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Comparison of nutrients degradation in small scale membrane bioreactors fed with synthetic/domestic wastewater

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Abstract

Two membrane bioreactors were operated with biological phosphorus removal, carbon degradation and denitrification to check how comparable and representative they were compared to full-scale plants. One was fed with synthetic municipal wastewater and was switched from pre- to post-denitrification without carbon dosing. The influent of the second plant was drawn from a separate sewer. This plant worked the whole time with post-denitrification without carbon dosing. The synthetic wastewater was designed to achieve a realistic COD:TN:TP ratio and tested for long time biodegradability. The eliminations were >94% (COD) and >97% (TP) for both plants. This was within the range of commercial plants, as well as the TN elimination for the pre-denitrification of plant I (>75%). The eliminations of TN for post-denitrification were above 80% for both plants despite the high influent concentrations and the missing carbon source for post-DN. A calculation of the nitrification rates gave values similar to those found in literature (1–6 mgN/(gMLVSS h)). A comparison of the denitrification showed expected rates for pre-denitrification (7.5 mgN/(gMLVSS h)) for plant I. The values (on average 1.8 mgN/(gMLVSS h)) for post-denitrification in plant II were higher than endogenous denitrification rates which are commonly reported as 0.2–0.8 mgN/(gMLVSS h). The rates for post-denitrification in plant I were only slightly higher than endogenous ones (0.9 mgN/(gMLVSS h)).

Keywords: Membrane bioreactor; Post-denitrification; Biological phosphorus removal; Carbon degradation; Synthetic wastewater

1. Introduction

Denitrification is a process widely used in wastewater treatment. Smaller basin sizes due to higher denitrification rates and the advantage of oxygen savings for carbon removal due to the carbon degradation of the denitrifying heterotrophic bacteria are the main reasons that the plants are usually operated with pre-denitrification. When stringent effluent requirements for NO_3-N exist, also downstream denitrification (post-denitrification) but with dosage of a carbon source (e.g. methanol) is common. Without any carbon source the denitrification rates are endogenous and range only between 0.2 and 0.8 mgN/(gMLVSS h) [1].

A bench scale plant operated earlier at the Department of Chemical Engineering showed surprisingly high denitrification rates (up to 2.8 mgN/(gMLVSS h)) even though working with post-denitrification without any additional carbon [2]. The bench scale plant was a membrane bioreactor (MBR) for carbon, nitrogen and biological phosphorus removal connected to a combined sewer. For a more detailed investigation of this special modification of the post-denitrification process, two membrane bioreactors at laboratory (plant I) and pilot scale (plant II) were operated. To ensure a well-defined and steady input for this fundamental study, plant I was operated with synthetic wastewater, which as a first step had to be optimized to meet the needs of the particular process while still ensuring comparability. Plant II was fed with domestic wastewater from a sewer.

In order to confirm the comparability of the synthetic wastewater composition to real wastewater in terms of e.g. BOD_t, COD:TN:TP ratio as well as the comparability of plant I with commercial plants, the test facility has been operated first in a classical set-up with pre-denitrification, which is commonly used in commercial MBRs and common wastewater treatment plants (WWTP). In a second step, the classical set-up was modified to post-denitrification. Results (eliminations, effluent concentrations, nitrification and denitrification rates) with synthetic wastewater were compared to results from domestic

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Nomenclature

AE	aerobic
AN	anaerobic
AX	anoxic
BOD_t	biochemical oxygen demand after *t* days (mg/L)
COD	chemical oxygen demand (mg/L)
DNR	specific denitrification rate $\frac{\Delta NO_3-N}{\Delta t \cdot MLVSS}$ ($mgNO_3$-N/h gMLVSS)
HRT	hydraulic retention time (h)
MBR	membrane bioreactor
ML(V)SS	mixed liquor (volatile) suspended solids (g/L)
NH_4-N	ammonia-nitrogen (mg/L)
oPO_4-P	ortho-phosphat (mg/L)
Post-DN	post-denitrification
Pre-DN	pre-denitrification
TN	total nitrogen (mg/L)
TP	chemical oxygen demand (mg/L)
NR	specific nitrification rate $\frac{\Delta NH_4-N}{\Delta t \cdot MLVSS}$ ($mgNO_3$-N/h gMLVSS)
SRT	solids retention time (d)
V_{DN}	volume denitrification zone (L)
V_N	volume nitrification zone (L)
WWTP	wastewater treatment plant

wastewater treated in a similar set-up in plant II and to results from typical commercial plants.

