Original Research

The Influence of Hydraulic Retention Time and Sludge Age on the Kinetics of Nitrogen Removal from Leachate in SBR

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Abstract

In the present study the influence of hydraulic retention time (HRT) in SBR and sludge age (SRT) on the effectiveness of nitrogen removal from leachate was investigated. Two series were performed. In each series, experiments were carried out in four SBR operated in parallel at HRT 12, 6, 3 and 2d, respectively. Each series differed in sludge age. In series 1, SRT decreased from 51 to 17d with shortening of HRT, while in series 2 it was about 2 times shorter.

In each series the amount of nitrogen used on biomass synthesis, removal of nitrogen in denitrification and losses of ammonium during the aeration phase were estimated on the basis of material balance for all nitrogen forms in SBR cycle. It was shown that nitrogen consumption on biosynthesis was decreased linearly to the HRT increase. In series 1, the rate of ammonium nitrogen removal increased from 5.38 mgN_{NH4}/g VSS·h to 7.36 mg N_{NH4}/g VSS·h with HRT shortening from 12 to 3d, respectively. In series 2 the rate of ammonium nitrogen removal was nearly constant, regardless of HRT and on average it was 1.74 mg N_{NH4}/g VSS·h. The losses of ammonium nitrogen in series 1 increased from 8.2 mg N/dm^3·cycle in SBR 1 to 33.4 mg N/dm^3·cycle in SBR 4, while in series 2 it was almost 2 times lower.

Keywords: landfill leachate, SBR, hydraulic retention time (HRT), sludge age, nitrogen removal, losses of ammonium

Introduction

Leachates from active and closed municipal waste landfills are often a source of contamination to groundwater and surface water. Their exposure to the environment may occur as uncontrolled overflow, rainfall runoff, subsidence and infiltration. The most common method of prevention is collecting and treating leachate.

The chemical composition of leachates differs markedly, depending on the type of waste deposits and the age of the landfill. Leachate from young landfills includes significant amounts of readily biodegradable organic matter, while from the maturation phase it contains mainly refractory organic compounds. Moreover, they are rich in ammonia nitrogen and have a hazardous nature. For these reasons, designing unified systems of treatment is difficult.

Landfill leachate can be treated together with municipal wastewater [1, 2]. However, stricter regulations for nitrogen discharge and growing concern over the potential effect of recalcitrant and toxicity leachate constituents have led to increased demands for separate treatment plants to be included in landfill infrastructure. For this purpose, many methods are used: biological [3-8], physicochemical [9, 10] and two- or multistage systems [11-14]. According

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to Leitzke [15], Kaczorek et al., [16] even when physico-chemical processes are used for organics removal, biological nitrification and denitrification as a preliminary stage of treatment is advisable.

Nitrogen removal from wastewater is based on processes of ammonium oxidation to nitrite and nitrate (nitrification) and reduction of nitrification products to N_2 (denitrification). In activated sludge, the ammonium oxidation proceeds slowly because of slow nitrifying bacteria growth. To provide sufficient nitrifying cells residence time there has been an on-going search for new technologies. One of these is the suspended-carrier biofilm process (SCBP). The SCBP has come into extensive use for nitrification and denitrification of municipal sewage. Research devoted to nitrification of landfill leachate using suspended-carrier biofilm technology was carried out by Welander et al. [17]. The authors found that the effectiveness of nitrification is dependent on carrier media type as well as its surface structure. It was observed that in reactors containing small pieces of polyethylene and pieces of polyethylene with 5% NH_4Cl addition, the nitrification rate was clearly lower (from 5.6 to 11 mg $N_{NH4}/dm^3 \cdot h$) than in a reactor with cellulose packing (about 40 mg $N_{NH4}/dm^3 \cdot h$).

During landfill exploitation, changes of leachate composition and a decrease in C/N ratio are observed. In young landfills, organic acids occurring in leachate at high concentration may inhibit the rate of ammonium nitrogen oxidation, leading to a decrease in nitrification effectiveness (Takai et al., [18]). However, the longest and the most important (from the standpoint of biogas production) is the methanogenic phase, usually starting in the 5th year and lasting until the 17th year of landfill exploitation. In this phase, the content of degradable compounds decreases, while nitrogen concentration increases. Additionally, organic carbon can be insufficient and can cause a decrease in denitrification efficiency. The requirement of added organics such as methanol is necessary [19, 20].

