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Nitrogen removal in wastewater stabilisation ponds

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ABSTRACT

The objective of this work was to evaluate the nitrogen removal in a wastewater stabilisation pond (WSP) system and to assess the relative contributions of ammonia volatilisation and nitrificationdenitrification to the overall N-removal. The water quality at the inlet and outlet of each treatment stage of the Mèze-France WSP system was monitored fortnightly, from July 2003 to August 2004. The mean nitrogen removal was 65%. Nitrogen removal was higher in summer (89%) than in winter (38%). Maturation ponds provided the bulk of the nitrogen removal. The water quality was monitored from April 1st to July 13th 2005 at the inlet and outlet of the third 2 ha maturation pond. Volatilization has been assessed through literature data complemented by specific experiments. Ammonia volatilisation was low. Nitrification took place during warm periods. In June, oxidised nitrogen content reached 30 mg L⁻¹, then decreased when denitrification occurred. Nitrification–denitrification seems to be the major pathway for nitrogen removal in this maturation pond in hot seasons. A dynamic mathematical model was elaborated to simulate the nitrogen transformations. Organic nitrogen, ammonia and oxidised nitrogen contents in the pond effluent were simulated successfully. The model showed that during the period considered nitrificationdenitrification and volatilization were respectively responsible for 82 and 18% of the overall nitrogen removal.

Keywords: Ammonia volatilisation; Denitrification; Nitrification; Nitrogen removal; Wastewater stabilisation pond

1. Introduction

While pond systems are well known for their ability to achieve high removal of pathogens and organic pollutants, not enough attention has been paid to their potential as regards nutrient reduction. This has become a real concern for many communities which used to dispose of their treated effluents in receiving bodies that,

Many mechanisms are involved in the fate of nitrogen in ponds: uptake of inorganic nitrogen by algae and bacteria, mineralisation, sedimentation, volatilisation of ammonia and nitrification coupled with denitrification [2,3]. For a long time, contradictory assumptions on the main mechanism responsible for nitrogen removal were proposed. Some researchers believed that ammonia vola-

due to a more strict enforcement of the European Directive 91/271 EEC [1], have recently been classified as sensitive.

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tilization largely explains nitrogen removal from WSPs [4-7]. Other authors [8-11], however, claimed that ammonia volatilisation accounts for only a small fraction of N removal. On the basis of the low prevailing nitrate concentrations in ponds systems, several studies concluded that nitrification does not take a significant place and consequently denitrification does not play a major role in nitrogen removal [11–13]. Pano and Middlebrooks [12] as well as Nurdogan and Oswald [14], Shelef et al. [15], El Halouani et al. [16], El Hafiane and El Hamouri [17] and Garcia et al. [18], who investigated high rate algal ponds, assumed that ammonia losses are mainly attributable to biological activity, which includes assimilation into algal biomass, and to volatilization. However recent data [19-21] showed that nitrate concentrations can reach significant values during several weeks in maturation ponds simultaneously with steep variations of ammonia content. Several investigations showed or suggested that nitrification/denitrification can be a major process of nitrogen removal in warm periods in maturation ponds [19,21,22]. Sedimentation and denitrification were the two main fluxes responsible for removing nitrogen in a primary facultative pond [23] and in a 4 pilot-scale ponds in series [24].

The aim of this work was to evaluate nitrogen removal in the Mèze WSPs and, focusing on the maturation ponds, to determine the mechanisms responsible for this nitrogen removal. Volatilization has been assessed through literature data complemented by specific experiments. A mathematical modeling of the fate of nitrogen was undertaken in order to verify whether the dramatic drop in nitrogen content observed in spring season can be explained by nitrification combined with denitrification.

