

DESIGN CONSIDERATIONS FOR A NITRIFICATION-DENITRIFICATION PROCESS USING TWO FIXED-BED REACTORS IN SERIES

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ABSTRACT

This paper concerns design considerations (kinetics and hydraulic criteria) for a biological nitrification-denitrification attached culture process for sewage treatment.

The treatment system is composed of two submerged filters. The first one, an anoxic reactor, denitrifies and retains wastewater suspended solids. The second one oxidizes organic carbon and ammonia nitrogen.

It was found that the main parameter for the system is the recycle ratio since it sets the real hydraulic detention time in reactors and determines the quantity of nitrogen available to the denitrification process.

KEYWORDS

Biological nitrification-denitrification. Attached growth sewage treatment. Submerged filters.

NOTATION

Symbol	Quantity name	Dimension	Symbol	Quantity name	Dimension
H	Height	L	v	Superficial velocity	$L^3 T^{-1}$
A	Area	L^2	Q	Liquid volumetric flow rate	$L^3 T^{-1}$
V	Volume	L^3	Q_G	Gas volumetric flow rate	$L^3 T^{-1}$
ϵ	Void volume fraction	-	R	Recycle ratio	-
S	Soluble material concentration	$M L^{-3}$	HEC	Height of efficient column	L
τ	Space time (volume of reaction phase divided by volumetric flow rate of phase entering the reactor)	T			

INTRODUCTION

Up to today, several patterns of nitrification - denitrification systems have been proposed for suspended cultures. In the case of attached cultures, only a

few alternatives exist. As different works have demonstrated successful applicability of submerged filters to specific cases of sewage treatment (Haug and Mc Cartý, 1972 ; Mc Harness *et al.*, 1975 ; Gaid *et al.*, 1982 ; Faup *et al.*, 1982), etc... it was decided to study a nitrification-denitrification system using these types of reactors.

Figure 1 shows the selected system for treatment using column biological filters. The first reactor achieves filtration and denitrification. Flow in this filter has to be down-ward in order to prevent the entrainment of filtered solids by nitrogen bubbles, as previously demonstrated (Jimenez *et al.*, 1984)

The second reactor oxidizes organic matter and ammonia nitrogen. Oxygen requirements in this upflow reactor are satisfied by direct bubbling aeration.

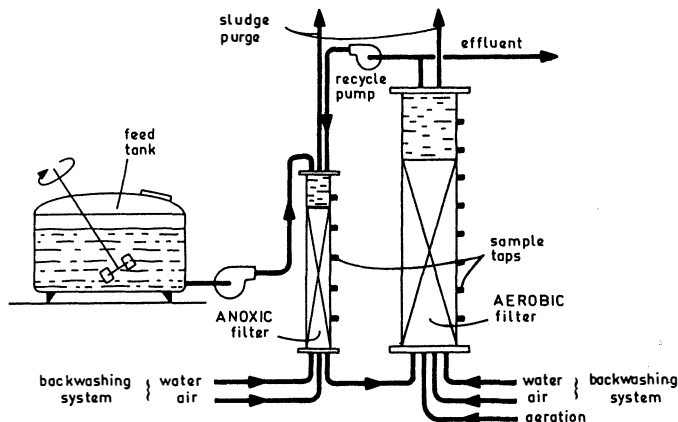


Fig. 1. Selected system for sewage nitrification-denitrification

Design of pilot plant facilities

The anoxic filter is made of a 1.8 m tall PVC column having a 53.7 mm inside diameter. The aerobic column reactor was 2.5 m tall and had a 150.6 mm inside diameter. The bottom in this last reactor was constructed to feed wastewater and air uniformly.

Samples taps were located at 0.28 m intervals along the length of the columns : 5 in the downflow reactor and 8 in the upflow reactor.

Each column was packed with media with an effective particle diameter (d_{10}) of 2.8 mm and a 50 % porosity. Fill depth was 1.42 m in the anoxic reactor and 2.1 m in the aerobic reactor, so water void volumes were 1.6 l and 18.7 l respectively.

Influent quality

Wastewater used in this study came from an extended aeration treatment plant (10 000 eq. inhab.). Some characteristics of the non-settled sewage are presented in Table 1.