2. Materials and methods

2.1. Plant design

2.1.1. Plant 1, laboratory scale MBR

The cascaded bioreactor (Fig. 1) was operated at room temperature (18–23 °C) and fed with synthetic wastewater. For approximately half a year the reactor worked with pre-denitrification (pre-DN). By switching the 3 anoxic (AX1-3) and 2 aerobic (AE1-2) chambers (compare Fig. 1), the operation mode was changed to post-DN later on. Both anaerobic chambers (AN1, AN2), the membrane filtration chamber (MF) and the recirculations (R1, R2) were left in their original configuration. A small mixing basin (MB) protected the first anaerobic zone against oxygen (for pre-denitrification) or nitrate (for post-denitrification) entrainment. A small oxygen degradation zone (not shown) was situated between the last aerated chamber and the first chamber for denitrification in post-DN. Excess sludge was withdrawn three times daily. The sludge retention time (SRT) was 20 days for pre-DN and 23 days for post-DN. For other operating parameters see Table 1. The plant was seeded with sludge from the WWTP Berlin-Wassmannsdorf, which operates with pre-denitrification and enhanced biological phosphorus removal (EBPR). For the separation of biomass and treated water, an immersed plate and frame module was used (GKSS, Germany, polyacrylic nitril, 37 nm, 0.6 m^2). The recirculation (R_2) from the membrane chamber was 300% of the inflow during pre- and 400% during post-denitrification.

Table 1
Constant operating parameter I

	Plant I	Plant II
Flow rate	(3.7–5.0) L/h	13 L/h
HRT	(12.2–13.7) h	10.8 h
V	50.3 L	170 L

2.1.2. Plant II, pilot scale MBR

Plant II (Fig. 2) was operated for 1.5 years with wastewater from a pumping station in Berlin, serving 800 inhabitants. This pumping station is located in a remote area with separate sewer and therefore the influent consists only of domestic wastewater devoid of industrial and storm water. The wastewater was drawn through a 1 mm slit screen directly from the mains of the pumping station following the daily flow profile of the station, and pumped into a buffer tank (156 L maximum) in order to level out hydraulic and concentration peaks and allow for a constant hydraulic influent [3]. Plant II was seeded with sludge from the WWTP Berlin Wassmannsdorf like plant I. The plant was cascaded into six zones: one anaerobic, two aerobic and anoxic zones and one aerated membrane tank where a 1.4 m^2 plate and frame module (GKSS, Germany, polyacrylic nitril, 37 nm) was implemented. A channel was installed between aerobic and anoxic zones with a top down flow, which minimized oxygen entrainment from the aerobic to the anoxic zones.

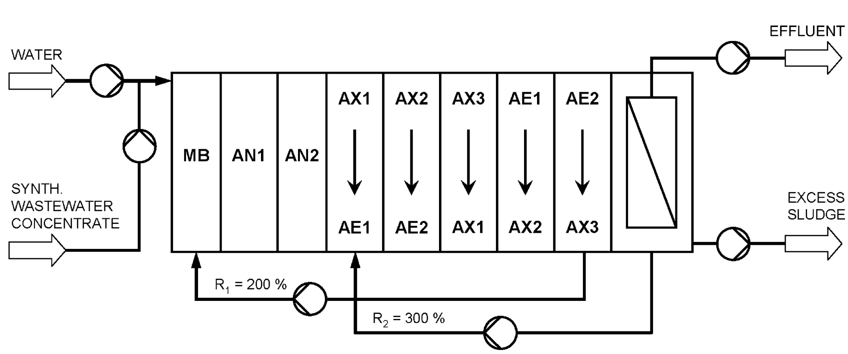

Fig. 1. Flow sheet plant I, laboratory MBR.

