INTENSIFICATION OF THE DENITRIFICATION PROCESS BY ADDITION OF ORGANIC MATERIAL

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ABSTRACT

The most widespread method of nitrogen removal in Sweden is pre-denitrification. At wastewater treatment plants with low, unsufficient C/N ratio addition of an external carbon source is necessary for intensification of the denitrification process. In this way an increase of the C/N ratio, denitrification rate and a fraction of easily biodegradable organic compounds occurs in the influent to the biological step. Studies of the effects of ethanol addition on nitrogen removal in a pre-denitrification system were carried out at pilot plants located at the Uppsala and Stockholm municipal wastewater treatment plants. Most studies were focused on use of ethanol to improve the nitrogen removal. The process performance in the pilot plant studies was monitored with on-line measurements, 24-hour composite samples and numerous grab samples. The sludge activity was monitored through measurements of nitrate uptake rate (NUR) and oxygen uptake rate (OUR). The activity of sludge that was already adapted to ethanol was studied with respect to other carbon sources. Monod kinetics was used to describe the sludge activity as a function of the substrate concentration. The paper summarises the results of studies of the addition of glycol and acetic acid as potential easily degradable organic materials for intensification of the denitrification process.

KEYWORDS

Activated sludge; carbon source; denitrification; ethanol; nitrogen removal; simulation

INTRODUCTION

Many Swedish wastewater treatment plants with biological nitrogen removal have problems with the process efficiency due to a low organic material content in the influent. Pre-precipitation widely used for phosphorus removal results in removal of organic carbon in suspended and colloidal form. Chemical pre-precipitation causes also a decrease of soluble carbon content, C/N ratio and the denitrification rate (Henze et al. 1992). To meet more stringent effluent standards for nitrogen removal many treatment plants need to upgrade their technological process. In Sweden the actual total nitrogen effluent standards are in the range of 8 - 15 mg N/l so an intensification of the denitrification process is necessary. Addition of an external carbon source is a possible way to overcome these problems. It increases both the denitrification rate and C/N ratio as well as the fraction of easily biodegradable organic materials. The process allows also to obtain a higher nitrogen removal efficiency in the existing basin volumes. Pre-denitrification is the most widespread method of nitrogen removal in Sweden and is used at about 70 -80 % of wastewater treatment plants with nitrogen removal (Eklund, 1995). Swedish experience with ethanol and methanol used in full-scale were reported by Plaza (1990), Andersson et al. (1995) and Nyberg et al. (1995). Christensson et al. (1994) in their laboratory-scale studies, confirmed that ethanol is considerably more readily available as a carbon source for denitrification than methanol. Moreover, a shorter adaptation time was needed. The growth rate of denitrifiers with ethanol was 2-3 times higher than with methanol and a substrate consumption per kg NO_3-N

removed is about 20 % lower. Ethanol can be obtained as a relatively inexpensive waste product from the pharmaceutical, chemical and brewing industry.
The oxygen uptake rate (OUR) during nitrification inhibition with allylthiourea (ATU) may be described using the Monod equation:

$$OUR = \frac{OUR_{max} * S}{K_S + S} + b \qquad (1)$$

where: OUR = oxygen uptake rate
OUR_{max} = maximum oxygen uptake rate
K_S = half saturation constant
S = concentration of organic material
b = respiration rate due to endogenous respiration

There are three different phases of the oxygen uptake rate that can be distinguished during adding of an easily biodegradable organic substance in batch tests. In the first phase, a rapid linear initial oxygen uptake rate is obtained due to oxidation of the easily biodegradable organic material. Following this process, a second phase is observed with a much slower oxygen uptake rate in which bacteria consume hydrolysis products. Finally, there is the third phase with a slow oxygen uptake rate due to the endogenous respiration.

In order to evaluate the effects of adding an external carbon source on the pre-denitrification process performance several factors must be considered as:
- type of organic material based on classifications as in the IAWQ model (Henze et al.,1994)
- rate constants in the Monod formula;
- adaptation of microorganisms to the carbon source;
- denitrification rate;
- effects on sludge production and properties
- costs of organic materials

MATERIALS AND METHODS

Most of the studies were performed at pilot plants located at the Uppsala and Stockholm wastewater treatment plants. Some complementary investigations were carried out both as continuous and batch tests.
on a laboratory scale with synthetic wastewater.

Pilot plant study - Uppsala

The pilot plant at Uppsala consisted of two separate lines (P1 and P2). Both lines operated as a single sludge system with pre-denitrification. The volume of the activated sludge tank in each line was 2.35 m^3. The tank was divided into three zones: anoxic zone, aerated zone and anoxic mixing zone. The volumes of the particular zones were: 18%, 67% and 15% of the total reactor volume, respectively. A settling tank had a volume of 0.55 m^3. An external carbon-source was added directly to the pre-denitrification tank. The pilot plant was equipped with various on-line meters as shown in Fig. 1 and specified in Table 1. A novel computerized control and supervision system (CCS) was designed specially for the pilot plant by researchers from the Uppsala University. It included the monitoring and control of flows and oxygen concentration in the nitrification tank. The on-line measurements data were collected and stored by the computer system and were evaluated by Matlab, Excel and a special program written in Turbo Basic (Carlsson et al., 1997). *Experimental strategy and operational conditions*. Both lines were supplied with the same wastewater pumped from the full scale plant after pre-precipitation with ferric chloride. Line P1 was operated with ethanol dosing, and P2 was used as a reference line. The characteristic process parameters are summarized in Tables 2 and 3. For the flow rate of Q = 220 l/h the wastewater retention time was 11.4 h.

