CHAPTER 6

MODEL-BASED ASSESSMENT OF THE BACKWASHING FREQUENCY AND INTENSITY REQUIREMENTS IN THE PACKED BED REACTOR

Foreword

Studies of the steady state achieved after long term operation of the nitrifying packed bed reactor, both experimental (Chapter 3) and using a mathematical model (Chapter 5) provided relevant information on the macroscopical distribution of the two bacterial strains involved in the nitrification process along the packed bed vertical axis. Further uses of the model are explored in the present chapter, where the model is calibrated for its application to the prediction of the reactor behaviour under dynamic conditions and subsequently used to evaluate the effect of a perturbation on the overall reactor operation. The progressive clogging of the packed bed, as previously described, requires of periodical maintenance operations to remove excess biomass from the reactor .The focus of this chapter is the evolution of the different parameters during a backwash event and their effect on the overall nitrifying efficiency. Special emphasis is on elucidating the best operational conditions to minimise the nitrite concentration in the reactor in order to avoid its subsequent accumulation in the MELiSSA loop.

6.1 INTRODUCTION

A one-dimensional multi-species biofilm model was developed (Chapter 4) to allow the prediction of the reactor behaviour under different scenarios and to evaluate the effect of a wide range of parameters on the nitrifying efficiency and on the biofilm composition at different heights of the packed bed.

The model was capable of the accurate prediction of the results yielded by the Q-PCR analysis of the nitrifying biofilm without previous optimisation of any kinetic or stoichiometric parameters. The results of the simulation of the nitrifying efficiency of the reactor at the different steady states achieved throughout the operational period were also highly accurate. The application of the model to the final steady state achieved in the reactor (1750 days) revealed that the main parameters contributing to the segregation of *N. europaea* and *N. winogradskyi* along the packed bed vertical axis were the oxygen mass transfer and the liquid phase flow pattern in the reactor (Montràs et al., 2008).

One of the main issues to be taken into account in the operation of a packed bed reactor is its progressive clogging by biomass accumulation and the implications this has in terms of oxygen mass transfer as well as the alteration in the hydraulic residence time in the reactor due to the increasing biofilm thickness (and consequent decrease of the bulk volume). To keep the biofilm thickness below certain limits in order to prevent clogging, the common procedure in packed bed reactors is to carry out periodical backwash operations i.e. the liquid flow in the reactor is temporarily reversed. A backwash event leads to the detachment and removal of a fraction of the biomass from the biofilm through a combination of detachment mechanisms (Morgenroth and Wilderer, 2000).

The execution of these maintenance events inevitably has an impact on the nitrification efficiency in the reactor (Choi et al., 2007; Sastry et al., 1999; Laurent et al., 2003). Backwashing intervals that range from days (Boller et al., 1997) up to months (Brown et al., 2003) are well reported in the literature. Given the critical role of nitrite accumulation in the MELiSSA loop, the study of the effect of a backwash event on the biofilm reactor performance was of special relevance. However, evaluating the effect of backwashing is not as straightforward as it may seem since several process conditions are affected, including the amount of biomass, the biofilm structure, the liquid volume and other parameters that may be affected as a consequence. The availability of a mathematical model, as developed in Chapter 4, provided the necessary tools to investigate the response of the reactor to backwashing.

6.2 OBJECTIVES

The main goal of this chapter is the exploitation of the mathematical model developed in Chapter 4 under dynamic conditions. The following objectives have been defined:

- Calibration of the mathematical model parameters using experimental data of reactor operation, for the use of the model under dynamic conditions.
- Use of the optimised model to predict the reactor behaviour during the transient state following a backwash event.
- Evaluation of the possibility to design a backwashing strategy that minimises the impact of these maintenance cycles on the reactor performance (e.g. nitrite accumulation).

6.3 CALIBRATION AND UTILISATION OF THE MATHEMATICAL MODEL UNDER DYNAMIC CONDITIONS

The study of the effect of different parameters on the relative abundance of the two nitrifying species present in the reactor at steady state, revealed that the profile along the packed bed was most sensitive to three parameters: (i) the volumetric oxygen transfer coefficient ($K_L \cdot a$), (ii) the back mixing component included in the definition of the flow model (Q_{bmix}) and (iii) the ratio between the growth yields of *N. europaea* and *N. winogradskyi* (sections 5.4.3.4 – 5.4.3.6). The $K_L \cdot a$ coefficient and the back-mixing component of the liquid flow affected most notably the lower parts of the reactor, while the effect of the ratio between the growth yields of the two bacterial species involved in nitrification had a greater effect on the biomass profile in higher fractions.

The accuracy of some of the parameters used for the application of the model to the final steady state, and the possible simplifications that were applied, had to be reconsidered when the model was adapted for use in simulating dynamic conditions. To this effect, all parameters that were known to be affected by time, or related to other parameters that directly depended on time, were defined as state variables.

Therefore, such simplifications as the assumption of a $K_L \cdot a$ coefficient profile that was constant with time, could not be applied to the dynamic study. Instead, a $K_L \cdot a$ coefficient profile that was dependent on time was implemented in the model definition for all simulations presented hereafter, as specified in the general definition of the model (section 4.2.3.1, Chapter 4).

Besides the time dependent evolution of some parameters such as the $K_L \cdot a$, the suitability of the literature values assumed for certain stoichiometric and kinetic parameters had to be reconsidered, and those that were known to have an effect on the nitrifying efficiency were submitted to optimisation. The presence of heterotrophic bacteria was neglected due to the low abundance detected and because the reactor was operating axenically for a long period of time.

6.3.1 INTEGRATION AND PARAMETER ESTIMATION SETTINGS

The shorter operational periods analysed during the study of the system dynamic response allowed the use of a slightly lower discretisation of the biofilm thickness with reasonable simulation times. A grid with 30 to 40 divisions was used for the study of the dynamic response. The lower discretisation (40-division) was applied to the bottom fraction of the packed bed (B_2), which was significantly thicker, while a 30-division grid was used in the remaining fractions (B_3-B_7). A computational time of 50 minutes for 60 days of operation was obtained with these settings on an Intel® Pentium® D CPU at 3.00 GHz.

Model parameters represented by constant variables (p) are estimated by AQUASIM by minimising the sum of the squares (χ^2) of the weighed deviations between the measurements and the calculated model results, according to Equation 6.1.

$$\chi^2(p) = \sum_{i=1}^{n} \left(\frac{y_{meas,i} - y_i(p)}{\sigma_{meas,i}} \right)^2 \qquad \textbf{6.1}$$

In this expression $y_{meas,i}$ represents each one of the experimental measurements used for the parameter optimisation (for i=1,…, n) and is represented by a real state variable; $y_i(p)$ is the model calculated value of the variable estimated at the time and location of the measurement (i), which is estimated based on a number of model parameters ($p=p_1,\ldots, p_m$); Finally, $\sigma_{meas,i}$ is the standard deviation of each one of the i measurements, which was defined in this study as a global value for all the measurements listed under the same real list variable.

AQUASIM performs a minimisation of the sum of the least squares (Equation 6.1) with the constraints ($p_{min,i} \leq p_i \leq p_{max,i}$), where $p_{min,I}$ and $p_{max,i}$ are the minimum and maximum possible values of the parameter p_i whose value is subject to optimisation. The secant algorithm (Ralston and Jennrich, 1978) combined with the active set technique (Gill et al., 1981) was selected to perform the numerical minimisation of the function object (Equation 6.1). A tolerance of 0.001 and a maximum number of iterations equal to 1000 was set. A value of the estimated parameter, together with an estimation of the standard deviation of the residual error term is provided by AQUASIM (standard error of the estimation). The confidence interval for a estimated parameter (p) was calculated, with 95% confidence, using Equation 6.2, where p is the parameter, s_E is the standard error of the estimation as calculated by AQUASIM, and $(1\text{-}\alpha)_{95\%}$ is the 0.95 quartile of Student's distribution.

$$\left[p - s_E \cdot (1-\alpha)_{95\%}\ ,\ p + s_E \cdot (1-\alpha)_{95\%}\right] \qquad \mathbf{6.2}$$

6.3.2 MODEL CALIBRATION

6.3.2.1 Parameter identification

As previously stated, the analysis of long term steady state results by means of the mathematical model did not contemplate any parameter optimisation procedure. Although the effect of a selected number of parameters was evaluated, actual optimisation of kinetic and other operational parameters was not carried out. However, the application of the model to the prediction of the reactor behaviour during the transient states requires a more thorough study that involves identification of the possible parameters affecting the nitrification efficiency and their subsequent optimisation using experimental data for the model calibration.

To evaluate the suitability of the literature values assumed for the different parameters, the model was applied to the simulation of the reactor start up, and the modelled results obtained using the parameters listed in Table 4.2 were compared to the experimental measurements of ammonium, nitrite and nitrate during the first 85 days of reactor operation (i.e. start up period). Certain discrepancies were observed between the modelled and the experimental values of the nitrogen species concentrations in the transient states following a change of ammonium load in the reactor feeding. In order to identify the parameters that most likely have an effect on the dynamic response of the system, a screening was performed in which the sensitivity of the modelled results toward a change of each one of the different kinetic and structural parameters was evaluated.

The nitrite peak achieved during the transient states following a step in ammonium load (either by increasing the liquid flow rate or the ammonium concentration in the feeding solution) was found to be sensitive to several parameters. Among them, the inhibition constant of *N. winogradskyi* due to FA ($K_{FA}^{In,Ntb}$) and the growth yields of both nitrifying strains on their respective substrates ($Y_{X/S}^{Nts}$ and $Y_{X/S}^{Ntb}$) were selected for further detailed study due to the significant sensitivity of the system towards a variation in their values.