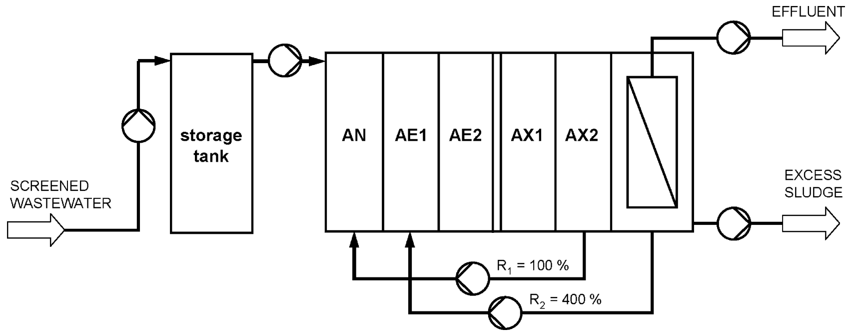

Fig. 2. Flow sheet plant II, pilot MBR.

2.2. Analyses

Anions were measured using a Dionix DX 100 ion chromatograph with an IonPac AS 4a column for NO_3-N, NO_2-N, oPO_4-P (ortho-phosphat) and an IonPac CS12a column for ammonia-nitrogen. For the determination of chemical oxygen demand (COD), total phosphorus (TP) and total nitrogen (TN) Dr. Lange cuvette test kits LCK 114, 314, 414, 350, 349, 338, 238 were used. All cuvette tests comply in calibration, detection and quantitation limits with ISO 8466-1, DIN 38402 A51 and DIN 32645.

For suspended solids, 100 mL sludge samples were taken and dried at 105 °C until constant weight was reached. The dried sample was heated to 600 °C for 3 h, and the amount of volatile suspended solids was calculated from the weight of the residue (compare DIN 38409 part 1).

The BOD (biological oxygen demand) analysis was done with the OxiTop system by WTW, Germany, following the European Standard EN 1899-1. For seeding, strongly diluted sludge (10 mL supernatant per liter dilution water) from the respective research plant was used. The wastewater from a pumping station in Berlin connected to a combined sewer was analysed with sludge from plant I.

2.3. Synthetic wastewater

2.3.1. Composition

There are numerous synthetic wastewater compositions given in literature (e.g. [4–9]), each of them designed for special research. The composition of the synthetic wastewater was chosen according to Nopens et al. [4]. This wastewater composition is supposed to be close to the composition of municipal wastewater. It especially had a wide range of different carbon sources and contains polysaccharide, proteins and lipid components. However, the original recipe had to be modified because the original composition hampered biological phosphorus removal. Particularly mineral and trace metal contents were therefore adapted according to Brand [10]: H_3BO_4 300 μg/L, $CuCl_2$ 40 μg/L, KI 60 μg/L, $MnSO_4 \cdot H_2O$ 320 μg/L, $NaMoO_4 \cdot 2H_2O$ 120 μg/L, $ZnCl_2 \cdot 2H_2O$ 140 μg/L and $CoCl_2 \cdot 6H_2O$ 300 μg/L. A concentrate of the synthetic wastewater was adjusted to pH 2 and stored for one week in a concentrate tank. Tap water (stored in a fresh water tank) and concentrate were pumped into the mixing basin of the research plant. The pH level was kept above 7.3, controlled in the first anaerobic chamber by NaOH dosing.

The composition of the synthetic wastewater (Table 2) was changed when operation was switched from pre-DN to post-DN to achieve a better comparability to plant II (see also section Operating parameters, Post-denitrication).

2.3.2. General parameters

Table 3 compares the influent parameters for plant I (pre- and post-DN), plant II and two selected full scale wastewater treatment plants. As full scale plants, the membrane bioreactor in Rödingen [11] for about 2000 p.e. and a classical WWTP near Berlin [12] for ≫500,000 p.e. were chosen. Both treatment plants are connected to combined sewers, work with pre-denitrification and cover a wide range of possible influent compositions. They are typical representatives for conventional wastewater treatment and MBR. In WWTP also EBPR is implemented.