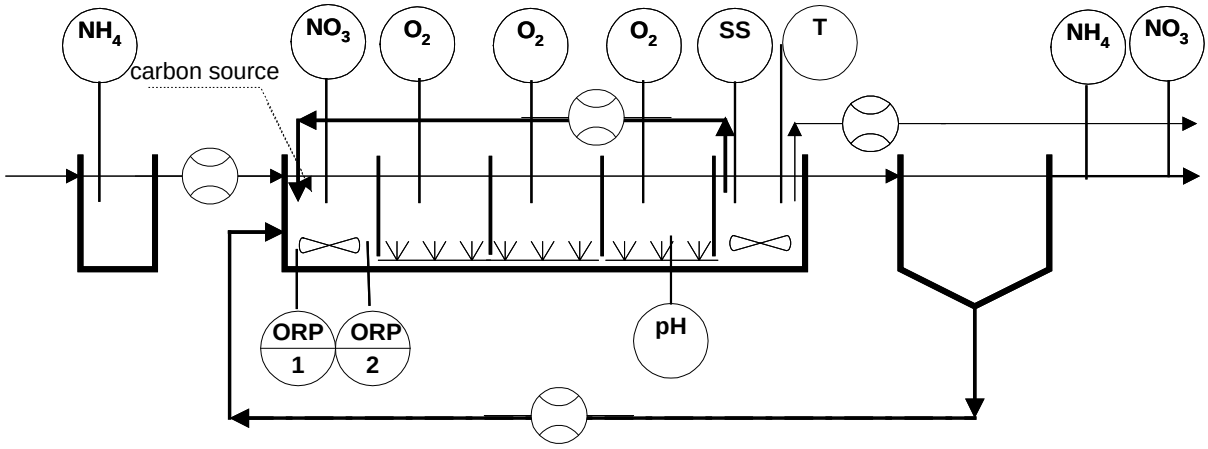


Fig. 1. Layout of the pilot-scale activated sludge plant.

Table 1. On-line mesurements in the pilot plant

Sensor (numbers)	Location
DO (6)	Cell 2-4 in P1 and P2
Ammonia (1)	Influent or effluent
Nitrate (2)	Cell 1 in P1 and effluent of P1 and P2
Susp. solids (2)	Cell 5 in P1 and P2
pH (2)	Cell 4 in P1 and P2
Redox (4)	Cell 1 in P1 and P2
Temp. (1)	Cell 5 in P1

Table 2. Capacity of pumps in the pilot plant and flow rates used in most experiment

Parameter	Uppsala		Stockholm
Flow (numbers)	Capacity (l/h)	Average used during the study, (l/h)	Average used during the study, (l/h)
Influent (2)	100-500	220	120
Return sludge (2)	100-350	220	265
Internal recirculation (2)	300-1000	660	-
Excess sludge (2)	5-60	6	2.3
External carbon (solution)	0 - 8	0.73 (1:50)	9 (1:500 - 1:1000)

Table 3. Operation parameters

Parameter	Unit	Uppsala	Stockholm
Suspended solid concentration in the aeration tank	g/l	2.5	4.2
Aerobic sludge age	days	15	6.8-9
O_2 concentration in the first aeration chamber	mg/l	2	1.5-2
Ratio denitrification/nitrification zone		0.18	0.25
Added ethanol concentration to the influent calculated as COD	mg/l	100	130-160

Sampling procedure and analytical methods. Three types of measurements were performed at the pilot plant: on-line measurements (stored every 10 minutes), 24-hour samples and batch tests. Twenty-four hours composite samples were collected twice a week from the influent and effluent from both lines. The following

parameters were measured: COD, filtered COD, Tot-N, NH_4-N, NO_2+NO_3-N, HCO_3 and SS. During certain periods of an intensive sampling program twenty-four hour composite samples were collected every day. All analytical procedures were performed according to the Swedish Standards (SIS). Batch tests were used to measure sludge activity in both experimental lines. Potential denitrification rates were determined directly by measuring the nitrate uptake rate (NUR) and indirectly by oxygen uptake rate (OUR) tests. Measurements of NUR and OUR were made according to Kristensen et al. (1992).

Pilot plant study - Stockholm

The research study on intensification of the denitrification process by addition of glycol and acetic acid was conducted at a pilot plant located at the Himmerfjärden municipal wastewater treatment plant, south of Stockholm. The pilot plant consisted of a biological reactor with pre-denitrification and a secondary clarifier. The volume of the biological reactor was 0.5 m^3. The denitrification zone occupied 25 % of the total reactor volume. Process operating parameters are presented in Tables 2 and 3. On-line measurements included: Total Organic Carbon (TOC) in the influent, MLSS and periodically NH4-N and NO3-N in both influent and effluent. Grab samples of wastewater were taken 3 times a day and the same analysis as at the Uppsala pilot plant were performed.

Laboratory-scale study

A laboratory-scale plant consisted of a biological reactor with a total volume of 70 l. The volume of the anoxic zone was 20 l. The plant was supplied with a synthetic wastewater (Q = 200 l/d) with the following characteristics: COD - 300 mg/l, P_{tot} - 6.5 mg/l and N_{tot} - 50 mg/l. Adaptation of sludge to ethanol dosages was evaluated using OUR tests. The sludge already adapted to ethanol was used to study the Monod kinetics. The endogenous respiration was measured as the oxygen respiration rate after aeration of the sludge for a few hours.

RESULTS AND DISCUSSION

C/N ratio and fraction of readily biodegradable organic materials in the wastewater

The influent and effluent characteristics at the Uppsala and Stockholm pilot plants are presented in Table 4. A weekly variation of total COD was observed during the experiments (Fig. 2 and 3). There were significant differences in the average COD and filtered COD values observed mainly in wastewater from Uppsala. The C/N ratio and the fraction of readily biodegradable organic material also varied weekly (Plaza, 1990; Xu and Hultman, 1996). A low C/N ratio results in poor denitrification that does not ensure a low nitrate concentration in the effluent.