6.3.2.2 Parameter optimisation

An underestimation of the *N. winogradskyi* growth yield was already demonstrated in section 5.4.3.4, where the relative abundance of *N. europaea* and *N. winogradskyi* was proved to be affected by the ratio between the growth yields of the two nitrifying strains, the results correlating better with the experimental biomass profiles when the ratio $Y_{X/S}^{Nts}/Y_{X/S}^{Ntb}$ was decreased. While the value assumed as the growth yield of *N. europaea* (Hunik et al., 1993; Table 4.2) was expected to be accurate, the growth yield of *N. winogradskyi* had probably been incorrectly estimated (see section 5.4.3.4). Therefore, a new value needed to be assumed for this system.

Following these observations, a literature search was carried out to obtain the most recent data on true growth yield of *N. winogradskyi*, and a new value of this parameter was assumed taking into account the evidence discussed in section 5.4.3.4 and the latest information available in the literature. Vadivelu et al., (2006) emphasized the relevance of the significant amount of energy utilised by *N. winogradskyi* on maintenance, which makes it essential to make a distinction between the true- and the apparent- growth yields.

In the present study, as previously stated, the maintenance is taken into account as a separate dynamic process in the formulation of the model, and thus the true growth yield should be used in the model definition, which is independent on the growth conditions. Most parameters found in the literature correspond to the apparent rather than the true growth yield, leading to a wide range of different values for this parameter, most probably due to the different amounts of maintenance energy required under the growth conditions in which the parameter value was obtained. Since a definitive value of the true growth yield was not available, a range of growth yield values reported in the literature (Vadivelu et al., 2006; Blackburne et al, 2007a), together with the information obtained in section 5.4.3.4, were used to select a value of the growth yield to be used in this study.

Blackburne et al. (2007b) reported a value of 0.072 g VSS·g^{-1} N-NO_2^- ; Alleman (1984) and Beccari (1979) reported values between 0.02 and 0.084 g VSS·g^{-1} N-NO_2^-. The relative abundance of *N. europaea* and *N. winogradskyi* at steady state presented in section 5.4.3.4 and published in Montràs et al. (2008) suggest that the true growth yield of *N. winogradskyi* should be higher than the initially assumed value of 0.0414 g VSS·g^{-1} N-NO_2^- previously used in the model definition. Values of $Y_{X/S}^{Ntb}$ between 0.05 and 0.06 g VSS·g^{-1} N-NO_2^- were tested in section 5.4.3.4 providing a biomass profile along the packed bed that correlated better with the experimental values of the Q-PCR study. An intermediate value of 0.054 g VSS·g^{-1} N-NO2 was finally assumed to perform the model calibration using experimental start-up data.

The value of the inhibition constant of *N. winogradskyi* by FA assumed in the initial definition of the model was a rough estimate based on other nitrification studies (Carrera et al., 2001). This parameter is highly sensitive to operational conditions and could very likely have been over- or under- estimated, making it unsuitable for the use in the the present system. Due to its importance in the nitrite accumulation in the transient state, and given the uncertainty of the assumed value, $K_{FA}^{In,Ntb}$ was selected for optimisation using the experimental start-up data.

Although the nitrite peaks obtained in the transient states were correctly predicted after the optimisation of the inhibition constant of *N. winogradskyi* by FA, the sole optimisation of the selected parameters could hardly explain the difference between the experimental and the model-predicted ammonium peaks achieved in the different transient states. While a good correlation was found between the experimental and modelled ammonium results once the steady state was achieved, the experimental peak attained during the transient state was in all cases higher than the model prediction, suggesting that the consumption of NH_4^+ by *N. europaea* following an increasing load step was much slower than predicted.

Discrepancies between model predictions and experimental observations during short-term batch experiments were first reported by Vanrolleghem et al., 2004 for a batch experiment with COD as substrate. The higher deviation from the experimental results was detected by the authors at the start of the experiment, i.e. after the substrate pulse addition. This transient phenomenon is known as wake-up or start-up process. A similar phenomenon, named acceleration, was observed by Guisasola et al. (2006) using ammonium as a substrate. In this case, oxygen uptake ratio was very low right immediately after the pulse addition and progressively accelerated until a maximum was reached. Although the set up used in both studies is significantly different from ours, the steps of increasing ammonium load can easily be assimilated to the pulse of substrate concentration applied to a batch experiment such as the one described by the authors of the aforementioned studies.

An exponential correction (Equation 6.3), applied to the kinetic equations of *N. europaea,* provided a good description of the ammonium transient states observed after each change in the ammonium load performed during start-up.

$$a = a_0 \cdot \exp\left[k_a \cdot (t')^2\right] \qquad \textbf{6.3}$$

The function is applied to the *N. europaea* kinetics while a<1. The deviation variable t' (d) defines the relative time elapsed between the last ammonium load step and the actual value of time. A value of 0.25 was set for a_0 (dimensionless). The parameter k_a (d^{-2}) was estimated with the least-squares method, using as experimental values the ammonium concentration data obtained during the reactor start-up (i.e. the first 85 days of operation, see Figure 6.1). Equation 6.4 shows the modified expression of the function describing the comsumption of ammonium by *N. europaea*. Maintenance and decay equations were also modified accordingly.

$$\frac{dC_{NH_4^+}}{dt} = \frac{1}{Y_{X/S}^{Nts}} \cdot \mu_{max}^{Nts} \cdot \frac{C_{NH_4^+}}{K_{NH_4^+}^{Nts} + C_{NH_4^+} + \frac{C_{NH_4^+}^2}{K_{FA}^{In,Nts}}} \cdot \frac{C_{O_2}}{K_{O_2}^{Nts} + C_{O_2}} \cdot a_0 \cdot \exp\left[k_a \cdot (t')^2\right] \qquad \textbf{6.4}$$

The inhibition constant of *N. winogradskyi* by FA ($K_{FA}^{In,Ntb}$) was subsequently estimated using the same numerical method, through the least-squares method using nitrite experimental data obtained during the same period of time as experimental values. The model description obtained after the implementation of the wake-up phenomenon in the definition of the dynamic processes, presented a good correlation of the ammonium, nitrite and nitrate concentrations predicted by the model with the experimental values (Figure 6.1). The estimated values of the adjusted parameters (k_a and $K_{FA}^{In,Ntb}$), together with the absolute error estimated with 95% confidence, are listed in Table 6.1.

Table 6.1 Optimised values of the parameters affecting the ammonium and nitrite accumulation after optimisation. Absolute errors estimated with a 95% confidence level.

Parameter	Units	Value
k_a	d^{-2}	$0.045 \pm 6 \cdot 10^{-3}$
$K_{FA}^{In,Ntb}$	$g\ FA \cdot L^{-1}$	$0.00146 \pm 9 \cdot 10^{-5}$

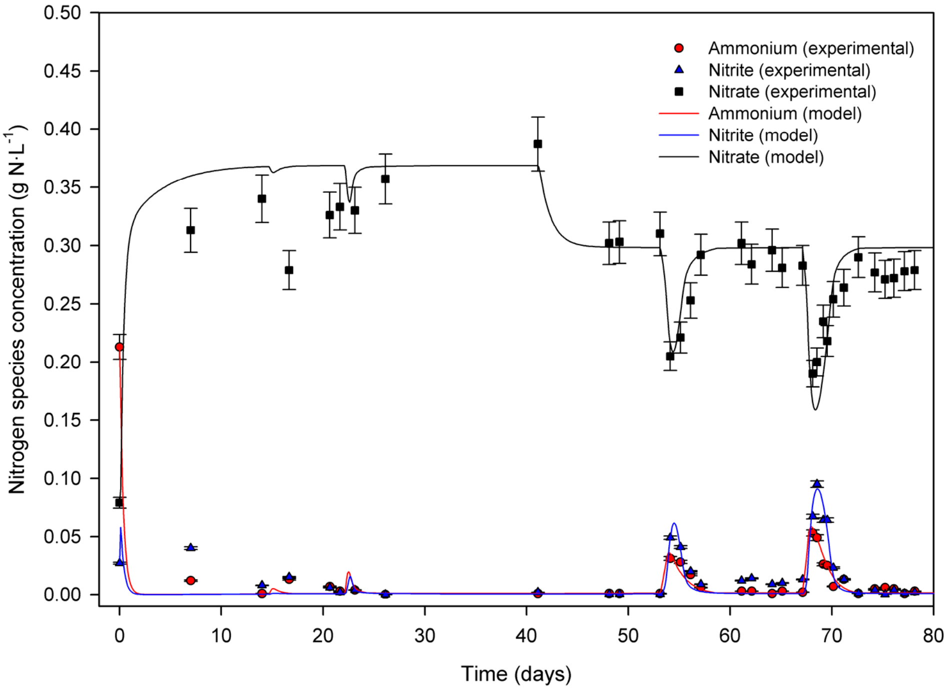


Figure 6.1 Results of the optimisation of the reactor start-up. The *N. europaea* kinetics were modelled taking into account the wake-up phenomenon and subsequently estimating the parameters of the Gauss-type expression used to model this phenomenon. The inhibition constant of *N. winogradskyi* by FA was subsequently optimised.

6.3.3 ANALYSIS OF THE DYNAMIC RESPONSE DURING A BACKWASH EVENT: EFFECT ON THE NITRIFYING EFFICIENCY

The periodic execution of maintenance operations in biofilm reactors is essential, as previously mentioned, in order to avoid the eventual clogging of the packed bed and the consequent failure of the reactor operation. The decrease of the biofilm thickness following a backwash event leads to a higher bulk volume availability, thus increasing the residence time in the reactor. While cyclic backwash events are necessary to guarantee the reactor performance, the negative effect of the increasing volume on the mass transfer cannot be overlooked due to the dependence of the volumetric oxygen mass transfer coefficient ($K_L{\cdot}a$) on the gas superficial velocity. Therefore, the performance of the reactor following a backwash event is a compromise between the positive effect of a lower biofilm thickness and the negative effect of a reduced oxygen mass transfer coefficient.

