal of nitrite nitrogen). During the experimental period, the average removal of nitrite and ammonium nitrogen amounted to 124 g NO_2-N·m^{-3} and 98 g NH_4-N·m^{-3}, respectively. It corresponded to an average influent ratio of nitrite-to-ammonium equal to 1.2 (Figure 8 and Table 3). Such a reaction course is characteristic for the Anammox process. Additionally, the average nitrate nitrogen production corresponds to 6.1% of the average total inorganic nitrogen removal. It is proof of the underproduction of nitrate. In accordance with the stoichiometry of the Anammox process, the maximum efficiency that can be obtained in the Anammox reactor amounts to 88.2%. Therefore, 11.2% production of nitrate compared to the sum of removed ammonium and nitrite is expected. The difference in nitrate

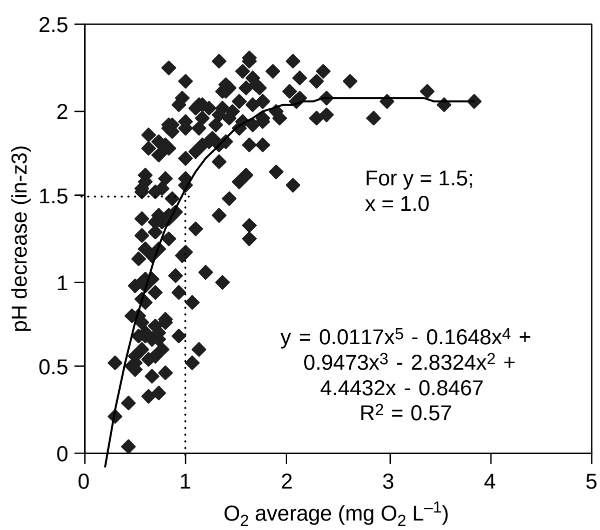

Figure 4 The pH decrease and the average dissolved oxygen concentration in R1

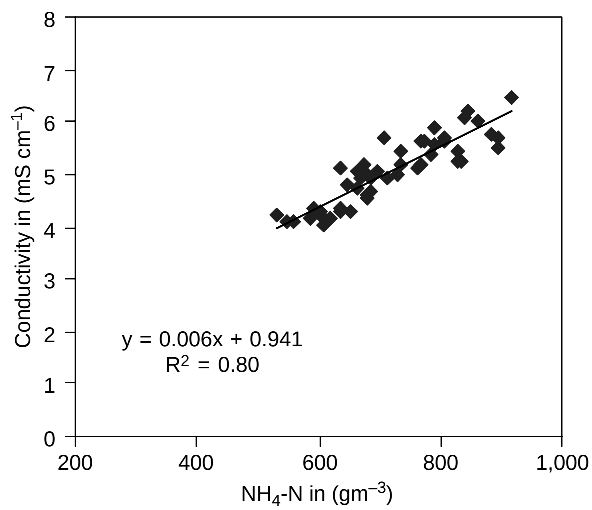

Figure 5 NH_4-N concentration and conductivity in the influent to the pilot plant

production between the calculated and theoretical values reveals that there are other reactions present in the Anammox reactor. Simultaneous denitrification with some available COD cannot be excluded. The proceeding phases of the experiment on increase in the influent nitrogen load led to a stable and very low effluent nitrite nitrogen concentration (on average 5.1 g NO_2-N·m^{-3}). The nitrogen removal efficiency was maintained at a stable level for the last 10 months of the study period (Figure 8), with an average value oscillating around 88% (maximum value of 97%).

The aim of the experiment was assessed by examining the relationship between the removed values of nitrite and ammonium nitrogen in the Anammox reactor (Figure 9). A high correlation coefficient reaching the value of 0.85 and a slope of the correlation line amounting to 1.22 (very close to theory) prove that the Anammox process was established effectively. The following relationship (Figure 10) between the influent NAR and nitrogen removal efficiency in the Anammox reactor shows an apparent dependence of the Anammox process performance on the proper quotient between

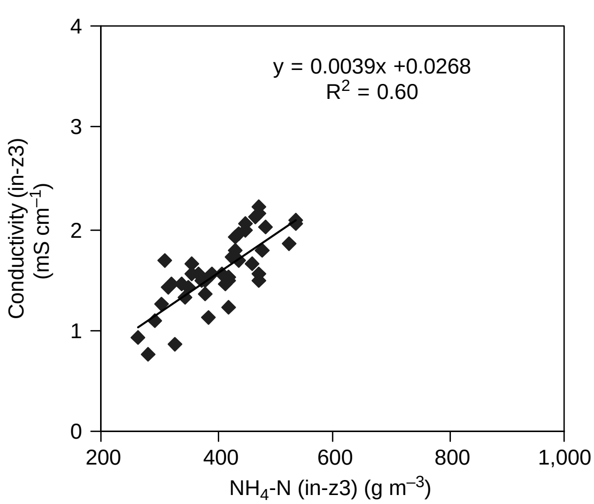

Figure 6 Removed ammonium and removed conductivity in R1

Table 3 Experimental results for the Anammox reactor (R2)