Table 4. Influent and effluent characteristics for both pilot plants (average values)

Pilot plant	Uppsala				Stockholm		
Parameter (mg/l)	Influent	Range	Effluent P1	Effluent P2	Influent	Range	Effluent
COD	307	150-590	77	64	150	75-310	59
CODf	105	40-200	45	40	130	55-280	48
TOC					38	18-90	15
Tot-N	33.3	23.3-45.6	11.2	22.5	25.2	17.5-30.0	9.7
Tot-Nf	26.8	14.6-36.4	8.9	20.1	23.1	13.8-26.1	8.6
NH_4-N	23.3	10.5-31.4	0.22	0.13	20.2	14.6-25.0	0.9
NO_3+NO_2-N	0,43	0.02-2.0	6.5	18.1	0.1	0.0-1.1	4.0
SS	98	50-270	33	17	72	35-180	28

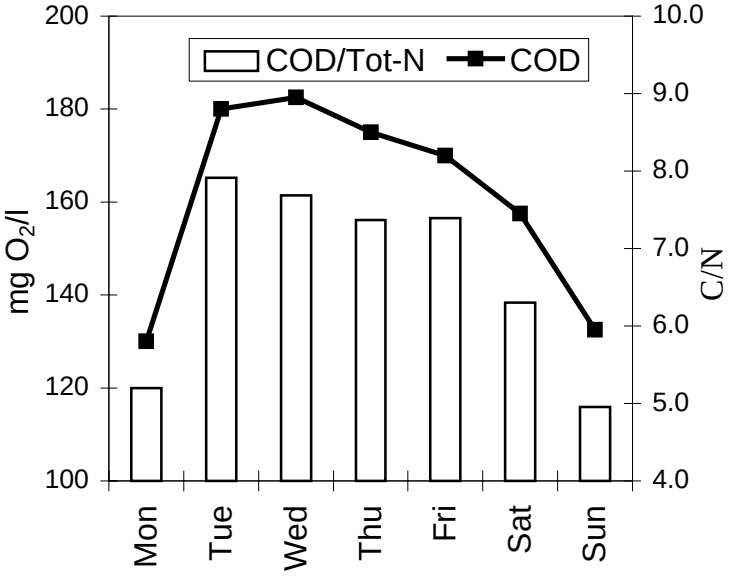


Fig. 2. Weekly variation of COD and COD/Tot N in the influent - Stockholm.

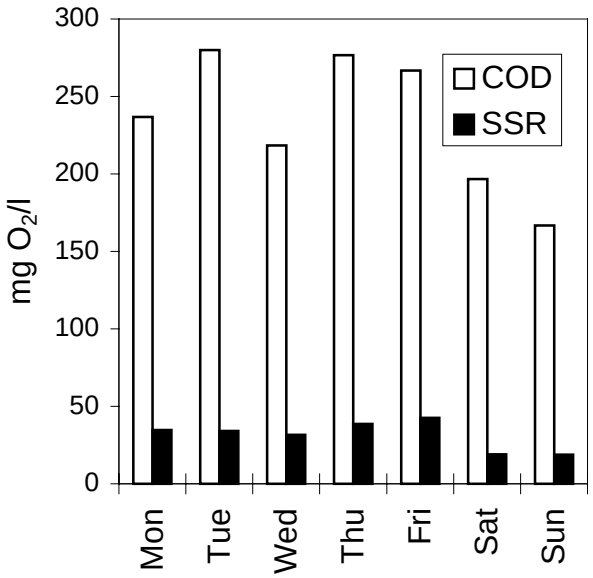


Fig. 3. Weekly variation of COD and fraction of readily biodegradable COD (S_{SR}) in the primary effluent in Uppsala.

Ethanol

Addition of ethanol to the reactor increased significantly the efficiency of nitrogen removal. Concentration of the total nitrogen decreased from 22.5 mg N/l for the reference line to 11.2 mg N/l for the line with ethanol. The varying concentration of total nitrogen in the influent to the Uppsala pilot plant and in the effluent from line P1 and P2 are presented in Fig. 4.

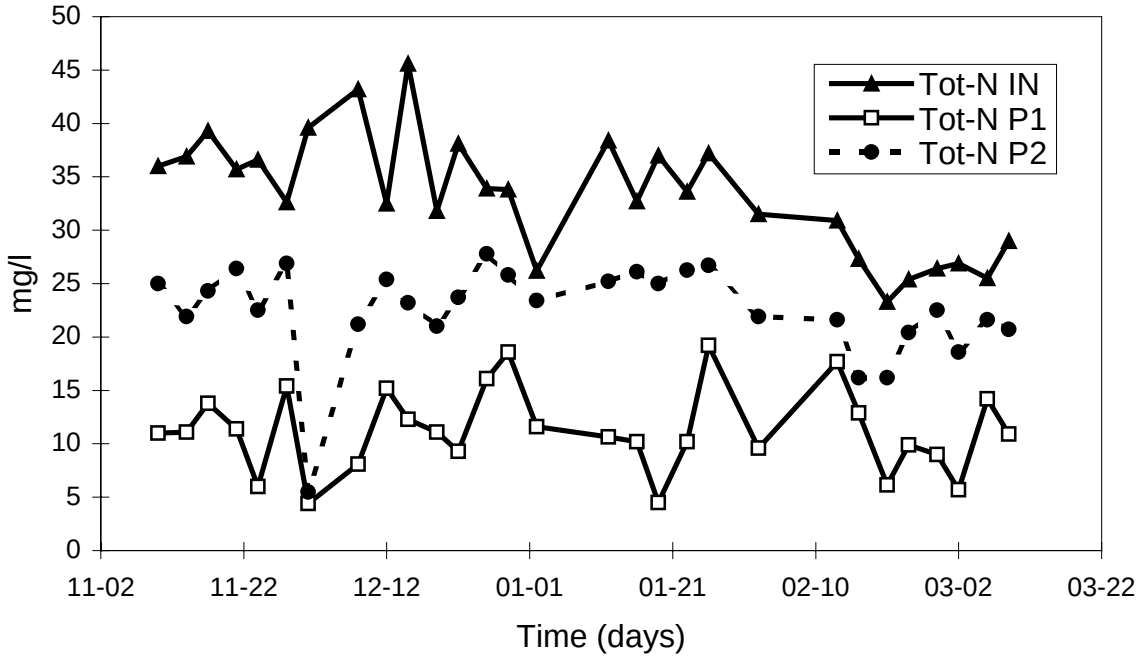


Fig. 4. Total nitrogen concentration in the influent and effluent - P1(addition of ethanol) and P2 (without addition of ethanol).