With the goal to assess the effect of the different parameters on the nitrifying efficiency during backwashing and after the reactor operation is restored, a detachment event is simulated using the mathematical model. The reactor start-up strategy is modelled in the same way as in the previous simulations, which corresponded to the last period of operation of the reactor. Following the start-up, the simulation is run with a constant ammonium loading rate of 0.53 g $N\text{-}NH_4^+ \cdot L^{-1} \cdot d^{-1}$ and the backwash event is implemented when the biofilm thickness profile had achieved a value very close to the steady state in all the fractions of the packed bed (200 days of operation).

Due to the lack of experimental data on biomass concentration, detachment during the events in each fraction of the packed bed is modelled using a correlation based on literature reports and which described detachment rate (u_{det}) as a function of biofilm thickness (LF) and biofilm growth velocity (u_F). The usual detachment expression (Equation 6.5), as described in Chapter 4, section 4.2.8, is multiplied by a constant dimensionless factor k (Equation 6.6) to simulate the detachment increase during backwashing. This approach allows modelling the heterogeneous effect of a backwash event on the different fractions of the packed bed and has been used in previous studies (Wanner and Morgenroth, 2004).

$$u_{det} = (LF/LF_{max})^2 \cdot u_F \quad \text{when } u_F > 0 \text{ (otherwise } u_F = 0) \qquad \textbf{6.5}$$

$$u_{det} = k \cdot (LF/LF_{max})^2 \cdot u_F \quad \text{when } u_F > 0 \text{ (otherwise } u_F = 0) \qquad \textbf{6.6}$$

The duration of the backwash events is set to 0.05 d (1.2 h) for all the performed simulations, and the intensity of the event is modified by changing the value of the factor k when necessary. The feeding of the reactor is stopped for the duration of the event, after which a step in the loading rate is applied.

For the event presented in this section, the factor k is set to 1500 and the maintenance operation is carried out with a frequency of 45 days. The effect of these two parameters (intensity and frequency) on the reactor performance and their optimisation will be discussed in detail in section 6.3.4. This combination of frequency and intensity was selected on a first approach because this intensity (k=1500) produced a reasonable biomass loss, while the frequency of 45 days allowed monitoring the reactor performance up to the point in which the original thickness was recovered, as seen in Figure 6.2.In this figure the effect of the successive backwash events, carried out with a frequency of 45 d, on the biofilm thickness of the different fractions is assessed through simulation. The figure shows how the fractions are differently affected by the reversal of the flow during backwashing. The fractions situated closer to the top of the packed bed (especially B_7) are hardly affected by detachment due to the lower growth rate of the biofilm in these fractions, while the lower fractions are subjected to an important biomass loss.

Following the detachment event induced in the experimental set-up during backwashing, and after the original ammonium load is restored, the biofilm thickness progressively tends to its original state. Focusing on the simulated results of one of the cycles, the effect of backwashing on the overall nitrifying process can be studied.

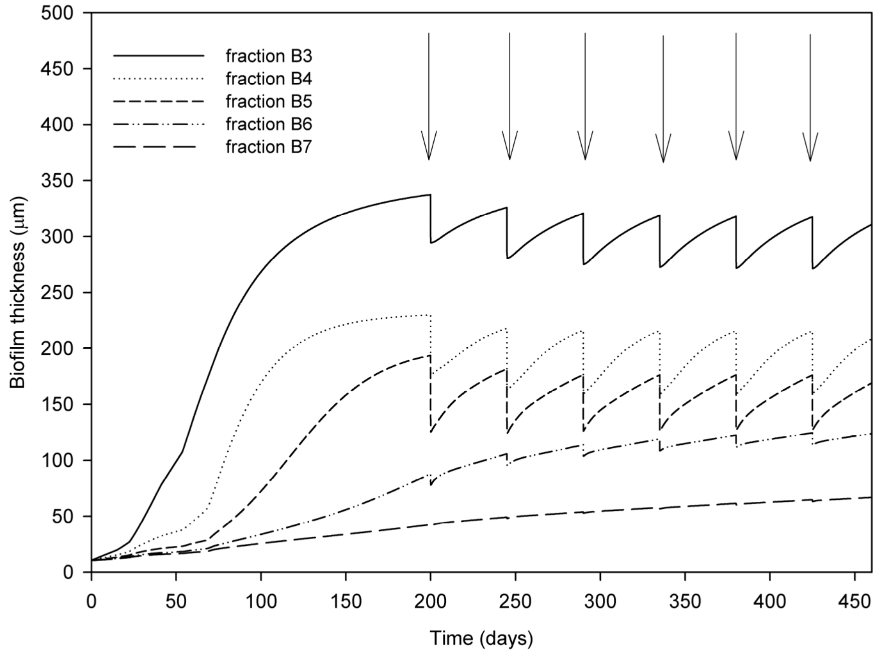
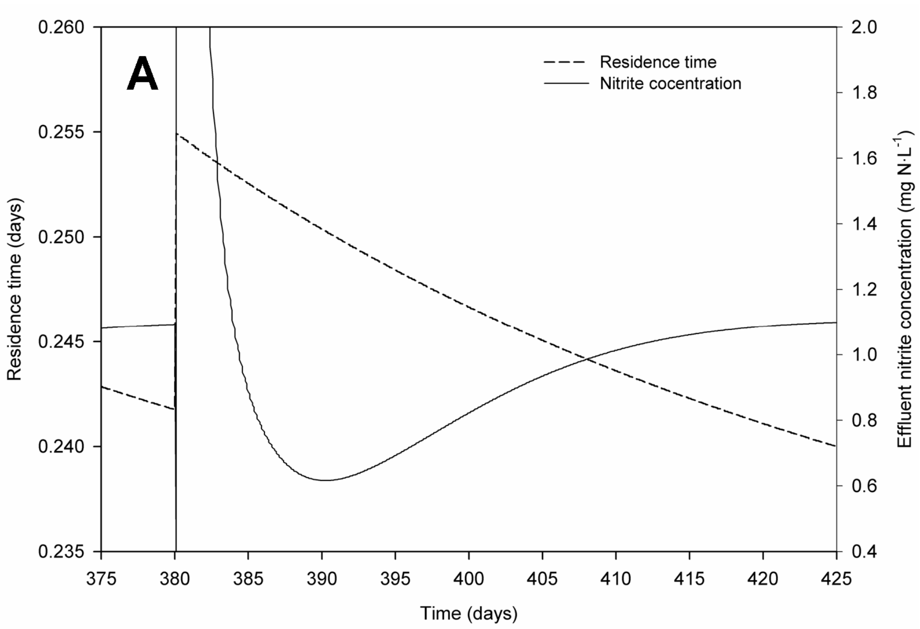


Figure 6.2 Time-course of the biofilm thickness during several backwash events at different heights. Fraction B_2, where the biofilm grows following the direction of the packed bed vertical axis has been omitted due to the different scale (0.1-1.2 cm). The arrows indicate the start of a backwash event.

In Figure 6.3 the decrease of the overall residence time in the reactor due to biofilm growth in between two backwash events can be observed as well as the evolution of the nitrite concentration in the effluent (Figure 6.3A) and the nitrifying efficiency (total conversion to NO_3^-, Figure 6.3B). The nitrite concentration reaches a minimum value of 0.6 mg $N\text{-}NO_2^-\cdot L^{-1}$ 10 days after the maintenance operation was finished and the operation re-started, leading to an improvement in the nitrifying efficiency (Figure 6.3B).

The trend is then reversed and the nitrite concentration starts to increase when the residence time, which had reached a value of 0.255 days after the event, diminishes to a value of 0.250 days. However, the evolution of the nitrite concentration, and thus the nitrifying efficiency in the reactor, can not be evaluated simply in terms of the residence time in the reactor. The effect of oxygen mass transfer and the dissolved oxygen (DO) concentration in the bulk liquid of the biofilm reactor have an important role in nitrification processes, and thus the trend of the nitrifying efficiency is most probably the result of several combined effects.



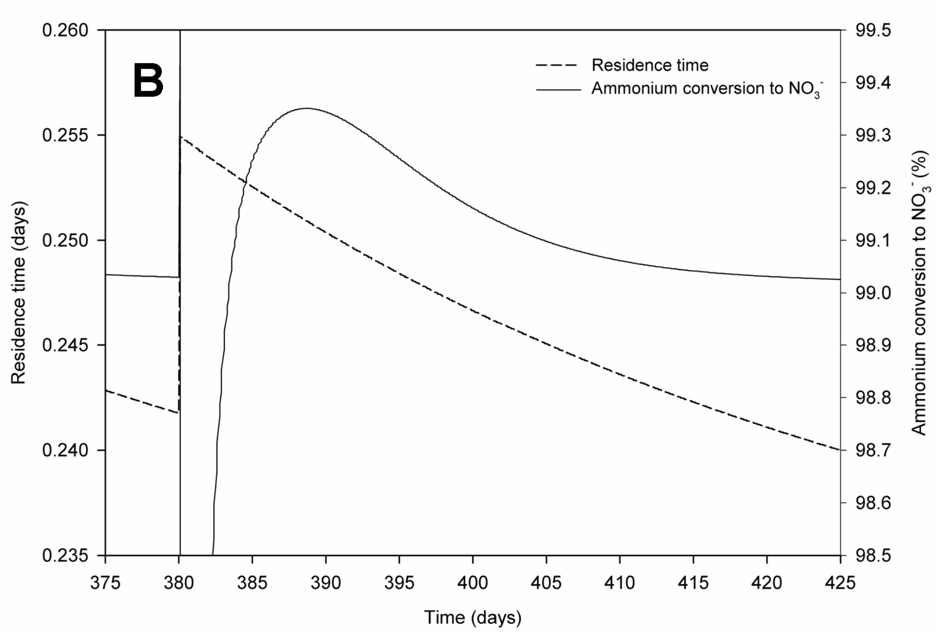


Figure 6.3 Profile of the overall liquid residence time in the reactor following a backwash event performed at 380 days of reactor operation. Effect on the nitrite concentration (A) and on the total nitrifying efficiency to nitrate (B).