To overcome existing limitations, several novel concepts of microbial treatment processes for nitrogen removal have been developed [21]. They are Sharon, Anammox or CANON [22-26]. They are based on a new concept in microbiology: partial nitrification, nitrifier denitrification and anaerobic ammonia oxidation.

The present study was devoted to nitrogen removal from leachate by activated sludge treatment in SBR. The influence of hydraulic retention time (HRT) and sludge age on nitrogen consumption on biosynthesis, effectiveness of dissimilative denitrification and ammonium losses in the aeration phase were analyzed. Following this, the rate of ammonium removal and nitrification rate in the SBR cycle were estimated.

Materials and Methods

Leachate Feed

The leachate used in this study was collected from a municipal landfill located in Wysieka near Bartoszyce in the Warmia and Mazury Province. The landfill has been in operation since January, 1996. Leachate is collected by drainage, 1,200 m in length and 80 mm in diameter, and stored in a retention tank.

The characteristics of the landfill leachate investigated are shown in Table 1.

Table 1. Characteristics of landfill leachate used in series 1 and 2.

Constituent	Values	
	series 1	series 2
Biochemical oxygen demand (mg BOD_5/dm^3)	457	622
Chemical oxygen demand (mg COD/dm^3)	1237	1596
Total nitrogen (mg N/dm^3)	189	154
Ammonium nitrogen (mg N_{NH4}/dm^3)	141	113.3
Organic nitrogen (mg N_{org}/dm^3)	47.2	38.8

Process Configuration and System Design

The investigations were carried out on a laboratory scale in four SBR operated in parallel (SBR 1-SBR 4). The reactors with the working volume of 6 dm^3 were made of plexiglass and were fitted with a stirrer at the regulated rotation speed (36 r/min). Dissolved oxygen was supplied using porous diffusers placed at the bottom of reactors. In all series, the amount of oxygen supplied to the reactor in the aeration phase was regulated in order to maintain oxygen concentration at the end of the phase at the level of 2.5-4.0 mgO_2/dm^3. The system was in operation at room temperature for 2 months for each series.

Two series were performed. The amount of the leachate supplied within 24h to subsequent reactors changed from 0.5 dm^3 (SBR 1) to 3 dm^3 (SBR 4), as a result of which the hydraulic retention time (HRT) ranged from 12d (SBR 1) to 2d (SBR 4). The volumetric exchange rate, defined as the ratio of the volume of leachate supplied to the reactor in a cycle to the working volume of the reactor, changed from 0.083 $dm^3/dm^3 \cdot d$ (SBR 1) to 0.5 $dm^3/dm^3 \cdot d$ (SBR 4) (Table 2).

The effect of sludge age on nitrogen removal was explored. In both series the reactor operating cycle consisted of five phases: filling (0.08h), mixing (3h), aeration (18h), settling (2.84h), and decanting (0.08h). Series differed in the sludge age (SRT). In series 1, the sludge ages were 51d (SBR 1), 29d (SBR 2), 20d (SBR 3) and 17d (SBR 4). The SRT in this series was maintained by carrying off the excess sludge in the amounts of 50 cm^3/d in SBR 1 and 2 (which corresponds to 10 and 5% of leachate contents) and 100 cm^3/d in SBR 3 and 4 (5 and 3.3% of leachate contents, respectively). In series 2, the sludge age was nearly 2 times shorter than in series 1 and changed from 24d in SBR 1 to 8d in SBR 4. In SBR 2 and 3 the SRT were 16d and 10d, respectively. It was obtained by carry-

Table 2. The operational conditions in series 1 and 2.

Operational conditions	Values			
	SBR 1	SBR 2	SBR 3	SBR 4
Volume of leachate influent in SBR operating cycle (dm^3)	0.5	1.0	2.0	3.0
Hydraulic retention time (HRT) (d)	12	6	3	2
Volumetric exchange rate (n) ($dm^3/dm^3 \cdot d$)	0.083	0.167	0.33	0.5

ing of the volume of the excess sludge, i.e. 125, 225, 425, and 600 cm^3, respectively (25 – 20% of leachate contents supplied in the cycle).