The parameters of the synthetic influents for pre-DN and post-DN were chosen to range between those of the commercial MBR and WWTP plants. The relatively high portion of organic nitrogen caused the lower ammonia-nitrogen concentration in the synthetic influent compared to the real wastewaters. Judging the COD:TN:TP ratios of all plants a lower COD:TN ratio is obvious in the synthetic influent pre-DN. The high nitrogen concentration, however, was necessary to avoid anaerobic conditions in the last anoxic chamber due to good denitrification rates in the first anoxic chamber. For the assessment it should be noted that the COD:TN ratios of the commercial plants were averaged. The

Table 2
Composition of the synthetic wastewater

Ingredients	Concentration original (mg/L)	Concentration pre-DN (mg/L)	Concentration post-DN (mg/L)
Peptone	17.4	28	25
Yeast extract	52.2	83	80
Milk powder	116.2	185	160
Starch	122	194	200
Sunflower oil	29	46	35
Ammonium acetate	79.4	126	150
Propionic acid		127	90
KH_2PO_4	23.4	21	26
$MgHPO_4 \cdot 3H_2O$	29	6	6
K_2HPO_4		21	26
Urea	91.7	146	50
$(NH_4)Cl$	12.8		
$FeSO_4 \cdot 7H_2O$	5.8	8.0	8.0

Table 3
Inflow concentrations of the different compared wastewater treatment plants

Parameter (mg/L)	Plant I, synth. wastewater pre-DN (min–max)		Commercial MBR (min–max)		Commercial WWTP (min–max)		Plant I, synth. wastewater post-DN (min–max)		Plant II (min–max)	
COD	663	(571–756)	430	(400–800)	985	(481–1550)	747	(697–808)	1275	(613–3142)
BOD_5	–	–	250	(150–450)	424	(222–560)	–	–	–	–
TN	101	(76–134)	58	(40–80)	81	(47–100)	71	(51–78)	122	(91–188)
NH_4-N	30	(23–41)	35	(30–60)	59	(34–73)	28	(26–32)	82	(70–113)
TP	12	(10–12)	11	(5–12)	11.7	(7–14)	15	(13–16)	21	(11–45)
oPO_4-P	9	(6–13)		(<10)	7.5	(4–9)	13	(11–15)	8.5	(6–11)
COD:TN:TP	100:15:1.7		100:13:2.6		100:8:1.2		100:9.4:2		100:9.6:1.6	

nitrogen and carbon influent peaks occur usually at different day times in commercial wastewater treatment plants which causes changing COD:TN ratios during a day.

2.4. Operating parameters

2.4.1. Pre-denitrification

The relatively high influent concentrations into plant I lead to high COD, TN and TP loads despite the high MLSS (Table 4). The COD load for plant I was significantly higher compared to commercial membrane bioreactors (MBR—COD load 0.03 kg/(kg d)), which are usually operated at COD loadings <0.1 kg/(kgMLSS d) for aerobic sludge stabilization. The specific sludge production of plant I was within the expected range (0.4 kg/(kg d)). The high MLVSS of plant I was due to the low input of mineral substances.

The pre-denitrification zone in MBRs has to be bigger because of the oxygen entrainment through the recirculation. Usually a V_{DN}:V_N ratio of 1:1 like in the MBR Rödingen is employed. It has been shown that the membrane chambers take part in the biological degradations, which is therefore considered in calculations as an aerated zone (nitrification).

2.4.2. Post-denitrification

The operation parameters of plants I and II were kept as similar as possible (Table 5) to ensure comparability of results. The influent concentrations were adapted within the same COD:TN:TP ratio (Table 3), in order not to repeat the exceptionally high loads of plant II.

Table 4
Operating parameter for pre-denitrification

	Plant I, pre-DN	MBR	WWTP
COD load (kg/(kg d))	0.12	0.03	0.23
TN load (kg/(kg d))	0.02	0.003	0.02
TP load (kg/(kg d))	0.002	0.0005	0.003
SRT (d)	20	>40	10
Specific sludge prod. (kg/(kg d))	0.4	0.5–0.7	0.4
V_{DN}:V_N	1:1.5	1:1	1:1.6
MLSS (g/L)	10	12	3.9
MLVSS (%)	82	50	77
HRT ($V_{DN}+V_N$) (h)	9	4–40	3.5–50 (10)

Table 5
Operating parameter for post-denitrification

	Plant I, post-DN	Plant II
COD load (kg/(kg d))	0.14	0.33
TN load (kg/(kg d))	0.013	0.03
TP load (kg/(kg d))	0.003	0.005
SRT (d)	23	28
Specific sludge prod (kg/(kg d))	0.6	0.1
V_{DN}:V_N	1.7:1	1.5:1
MLSS (g/L)	11	11.7
MLVSS (%)	80	75
HRT ($V_{DN}+V_N$) (h)	9	7