Fig. 5 shows the influence of the C/N ratio on the effluent NO_3-N concentration from both pilot plants (P1 and P2). The data from the two lines shows a decreasing trend for the effluent NO_3-N concentration with increasing values of the C/N ratio. However, a substantial difference in the effluent NO_3-N concentrations

between the two plants is caused by the dosage of ethanol. Over a 4 - month long period of study a dosage of ethanol corresponding to 100 mg COD/l resulted in an improved removal of NO_3-N and a decrease of the effluent concentration from 18.1 mg/l to 6.5 mg/l (average 11,6 mg/l). During that time the COD/N ratios for line P1 with ethanol and for line P2 (reference line) averaged 12.1 and 9.0, respectively. An increase of the COD/N ratio resulted also in a higher nitrogen removal. The average values for the reference line and the ethanol line were 32.4 to 66.4 %, respectively. Fig. 6 presents the influence of the COD/N ratio on the total nitrogen removal in both lines.

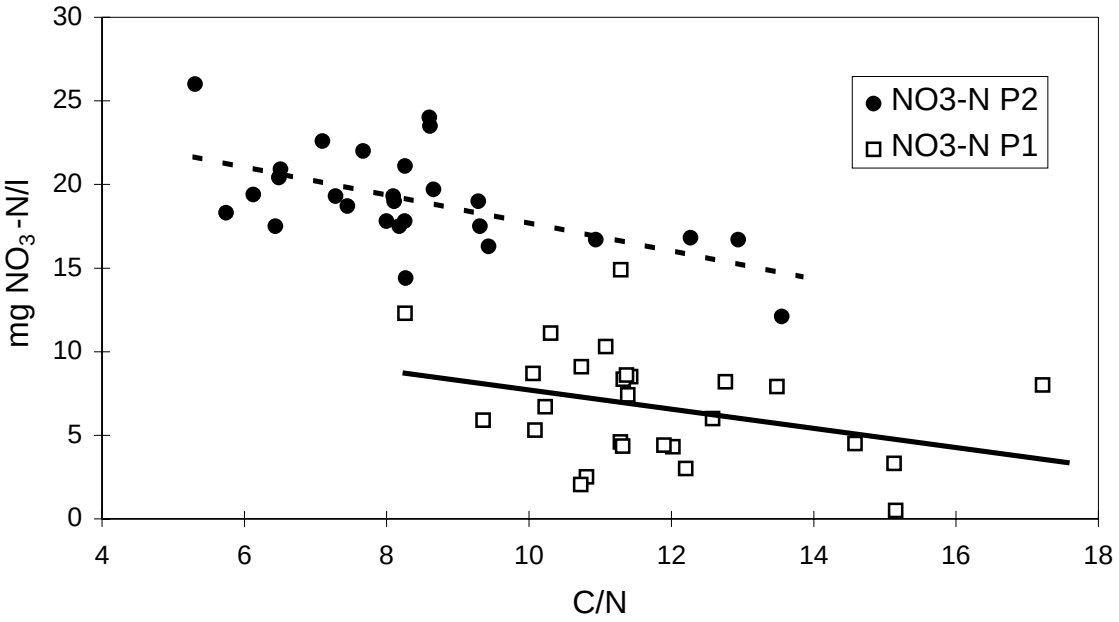


Fig. 5. C/N ratio in the influent vs. the effluent NO_3-N concentration.

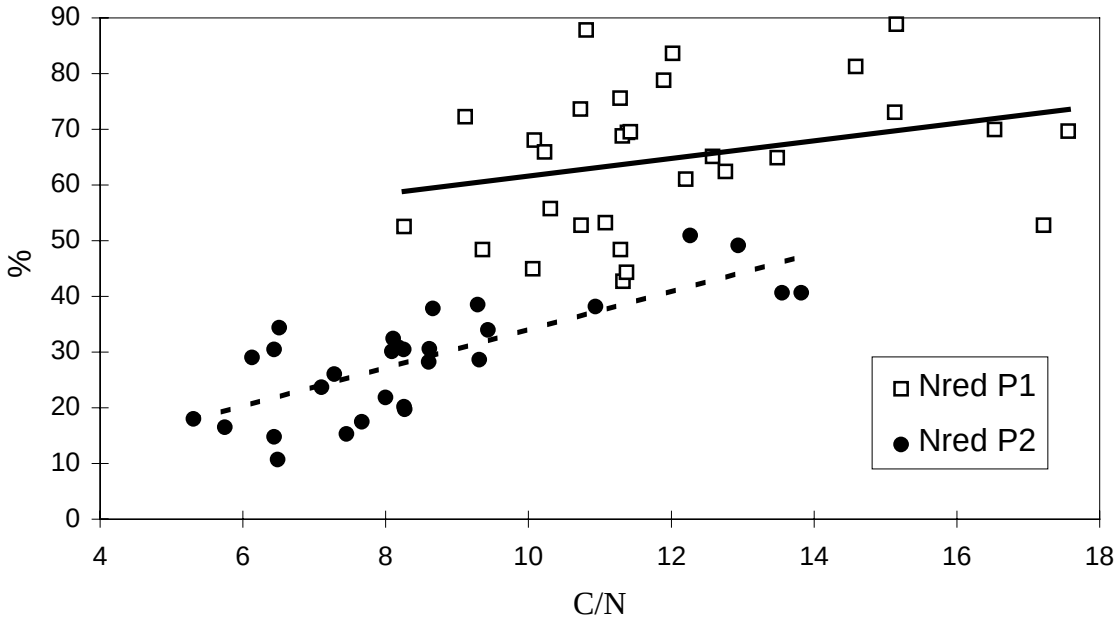


Fig. 6. Nitrogen removal efficiency vs. COD/N ratio. P1 and P2 with and without ethanol addition, respectively.