The volumetric oxygen mass transfer coefficient ($K_L \cdot a$) was calculated as a function of the gas superficial velocity, and thus is directly dependent on the liquid residence time in each fraction of the packed bed. The oxygen mass transfer is enhanced by the increase in biofilm thickness, the effect being more important in the lower fractions, where the biofilm growth is faster and the bulk volume decreases at a faster rate (Figure 6.4). On the other hand, the biomass growth in the biofilm has an influence on the oxygen uptake and thus the DO profiles need to be evaluated in addition to the mass transfer.

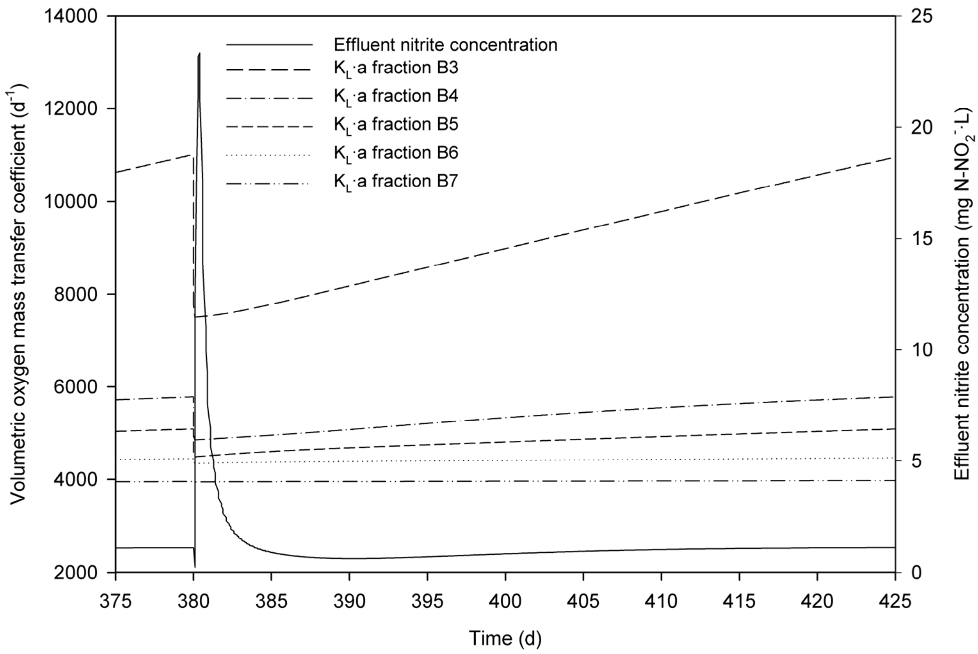


Figure 6.4 Trends in the nitrite concentration in the effluent and the oxygen volumetric mass transfer coefficient at different heights along the packed bed.

The nitrite concentration is represented alongside the DO concentration (Figure 6.5) and the DO/N-NH_4^+ ratio (Figure 6.8) for each one of the packed bed fractions during the time span elapsed between two backwash events (380-425 days).

The DO concentration increases in all fractions for the duration of the backwash event due to the lower consumption during these 0.05 days of batch operation. When the reactor operation is re-started (380.05 days) and following the transient state in which accumulation of NH_4^+ and NO_2^- takes place in the effluent, the DO concentration progressively tends to its steady state levels.

The dissolved oxygen concentration in the packed bed is influenced by (i) the oxygen mass transfer and (ii) the oxygen uptake by the biomass. These two processes are affected by the bulk volume in opposite ways: while the progressive decrease of the bulk volume due to biomass growth has a positive effect on the K_L·a, the same biomass growth requires an increasing amount of oxygen until the steady state is achieved. In the early stages following the backwash event (first 5-10 days) the DO concentration clearly decreases with time in the lower fractions (B_2,

B_3, B_4) due to the significant oxygen uptake, while an increase is observed in fractions B_5, B_6 and B_7 (Figure 6.5). The DO concentration then remains stable in fractions B_6 and B_7 (only a slight decrease is observed in B_6) until the next cycle is started.

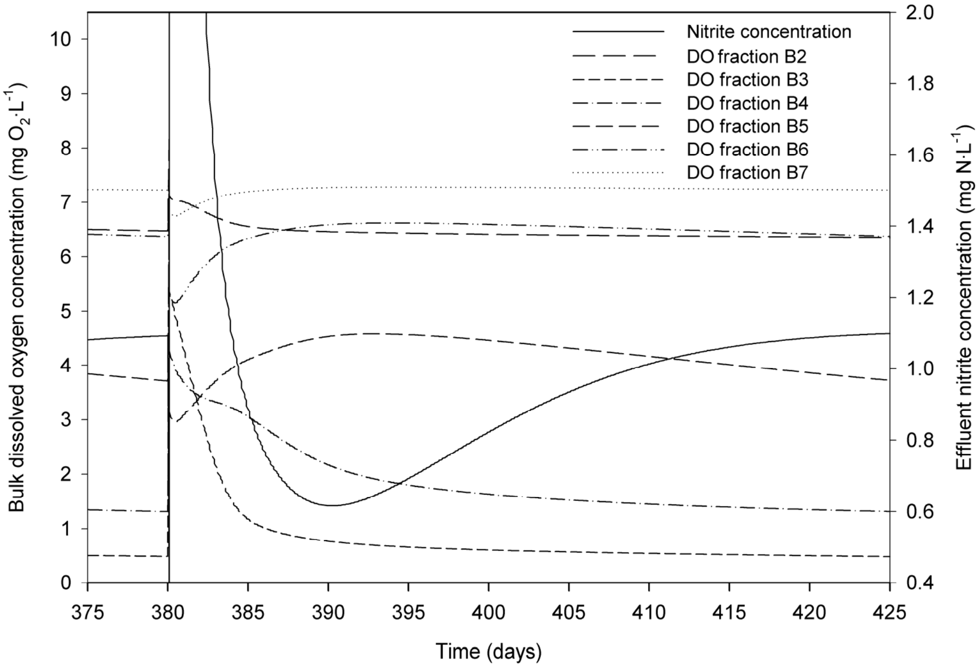


Figure 6.5 Compared trends of the nitrite concentration in the effluent and the dissolved oxygen concentration at different heights of the packed bed. Start of backwash event: 380 days.

This temporary increase of the DO in the higher fractions could be explained by the improvement in the $K_L·a$ due to growth. Since the oxygen uptake in these fractions is low due to the the lower availability of substrates, the effect of the mass transfer improvement can be observed. In fraction B_5 and B_6 a change of trend in the DO concentration can be observed around 395 days. This indicates a turning point in which the improving oxygen mass transfer is not enough to compensate the higher oxygen demand that occurs in this fraction when the nitrite produced in the lower fractions is consumed by *N. winogradskyi*. In Figure 6.6B a significant decrease of the nitrite concentration between 0.8 d and 10 d can be observed which would be responsible for this decrease of the DO concentration observed in fractions B_5 and B_6.

It has been reported (Bernet et al., 2005; Sinha and Annachhatre, 2007) that the ratio between DO concentration and ammonium concentration in the bulk liquid, rather than the actual DO concentration alone, are responsible for the nitrite accumulation in a nitrification process. Bernet et al. (2005) observed that when the loading rate was kept constant, the ratio $N\text{-}NO_2^-/N\text{-}NH_4^+$ depended only on the DO concentration. Similar results were obtained by Picioreanu et al. (1997), when investigating the effect of variations in the superficial loading rate and the liquid-biofilm mass transfer coefficient on nitrite accumulation.