3. Results and discussion

3.1. Biological degradability of the synthetic wastewater

Wastewaters can easily be characterised and synthetic wastewater be designed considering the COD:TN:TP ratios. Nevertheless, for a long study not only the complete carbon but also the biodegradability is important. A comparison of the demands of biological oxygen of the synthetic wastewater, domestic wastewater of plant II and municipal wastewater from a pumping station in Berlin showed that the time related evolution of the BOD_t in the synthetic was not as smooth as for the real wastewaters (Fig. 3). Particularly the initial slope is much steeper for the synthetic wastewater than for the real wastewaters. Also the BOD_1 (synthetic wastewater) reaches already

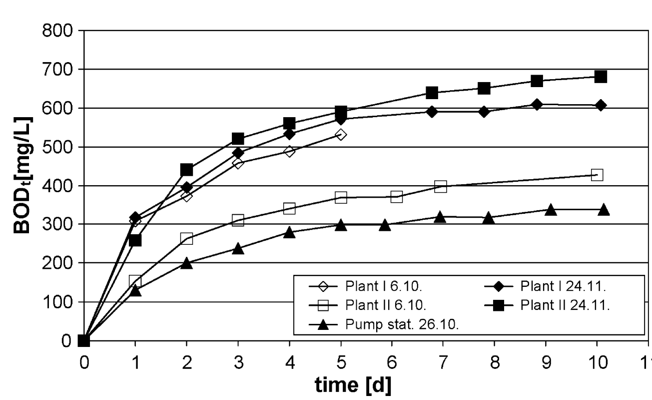

Fig. 3. BOD developments for real and synthetic wastewater.

Table 6
Outflow concentrations of the different compared wastewater treatment plants

Parameter (mg/L)	Plant I, synth. wastewater pre-DN (min–max)		Commercial MBR (min–max)		Commercial WWTP (min–max)		Plant I, synth. wastewater post-DN (min–max)		Plant II (min–max)	
COD (mg/L)	25	(13–39)	22.5	(15–30)	49	(37–58)	25	(13–28)	48	(35–79)
$Elim_{COD}$ (%)	96	–	95	–	95	–	97	–	94	–
TN (mg/L)	23	(11–30)	10	(5–17)	15	(10–21)	6	(3–21)	23	(18–29)
NH_4-N (mg/L)	0.2	–	0.2	–	0.3	–	0.1	–	1.3	–
$Elim_{TN}$ (%)	76		83		82		92		81	
TP	0.3	(0.05–1.3)	0.3[a]	(0.2–0.4)	0.4	(0.2–0.6)	0.2	(0.04–1.4)	0.2	(0.09–0.9)
oPO_4-P	0.2	–	0.2	–	0.1	–	0.1	–	0.1	–
$Elim_{TP}$ (%)	97	–	97[a]	–	97	–	99	–	98	–

[a] Phosphorus precipitation.

57% of BOD_5 compared to only 41% for real wastewaters, i.e. the concentration of easily degradable organic compounds was significantly higher than that of slowly degradable compounds in the synthetic wastewater. After 5 days already 94% of the BOD_{10} was reached with synthetic influent compared to 87% for the wastewater from the sewers. The BOD_5:COD ratio of 0.9 for the synthetic inflow confirmed this observation. Plant II and data from literature [13] gave BOD_5:COD values of 0.75 and 0.5–0.66 respectively.

Nevertheless the variation between two samples of real wastewater from the same source on different dates (plant II, 6.10. and 24.11) was observed to be greater than the variation between the synthetic water samples against the real wasterwater samples.

3.2. Elimination

3.2.1. Elimination and outflow concentration of COD, TN, TP

Table 6 gives an overview of all outflow concentrations and their minimum and maximum values for the four compared plants and the configurations pre-DN and post-DN.