2. Methodology

2.1. Site description

The Mèze WSPs are located on the Mediterranean coast (03°35′06″E, 43°25′10″N). They were constructed in 1980, enlarged and upgraded in the years 1996–1999. Mèze WSP plant has nowadays four treatment stages: 2 anaerobic ponds (AP) in parallel, 4 step fed facultative ponds in series (R_1 , R_2 , R_3 and R_4) with recirculation and aeration (SFP)[®] [25,26], 3 maturation ponds (M_1 , M_2 and M_3) in series and 2 polishing ponds (P_1 and P_2) (Fig. 1). The total area of the ponds is 14.4 ha, and the organic load amounts to 1,120 kg BOD per day, being equivalent to 19,000 p.e.. A more detailed description of the ponds was presented by Picot et al. [21] and Brissaud et al. [27]

2.2. Sampling and analysis

Analysing fortnightly from July 2003 to August 2004 the water quality at the inlet and outlet of (i) the anaerobic ponds in parallel, (ii) the step fed ponds, (iii) the 3



Ponds	Туре	Area (ha)	Depth (m)	
A ₁ , A ₂	Anaerobic ponds	2 x 0.23	3.1	
R_1, R_2, R_3, R_4	Step fed facultative ponds with recirculation (SFP) [®]	4 x 0.67	1.8	
M_1, M_2, M_3	Maturation ponds	4 + 2 + 2	1.4 -1.7	
P ₁ , P ₂	Polishing ponds	1.9 + 1.2	0.8 -1.3	

Fig. 1. Layout of the Mèze waste stabilisation ponds system.

first maturation ponds and (iv) the two last polishing ponds allowed specifying the role of each treatment stage in the overall performance. Composite 24 h samples were taken and analysed for suspended solids (SS), total chemical oxygen demand (COD), filtered COD (Whatman GF/C filters), total Kjeldhal nitrogen (TKN), ammonia (NH₃₋₄-N), nitrate (NO₃-N), nitrite (NO₂-N), total phosphorus (TP) and orthophosphate (PO₄-P) according to Standard Methods [28]. Biological oxygen demand (BOD) were measured with a respirometer Oxytop WTW. *Escherichia coli* (*E. coli*) and faecal enterococci were enumerated using microplate methods. Removal efficiency was expressed in percents of mass flow as follows:

100 [(inlet mass flow - outlet mass flow)/ inlet mass flow]

Additional analyses were made weekly, from April 2005 until July 2005, at the outlet of ponds A_1 , R_4 , M_1 , M_2 , and M_3 in order to identify the mechanisms of nitrogen removal.

2.3. Volatilisation assessment

The production of biogas was measured with 2 floating gas collectors specially developed by Paing et al. [29] and Picot et al. [30] and positioned at the M_3 pond surface. They were made with a circular Plexiglas cap with a bottom area of 0.385 m^2 . The daily biogas production rate was measured and the biogas sampled in a Tedlar bag. NH₃ concentration was measured by bubbling the biogas into a solution of HCl.

Independent ammonia volatilization tests were conducted according to the experimental method employed in Stratton [31,32]. Ammonia losses were measured in outdoor jars filled with 15, 10 and 6 cm of 40 mgL⁻¹ ammonium sulfate solution, the pH varying between 7.5 and 9, the mean daily temperature between 15 and 25°C and the daily mean wind velocity between 2.5 and 6.3 m.s⁻¹ [31].

2.4. Mathematical model

The model for nitrogen transformation and removal was adapted from Mayo and Mutamba [2], Senzia et al. [23] and Fritz et al. [34]. Pond M₃ inlet and outlet flow rates were calculated at a daily time step from the records of the plant inlet flow rate, water depth observations and rainfall and evaporation derived from local meteorological records; water losses due to leakage were considered negligible. Pond M₃ was assumed to behave like a perfectly mixed reactor. The model simulates the concentrations of ammonia [NH_{3.4}-N], organic nitrogen [Org-N] and nitrite and nitrate nitrogen [NO_{2.3}-N] in the effluent, the respective inlet concentrations being observed or interpolated from observed values. Seven processes of nitrogen transformation were considered, including ammonia assimilation (a), ammonia volatilization (v), organic matter sedimentation (s), benthic sludge regeneration (r), organic matter mineralization (m), nitrification (*n*) and denitrification (*d*). $r_{a'}$, $r_{r'}$, $r_{s'}$, $r_{r'}$, r_{m} , r_{n} and r_{d} are the respective nitrogen transformation rates expressed in mg.L⁻¹d⁻¹ (Fig. 2). Ammonia content was high enough to admit that NO₃-N assimilation was negligible. Expressions of the transformation rates are shown in Table 1. They were derived from the works cited in the Reference column.