Analytical Method

In raw leachate, analyses of COD (according to the methodology described by Hermanowicz et al., [27]), BOD_5 (with use of the OxiTop® by the method given by WTW firm), total nitrogen and ammonium (according to the methodology described by Hermanowicz et al., [27]) were performed.

The effluents from SBR were subjected to daily measurements of total nitrogen, ammonium, nitrite and nitrate (according to the methodology described by Hermanowicz et al., [27]).

The mixed reactor content was measured for volatile suspended solids (VSS) and total suspended solids (TSS) (according to the methodology described by Hermanowicz et al., [27]) and oxygen concentration (using an oxygen controller HI 9142).

Results

The influence of HRT and sludge age on nitrogen removal was analyzed on the basis of balance of all nitrogen forms in the influent and effluent in the operational cycle of SBR. It was assumed that nitrogen removal from leachate is a result of:

- biomass production in duration of the cycle period ($C_{N\text{-}syn}$),
- dissimilative nitrate and nitrite reduction in the mixing phase (denitrification) ($C_{N\text{-}den}$),
- losses of ammonium in the aeration phase ($C_{N\text{-}A}$).

Calculation of nitrogen concentration consumed on biomass production ($C_{N\text{-}syn}$). Nitrogen concentration used for synthesis of activated sludge in the operational SBR cycle may be expressed by the following equation (1):

$$C_{N-syn} = (Y_{obs} \cdot (C_0 - C_e) \cdot X_N) / t_c \qquad (1)$$

where:
$C_{N\text{-}syn}$ - nitrogen concentration consumed on biomass production (mg $N/dm^3 \cdot$cycle),
Y_{obs} - observed coefficient of biomass production (mg VSS/mg COD),
C_0 - organics concentration in leachate at the beginning of the SBR cycle (mg COD/dm^3),
C_e - organics concentration in the effluent (mg COD/dm^3),
X_N - nitrogen concentration in biomass dry matter (mg N/mg VSS),
t_c - duration time of SBR operating cycle (cycle).

The observed coefficient of biomass yield (Y_{obs}) and nitrogen content in dry mass of activated sludge (X_N) were determined earlier by Kulikowska [28]. In the present study, the Y_{obs} and X_N values were used for calculations of $C_{N\text{-}syn}$.

The experimental data on nitrogen concentration consumed in biomass production during the operational SBR cycle versus volumetric exchange rate of leachate are illustrated in Fig. 1. From the data it can be concluded that in both series $C_{N\text{-}syn}$ increased linearly with an increase in volumetric exchange rate, but in series 2 (with shorter sludge age) the regression line coefficient was about 1.5-fold higher.

The nitrogen balance in activated sludge during SBR cycle. The experimental data of ammonium, nitrate and nitrite concentration profiles in duration of SBR cycle obtained in mixing and aeration phases at varied HRT (SBR 1-4), are shown in Figs. 2a-d. It was observed that during the mixing phase the ammonium concentration was nearly stable in all reactors. Nitrogen removal was a result of dissimilative reduction of nitrate, which remained in the SBR reactor after the previous cycle. The concentration of nitrate reduced in mixing phase increased from 2.71 N_{NO3}/dm^3 (SBR 1) to 5.39 mg N_{NO3}/dm^3 (SBR 3).

On the basis of analysis of nitrogen concentration profiles $C_N = f(t)$ it was stated that removal of ammonium nitrogen rate and nitrification rate were followed according to first-order kinetics.

During the start-up of the aeration phase, a high concentration of ammonium was obtained. It was observed

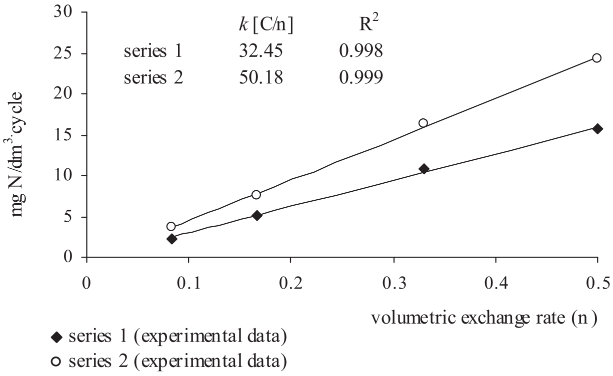

Fig. 1. Nitrogen amount used for biomass synthesis in SBR operating cycle.

