Modeling and optimization in membrane bioreactor and moving bed biofilm reactor-membrane bioreactor

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Abstract

A membrane bioreactor and two hybrid moving bed biofilm reactor-membrane bioreactor systems were studied regarding the process modeling and operation optimization through different mathematical models for organic matter and nitrogen removal. A multiple linear regression method and a multivariable statistical analysis were applied. The process variables were hydraulic retention time, total biomass concentration, temperature, chemical oxygen demand of the influent and total nitrogen of the influent. In general, the values of coefficient of determination (R²) for the different model fittings were higher for the hybrid MBBR-MBR processes than those obtained for the MBR, probably due to the presence of suspended and attached biomass in the hybrid MBBR-MBR systems.

Keywords: Membrane bioreactor, Microbial kinetics, Modeling, Moving bed biofilm reactor, Optimization.

1. Introduction

Membrane bioreactor (MBR) and hybrid moving bed biofilm reactor-membrane bioreactor (hybrid MBBR-MBR) constitute some of the most advanced technologies concerning wastewater treatment used today [1-5], becoming an excellent alternative to the conventional activated sludge process [6-8]. These systems allow to comply with effluent limits and water quality guideline [3,9], showing important advantages [10-13].

Despite their growing implementation, to the best of our knowledge there has been limited research on MBR and hybrid MBBR-MBR systems concerning process modeling and operation optimization. In this regard, modeling and optimization are required to explore the fundamental processes that govern reactor behavior [3,14,15]. Thus, these tools could improve the basic activated sludge models through a more accurate characterization of the biological processes, and a more precise control and optimization of the operational parameters of the wastewater treatment plants (WWTPs) [16-18].

In light of this, modeling and optimization of biological wastewater treatment systems such as MBR and hybrid MBBR-MBR have to be focused on the removal processes of organic matter and nitrogen [14]. They constitute invaluable tools in terms of time and effort required to assess different
alternatives for providing knowledge on underlying mechanisms affecting process performance and reducing energy operational costs [3,19].

Multiple linear regression method is a mathematical and statistical technique that can be used to model and analyze wastewater treatment processes regarding organic matter and nitrogen removal depending on several independent variables [20,21]. In this research, the independent operation variables were hydraulic retention time (HRT), total biomass concentration ($X_t$), temperature (T), chemical oxygen demand of the influent (COD$_\text{influent}$) and total nitrogen of the influent (TN$_\text{influent}$). Furthermore, three models were proposed for the kinetic parameters, i.e. maximum specific growth rate ($\mu_m$), substrate half-saturation coefficient ($K_S$) and yield coefficient ($Y$), to assess the heterotrophic and autotrophic kinetic behavior based on a mass balance for substrate applied to a bioreactor. This experimental design is based on fitting the model by the least squares technique [22].

The increase of the efficiency of the processes of organic matter and nitrogen removal without increasing the cost is a fundamental method used to get a higher operation performance, which is called optimization. Optimization consists on choosing the values for independent variables to obtain the best system response from sets of available alternatives [23].

In light of this, the design procedure includes (i) the design of a series of experiments for measurement of the response of interest, (ii) the development of a mathematical-statistical model for the organic matter and nitrogen removal through the application of a multiple linear regression method and a multivariable statistical analysis, (iii) the obtention of the optimum experimental process variables that produce a maximum value of response, (iv) the development of mathematical models for the heterotrophic and autotrophic kinetic parameters, (v) the assessment of the optimum kinetic parameters by considering the optimum experimental process variables and the models proposed, and (vi) the evaluation of the substrate degradation rate for organic matter and nitrogen removal by applying the optimum kinetic parameters.

Thus, this methodology gives a systematic preview to achieve the optimum operation conditions for desirable responses regarding organic matter and nitrogen removal efficiency and kinetic performance as well as reducing the number of experimental runs in an effective manner [14].

In this study, chemical oxygen demand (COD) and total nitrogen (TN) removal as well as the heterotrophic and autotrophic kinetic parameters were assessed as a function of different operation variables such as HRT, $X_t$, T, COD$_\text{influent}$ and TN$_\text{influent}$ in MBR and hybrid MBBR-MBR. The aim of this work was to develop mathematical models in terms of organic matter and nitrogen removal from municipal wastewater and kinetic performance to optimize the operation conditions.