TABLE 1 Characteristics of influent wastewater

PARAMETER	MAXIMUM	MEAN	MINIMUM
TOCs (mgC/l)	115	71	26
TOCc (mgC/l)	178	122	64
CODs (mgO ₂ /l)	373	263	95
CODc (mgO ₂ /l)	795	568	245
NH ₄ (mgN/l)	45	34	21
TKNc (mgN/l)	60	47	26
Pc (mgPO ₄ /l)	91	74	62
SS (mg/l)	297	184	103
pH	8.1	7.5	7.2

Backwashing

The filter cleaning operation was initiated when head loss (created both by solids retention and by biomass production) reached 3 meters water column (m H₂O). Backwashing conditions were as follows :

Combined water and air were introduced for ten minutes at flow rates of 25 m/h and 55 m/h respectively. This was followed by five minutes of water rinsing at a flow rate of 25 m/h.

Analytical techniques

The following chemical analyses were measured according to Standard Methods (1980) and "Normes Afnor" :

- Nitrogen series (Nitrate, nitrite, ammonia nitrogen and total Kjeldahl nitrogen).
- Organic matter series (COD and TOC)
- Suspended solids (SS)
- Total phosphorus
- Dissolved oxygen

Experimental program

The operating conditions are presented in Table 2.

TABLE 2 Operating conditions of recycle ratio influence study

Q l/h	R	vf m/h	h	
4	1.0	3.54	5.0	
4	2.0	5.31	5.0	hydraulic residence time of
4	2.5	6.19	5.0	influent flow in the system
6	1.0	5.31	3.4	anoxic + aerobic filter)
6	2.0	7.96	3.4	= 50 %
6	2.5	9.29	3.4	
6	3.0	10.62	3.4	Total superficial velocity in
6	3.5	11.65	3.4	anoxic reactor

At a fixed flow rate (4 or 6 l/h) the recycle ratio was increased until the nitrate concentration of the effluent decreased to 5 mg N-NO_x/l. (N-NO_x = N-NO₂ + N-NO₃). This increment in the recycle ratio allowed an increase in the oxidized nitrogen load into the anoxic reactor, thus permitting the study of filtration behaviour at different hydraulic loads but at constant influent suspended solids loading.

RESULTS AND DISCUSSION

Anoxic reactor

Filtration. As shown in Figure 2, solids retention takes place only in the top of the reactor. While it is difficult to extrapolate to full scale units it is reasonable to assume that the anoxic reactor should be modified to have better head losses distribution. An optimization study should be done to determine backwash conditions when total clogging of filter media and settled pressure losses are reached simultaneously.

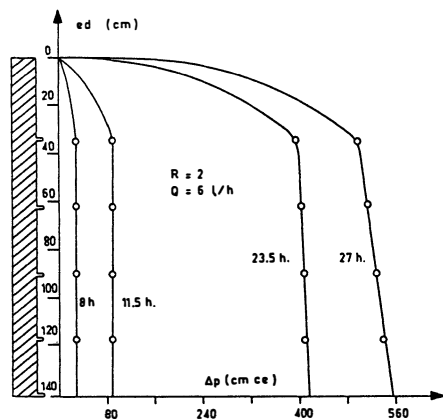


Fig. 2. Head losses distribution in anoxic submerged filter

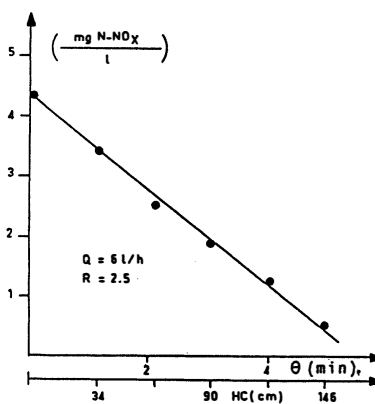


Fig. 3. Example of an oxidized nitrogen profile in anoxic reactor

Denitrification. Figure 3 shows an oxidized nitrogen profile. Linear profiles were systematically obtained for concentrations of the order of 4 to 6 mgN-NOx/l. The tracer studies made with Dextran blue colorant (Jimenez, 1985), showed that flow in both reactors was near plug flow.