that nitrate were formed in amounts clearly less than the ammonium consumed. As a consequence in series 1, correspondingly with the decrease of HRT in 1-3 SBR reactors, the ammonium nitrogen removal rate increased from 5.38 mg N_{NH4}/g VSS·h (SBR 1) to 7.36 mg N_{NH4}/g VSS·h (SBR 3), whereas the nitrification rates in SBR 1 and SBR 3 were over 7-fold, and in SBR-2 almost 12-fold lower than the ammonium removal rate. In SBR 4, at HRT = 2d, a decrease in ammonium removal rate to 1.45 mg N_{NH4}/g VSS·h and nitrification rate to 0.05 mg N_{NO3}/g VSS·h was observed. In series 2, except for SBR 4, shortening of sludge age caused a decrease in ammonium removal rate as well as nitrification rate. Consequently, in this series, the mean rate of ammonium removal was 1.73 mg N_{NH4}/g VSS·h and the nitrification rate was 0.09 mg N_{NO3}/g VSS·h.

It is evident that the amount of removed nitrogen as a result of denitrification was much lower than the amount of nitrogen utilized for biomass production. Additionally, in all reactors, the gradient of ammonium concentration at the beginning and the end of the aeration phase was clearly higher than gradient of nitrate concentration. This means that unbalanced ammonium oxidation-to-nitrate or nitrite and ammonium removal from leachate in the aeration phase were observed.

It was assumed that the lack of balance between removed ammonium amount and nitrate formed in both series could be a result of partial nitrification, nitrifier denitrification or deamonification in the aeration phase. The nitrogen losses (C_{N-A}) were calculated as the difference between total nitrogen concentration at the beginning and the sum of nitrogen concentration consumed on biomass synthesis in the SBR operating cycle and ammonium, organic, nitrite and nitrate nitrogen concentrations in the effluent:

$$C_{N-A} = C_{0,N} - (C_{k,N_{NH4}} + C_{k,N_{org}} + C_{k,N_{NO2}} + C_{k,N_{NO3}} + C_{N-syn}) \quad (2)$$

where:

- $C_{0,N}$ - total nitrogen concentration at the beginning of the aeration phase (mg N/dm^3),
- $C_{k,N-NH4}$ - ammonium nitrogen concentration at the end of the aeration phase (mg N_{NH4}/dm^3),
- $C_{k,Norg}$ - organic nitrogen concentration at the end of the aeration phase (mg N_{org}/dm^3),
- $C_{k,N-NO2}$ - nitrite nitrogen concentration at the end of the aeration phase (mg N_{NO2}/dm^3),
- $C_{k,N-NO3}$ - nitrate nitrogen concentration at the end of the aeration phase (mg N_{NO3}/dm^3),
- C_{N-syn} - nitrogen concentration used for biomass synthesis in the SBR operating cycle (mg N/dm^3).

In series 1 the nitrogen losses increased from 8.2 mg N_{NH4}/dm^3 in SBR 1 to 33.4 mg N_{NH4}/dm^3 in SBR 4, while in series 2 it was nearly 2 times lower and changed