Glycol

The study conducted at the pilot plant in Stockholm was focused on the intensification of the denitrification process by addition of glycol and acetic acid. Glycol was a waste product from the Stockholm airport, where it is used for plane deicing. For 7 days of June acetic acid was fed to the plant in amount equivalent to 130 mg COD/l. It raised the COD/N ratio from 6.1 to 11.9. The average nitrogen removal efficiency was 72.1 %. For 12 days of August glycol was dosed to the rector in amount corresponding to 160 mg COD/l, increasing the COD/N ratio in the influent from 7.1 to 14.5. The average nitrogen removal was 69.3 %. Removal of nitrogen at low temperatures was investigated from October 11 to November 12. A dosage of glycol corresponding to 100 mg COD/l caused an inrcrease of the COD/N ratio from 6.3 to 10.2. The period of the study was divided into 2 stages with different wastewater temperatures. The average nitrogen removal at 13 ^{o}C was 67.4 % while at 8 ^{o}C it was slightly lower (63.1 %). The drop of temperature was accompanied by a decrease of the nitrification efficiency. It resulted in a higher ammonia nitrogen concentration in the effluent. Figure 7 presents total nitrogen concentrations in the influent and effluent discharged from the pilot plant where both acetic acid and glycol were dosed at different temperatures. Process parameters for nitrogen removal with addition of different external carbon sources are summarized in Table 5.

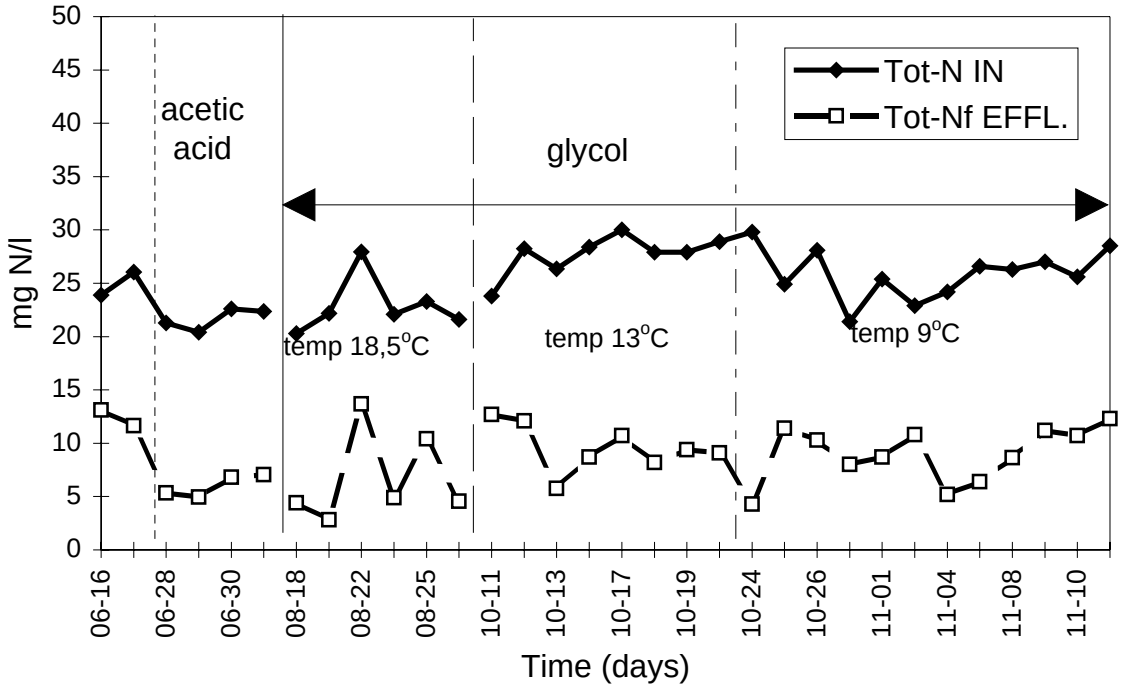


Fig. 7. Total nitrogen in the influent and effluent - the pilot plant in Stockholm.

Table. 5. Process parameters for nitrogen removal with addition of different external carbon sources (average values)

Carbon source	Temp. ^{o}C	Influent COD unf. (filtered) (mg O_2/l)	COD/Tot-N (COD_f/Tot-N)	Influent COD unf.(filtered) + org.material (mg O_2/l)	COD+ org.material/Tot-N (COD_f+org.material/Tot-N)	Nitrogen removal %
acetic acid*	20.5	132 (111)	6.1 (5.1)	261 (240)	11.9 (11.1)	72.1
glykol 1*	18.5	161 (135)	7.1 (5.9)	329 (303)	14.5 (13.3)	69.3
glykol 2*	11.5	166 (141)	6.3 (5.5)	269 (248)	10.2 (9.4)	63.5
ethanol**	12-15	307 (105)	9.0 (3.1)	407 (205)	12.1 (6.0)	66.4

* Stockholm ** Uppsala

Adaptation

Ethanol. Adaptation of denitrification bacteria to ethanol may be measured as a direct effect on the nitrogen removal efficiency or as the influence on the maximum denitrification rate in batch studies. It was found that addition of ethanol assures a rather constant nitrogen removal efficiency and low nitrate concentrations in the effluent just after 30-40 hours. The maximum denitrification rate was, however, not obtained until after 12 days of ethanol addition. Values of the maximum denitrification rate above a certain threshold value seemed to have only a little influence on the nitrogen removal efficiency.