At these conditions denitrification phenomena can be represented by a zero order reaction in a plug flow system. Thus we have :

$$\bar{z} = (1/kd) (NO_{xed} - NO_{xsd}) \quad (1)$$

With : NO_x : oxidized nitrogen concentration (Nitrites + nitrates)

ed : concentration at anoxic influent

sd : concentration at anoxic effluent

kd : zero order reaction coefficient, equal to 1.22 mgN/l.mn

Since biomass values are not available in reactor due to the presence of filtration solids in the system, the kd value is given only in terms of N-NO_x concentration and time.

Time is measured as a function of the water volume in the reactor (filled with clean packing) and the total flow traversing reactor. The convenience of this has been described by Francis, (1977). Thus the residence time is given by :

$$\bar{z} = V_w/Q(1+R) = \epsilon \cdot V_r/Q(1+R) \quad (2)$$

or in terms of pilot dimensions

$$\bar{z} = \epsilon \cdot HC \cdot Ad/Q(1+R) \quad (3)$$

where : V_w : is the water volume contained in the reactor filled with clean packing (ε = 50 %)

V_r : reactor volume to packing height

Hc : packing height

Ad : surface of anoxic reactor

Relating expression 3 to 1 we obtain :

$$HC = \frac{Q(1+R)}{Ad \cdot \epsilon} \cdot \frac{1}{kd} (NO_{xed} - NO_{xsd}) \quad (4)$$

At a denitrification efficiency of 100 % (HECd 100 %) NO_{xsd} equals zero and the required filter depth is given as :

$$HECd_{100\%} = \frac{Q(1+R)}{Ad} \cdot \frac{1}{kd} (NO_{xed}) \quad (5)$$

The first ratio on the right hand side of equation 5 is equivalent to the superficial velocity :

$$vf = Q(1+R)/Ad \quad (6)$$

Carbon requirement for denitrification.

The organic carbon uptake during denitrification was 5.6 ± 1 mg COD/mgN-NOx or 1.16 ± 0.2 mgTOC/mgN-NOx. For sewage, Moreaud (1979) reported carbon uptake values between 5 - 7 mg COD/mgN. These influent values do not ensure good denitrification yields. In such a system only values greater than 14 mg COD/mgN (or 4.5 mgTOC/mgN) in anoxic influent will keep denitrification efficiencies above 90 %.

Aerobic reactor

Elimination of organic matter.

A TOC profile of the aerobic reactor is presented in Figure 4. Elimination of organic matter is accomplished mainly in the bottom part of the filter by heterotrophic population.

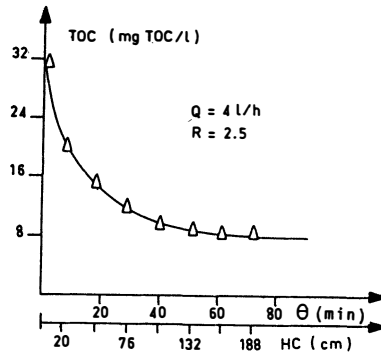


Fig. 4. TOC profile in aerobic reactor.

The elimination of organic matter measured as TOC for a first order reaction and a plug flow reactor may be written as :

$$(TOC)_z = TOC_{in} \cdot e^{-k_{TOC} \cdot z} \quad (7)$$

where $z = \epsilon \cdot Vr / Q(1+R)$ as in the anoxic reactor

TOC_{in} : is the TOC value in aerobic reactor influent

k_{TOC} : is the first order rate constant of organic carbon elimination.

Evaluation of the rate constant yield values of approximately 0.019 mn^{-1} (1.14 h^{-1}).

In terms of the size of the reactor equation 7 becomes :

$$(\text{TOC})_{\text{HC}} = \text{TOCin} \cdot e^{-(k_{\text{TOC}} \cdot \epsilon \cdot \text{HC})/V_p} \quad (8)$$

where : v_p : is the superficial velocity in aerobic reactor
 $(\text{TOC})_{\text{HC}}$: TOC concentration as a fonction of the packing height.
 TOCin : TOC concentration at aerobic influent

It must be taken into account in equations 7 and 8 that the organic carbon efficiencies are limited by the whole nitrification-denitrification system. Efficiencies greater than 65 % TOCs elimination are unattainable in this aerobic reactor.