2. Experimental

2.1. Description of the wastewater treatment plants

The pilot-scale WWTPs, which were designed for organic matter and nitrogen removal, consisted of an MBR (Fig. S1a, Supporting Information), a hybrid MBBR-MBR, which had carriers in the anoxic and aerobic chambers of the bioreactor (Fig. S1b, Supporting Information), and a hybrid MBBR-MBR, which contained carriers only in the aerobic zone of the bioreactor (Fig. S1c, Supporting Information). The description of these WWTPs is given in Text S1 in the Supporting Information.

2.2. Operation conditions

The variables analyzed in this study were the type of wastewater treatment plant (WWTP), HRT, $X_t$ in the bioreactor, as mixed liquor suspended solids (MLSS) for suspended biomass and/or biofilm density (BD) for attached biomass, T, COD$_\text{influent}$ and TN$_\text{influent}$. TN$_\text{influent}$ was the sum of organic nitrogen, ammonium-nitrogen, nitrite-nitrogen and nitrate-nitrogen. These operational conditions...
are shown in Tab. S1 in the Supporting Information. Operational conditions number 8 and 10 are not published, although the analysis was carried out globally. The value ranges of the operational conditions are specified in Text S2 in the Supporting Information.

The COD and TN removals and the kinetic parameters for heterotrophic and autotrophic biomass under the different operational conditions are indicated in Tab. S2 in the Supporting Information. In this regard, the following kinetic parameters were evaluated: maximum specific growth rate for heterotrophic biomass \( \mu_{m,H} \), half-saturation coefficient for organic matter \( (K_m) \), yield coefficient for heterotrophic biomass \( (Y_H) \), maximum specific growth rate for autotrophic biomass \( \mu_{m,A} \), half-saturation coefficient for ammonia nitrogen \( (K_{NH}) \), yield coefficient for autotrophic biomass \( (Y_A) \).

2.3. Statistical analysis

A multivariable statistical analysis applying the software Canoco for Windows v. 4.5 (ScientiaPro, Budapest, Hungary) was used to quantify the influence of the environmental variables (HRT, \( X_T \), \( T \), CODinfluent and TNinfluent) on the species data (COD and TN removal and the kinetic parameters for heterotrophic and autotrophic biomass), and to obtain the variables with the highest influence on the behavior of the different systems studied [24]. For this, the length and the angles between the different vectors symbolizing the variables and species were considered. This analysis is described in Text S3 in the Supporting Information.

2.4. Mathematical modeling and optimization

In order to fit the functions that describe the removal of organic matter and total nitrogen, \( Y_{OM} \) and \( Y_{TN} \), respectively, depending on the HRT, \( X_T \), \( T \), and CODinfluent (for the organic matter removal) or TNinfluent (for the total nitrogen removal), a multiple linear regression method was used [21].

In light of this, the multiple linear regression method analyzes four independent variables (HRT, \( X_T \), \( T \), and CODinfluent or TNinfluent) and another dependent one (\( Y_{OM} \) for organic matter removal or \( Y_{TN} \) for total nitrogen removal). For the formulation of this mathematical model regardless of the organic matter or TN removal, CODinfluent and TNinfluent are represented as \( S_o \), and the response variable was named as \( Y_{OM/TN} \).

\( Y_{OM/TN} \) can be expressed by a linear function of the variables \( HRT, X_T, T \) and \( S_o \) through the coefficients \( \beta_{0,H/A}, \beta_{1,H/A}, \beta_{2,H/A}, \beta_{3,H/A}, \beta_{4,H/A} \) according to Eq. (1):

\[
Y_{OM/TN} = \beta_{0,H/A} + \beta_{1,H/A} HRT + \beta_{2,H/A} X_T + \beta_{3,H/A} T + \beta_{4,H/A} S_o
\]  (1)

For that, a normal probability model is considered, as appears in Eq. (2):

\[
Y_{OM/TN} = \beta_{0,H/A} + \beta_{1,H/A} HRT + \beta_{2,H/A} X_T + \beta_{3,H/A} T + \beta_{4,H/A} S_o + U
\]  (2)