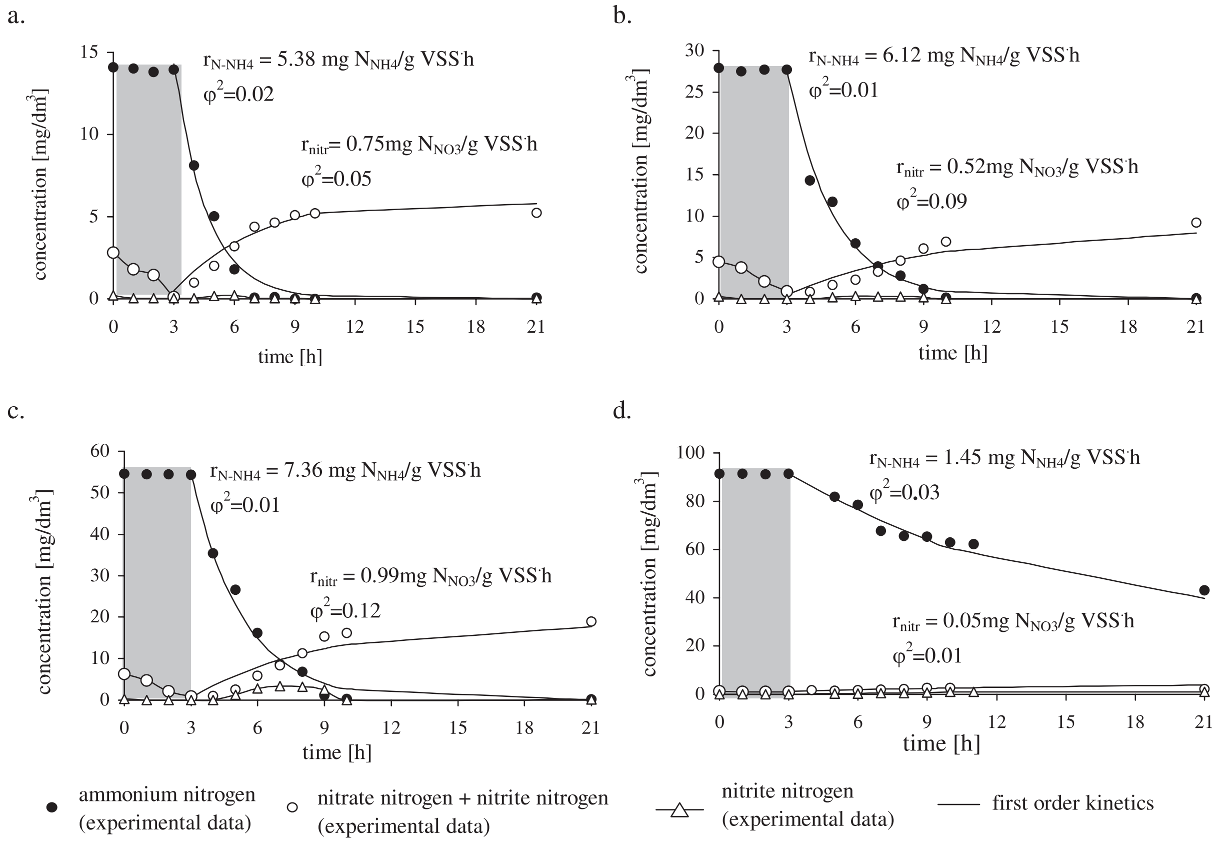

Fig. 2. Concentrations of ammonium, nitrite and nitrate nitrogen during SBR operating cycle (series 1); a. SBR 1, b. SBR 2, c. SBR 3, d. SBR 4.

Table 3. Nitrogen concentration in the effluent from SBR reactors in series 1 and 2.

Nitrogen concentration		SBR 1	SBR 2	SBR 3	SBR 4
Ammonium nitrogen (mg N_{NH4}/dm^3)	Series 1	0.8	0.7	0.9	46.4
	Series 2	2.3	11.9	20.5	36.0
Organic nitrogen (mg N_{org}/dm^3)	Series 1	26.4	27.6	28.3	37.7
	Series 2	25.2	26.5	31.8	35.0
Nitrite nitrogen (mg N_{NO2}/dm^3)	Series 1	0.08	0.09	0.15	1.48
	Series 2	1.46	1.37	0.52	1.49
Nitrate nitrogen (mg N_{NO3}/dm^3)	Series 1	4.1	8.9	18.7	1.4
	Series 2	2.04	5.92	7.32	1.47

from 5.1 mg N_{NH4}/dm^3 in SBR 1 to 15.7 mg N_{NH4}/dm^3 in SBR 4. The obtained data indicates that in both series, the losses of ammonium nitrogen during aeration phase for any volumetric exchange rate can be found from $C_{N\text{-}A,\max}$ value. Mathematically, this can be expressed as follows:

$$C_{N-A} = C_{N-A,\max}(1-e^{-k\cdot n}) \qquad (3)$$

where:

$C_{N\text{-}A,\max}$ - the maximum losses of ammonium nitrogen for established operation condition in SBR (mg N/dm^3),

k - reaction constant ($d\cdot dm^3/dm^3$),

n - volumetric exchange rate ($dm^3/dm^3\cdot d$).