Elimination of ammonia nitrogen

Elimination of ammonia nitrogen is achieved in aerobic reactor in two steps. In the bottom portion of the filter, where organic carbon oxidation is taking place, ammonia is removed by heterotrophic assimilation (production of new cellular material). At this step, the limiting substrate is organic carbon and the nitrogen elimination profiles are of zero order reaction type (i.e. linear).

At the top of the filter, where biodegradable carbon has been exhausted, autotrophic microorganisms oxidize ammonia nitrogen into nitrites and nitrates. In this reaction the main substrate is the ammonia nitrogen, so these profiles are exponential (Figure 5).

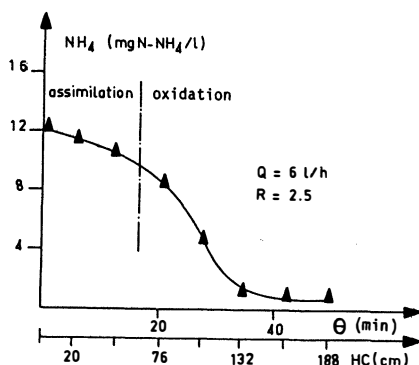


Fig. 5. Ammonia nitrogen elimination profile.

The linear zone corresponds to N-assimilation phenomenon while the exponential zone corresponds to the ammonia nitrogen oxidation.

In formulating a mathematical model for the elimination of Ammonia Nitrogen, it was assumed that these first and zero order reactions at the top and bottom of the filter respectively, were, independent thus :

1. For linear profile zone (assimilation phenomenon)

$$(\text{NH}_4)_z = \text{NH}_4 \text{ in} - k_{\text{as}} \cdot z_1 \quad (9)$$
 (Zero order reaction).

2. For exponential profile zone (oxidation phenomenon)

$$(\text{NH}_4)_z = \text{NH}_4 \text{ ox} e^{-k_{\text{ox}} \cdot z_2} \quad (10)$$

(First order reaction in a plug flow reactor)

where : $\text{NH}_4 \text{ in}$: is the ammonia nitrogen concentration in the aerobic reactor influent.

$NH_4\text{ox}$: concentration of ammonia nitrogen available to nitrification
 k_{as} : assimilation zero order rate constant. Equal to 0.149
 $\text{mgN-NH}_4/\text{l.mn}$ in this work.
 k_{ox} : Ammonia nitrogen elimination first order rate constant. Equal
to 0.087 mn^{-1} (5.22 h^{-1}).

In the filter, autotrophic bacteria are located after heterotrophic bacteria. The quantity of nitrogen available for nitrification ($NH_4\text{ox}$) is assumed to be equal to the ammonia nitrogen concentration in aerobic influent minus the ammonia nitrogen assimilated by heterotrophic bacteria (autotrophic assimilation is neglected). As long as the $NH_4\text{as}$ value is proportional to the carbonaceous consumption the following relation holds :

$$NH_4\text{ox} = NH_4\text{in} - NH_4\text{as} = NH_4\text{in} - f Y_h (\text{COT}_{in} - \text{COT}_{sn}) \quad (11)$$

where : f : is the conversion factor converting SSV into N, 0.108 mgN/mg SSV (Metcalf (1979)).

Y_h : observed cell yield coefficient for heterotrophic bacteria (0.4 mgSSV/mg COD, 1.42 mgSSV/mg TOC).

Taking HEC definition as the filter height necessary to obtain a desired degree of elimination (Elmaleh, 1978), the total height of the aerobic reactor for a fixed superficial velocity is given by the sum of the height required for assimilation plus the height required for ammonia oxidation :

$$HEC_n = HEC_{as} + HEC_{ox} \quad (12)$$

Substituting $Z = HC \cdot (\epsilon / v_p)$ in equation 9 and using the definition for

HEC, leads to the following expression :

$$HEC_{as} = \frac{f Y_h}{\epsilon \cdot k_{as}} \cdot \frac{Q(1+R)}{A} (\text{S}_{in} - \text{S}_{sn}) \quad (13)$$

and

$$HEC_{ox} = \frac{Q(1+R)}{A} \cdot \frac{1}{\epsilon \cdot k_{ox}} \cdot \text{Ln} \frac{NH_4\text{in} - NH_4\text{as}}{NH_4\text{sn}} \quad (14)$$

where : S : Is the soluble organic matter, measured as COD or TOC depending on the dimensions of Y_h .