The parameters \( \beta_{0,H/A}, \beta_{1,H/A}, \beta_{2,H/A}, \beta_{3,H/A}, \beta_{4,H/A} \) set the values of the independent variables \( HRT, X_T, T \) and \( S_o \), and allow to obtain the corresponding values of \( Y_{OM/TN,i} \) according to Eq. (3):

\[
Y_{OM/TN,i} = \beta_{0,H/A} + \beta_{1,H/A} HRT_i + \beta_{2,H/A} X_{T,i} + \beta_{3,H/A} T_i + \beta_{4,H/A} S_{o,i} + u_i \quad (i = 1, 2, ..., n)
\]  (3)

\( Y_{OM/TN,i} \) \( \sim \) \( N(\beta_{0,H/A} + \beta_{1,H/A} HRT + \beta_{2,H/A} X_T + \beta_{3,H/A} T + \beta_{4,H/A} S_o, \sigma^2) \) independent, \( (i = 1, 2, ..., n) \)

The matrix of random error \( U \) is determined as observed in Eq. (4):

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Eqs. (5) and (6) represent the mathematical model as a matrix form:

$$Y_{OM/TN} = X \beta_{H/A} + U \quad (5)$$

where $X$ is the design matrix.

The estimations of the parameters corresponding to matrix $\beta_{H/A}$ are assessed by Eq. (7):

$$\hat{\beta}_{H/A} = (X'X)^{-1} X' Y_{OM/TN} \quad (7)$$

where $X'$ is the transpose of matrix $X$.

The terms $X'X$ and $X'Y_{OM/TN}$ corresponding to Eq. (7) are developed in Eqs. (8) and (9):

$$X'X = \begin{pmatrix}
\sum_{i=1}^{n} HRT_i^2 & \sum_{i=1}^{n} HRT_i X_{T,i} & \sum_{i=1}^{n} HRT_i T_i & \sum_{i=1}^{n} HRT_i S_{o,i} \\
\sum_{i=1}^{n} X_{T,i} HRT_i & \sum_{i=1}^{n} X_{T,i}^2 & \sum_{i=1}^{n} X_{T,i} T_i & \sum_{i=1}^{n} X_{T,i} S_{o,i} \\
\sum_{i=1}^{n} T_i HRT_i & \sum_{i=1}^{n} T_i X_{T,i} & \sum_{i=1}^{n} T_i^2 & \sum_{i=1}^{n} T_i S_{o,i} \\
\sum_{i=1}^{n} S_{o,i} HRT_i & \sum_{i=1}^{n} S_{o,i} X_{T,i} & \sum_{i=1}^{n} S_{o,i} T_i & \sum_{i=1}^{n} S_{o,i}^2
\end{pmatrix} \quad (8)$$

$$X'Y_{OM/TN} = \begin{pmatrix}
u_1 \\
u_2 \\
\vdots \\
u_n
\end{pmatrix} \quad (9)$$
Each of the coefficients $\beta_{i,H/A}$ represents the effect of the independent variable on the dependent variable. The estimated value $\hat{\beta}_{i,H/A}$ indicates the variation that the dependent variable experiments when the independent variable varies in one unit and the rest of them keep constant.

Having obtained the functions that relate the removal of organic matter and total nitrogen to the HRT, $X_T$, $T$ and $S_o$ for the MBR, hybrid MBBR-MBRa and hybrid MBBR-MBRb, the coefficient of determination ($R^2$) was evaluated in order to obtain a measurement regarding the goodness of fit, defined as indicated in Eq. (10):

$$R^2 = \frac{\sum_{i=1}^{n}(Y_{OM/TN,i} - \bar{Y}_{OM/TN})^2}{\sum_{i=1}^{n}(Y_{OM/TN,i} - \bar{Y}_{OM/TN})^2}$$

Therefore, for each of the three systems, two functions were obtained to describe the performance of these systems concerning the removal of organic matter and total nitrogen removal as a function of the HRT, $X_T$, $T$ and $S_o$. These functions were optimized by analyzing the common operation ranges of HRT (9.5 – 30.4 h), $X_T$ (2,798.05 – 3,768.44 mg L$^{-1}$), $T$ (14.9 – 23.3 ºC), COD$_{influent}$ (257.47 – 437.73 mg O$_2$ L$^{-1}$) and TN$_{influent}$ (95.48 – 147.76 mg N L$^{-1}$) for the MBR, hybrid MBBR-MBRa and hybrid MBBR-MBRb (Tab. S1, Supporting Information).