The maximum losses of ammonium nitrogen ($C_{N\text{-}A,\max}$) were in correlation with sludge age and in series 1 were 2.5-fold higher in comparison to series 2 (Fig. 3).

The influence of HRT and sludge age on the quality of treated leachate. The research results showing the concentration of nitrogen forms in the effluent from SBR are presented in Table 3. In series 1, with a long sludge age, complete nitrification and ammonium concentration in treated leachate below 1 mg N_{NH4}/dm^3 were obtained in SBRs

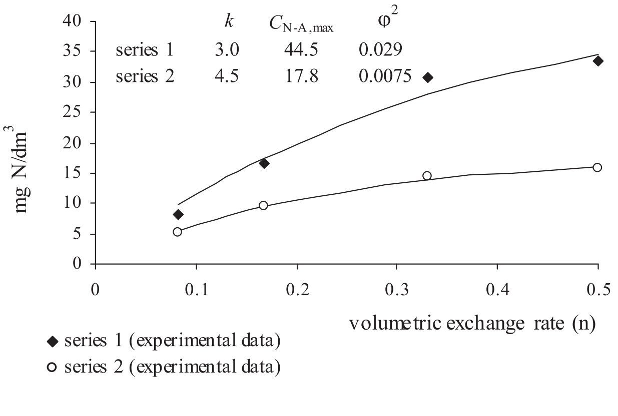

Fig. 3. Losses of ammonium nitrogen during aeration phase in SBR operating cycle.

1-3. The nitrate concentration in these reactors increased from 4.1 mgN_{NO3}/dm^3 to 18.7 mgN_{NO3}/dm^3 and nitrite from 0.08 mgN_{NO2}/dm^3 to 0.15 mgN_{NO2}/dm^3, respectively. In SBR 4, at HRT = 2d, the ammonium concentration in the effluent was 46.6 mgN_{NH4}/dm^3, while nitrate and nitrite concentrations did not exceed 1.5 mgN_{NO3}/dm^3. In the effluent from series 1, organic nitrogen was a dominant form and ranged between 26.4 mgN_{org}/dm^3 (SBR 1) and 37.7 mgN_{org}/dm^3 (SBR 4).

In series 2, with a shorter sludge age, the lowest concentration of ammonium (2.3 N_{NH4}/dm^3) was obtained in SBR 1, at HRT = 12d. The nitrate concentration was not significant and the highest value was observed in SBR 3. However, the nitrite concentration in this series was from slight to over a dozen-times higher than in series 1. It was found that the high concentration of organic nitrogen in the effluent ranged from 25.2 mgN_{org}/dm^3 (SBR 1) to 35 mgN_{org}/dm^3 (SBR 3).

Discussion

In the present work, the effect of hydraulic retention time and sludge age on nitrogen removal from landfill leachate was investigated. It was found that the amount of nitrogen consumed in biomass synthesis increased linearly with an increase of volumetric exchange rate. 2-fold shortening of the sludge age caused a 1.5-fold increase in k coefficient from 32.45 to 50.18 [C/n].

It was observed that ammonium concentration in the effluent did not exceed 1 mgN_{NH4}/dm^3 at HRT 3, 6 and 12d and sludge age 20, 29 and 51d, respectively. 2-fold shortening of sludge age caused the low concentration of ammonium nitrogen in the effluent (2.3 mgN_{NH4}/dm^3) to be obtained only at 12d HRT. However, at HRT 6 and 3d, the concentration of ammonium increased to 11.9 mgN_{NH4}/dm^3 and 20.5 mgN_{NH4}/dm^3, respectively. The results indicate that sludge age was the critical parameter for ammonium removal. Ammonium oxidation-to-nitrate nitrogen was successfully obtained at HRT above 20d.

Robinson and Maris [29] obtained concentrations of ammonium nitrogen in the effluent below 1 mgN_{NH4}/dm^3

at HRT about 5d. According to the authors, the total amount of removed ammonium nitrogen from leachate was consumed on sludge growth, which corresponded to 44% removal of organics (measured as COD) from leachate. Studies carried out by Bull et al. [30] made similar observations. The total nitrogen removal from leachate of 60% was a result of activated sludge growth. Research conducted by Zaloum and Abott [31] in SBR indicated that nitrogen elimination was caused by biosynthesis, air-stripping, nitrification and denitrification. At HRT = 20d, low concentrations of organic nitrogen (1.3 mg N_{org}/dm^3) and the sum of nitrate and nitrite nitrogen (6.5 mg/dm^3) in the effluent were obtained.