Studies showed that through the ethanol dosing an increase in the maximum denitrification rate was observed. This rate (expressed as mg NO_X-N/g VSS*h) was about 2.5 times higher in the ethanol line (P1) than in the reference line (P2) after the adaptation period was completed. The results of nitrogen removal rate (NUR) tests with ethanol, dosed at the concentration corresponding to 200 mg COD/l and performed at 15 ^{o}C, were compared with results obtained for acetate (Fig.8). A similar activity for ethanol and acetate was observed in the ethanol line (P1). The results are in good agreement with the study reported by Hallin et al. (1996).

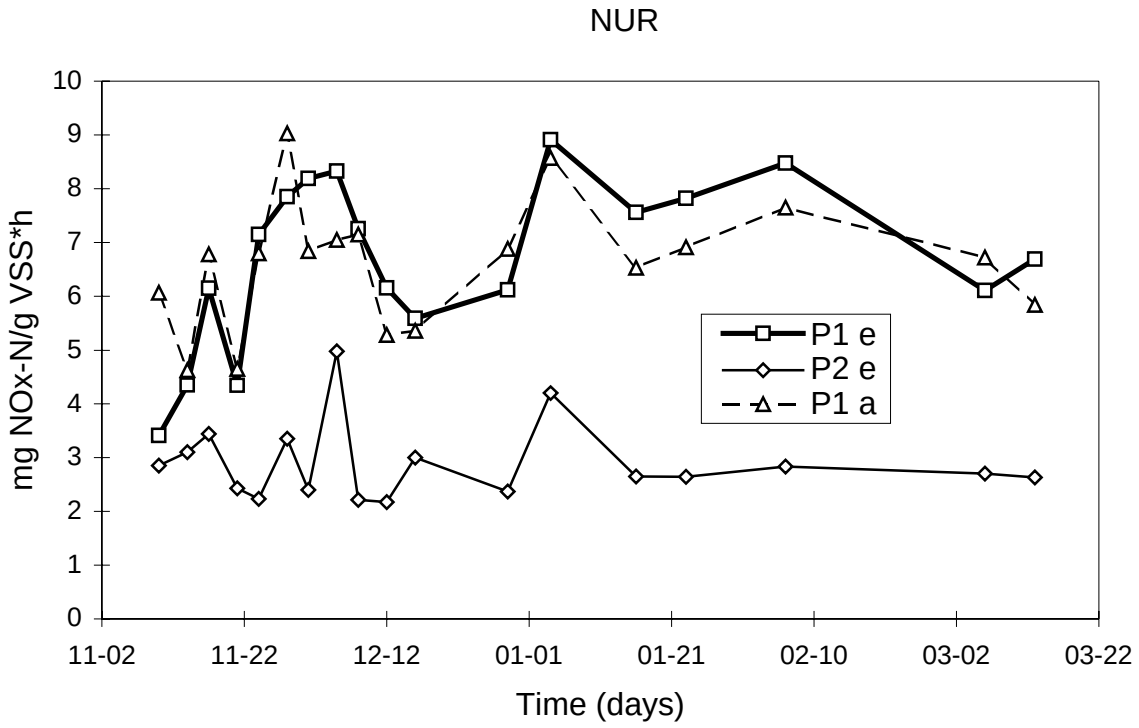
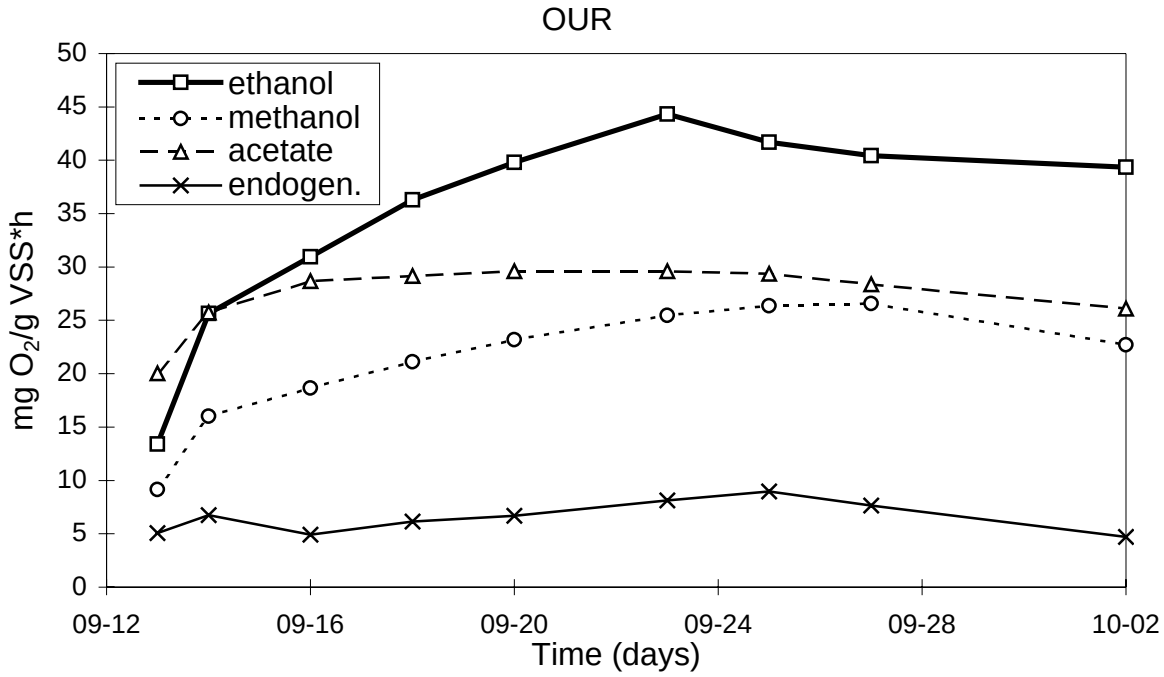


Fig. 8. Denitrification rates (NUR) for the ethanol line (P1) with ethanol (e) and acetate (a) addition at the NUR tests and for the reference line (P2) with ethanol addition to not-adapted sludge.