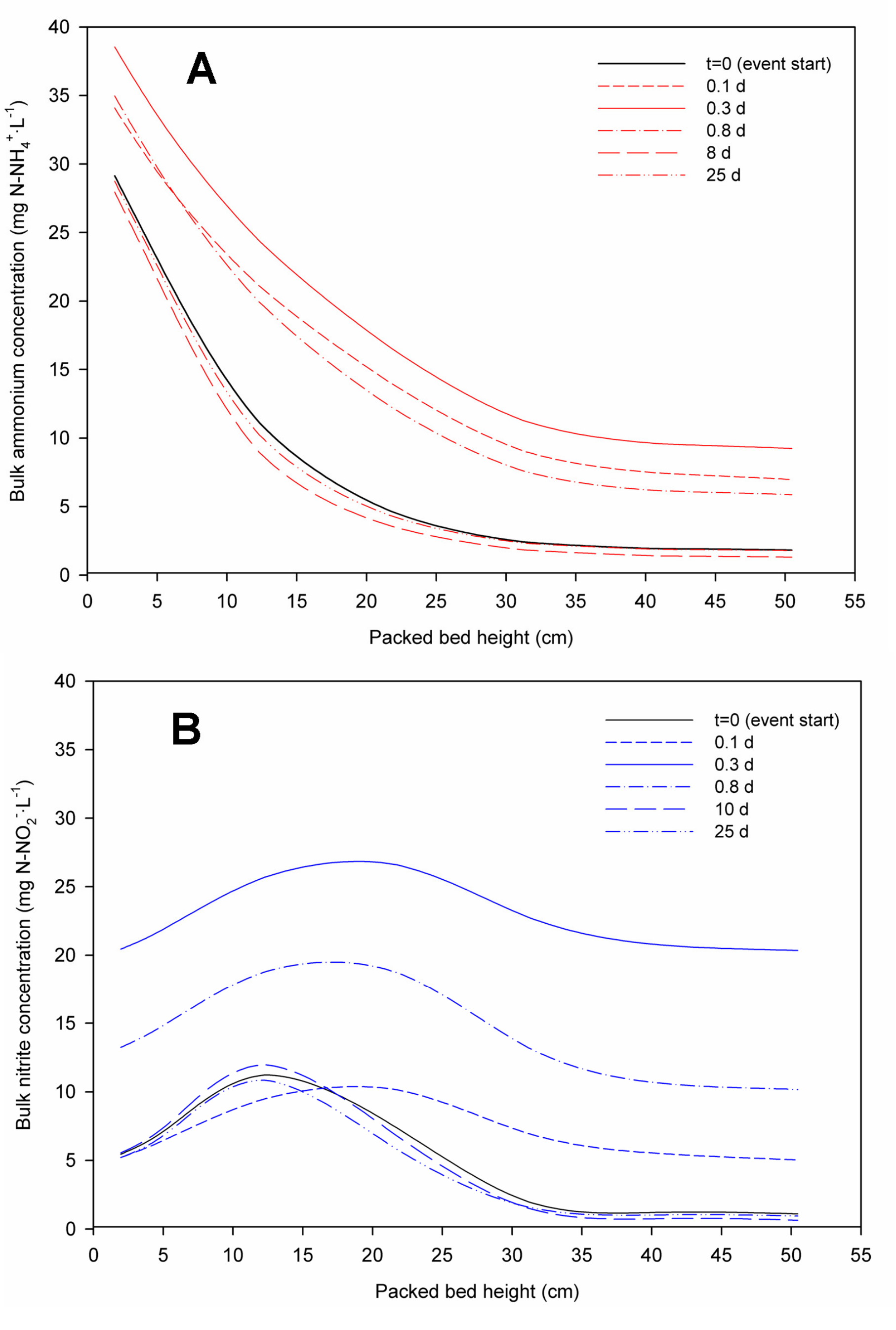


Figure 6.6 Ammonium (A) and nitrite (B) profiles at different times following a backwash event.

Higher loading rates lead to a decrease of the oxygen penetration in the biofilm, the oxygen becomes limiting within the biofilm and mass transfer controls the conversion rate. The lower penetration of oxygen and its consequent limiting effect in the lower fractions of the packed bed can be seen in Figure 6.7. The oxygen and the ammonium concentrations in the biofilm are represented as a function of the biofilm thickness. For a given time (8 days after the event), the oxygen penetration improves with height, due to the decreasing NH_4^+ load (Figure 6.6). The low penetration of oxygen in fraction B_2 (Figure 6.7A) also explains the higher accumulation of nitrite observed in this fraction (15 cm) in Figure 6.6B).

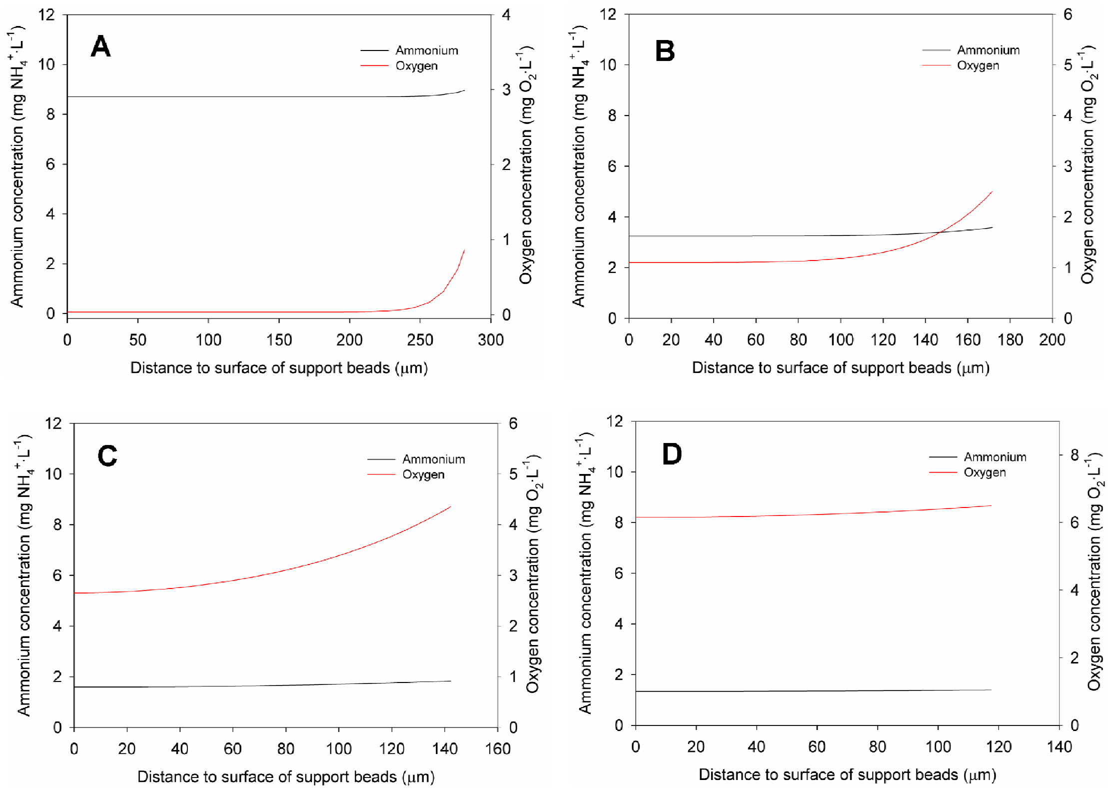


Figure 6.7 Ammonium and oxygen profiles inside the biofilm at time 8 days (following a backwash event) for different reactor heights: (A) fraction B2, (B) fraction B3, (C) fraction B4, (D) fraction B5.

The use of the DO/N-NH_4^+ ratio to evaluate the effect of the DO on the reactor performance makes the conclusions less dependent on the ammonium loading rate or, in this case, on the actual NH_4^+ concentration available in each fraction. In Figure 6.8 a peak in the DO/N-NH_4^+ ratio is observed in all fractions at a time of 388 days (i.e. 8 days after the event), which is followed by a minimum in the nitrite concentration 10 days after the event. At this point, the combined effect of the residence time starting to decrease due to growth (causing an increase on the NH_4^+ concentration) together with a higher demand for oxygen that is compensated by increasing $K_L \cdot a$, give as a result a lower DO/N-NH_4^+ ratio that eventually leads to an increase of the NO_2^- concentration in the effluent, until the original steady state value is achieved or a new maintenance cycle is started.

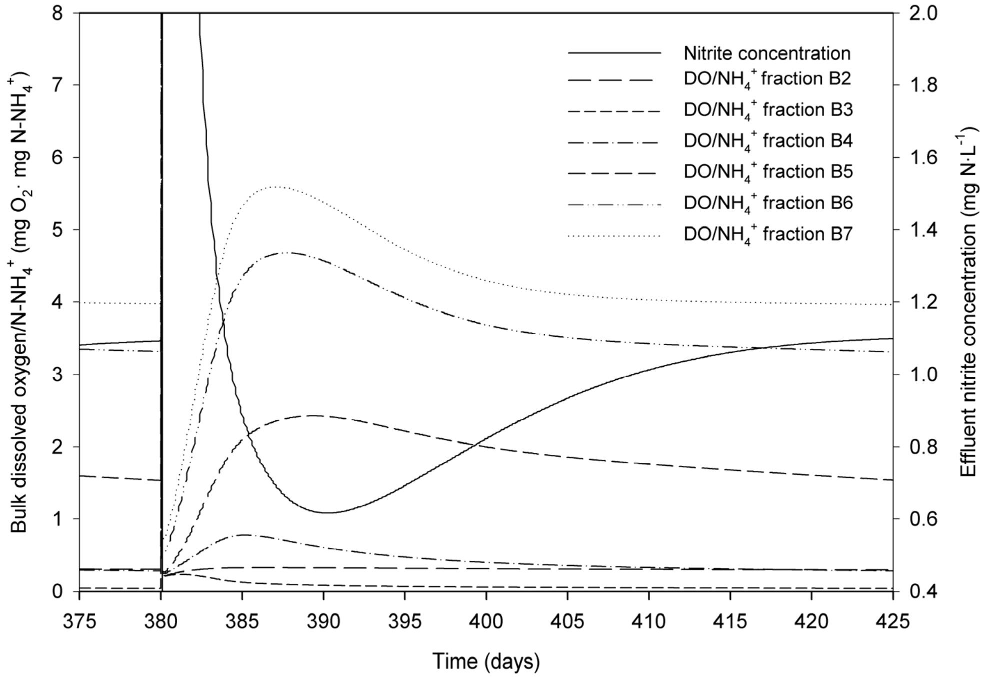


Figure 6.8 Compared trends of the nitrite concentration in the effluent and the ratio between dissolved oxygen concentration and ammonium concentration at different heights. Backwash event start: 380 days.

Figure 6.9 shows the oxygen profiles along the packed bed height obtained at different times following a backwash event. An increase in the ammonium and nitrite concentration in all the biofilm fractions starting at a time of 8 and 10 days respectively, can be observed in Figure 6.6A and Figure 6.6B., in which the trends in the ammonium and nitrite concentrations, respectively, as a function of reactor height are depicted.