The optimization was carried out by the Solver add-in facility of Microsoft Office Excel. This software allowed to determine the values of HRT, $X_T$, $T$ and $S_o$ that maximized the efficiencies regarding organic matter and total nitrogen removal for the MBR, hybrid MBBR-MBRa and hybrid MBBR-MBRb.

Regarding the assessment of the yields of organic matter and total nitrogen removal, the heterotrophic and autotrophic kinetic performances were analyzed by considering the data provided for the MBR, hybrid MBBR-MBRa and hybrid MBBR-MBRb in Tab. S1 and Tab. S2 in the Supporting Information. For this, functions that related the kinetic parameters $\mu_m$, $K_s$ and $Y$ to the operational variables HRT, $X_T$, $T$ and $S_o$ were proposed by analyzing the mass balances applied to the biological reactor of the MBR, hybrid MBBR-MBRa and hybrid MBBR-MBRb, according to Text S4 in the Supporting Information. It should be highlighted that the operational variable HRT was related to the sludge retention time (SRT) through Eq. (9) corresponding to Text S4 in the Supporting Information. Thus, the different models could also be expressed as a function of SRT.

In light of this, the following functions, Eqs. (11) – (13), are formulated to establish the relation of the dependent variables, $\mu_m$, $K_s$ y $Y$, to the independent variables, HRT, $X_T$, $T$ and $S_o$:

$$\mu_m = \frac{\lambda_{1,H/A}}{\text{HRT}} + \frac{\lambda_{2,H/A}}{X_T} + \frac{\lambda_{3,H/A}}{1 - e^{-\lambda_{4,H/A}T}} + \lambda_{5,H/A}S_o$$

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The empirical values of \( \mu_m, K_s, y \) for the heterotrophic (\( \mu_m^{H}, K_m^{H}, Y_H^{H} \)) and autotrophic (\( \mu_m^{A}, K_m^{NH}, Y_A^{NH} \)) biomass can be observed in Tab. S2 in the Supporting Information. The theoretical values of these kinetic parameters were evaluated by considering Eqs. (11) – (13) and the best-fit parameter values (\( \lambda_1^{H/A}, \lambda_2^{H/A}, \lambda_3^{H/A}, \lambda_4^{H/A}, \lambda_5^{H/A} \) for Eq. (11), \( \phi_1^{H/A}, \phi_2^{H/A}, \phi_3^{H/A}, \phi_4^{H/A}, \phi_5^{H/A} \) for Eq. (12) and \( \alpha_1^{H/A}, \alpha_2^{H/A}, \alpha_3^{H/A}, \alpha_4^{H/A}, \alpha_5^{H/A} \) for Eq. (13)) were determined by using the Solver add-in facility of Microsoft Office Excel. In this regard, an objective function was defined as the weighted sum of squares of differences between the empirical and theoretical values and this function was minimized to yield the most appropriate parameters for the considered functions. The values of \( R^2 \) were calculated according to Eq. (10).

The optimum values of \( \mu_m^{H}, \mu_m^{A}, K_m^{H}, K_m^{NH}, Y_H^{H}, Y_A^{NH} \) were assessed by considering the optimum operational conditions of HRT, \( X_T \), \( T \) and \( S_o \) for the MBR, hybrid MBBR-MBR\(_A\) and hybrid MBBR-MBR\(_B\). Finally, the values of substrate degradation rate for organic matter removal (\( r_{su,H} \)) and substrate degradation rate for total nitrogen removal (\( r_{su,A} \)) were determined to corroborate that were in the operational range for each of the systems analyzed.