Fig, 9. Oxygen uptake rates (OUR) for different added external carbon sources at the OUR tests during sludge adaptation to ethanol.

Adaptation studies were also performed in a laboratory scale. The adaptation was expressed as oxygen uptake rate (OUR). The oxygen uptake rate reached maximum values after about 12 days. The sludge showed a higher activity for ethanol than for acetate (Fig. 9). The oxygen uptake rate for methanol also increased 2.5 times once the adaptation period had been completed both in the laboratory and pilot experiments in Uppsala (Fig.10).

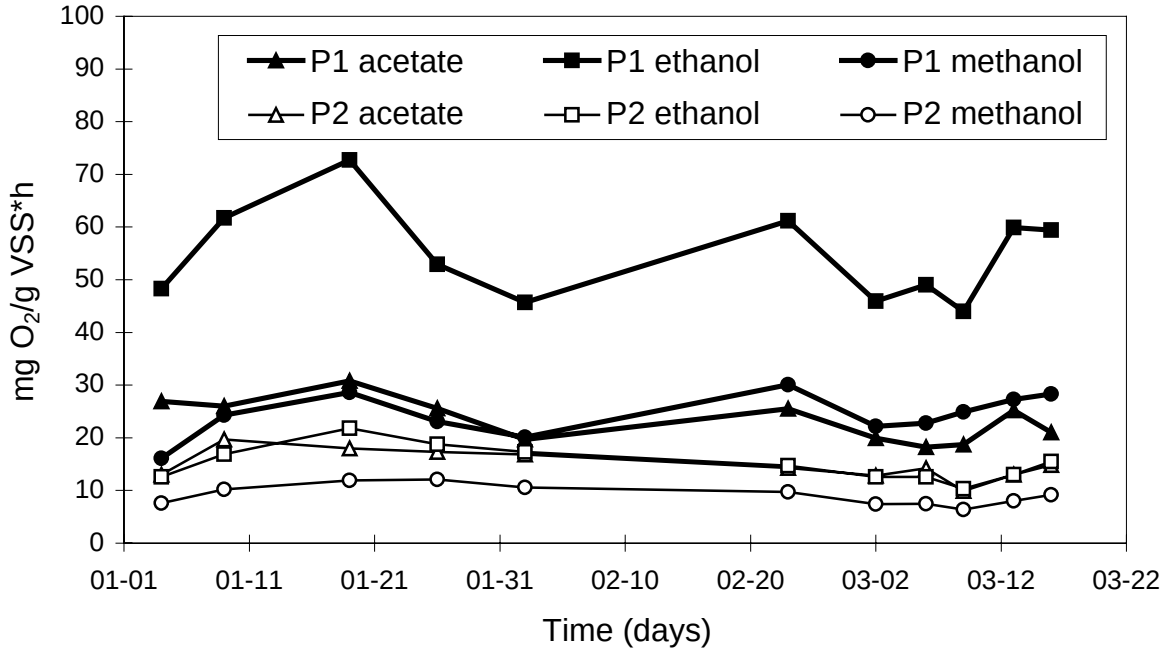


Fig. 10. OUR tests for different external carbon sources - line P1 and P2 at the Uppsala pilot plant.

Glycol The experiments were also conducted to evaluate how fast the nitrogen removal efficiency was affected by glycol addition.The results of the experiments including nitrogen removal efficiency, amount of nitrogen assimilated and denitrified are summarized in Table 6. Only 20 h after the glycol was introduced to the process the nitrogen removal efficiency reached 80%. After the adaptation period, the denitrification rate for the glycol dosage corresponding to 100 mg O_2 /l was 6.5 - 8.4 mg NO_x -N/g VSS*h.

Table 6. Nitrogen removal efficiency with glycol

Time of dosing	Influent COD (mg O_2/l)		Influent COD + organic material (mgO_2/l)		Nitrogen removal	Nitrogen assimilated		Nitrogen denitrified
h	unf.	filtered	unf.	filtered	%	mg N/l	%	mg N/l
0	110	99	110	99	13	2.4	10	0.7
3	155	112	360	320	36	3.1	11	7.6
6	160	143	365	349	47	2.3	9	9.9
20	203	176	405	380	81	2.5	11	15.9
23	154	145	360	351	76	2.6	12	14.2
26	116	110	324	319	76	2.7	12	13.7

Monod kinetics for methanol and ethanol additions

Monod kinetics was studied using the oxygen uptake rate (OUR) test for the activated sludge from the laboratory activated sludge pilot plant. The plant included denitrification and the sludge was already adapted to ethanol. The addition of ethanol resulted in a respiration curve with two different oxygen uptake rates while during methanol dosing a constant respiration rate was observed without an initial rapid phase (Fig. 11).

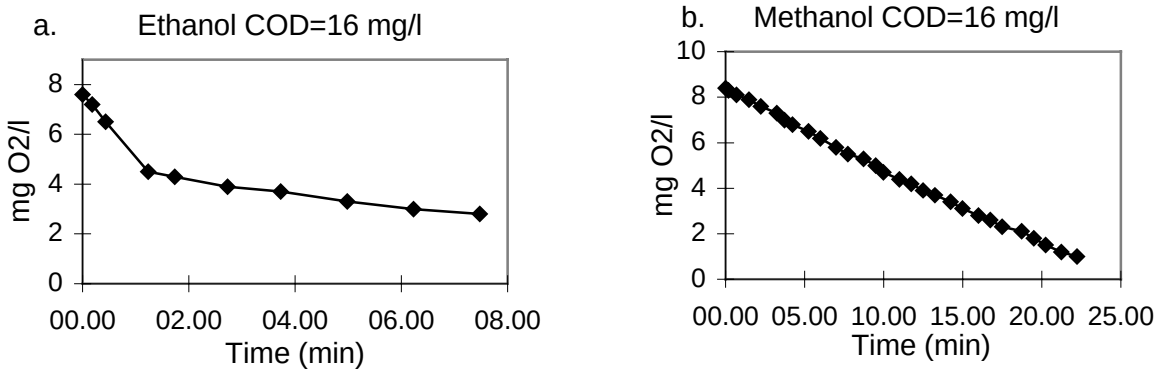


Fig. 11. OUR tests with ethanol (a) and methanol (b) added in amount of 16 mg O_2/l as COD.