Parameter	Unit	Average	Range
HRT	d	3.0	2.6–3.5
Influent inorganic N	$g \cdot m^{-3}$	243.9	109.7–399.3
Effluent inorganic N	$g \cdot m^{-3}$	34.2	4.8–98.6
Removal of N	%	83.8	25.0–97.4
NO_2-N/NH_4-N ratio	–	1.2	0.5–1.9
NO_2-N (Z1 + Z2)	$g \cdot m^{-3}$	16.0	0.4–57.9
pH value–influent (median)	–	7.2	–
pH value–effluent (median)	–	8.2	–
DO concentration	$g\ O_2 \cdot m^{-3}$	Z1 + Z2* 0.13 Z3 0.13	Z1 + Z2 0–0.50 Z3 0–0.37
Conductivity	$mS \cdot cm^{-1}$	In 1.50 Z1 + Z2 0.79 Z3 0.74	In 0.72–2.42 Z1 + Z2 0.42–1.34 Z3 0.45–1.27
Temperature	°C	31.9	27.0–39.2

*Z1 + Z2, zones 1 and 2 are inter-connected

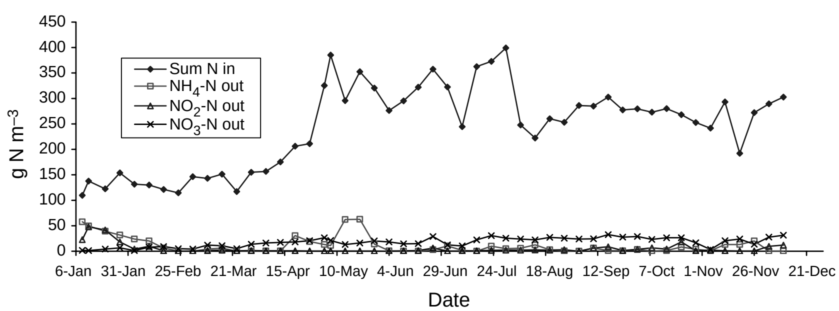

Figure 7 Inorganic nitrogen forms concentration in the influent and effluent (R2)

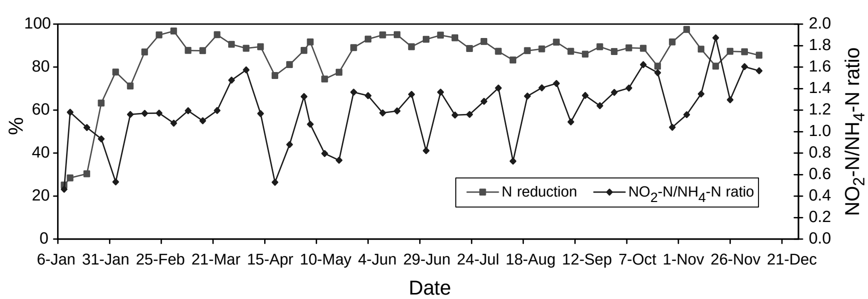

Figure 8 NO_2-N/NH_4-N ratio in the influent and total inorganic nitrogen removal efficiency (R2)

nitrite and ammonium nitrogen. The NAR values oscillating in the range 1.0–1.5 are appropriate to remove nitrogen in an efficient way reaching the average of 87% process efficiency. Figure 11 presents an increase in total inorganic nitrogen removal with an increase in the influent nitrogen load. Correlation coefficient amounting to as much as 0.95 provides evidence that a stepwise introduction of the enlarged nitrogen load allows an Anammox bacterial culture to cope with increased substrate concentration. It can be stated that the increase in the influent nitrogen load enhances the capacity of the Anammox reactor.