While ammonium decreases all along the packed bed axis for a particular time, a peak in the nitrite concentration is observed at a height around 15 cm (fraction B_3). This peak is produced because of oxygen limitation in the lower fractions of the packed bed (Figure 6.9), as assessed in section 5.4.3.2.

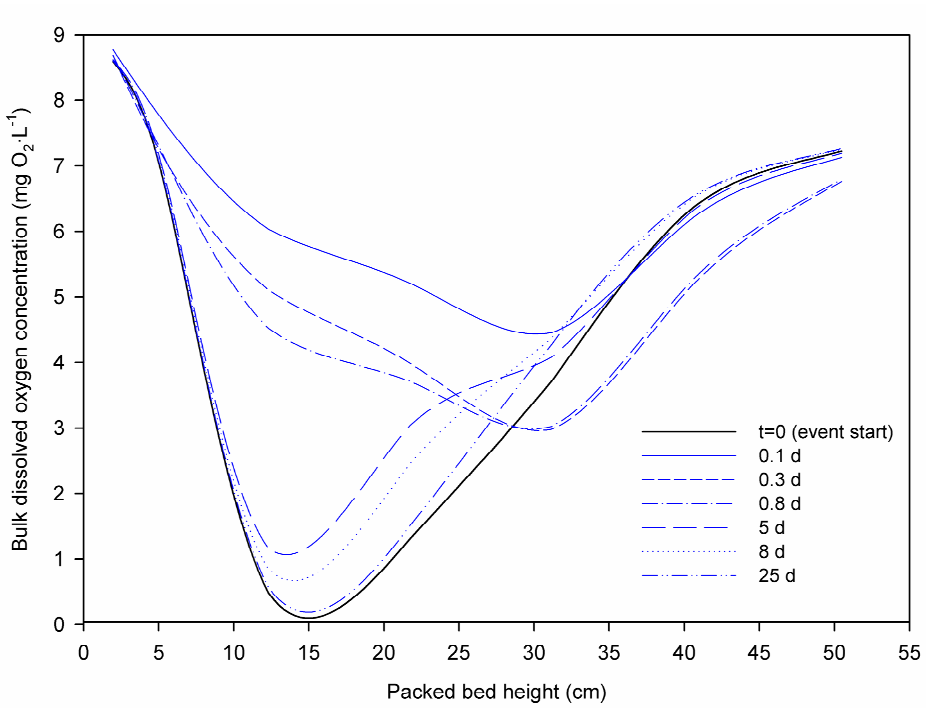


Figure 6.9 Dissolved oxygen concentration profiles obtained at different times following a backwash event, as a function of packed bed height.

Both the nitrite peak and the oxygen minimum observed with height, tend to be more prominent at longer times, which is in agreement with the significant increase of *N. europaea* in fraction B_3 starting 5 days after the event (Figure 6.10A).

The ammonium and nitrite profiles with height reach a maximum value at a time of 0.3 days (Figure 6.6B). At 5 days, the nitrite concentration in the effluent has already been reduced to a value around 1 mg $N\text{-}NO_2^{-}\cdot L^{-1}$, and a DO concentration close to the saturation concentration is reached in the upper fractions of the packed bed. The minimum ammonium concentration (1.3 mg $N\text{-}NH_4^{+}\cdot L^{-1}$) is reached at 8 days and the minimum nitrite concentration (0.6 mg $N\text{-}NO_2^{+}\cdot L^{-1}$) was attained 10 days after the event.

The benefits of the backwash event can be clearly appreciated in the increase of nitrifying efficiency (Figure 6.3). The increase of the residence time due to biomass detachment leads to a decrease of the ammonium and nitrite concentrations, thus improving the efficiency.

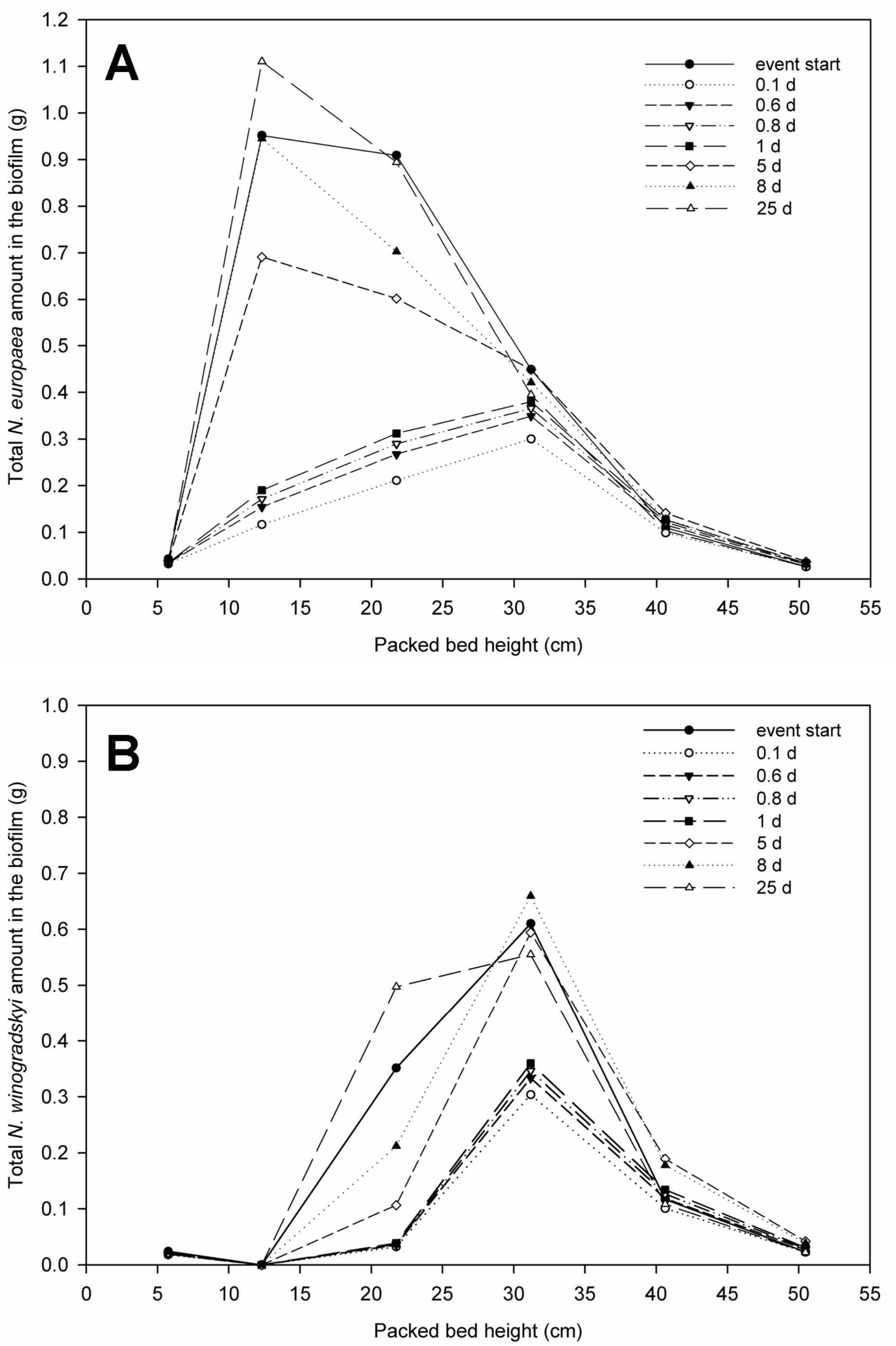


Figure 6.10 *N. europaea* (A) and *N. winogradskyi* (B) abundances along the packed bed height at different times following a backwash event.

6.3.4 POTENTIAL USE OF THE MATHEMATICAL MODEL TO OPTIMISE THE FREQUENCY AND INTENSITY OF THE BACKWASH EVENTS

A comprehensive study of a single backwash cycle (section 6.3.3) made it possible to assess the effect of backwashing on different process parameters along the transient state following the event. It became clear that the detachment event had an important immediate effect on the nitrifying efficiency. However, since the pilot reactor of the MELiSSA CIII has to be suitable for long term periods of operation, it is also important to assess the long term effect of periodical backwash operations on the overall reactor behaviour.

Backwash events should be scheduled taking into account the nitrifying efficiency in the reactor, and balancing the benefits and the disadvantages of both frequency and intensity. The design of an adequate maintenance strategy can take advantage of the availability of a mathematical model, as it allows for the simulation of different scenarios, predicting the behaviour of the system over an expanded period of time.

The lack of experimental data acquired during the backwash cycles does not allow for a comprehensive assessment of the optimal frequency and intensity of the events. The expression used to model detachment (Equation 6.6) has not been calibrated with experimental data. Therefore, in order to establish the real magnitude of the optimal intensity and frequency, it would be necessary to experimentally establish a relationship between event duration, backwash flow rate and amount of detached biomass.

The monitoring of some of these parameters requires further improvements in the reactor hardware and instrumentation, some of which were being developed in parallel with this study. In Chapter 7 the up-grade of the nitrogen species monitoring equipment in the effluent of the reactor will be addressed. Further improvements to the reactor hardware or other instrumentation, such as a biomass sensor that would allow the estimation of the detached biomass in each fraction during an event, are still under development and will be presented in Chapter 8. Therefore, only the principles for the use of the mathematical model in the development of a maintenance strategy will be outlined, and only general, qualitative conclusions will be presented.

6.3.4.1 Effect of successive backwash events on the reactor performance

Choi et al. (2007) studied the effect of backwashing in a packed bed used for percholarate removal. They observed that the history of the reactor had an influence on the efficiency of the backwash events. When weak events were used rather than backwash events with higher intensities, the effect of the successive maintenance operations was not as effective and higher perchlorate concentrations were detected in the effluent.