The difference in the initial phase of the Monod kinetics for ethanol and methanol are shown in Fig. 12 . The analysed sludge was taken from the pilot plant and was previously adapted to ethanol. The experiments were conducted for ethanol doses above 8 mg COD/l. At the lower concentration of COD it was difficult to distinguish the first phase of OUR for the actual sludge concentration of 2.5 g/l. During the experiments a constant value of the OUR was obtained equal to 73 mg O_2/g VSS*h (including endogeneous respiration). The influence of the methanol dosage could be described by the Monod relationship. The maximum OUR value (including endogenous respiration) was 39 mg O_2/g VSS*h and the half saturation constant was 48 mg COD/l.

Complementary studies were made to investigate the influence of low dosages of ethanol on the sludge activity. The sludge samples were taken from the laboratory pilot plant installed at the Div. of Water Resources Engineering (Fig. 13). The sludge was adapted to ethanol. To measure the first phase of the oxygen uptake rate the sludge had a concentration of 0.5 g/l. In the first phase the maximum oxygen uptake rate for the (including endogenous respiration) was 55 mg O_2/g VSS*h and the half saturation constant was only 1 mg COD/l. In the second phase the oxygen uptake rate was much lower and increased slightly with the increase of ethanol dosage.

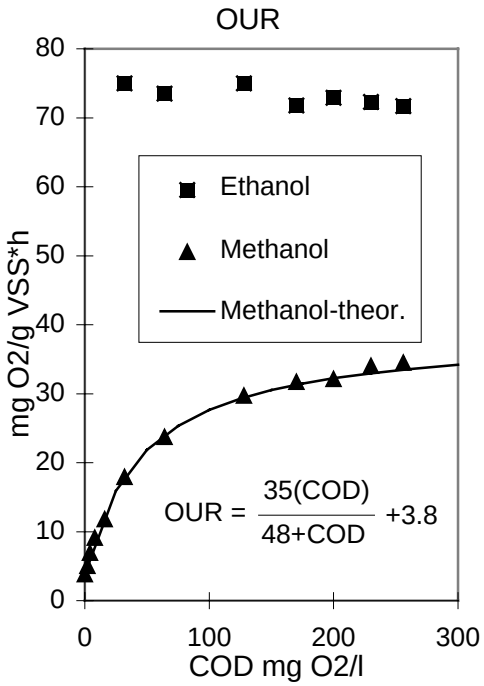


Fig. 12. OUR vs COD. described by the Monod kinetics - added COD concentrations from 0 to 300 mg O_2/l.

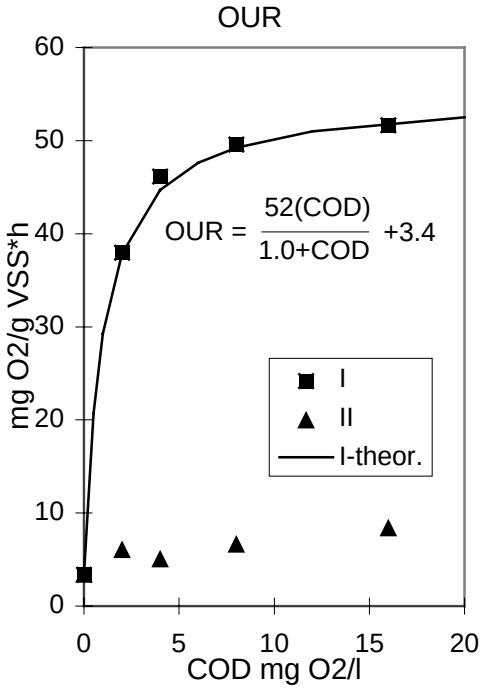


Fig. 13. OUR vs. COD described by the Monod kinetics - initial and second phase for added COD concentrations from 0 to 16 mg O_2/l.

CONCLUSIONS

Studies have been performed both in laboratory and pilot scale to evaluate the use of ethanol and glycol as an external carbon source for the intensification of the denitrification process. The studies have shown:

Ethanol is a readily biodegradable organic substance for adapted sludges and its dosing results in denitrification rates comparable with acetate dosing. Acetate is readily biodegradable for the denitryfying bacteria.

Addition of ethanol to a pilot plant improved the nitrogen removal efficiency after 40 hours of operation. However the maximum denitrification rate measured in batch tests was, not obtained until about 12 days of ethanol addition.

Glycol turned out to also be an easily biodegradable organic material for bacteria and only one day was required to achieve a high nitrogen removal efficiency. The denitrification rates were from 6.5 to 8.4 mg/ NO_3-N/g VSS*h.

The influent to the pilot plants had a low fraction of readily biodegradable organic materials. The COD/N ratio does not provide enough information on the efficiency of the biological nitrogen removal process. The nitrogen removal efficiency was only slightly improved by an increasing C/N ratio. The addition of ethanol and glycol improved significantly the nitrogen removal efficiency mainly due to its function as readily biodegradable organic materials and to less extent due to an increase of the C/N ratio.

The oxygen uptake rate tests (OUR) performed with ethanol showed a rapid initial phase followed by the second phase with a much lower oxygen uptake rate. The initial oxygen uptake rate followed the Monod relationship with a very low value of the half saturation constant (K_S = 1 mg COD/l).

The studies on methanol addition to sludges previously adapted to ethanol showed a much less maximum oxygen uptake rate, if compared with the addition of ethanol, and a high value of the half saturation constant (48 mg COD/l).

Sludge once adapted to ethanol shows an increased activity towards the other organic materials like methanol and acetic acid.

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