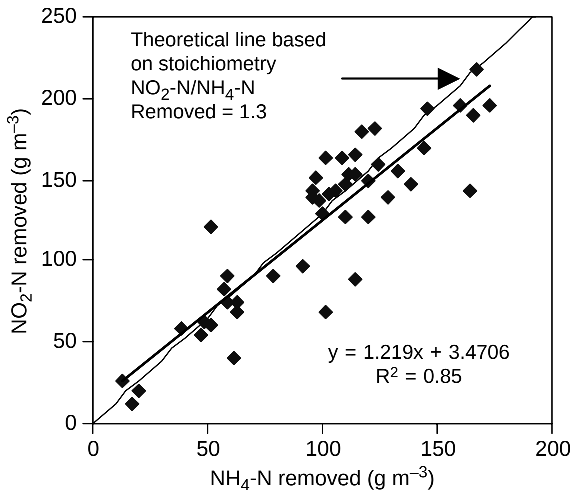

Figure 9 Removal of ammonium and nitrite nitrogen in R2

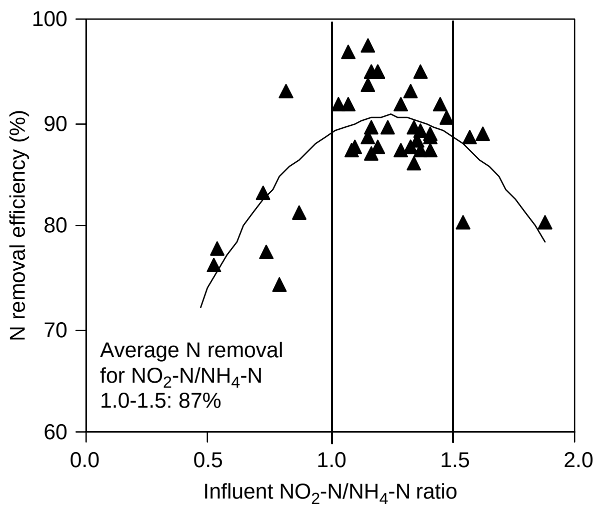

Figure 10 The influent NO_2-N/NH_4-N ratio and the nitrogen removal efficiency in R2

System approach to the deammonification process

The two-step partial nitritation/Anammox process for nitrogen removal could be treated independently as well as in an interconnected system. The interplay between the factors must be acknowledged in terms of simplicity of the system operation and the process efficiency. As the microorganisms responsible for the Anammox reaction are growing much slower than nitrifying bacteria, a suitable design of the reactors is required. The natural temperature of the digester supernatant is an advantage, as the medium must not be heated to a high degree. Adequate nitrite-to-ammonium ratio plays a key role in the deammonification system efficiency. A concurrent drop in the pH value by the amount of 1.5 and the adjustment of the air supply to obtain DO concentration of about 1.0 mg O_2 L^{-1} in the bulk liquid, combined with on-line conductivity measurements in the partial nitritation reactor, were the optimal control strategy in the studies. In the Anammox

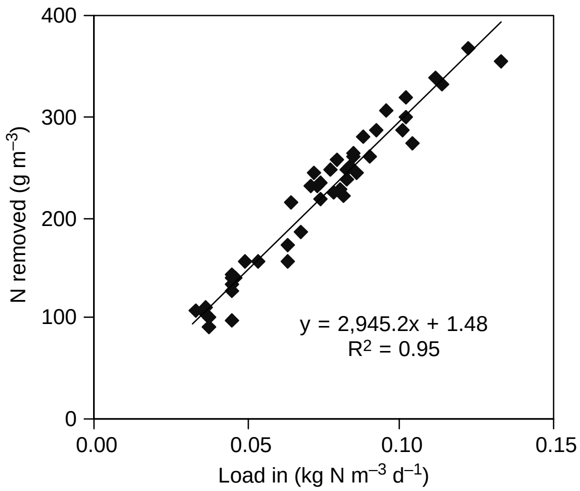

Figure 11 Total inorganic nitrogen removal and the influent load in R2

process, the influent NAR in the range 1.0–1.5 (the high efficiency of the system) strongly depends on the control strategy in the preceding step. On a daily basis, the progress in nitrogen conversions in the system can be checked by on-line conductivity measurements.

Conclusions

Over a period of 1 year it became feasible to successfully operate and maintain the combined partial nitritation/Anammox system for the treatment of the digester supernatant with high nitrogen removal efficiency. The analysis of data provided a valuable understanding of the studied processes. It resulted in recognition of control strategies that would enable an efficient operation of the system and future full-scale implementation. The complexity of the system is a challenge that should be assessed in terms of using all the available methods to simplify the operation and control of the combined nitritation/Anammox processes. The following conclusions can be stated.

1. The variability of the influent supernatant parameters highly affects the deammonification process performance.
2. Stable oxidation of ammonia to nitrite with only minor formation of nitrate nitrogen in the partial nitritation reactor was obtained. In the Anammox reactor, concomitant removal of ammonium and nitrite nitrogen with small nitrate nitrogen production proved that the process was established successfully.
3. Control strategies for the combined partial nitritation/Anammox process consist of directing a reaction course in the partial nitritation reactor to obtain a simultaneous drop in the pH value by the unit of 1.5 and maintaining the DO bulk-liquid concentration at a value of around 1.0 mg $O_2 \cdot L^{-1}$ in order to keep the effluent nitrite-to-ammonium nitrogen ratio close to the theoretical value of 1.3. The influent NAR to the Anammox reactor in the range of 1.0–1.5 grants conditions for the most efficient process.
4. Stepwise introduction of the enlarged nitrogen load allows bacterial culture to adjust to the increased substrate concentration. The increase in the influent load enhances the capacity of the Anammox reactor.
5. During a 1-year experimental period, the Anammox process reached a nitrogen removal efficiency of 84% on average and a maximum value amounting to as much as 97%.

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