Mass balance equations for water, organic nitrogen, ammonia nitrogen, nitrate nitrogen, autotrophic nitrifying bacteria and benthic sludge are detailed in Table 2. The modeling process was carried out using a fourthorder Runge–Kutta method and a calculation time step varying from 1 h to 1 d. The values of several coefficients were taken from the literature while the unknown values were determined through the calibration procedure and printed in bold in Table 1.

3. Results and discussion

3.1. Removal performance

Annual mean concentrations in the influent and effluent of each stage of treatment and overall removal efficiencies from July 2003 until June 2004 are shown in Table 3.

The annual mean temperature in the last pond was 17°C (minimum 5°C, maximum 28°C). Fig. 3 shows the role of each stage in the overall pollution removal and the fraction of pollutants remaining in the effluent of the WSP system. The first two treatment stages took the main part in the removal of SS, COD and BOD. The maturation ponds, third treatment stage, had a significant role in the elimination of SS, COD and BOD and a major one in nitrogen removal [21]. The polishing ponds contributed to the removal of nutrients, particularly phosphorus, but slightly increased the effluent SS content through algal growth and evaporation.

3.2. Fate of nitrogen

Annual mean influent nitrogen concentration was 51 mg L⁻¹, 71% of which being ammonia. The overall mean removal efficiency of nitrogen was 65%. Removal



Fig. 2. Nitrogen transformation in waste stabilization ponds.

Table	1

Nitrogen transformation	processes	equations and	parameters	description
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ρ = nitrifier concentration (mg L ⁻¹); Y = 0.15, yield coefficient		

Table 2 Mass balance equations

Substance	Mathematical expression
Ammonia	$\frac{d[NH_{3-4} - N]}{dt} = \frac{Q_i}{V}[NH_{3-4} - N]_i - \frac{Q_e}{V}[NH_{3-4} - N] - r_a + r_m - r_n - r_v + r_r$
Organic nitrogen	$\frac{\mathrm{d}[org-\mathrm{N}]}{\mathrm{d}t} = \frac{Q_i}{V}[org-\mathrm{N}]_i - \frac{Q_e}{V}[org-\mathrm{N}] - r_s - r_m + r_a$
Benthic sludge	$\frac{\mathrm{d}D_m}{\mathrm{d}t} = r_s - r_r$
Nitrate	$\frac{d[NO_{2-3} - N]}{dt} = \frac{Q_i}{V} [NO_{2-3} - N]_i - \frac{Q_e}{V} [NO_{2-3} - N] + r_n - r_d$
Nitrifier	$\frac{\mathrm{d}\rho}{\mathrm{d}t} = -Q_{i,j} \cdot \rho + Y \cdot r_n - b \cdot n^{(T-20)} \cdot \frac{[DO]}{0.5 + [DO]} \cdot \rho$
Water balance	$Q_e = Q_i + Q_r - Q_{ev}$

efficiency was higher in summer (89%) than in winter (38%). 26% of influent nitrogen was discharged into the receiving water body, the Thau coastal lagoon, in the form of ammonia, 6% as organic-N and 3% as nitrite or nitrate. The proportion of organic-N in the effluent was higher in summer and the beginning of autumn than in winter or spring when N-NH₄ effluent concentrations were less than 1 mg L⁻¹. The first two treatment stages, anaerobic ponds and step fed facultative ponds, removed

little nitrogen. Maturation ponds provided the bulk of nitrogen removal efficiency.