3.2.2. COD degradation

All compared experimental and commercial wastewater treatment plants have good and stable COD eliminations (Fig. 4). According to Kraume et al. [14], COD effluent concentrations of <30 mg/L for MBR and <50 mg/L for classical treatment plants can be expected. The higher effluent values of plant II are probably due to the very high influent concentrations of 1275 mg/L on average (Table 3). The elimination above 95% for both configurations of plant I can be explained by the relatively high amount of easily degradable organic compounds together with stable conditions during plant operation.

3.2.3. Biological phosphorus removal

Fig. 5 shows the elimination of TP in the four different plants. The commercial MBR is not shown because it is not operated with biological phosphorus removal. It also has to be noted that if required the WWTP Berlin applies phosphorus precipitations. The very good effluent concentrations of plant I (especially post DN) and II (0.3, and 0.2 mg/L, respectively) confirm results of an earlier research, where a membrane bioreactor showed a very good phosphorus removal despite high sludge ages above 15 days (compare Adam [2]).

3.2.4. Nitrogen removal

The eliminations for the treatment plants with pre-denitrification are governed by the recirculation of NO_3. The higher TN effluent concentration of plant I was probably due to the high influent concentrations (on average 101 mg/L, Table 3). With 76% the elimination in plant I is the result of the double denitrification—one small part in the mixing basin and the bigger portion in the anoxic zones. The effluent nitrogen compositions consisted almost completely of nitrate. Ammonium-nitrogen concentrations were 0.2, 0.2 mg/L and 0.3 mg/L for plant I, MBR and WWTP, respectively. Nitrite-

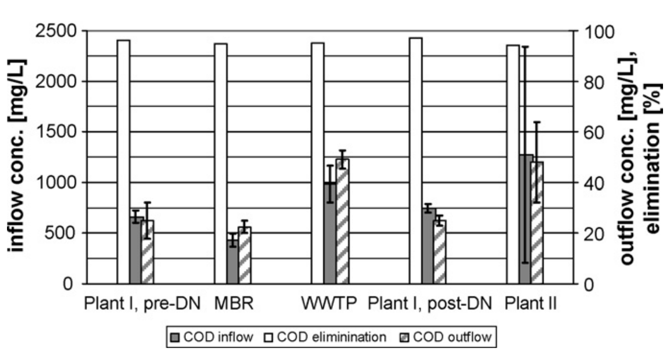

Fig. 4. COD inflow and outflow concentrations and eliminations.

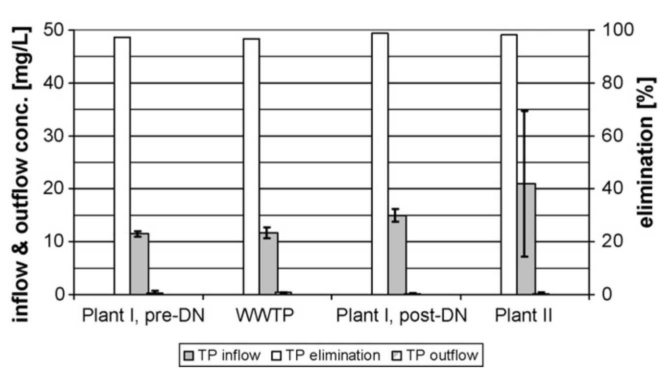

Fig. 5. TP inflow and outflow concentrations and eliminations.

Table 7
Nitrification rates for plants I and II

NR (mgN/(gMLVSS h))	Plant I, pre-DN (min–max)		Plant I, post-DN (min–max)		Plant II (min–max)	
AE1	4.1	(1.2–6.6)	3.2	(2.2–4.3)	4.1	(2.3–5.7)
AE2	2.0	(0–3.0)	1.2	(0–2.4)	2.7	(1.9–4)
MF	0.3	(0–1.0)	0.1	(0–0.5)	2.3	(1.0–6.0)

Table 8
Denitrification rates for plants I and II

DNR (mgN/(gMLVSS h))	Plant I, pre-DN (min–max)		Plant I, post-DN (min–max)		Plant II (min–max)	
AX1	7.5	(1.3–12.3)	0.9	(0.1–2.4)	1.8	(1.6–2.0)
AX2	0.7	(0–2.4)	1.0	(0–2.2)	1.1	(0.6–1.8)
AX3	0.3	(0–2.2)	0.6	(0.1–1.3)		

nitrogen was only measured for WWTP with 0.2 mg/L. Organic nitrogen content was not separately determined, but is likely to be lower than 2 mg/L.