The nitrogen balance data in the present study indicates that nitrogen losses occurred in the aeration phase of SBR cycle. In series 1, the loss values ranged from 8.2 mg N/dm^3 (SBR 1) to 33.4 mg N/dm^3 (SBR 4), while in series 2 they were lower (from 5.1 mg N/dm^3 in SBR 1 to 15.7 mg N/dm^3 in SBR 4). The data indicate that average ammonium losses were 1.9-fold higher with longer activated sludge age.

Im et al., [32] using the anaerobic-aerobic system, reported that the difference between ammonia removal rate and nitrification rate was a result of ammonia stripping and ammonia removal for biomass growth. Loukidou and Zouboulis [33], conducting research in SBR, filled to 50% of its empty volume with cube-shaped polyurethane particles, noted that denitrification achieves a sufficiently high degree, as nitrate concentrations in the effluent are substantially lower than those expected from the oxidation of initially present, high ammonia concentrations. This observation can be possibly explained by the mechanism of simultaneous nitrification and denitrification which occurs during the aeration phase.

It was observed that an ammonium nitrogen removal rate at HRT above 20d increased with HRT values from 5.38 mg $N_{NH4}/dm^3 \cdot h$ (0.13 kgN/$m^3 \cdot d$) to 7.36 mg $N_{NH4}/dm^3 \cdot h$ (0.18 kgN/$m^3 \cdot d$). At HRT below 20d, the rate was constant (about 1.6 mg $N_{NH4}/dm^3 \cdot h$; 0.04 kgN/$m^3 \cdot d$), regardless of HRT values. Consequently, the concentration of ammonium nitrogen in the effluent was high.

In studies with a sequencing batch reactor operated with ammonium-rich wastewater under oxygen-limited conditions at a suitable loading rate with aerobic nitrifying bacteria and Anammox bacteria, a nitrogen removal rate of up to 0.3 kgN/$m^3 \cdot$d was reported for the CANON process. In this reactor, heterotrophic denitrification did not occur, and no aerobic nitrite-oxidizing bacteria were detected [26]. The CANON process was carried out in gas lift reactors. Nitrogen removal rates up to 1.5 kgN/$m^3 \cdot$d were achieved [26].

The highest losses of ammonium nitrogen were obtained in reactors with a long sludge age. The increase of ammonium nitrogen losses with a corresponding increase in nitrification rate and sludge age indicates that nitrogen losses can be connected with the activity of nitrifying bacteria.

The literature also indicates that ammonium nitrogen removal in a single, aerated reactor is caused by complete autotrophic nitrogen removal over nitrite, known as the CANON process. The basis of this process is a combination of partial nitrification and Anammox. The partial nitrification is the oxidation of ammonium to nitrite in wastewater, but not to nitrate. The nitrifiers oxidize ammonia to nitrite, consume oxygen and create anoxic conditions, which the Anammox process needs. The Anammox process is the denitrification of nitrite with ammonia as the electron donor. The first step caused by too short sludge age could be a limiting factor in ammonium nitrogen removal

Conclusions

1. On the basis of nitrogen balance in a single SBR cycle, it has been shown that nitrogen removal was a result of biosynthesis and denitrification although a significant part of nitrogen was removed as a result of ammonium loss. In both series, ammonium losses were shown. However, in series 1 (with longer sludge age) the losses were nearly 2 times higher in comparison to series 2;
2. Corresponding with the decrease in hydraulic retention time (HRT), the nitrogen amount consumed on biomass synthesis increased linearly. The regression coefficient value was 2 times higher in the series with a shorter sludge age;
3. In reactors with 12, 6 and 3d HRT, the concentration of ammonium in the effluent did not exceed the value of 1 mgN_{NH4}/dm^3 at sludge ages above 20d;
4. The ammonium removal rate changed from 5.38 mg N_{NH4}/g $VSS \cdot h$ to 7.36 mg N_{NH4}/g $VSS \cdot h$, while the nitrification rate changed from 0.75 mg N_{NO3}/g $VSS \cdot h$ to 0.99 mg N_{NO3}/g $VSS \cdot h$.

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