Although average effluent $[NO_{2-3}-N]$ was low, with an annual mean of 1.6 mg L⁻¹, nitrate concentration was observed to reach 5 mg L⁻¹ in M₃ effluent for more than 15 d in October 2003 and 8 mg L⁻¹ in June 2004 (Fig. 4). Nitrification followed by denitrification could explain the low $[NH_{3-4}-N]$ observed in summer in pond M₃ effluent. In October, when the temperature was decreasing, deniTable 3

Annual mean concentration (standard deviation n = 23) in each stage of Mèze WSP and removal efficiency from July 2003 until June 2004

	Influent	Anaerobic ponds	Step fed facultative ponds SFP®	Maturation ponds	Effluent	Removal % mass flow
SS (mg L ⁻¹)	256 (85)	158 (99)	81 (47)	54 (39)	70 (62)	79
COD (mg L ⁻¹)	557 (208)	369 (169)	183 (72)	107 (46)	117 (66)	81
f COD (mg L ⁻¹)	237 (94)	161 (70)	86 (35)	51 (17)	55 (20)	77
BOD (mg L ⁻¹)	347 (133)	187 (82)	63 (22)	32 (26)	26 (24)	94
TKN (mg L ⁻¹)	51 (16)	43 (10)	39 (7)	20 (10)	16 (9)	68
NH4-N (mg L ⁻¹)	36 (9)	37 (7)	31 (5)	15 (13)	13 (11)	73
TP (mg L ⁻¹)	7.1 (1.7)	6.2 (1.5)	5.6 (1.2)	5.3 (0.7)	4.3 (1.2)	45
PO4-P (mg L-1)	4.3 (1.4)	3.5 (1.0)	3.0 (1.8)	4.0 (1.5)	3.0 (1.5)	36
<i>E. coli</i> (U.log 100 ml ⁻¹)	6.8 (0.4)	6.4 (0.6)	5.1 (0.8)	2.5 (0.7)	2.2 (0.7)	4.7 U.log



Fig. 3. Contribution of each treatment stage, respectively anaerobic $(A_1 + A_2)$, step fed facultative (SFP[®]), maturation $(M_1 + M_2 + M_3)$ and polishing $(P_1 + P_2)$, to overall removals (expressed as percentage of inlet load).



Fig. 4. Inorganic nitrogen and temperature of $\rm M_{3}$ effluent from July 2003 to August 2004.

trification declined and $[NO_3-N]$ increased. In November, water temperature was less than 13°C and a nitrate decrease came with an ammonia increase. $[NO_3-N]$ was <0.5 mg/L in winter and spring and increased again in June when temperature reached 22°C.

Sampling was carried out again from April to July 2005 to confirm the nitrification observed in 2004 in the third maturation pond M_3 simultaneously with temperature increase (Fig. 5). Indeed, pond M_3 had appeared to play a key role in the removal of nitrogen. The mean loading rate of the pond was 50 kg N ha⁻¹ d⁻¹ and the mean hydraulic retention time 16 d.

 M_3 effluent [NO_{2.3}-N] was virtually equal to zero until the end of May. At the same time, [NH_{3.4}-N] ranged from 35 to 25 mg L⁻¹. The nitrogen mass balance did not show a nitrogen removal higher than 5% of the inlet mass flow rate. The ammonia nitrogen balance suggested that ammonia volatilisation during this period was insignificant although pH value was 8.5. No biogas was collected in the floating gas collector.



Fig. 5. Nitrogen speciation and temperature in $\rm M_{3}$ effluent from April to July 2005.

At the start of June, when the pond temperature reached 21°C, nitrite concentration increased steeply, soon followed by nitrate concentration; NO₂-N and NO₃-N concentrations reached respectively 17 mg and 13.6 mg L⁻¹ on June 9th; at the same time $[NH_{3.4}$ -N] decreased and the mass balance of total nitrogen did not show any loss. From June 1st to 9th, nitrification rate was 76 kg N ha⁻¹ d⁻¹. DO content, with average daily values around 6 mg L⁻¹, was sufficient to allow the development of nitrification within the pond (Fig. 6). Temperature increase seems to have been the key factor of the nitrification development. The water temperature ranging between only 15.2 and 18.8°C under summer conditions in England, Camargo Valero and Mara [11] did not find nitrification in maturation ponds.