The evolution of several parameters over an extended period of time spanning several backwash cycles was studied in order to investigate the possible long term decrease of effectiveness described by Choi et al. (2007). In Figure 6.11 the evolution of the total liquid residence time in the reactor with an intensity simulated using a detachment factor of k=1500 and a frequency of 45 days, can be observed. The residence time presents a progressive trend to decrease along time, which could eventually have a negative effect on the nitrifying efficiency in long term operation. The same trend applies to the volumetric oxygen mass transfer coefficient. The trend of the backwash events to become less effective with time can also be observed in Figure 6.13 and Figure 6.14, where the evolution of the nitrifying efficiency and nitrite accumulation is presented.

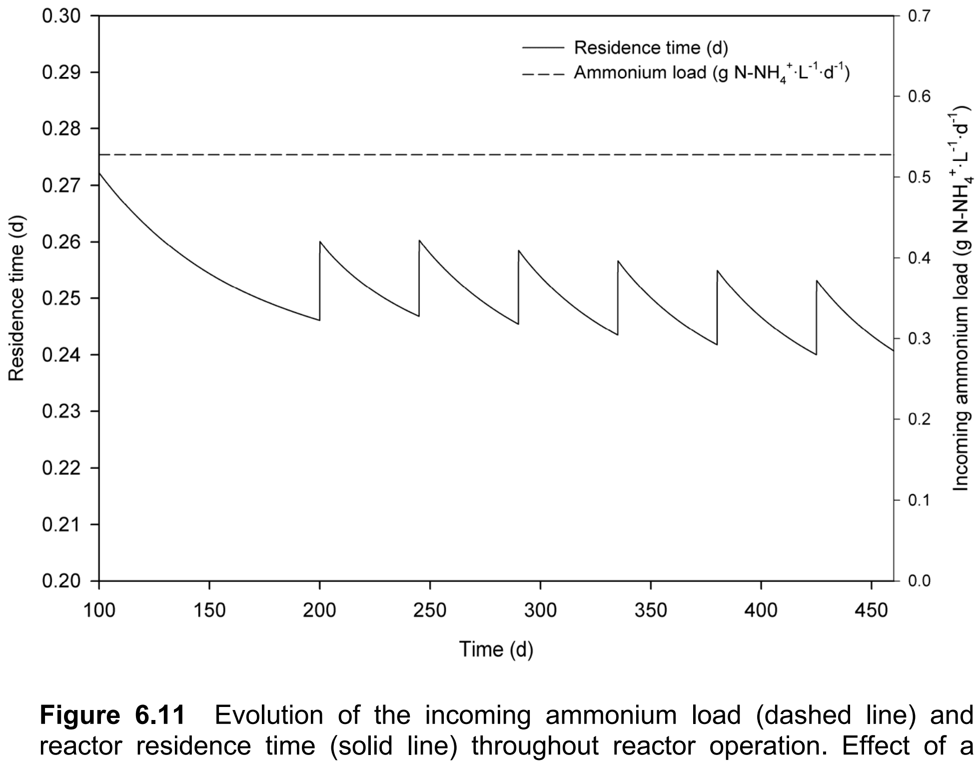


Figure 6.11 Evolution of the incoming ammonium load (dashed line) and reactor residence time (solid line) throughout reactor operation. Effect of a backwashing event with a frequency of 45 days and a simulated intensity using a detachment factor of k=1500.

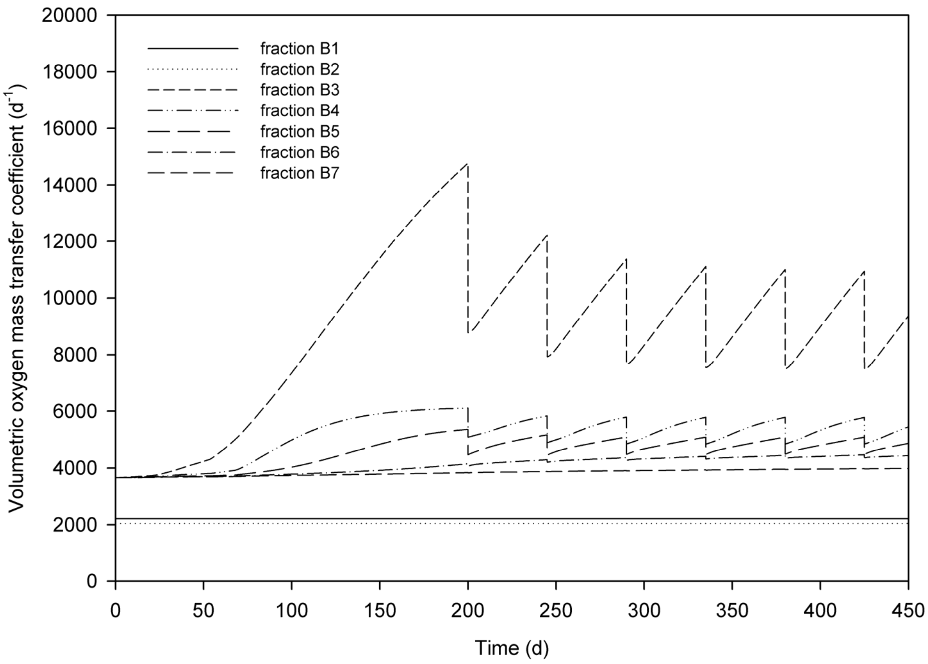


Figure 6.12 Evolution of the volumetric oxygen gas-liquid mass transfer with time in the different fractions of the packed bed. Effect of a backwashing event performed with a frequency of 45 days and a simulated intensity using a detachment factor of k=1500.

In order to evaluate the effect of frequency and intensity on the reactor efficiency, as well as on the loss of effectiveness of the maintenance cycles, the results obtained with the previously used backwashing configuration (k=1500, frequency=45 days) were compared to other frequency and intensity combinations.

6.3.4.2 Effect of backwashing intensity

Selecting three different values of the detachment factor (k) ranging from 1000 to 2000, the effect of the intensity of the backwashing events carried out in the packed bed was evaluated. In Figure 6.13 an event with a constant frequency of 45 days was used as a base to test the effect of different intensities on the effluent nitrite concentration and consequently on the nitrifying efficiency.

By increasing the backwash intensity from 1000 (A, B) to 1500 (C, D) and finally 2000 (E, F) two relevant effects can be observed: (i) the maximum efficiency, and the minimum steady state nitrite concentration, following a backwash event, were achieved with the strongest intensity (k=2000). Higher average efficiencies along the reactor operation were thus achieved with higher intensities. (ii) The effectiveness of the backwash events decreased slightly over time when weaker events were performed (Figure 6.13A, Figure 6.13B) as indicated by the higher slope of the successive peak values.

However, nitrite accumulation during the short transient state following the backwash event, when the reactor ammonium load is restored after maintenance, should not be overlooked. While the highest intensity (k=2000) appeared as most suitable to keep a high nitrifying efficiency in the reactor, the nitrite peak following the event had a value of 47.7 mg $N\text{-}NO_2^{-}\cdot L^{-1}$ with k=2000, 22.7 mg $N\text{-}NO_2^{-}\cdot L^{-1}$ with k=1500 and only 8.3 mg $N\text{-}NO_2^{-}\cdot L^{-1}$ with k=1000.