Optimal aeration rate

The effect of air influent flow ratio on the ammonia nitrogen elimination efficiency is illustrated in Figure 6.

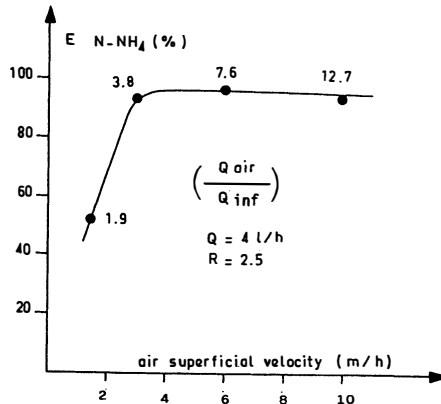


Fig. 6. Ammonia nitrogen elimination efficiency as a function of the air/influent flow ratio

The plotted curve indicates that the optimal air to influent flow ratio is 4. Higher ratios will cause greater dissolved oxygen concentrations in the aerobic effluent as a consequence of recycle.

Start-up.

The time necessary to reach "steady-state" in the anoxic and aerobic reactors after backwashing is shown in Figure 7a and 7b.

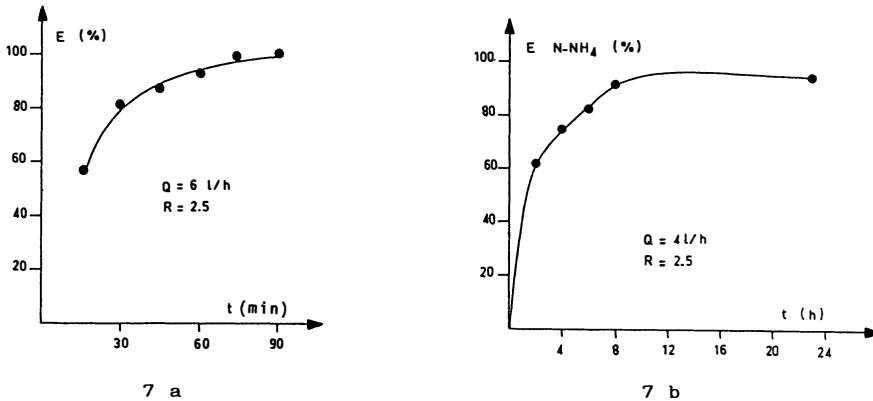


Fig. 7a, b. Start-up in anoxic and in aerobic reactor

The maximal operating efficiency is reached sooner in the anoxic reactor than in the aerobic reactor. This was expected since the specific growth rate of autotrophic bacteria is smaller than that of heterotrophic bacteria.

Nitrification-denitrification system

System efficiency. In Table 3 the calculated efficiencies at different operating conditions for the whole system and for each reactor are shown.

TABLE 3 Efficiencies at different operating conditions

Operating conditions	Q R	Whole system influent-effluent				Anoxic reactor	Aerobic reactor	Oxidized nitrogen
		TOCs	CODs	TKN	SS	denitrification	nitrification	in effluent*
4	1	87	92	95	97	100	98	7.8
4	2	90	93	99	95	93	93	6.3
4	2.5	90	93	98	97	99	95	4.0
6	1	89	92	96	95	96	98	12.0
6	2	90	91	97	95	95	96	9.4
6	2.5	89	94	96	98	87	92	7.5
6	3	88	93	96	98	45	92	6.9
6	3.5	87	84	96	98	38	89	6.8

*Concentrations in mgN-NOx/l considering oxydized nitrogen in anoxic effluent.

Denitrification efficiencies are above 90 % only for detention times (based on water void volume) : greater than ten minutes. Low denitrification efficiencies increased the oxidized nitrogen concentration in the effluent, while increasing the recycle ratio decreased the effluent concentration.

Recycle ratio. One of the main objectives of this treatment system is to reduce the nitrogen content in wastewater. In practice this means minimizing TKN and NOx concentrations in the effluent.