After June 10th, nitrite and nitrate concentrations decreased, which was not offset by any ammonia or organic nitrogen augmentation. Simultaneously, dramatic total nitrogen losses were observed. The most probable explanation of these losses was denitrification. DO records showed that high oxygen contents that promoted nitrification alternated everyday with very low values that allowed denitrification (Fig. 6). Biogas was not produced before June 10th; but after that date the collected biogas did not contain any measurable NH_3 even when pH value reached 9 in July; the presence of N_2O in biogas confirmed the existence of a denitrification process. At the end of June nitrification happened also in pond $M_{2'}$ increasing ammonia removal in this pond.

A dynamic mathematical model was elaborated to simulate the nitrogen transformations in maturation pond M₂. Ammonia volatilization was calculated using an expression derived from Stratton [31,32] with a coefficient 30×10⁻³ determined from the outdoor experiments we have carried out (Table 1). This estimate was preferred to the results of floating collector measurements which are suspected to under-evaluate ammonium volatilization [33]. Nitrogen species contents in the pond effluent were simulated successfully (Fig. 7). Modelling the fate of nitrogen resulted in the appraisal of the importance of its transformation routes (Fig. 8). Two different periods should be considered: before and after May 31st, i.e. before and after the nitrification manifestation. During the first period, assimilation increased with the temperature from 6 to 16 kg N d⁻¹ with a mean value of 12.8 kg N d⁻¹; sedimentation varied from 4.8 to 8.8 kg N d⁻¹ with a mean value of 7.5 kg N d⁻¹ and was fairly balanced by regeneration which increased from 6 to 10 kg N d⁻¹ and had a mean value of 7.8 kg N d⁻¹. Ammonia volatilization was 8.8 kg N d⁻¹ on average and the main route of nitrogen losses, while nitrification and denitrification were negligible (0.4 and 0.56 kg N d⁻¹ respectively).

The situation changed rapidly at the beginning of June and during the second period. Assimilation and volatilization fell down to less than 1 kg N d⁻¹ as a consequence of the decrease of the ammonia content, their mean val-



Fig. 6. Dissolved oxygen variations in pond M₃.



Fig. 7. $\rm M_3$ effluent nitrogen species: observed and simulated values.



Fig. 8. Simulated nitrogen transformation in $\rm M_{3}$ maturation pond

ues being respectively equal to 3.1 and 2.5 kg N d⁻¹. Sedimentation was around 4.7 kg N d⁻¹ and nearly balanced by regeneration, the mean value of the latter being 5.8 kg N d⁻¹. Nitrification was the paramount nitrogen transformation; as soon as the conditions could favor its development, nitrifying bacteria rapidly multiplied and could nitrify within a few days the bulk of the ammonia present in pond M₃. Afterwards, nitrification was limited to the oxidation of the ammonia entering the pond. Peak and average values of nitrification were respectively 260 and 76 kg N d⁻¹ during the second period. Denitrification, the mean value of which was 64 kg N d⁻¹, soon followed nitrification and eliminated most of the nitrate.

The model showed that from April 1st to July 13th 2005 nitrification-denitrification and volatilisation were respectively responsible for 82 and 18 % of total nitrogen removal. This confirms the preponderance of nitrification /denitrification to explain nitrogen removal in Mèze maturation ponds.

4. Conclusions

Total nitrogen removal in wastewater stabilisation ponds was season dependent; it was higher in summer than in winter. The major part of nitrogen removal occurred in the maturation ponds. Even if nitrite and nitrate concentrations were very often near zero, oxidised forms of nitrogen could reach a high level during a few weeks when denitrification did not go with nitrification. Monthly sampling of current monitoring procedures can miss the short periods when significant oxidized nitrogen content is appearing. Nitrification took place during warm periods from June to October when water temperature was higher than 20°C. In late spring, nitrification was followed by denitrification with a delay of about ten days. Denitrification stopped in October- November when water temperature decreased quickly.

Under summer conditions in the south of France nitrification-denitrification appears to be the major pathway for nitrogen removal in maturation ponds. Ammonia volatilization can be discounted as a significant mechanism for ammonia removal, even when pH and temperature are favourable to its occurrence.

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