With the change to post-denitrification, the influent concentration of plant I was adapted to lower values (101–71 mg/L). This explains the very good TN effluent concentration of 6 mg/L. Plant II also showed very good TN elimination. The high effluent concentration of 21 mg/L was still an acceptable value considering the high influent concentration of up to 188 mg/L (see Fig. 6, Table 3).

These results are especially remarkable since in plant II as well as in plant I no carbon was added to the anoxic zone. The effluent of the aerobic zone of plant II still contained ammonia, probably caused by a slightly overloading of the nitrification zone. In most cases complete nitrification was achieved in the membrane chamber.

3.3. Degradation rates

3.3.1. Nitrification rate, NR

The nitrification rates for plant I during pre-DN show considerable variation, due to the different concentrations within the single chambers of the cascaded aerobic zone (Table 7).

The average ammonium concentrations for AE1, AE2 and MF during pre-denitrification in plant I were 7, 2 and 0.2 mgNH_4-N/L. For post-denitrification they were 3.2, 1.2 and 0 mg NH_4-N/L. The nitrification rates for plant II probably reflect the higher nitrogen concentration in the plant and even in the membrane filtration chamber still significant nitrification occurs (see Table 7).

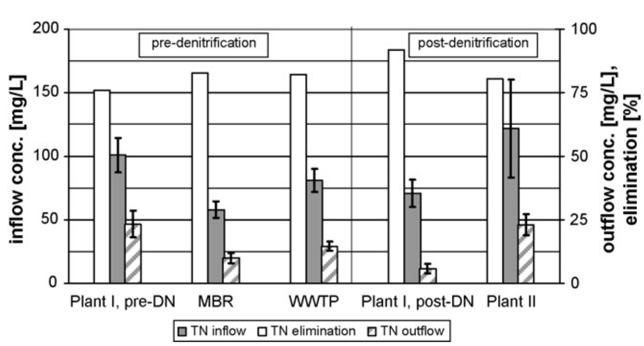

Fig. 6. TN inflow and outflow concentrations and eliminations.

3.3.2. Denitrification rate, DNR

Kuwaja and Klapwijk [1] listed denitrification rates for raw wastewater in the range of 1.0–6.0 mgN/(gMLVSS h), for acetate 2–20 mgN/(gMLVSS h) and endogenous rates of 0.2–0.8 mgN/(gMLVSS h). The denitrification rates for the pre-DN are in the upper range of these values (Table 8). The reasons could be the easily degradable carbon source (Fig. 3) and as well the relatively high and constant temperature. Interestingly, the DN-rates for the first mixing basin for pre- and post-denitrification are almost in the same range—4.4 and 3.7 mgN/(gMLVSS h), respectively. The denitrification rates for post-denitrification are significantly different in the two plants. Plant I showed slightly higher rates than for endogenous denitrification while plant II yielded much higher values of 1.8 and 1.1 mgN/(gMLVSS h) (Table 8) and thereby reproduced the good results as obtained by Adam [2], who indicates rates of 2.4 mgN/(gMLVSS h). If less disturbances, the more complex real wastewater, the build up of special storage compounds or some other influences are responsible for the higher denitrification rates in plant II is not clear yet.

4. Conclusions

The evaluated membrane bioreactors at laboratory scale and pilot scale show eliminations and effluent concentrations in the range of commercial plants and of published values for degradation rates. This is partly due to the large variety of operation conditions of a commercial plant (e.g. temperature, inflow concentration, recirculation schemes).

Tests for biodegradability of the different wastewaters (synthetic and domestic) showed a slightly disproportionate behaviour despite the same COD:TN:TP ratios.

With the pilot plant II (domestic wastewater) it was possible to repeat the very good denitrification rates for post-denitrification. For plant I (laboratory scale, synthetic wastewater) it was not possible yet.

In the future, more work needs to be directed to a better adaptation of synthetic wastewater compositions to domestic wastewater. Particularly the portions of different biodegradable carbon sources have to be reviewed. The aim of the whole research is the clarification of the reasons for the good post-denitrification without carbon dosing. The results of the study showed the comparability of the plant I against standard commercial plants despite the synthetic influent and thereby establish a fundamental basis for future research.

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