Generally stated, a Nitrogen mass balance can be expressed as :

$$\text{Nitrogen in - Nitrogen in} = \text{Denitrified} + \text{Assimilated} + \text{Filtered suspended}$$

$$\text{influent} \quad \text{effluent} \quad \text{nitrogen} \quad \text{nitrogen} \quad \text{solids nitrogen}$$

$$\quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \text{content}$$

The first term of this equation can be taken in practice as the Kjeldahl nitrogen concentration in influent (Nitrates and nitrites in influent taken as 0). In the effluent, N is composed of Kjeldahl nitrogen and oxidized nitrogen.

Alternatively, the denitrified nitrogen term may be expressed as a function of the denitrification efficiency and the anoxic influent oxidized nitrogen concentration :

$$\text{denitrified nitrogen (N}_2\text{)} = n_d \text{ NOxed} \quad (16)$$

or,

$$\text{NOxed} = (R \cdot \text{NOxsn}) / (1 + R) \quad (17)$$

If we introduce bacterial cell yield coefficients for heterotrophic denitrification Y_d , aerobic heterotrophic carbon oxidation Y_h , aerobic autotrophic nitrification Y_a and equations (16) and (17), the symbolic representation of the nitrogen mass balance is :

$$\begin{aligned}
 \text{TKNe.Q. } \Delta t & - \text{TKNsn.Q } \Delta t + \text{NOxsn.Q } \Delta t = R d' \text{NOxsn.Q } \Delta t + \\
 \text{Nitrogen in} & - \text{Nitrogen in} = \text{Denitrified} \\
 \text{influent} & \text{effluent} \text{nitrogen} \\
 & \text{denitrification organic carbon oxidation} \\
 + (f.Yd.R. n_d \text{NOxsn.Q } \Delta t) & + (f.Yh.(Se - Sn - b. n_d \text{NOxsn.R).Q. } \Delta t) \\
 + (f.Ya).(NH_4e - NH_4sn - f.Yh.(Se - Sn - b. n_d \text{NOxsn.R).Q } \Delta t & + \\
 & \text{autotrophic nitrification} \\
 & + \text{Assimilated} \\
 & \text{nitrogen} \\
 + n_f .fd.SSed.Q(1+R). \Delta t & + \text{Nitrogen} \quad (18) \\
 & \text{content in} \\
 & \text{filtered suspended solids}
 \end{aligned}$$

where : S : is the soluble organic matter measured as COD or TOC.
 b : is the factor of soluble organic matter uptake for denitrification process.
 n_{fd} : Anoxic reactor filtration efficiency.
 fd : Nitrogen content in filtered solids.
 SSed : Suspended solids concentration at anoxic influent.

The organic nitrogen content is related to the presence of suspended solids in the raw sewage. Because of this solids are retained in the first reactor, thus the influent has small TKN soluble/ NH_4 ratios, to approximate total N - mass balance with a mineral only N - mass balance. Accuracy in computations applying simplified mass balances instead of general mass balances will depend merely on the length of the filtration runs in the anoxic reactor. In the laboratory it has been observed that for filtration runs no greater than 24 h (and even 48 h), SS hydrolysis is negligible and the values of ammonia nitrogen concentration in anoxic influent (NH_{4ed}), ammonia nitrogen concentration in anoxic effluent (NH_{4sd}) and total Kjeldahl nitrogen concentration in anoxic effluent are very close.

So replacing TKN values by NH_4 values, eliminating filtration considerations and neglecting autotrophic assimilation, we can simplify equation (18) to :

$$NH_4e - NH_4sn - NOxsn - R. n_d \text{NOxsn} = f.Yd.R \text{NOxsn} + f.Yh.(Se - Ssn - b. n_d \text{NOxsn.R}) \quad (19)$$

Equation (19) which may be rewritten in the form :

$$R = \frac{NH_4e - NH_4sn - f.Yh.(Se - Ssn) - Noxsn}{n_d \text{NOxsn}.f.(1/f + Yd + b.Yh)} \quad (20)$$

and,

$$Noxsn = \frac{NH_4e - NH_4sn - f.Yh.(Se - Ssn)}{1 + b.R. n_d} \quad (21)$$

Equation 20 is useful in calculating a required recycle ratio when a given treatment degree is desired , while equation 21 can be employed to calculate oxidized nitrogen concentrations in the effluent.