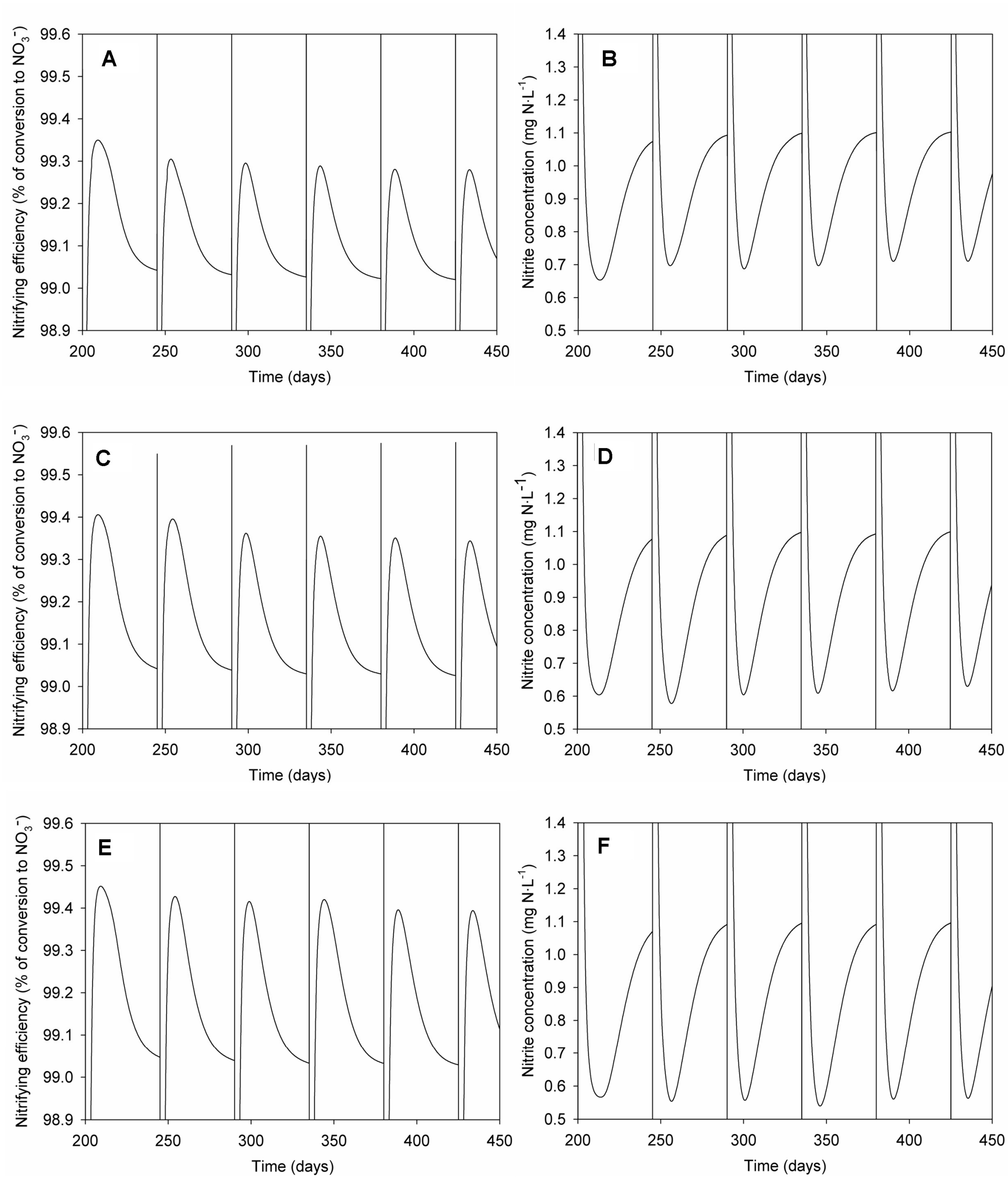


Figure 6.13 Effect of a backwashing event performed with the same frequency (45 days) and different intensities of detachment (A, B: k=1000; C, D: k=1500; E, F: k=2000). (A, C, E) Effect of the detachment intensity on the nitrifying efficiency in the reactor; (B, D, F) Effect of the detachment intensity on the nitrite concentration in the reactor effluent.

A stronger intensity leads to a higher overall nitrifying efficiency in the reactor and at the same time this efficiency is less affected by the history of maintenance events in the reactor. Taking into account the different factors, the highest intensity would be the most convenient option as long as (i) a good control strategy was implemented in the reactor to manage the nitrite accumulation during the transient state and (ii) the action of the control system does not increase the duration of the maintenance cycles significantly. Due to toxicity of nitrite, the use of a moderate intensity is regarded as the best solution in the studied reactor.

6.3.4.3 Effect of backwashing frequency

The effect of the frequency with which the events were performed was evaluated by performing several simulations with different backwashing frequencies (45, 60 and 90 days) and keeping a constant intensity (k=1500). The resulting effect on nitrifying efficiency and on the nitrite presence in the effluent can be seen in Figure 6.14.

The results obtained with three tested backwashing frequencies were very similar in terms of the maximum nitrifying efficiency (minimum nitrite concentration) achieved after a backwash event. However, analysing a wider time period it was observed that more frequent events (i.e. the period of time -90, 60, 45 days- decreased) eventually had a slight negative effect on the efficiency, leading to a progressive loss of effectiveness over time when the backwash events were performed every 45 days (Figure 6.14A and B).

When the nitrite peak reached in the transient state immediately following the event was analysed, the peak was higher for lower frequencies (i.e. 60 and 90 days) due to the higher biofilm thickness achieved and the stronger impact of the same detachment intensity on the biofilm structure. The nitrite peaks in the transient state ranged from 26 mg $N\text{-}NO_2^-\cdot L^{-1}$ (with a frequency of 45 days) up to 49 mg $N\text{-}NO_2^-\cdot L^{-1}$ (with a frequency of 90 days). Since the effect of frequency on the nitrifying efficiency attained was very weak (a maximum nitrifying efficiency of 94% was achieved for all frequencies), it is advisable to select a higher frequency (i.e. a shorter time period between cycles), which reduces nitrite accumulation, as long as it does not have a negative effect on long term effectiveness of the events. As this negative effect was not observed at all for frequencies of either 60 or 90 days, a frequency of 60 days would be chosen in this case due to the lower nitrite accumulation.

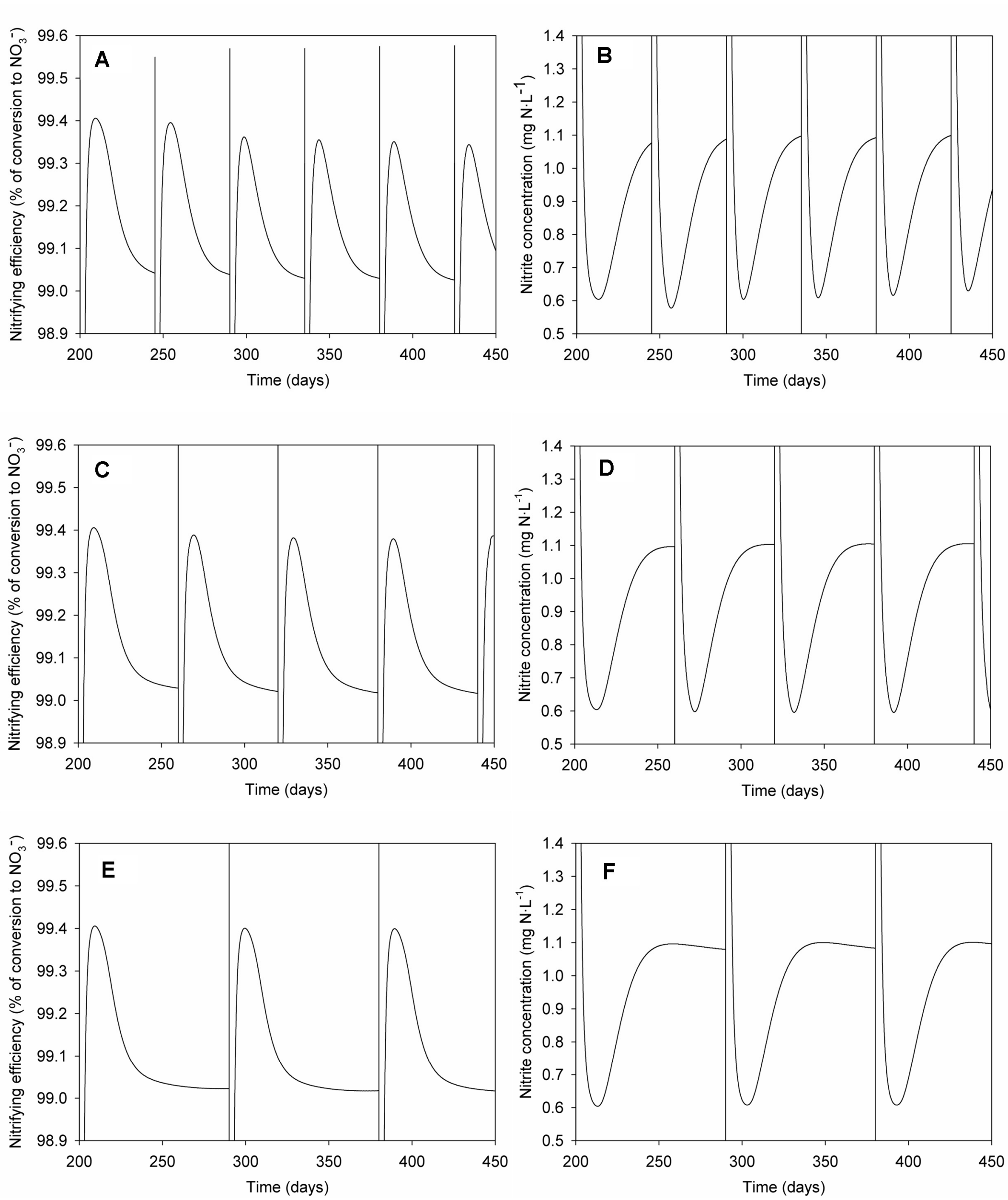


Figure 6.14 Effect of a backwashing event performed with a common intensity of detachment and different frequencies (A, B: 45 days; C, D: 60 days; E, F: 90 days). The intensity was simulated using a detachment factor of k=1500. (A, C, E) Effect of the frequency on the nitrifying efficiency in the reactor; (B, D, F) Effect of the frequency on the nitrite concentration in the reactor effluent.

Taking into account the effects of all parameters a suitable strategy can be outlined for biofilm growth control in the packed bed reactor. Several improvements were foreseen in the reactor hardware and instrumentation that, once implemented, should allow the optimisation of the backwashing strategy through the combination of the experimental information with the mathematical model. Some of these improvements include continuous monitoring of nitrogen species in the effluent of the reactor, as well as a nitrite control strategy (Chapter 7), and the design of a biomass sensor that will allow the monitoring of the biofilm thickness. The availability of a biomass sensor would allow establishing an experimental correlation between backwashing flow rate and detached biomass, providing a real measure of the intensity of the backwashing events that could be implemented in the model. The sensor is already under testing in the MELiSSA pilot plant, and its implementation in the reactor is already foreseen in the re-design of the reactor hardware (Chapter 8).

6.4 CONCLUSIONS

Further exploitation of the one-dimensional biofilm model developed in Chapter 4 was carried out in this chapter by evaluating the performance of the model under dynamic conditions. The model was applied to the study of the nitrifying efficiency and the parameters affecting it, with the focus on the reactor performance during the periodical backwash operations carried out in the packed bed to avoid clogging.

- The model was successfully calibrated for its use under dynamic conditions through optimisation of the following parameters with experimental data: (i) the parameters a_0 and k_a of the Gaussian equation used to model the observed wake-up effect of the subsequent steps of ammonium load and (ii) the inhibition constant of *N. winogradskyi* by FA ($K_{FA}^{In,Ntb}$).

- It was confirmed that persistent backwashing operations in the reactor eventually led to a loss in effectiveness of these maintenance events. By regulating frequency and intensity this effect can be avoided, stronger backwash events being more convenient than weaker, more frequent, ones.