Comparison between experimental results and predicted values (from equation 19) is shown in Figure 8 for $Q = 4$ l/h and Figure 9 for $Q = 6$ l/h. Differences between values obtained with mineral N - mass balance and experimental results are not greater than 10 %. This difference is probably due to the hydrolysis of aerobic biomass.

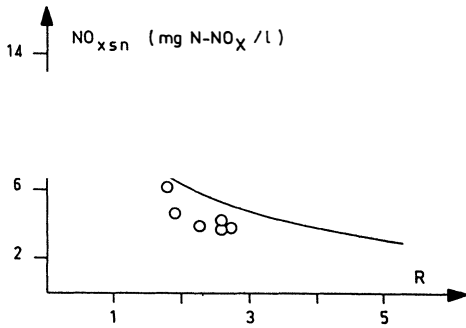


Fig. 8. Comparison of experimental results and predicted values for $Q = 4$ l/h

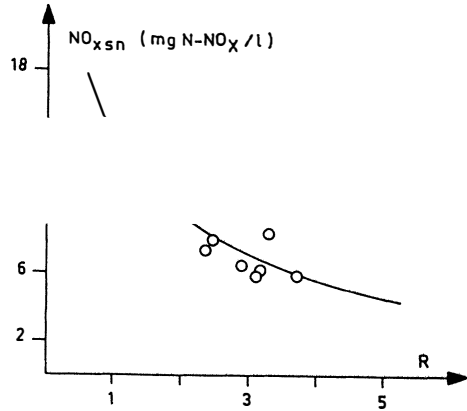


Fig. 9. Comparison of experimental results and predicted values for $Q = 67$ l/h

Sludge production. Sludge mass-balance was made when the aerobic reactor was cleaned. Backwashing frequency was very different in each pilot. It varied from 0.59 to 0.85 d^{-1} in the anoxic reactor and from 0.02 to 0.06 d^{-1} in the aerobic reactor. This means that backwashing in the anoxic filter was made on the average every 1.5 days, while in the aerobic filter this operation was only necessary every 29 days.

Considering filtration phenomena, solids production in the system was 0.22 kg SST/ m^3 of water treated or 0.42 kg SST/kg COD. This value agrees with the values proposed by Rusten (1984) : 0.3-0.55 kg SST/kg COD for a secondary submerged filter.

The biomass production was practically equal to the amount of solids present only in the aerobic filter (real sludge production). The production value calculated for this filter, 0.160 kg SST/kg COD, is comparable to the values given by Roques (1979) for an extended aeration activated sludge system.

Sludge characteristics. The sludge characteristics are shown in Table 4. The anoxic sludge has the same type of characteristics as a typical primary sludge, while aerobic sludges are similar to typical secondary sludge. As these sludges can not be disposed of directly to the environment, sludge treatment must be done.

TABLE 4 Characteristics of sludge and backwash water

Reactor	Water		Sludge	
	SVI	% organic matter	% N	Specific resistance
	ml/g			m/Kg (0.5 bar)
Anoxic	54	82	7.6	2.72×10^{14}
Aerobic	96	85	5.8	3.89×10^{13}

CONCLUSIONS

- Basic parameters for a successful design of a sewage nitrification-denitrification system have been elucidated.

- The efficiency of nitrogen elimination in a filter system was found to be mainly a function of the recycle ratio.

It was demonstrated that the filtration parameters will be limiting factors in anoxic reactor design. In order to combine denitrification and filtration phenomena, the anoxic reactor must be downflow.

- The aerobic reactor must be constructed for both carbon and nitrogen oxidation. The organic load applied in this reactor must be such that it enables autotrophic bacteria development at the end of the filter.

- Ammonia nitrogen elimination in the second reactor should be accomplished in two steps : assimilation and oxidation. The quantity of nitrogen assimilated or oxidized will depend on the organic load. As only oxidized forms of nitrogen are denitrified, effective N-elimination will depend on the percent of nitrogen eliminated by oxidation.

- Mineral N-balance allows an acceptable approximation in calculating recycle ratio.

The equations presented were developed for a plug flow reactor. In field conditions this may not be true and appropriated corrections must be done.

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