3. Results and discussion

3.1. Modeling and optimization of the process of organic matter removal

It should be noted that the efficiency of organic matter removal usually decreased when the HRT was lower for the MBR, hybrid MBBR-MBR\(_A\) and hybrid MBBR-MBR\(_B\) (Tab. S2, Supporting Information). This occurred as the organic loading rate was higher. Moreover, the increase of biomass concentration did not usually improve the COD removal (Tab. S2, Supporting Information). It could be due to a higher localized competition between the suspended and attached biomass, which could cause an inhibitory effect on the COD removal [25].

The results of the multivariable statistical analysis for organic matter removal are shown in Fig. 1. This analysis shows three triplot diagrams for the MBR (Fig. 1a), hybrid MBBR-MBR\(_A\) (Fig. 1b) and hybrid MBBR-MBR\(_B\) (Fig. 1c).

Figure 1

In general, the HRT presented a positive correlation with the COD removal and the kinetic parameters for heterotrophic biomass. It supported the fact that the efficiency of COD removal increased when the HRT was higher.

Regarding the biomass concentration, it should be noted that the MLSS and BD have almost no influence on the COD removal and the kinetic parameters for heterotrophic biomass as the angles between these vectors are of approximately 90°. This explained that the highest biomass concentrations did not usually improve the COD removal. However, the BD showed a positive correlation with the COD removal and heterotrophic kinetic parameters in the hybrid MBBR-MBR\(_B\) and the length of the BD vector was higher than in the case of the MLSS vector. Therefore, the attached biomass had a higher influence on the COD removal and kinetic parameters for heterotrophic biomass in the hybrid MBBR-MBR\(_B\); it could support the highest performance of COD removal under the HRTs of 9.5 h and 6 h in this system, as previously indicated.
The T did not practically affect the different systems studied regarding the organic matter removal (the angles were of approximately 90°) as its effect was decreased due to the fact that the WWTPs were not outside, but inside a laboratory.

Moreover, the COD removal and the kinetic parameters for heterotrophic biomass had a positive correlation in the three systems, so the kinetic study for heterotrophic biomass supported the results of organic matter removal.

The mathematical models, which express YOM as a linear function of the variables HRT, Xf, T and COD_{influent} are defined in Tab. 1 through the coefficients β_{0,H}, β_{1,H}, β_{2,H}, β_{3,H} and β_{4,H} for the MBR, hybrid MBBR-MBR_a and hybrid MBBR-MBR_b.

Table 1

In this regard, the coefficients β_{1,H} are positive and it corroborates the fact that COD removal and HRT are directly proportional. The same occurs with the coefficients β_{4,H} that show the direct proportion between COD_{influent} and COD removal. The slight influence of MLSS and BD on the COD removal (Fig. 1) is supported by the coefficients β_{2,H} which are close to zero. The effect of T was softened, as indicated previously, and it is confirmed by the low values of the coefficients β_{3,H} with a value of 0.0041 for the hybrid MBBR-MBR_b (Tab. 1).

The values of R^2 for the MBR, hybrid MBBR-MBR_a and hybrid MBBR-MBR_b, 0.9359, 0.9996 and 0.8397, respectively (Tab. 1), indicate that the mathematical models proposed show a high goodness of fit for the process of organic matter removal.

In general, the optimum operational conditions regarding HRT, Xf, T and COD_{influent} for the three systems correspond to the highest value of the common operation interval (30.4 h, 3,768.44 mg L^{-1}, 23.3°C and 437.73 mg O_2 L^{-1}), with the exception of the hybrid MBBR-MBR_b that showed an optimum value for X_f of 2,798.05 mg L^{-1} (Tab. 1). This was probably due to the higher effect of BD on the COD removal and heterotrophic kinetic parameters for this system as compared with the MBR and hybrid MBBR-MBR_a.

Regarding the mathematical models to fit the heterotrophic kinetics depending on HRT, X_f, T and COD_{influent}, it should be noted that the values of R^2 corresponding to the hybrid MBBR-MBR_a and hybrid MBBR-MBR_b are higher than those obtained for the MBR (Tab. 1). This could be probably due to the coexistence of suspended and attached biomass in the hybrid MBBR-MBR systems, which could lead to a modification in the goodness of fit of the kinetic parameters.

In relation to the MBR, μ_{m,H} and Y_H showed a positive correlation with HRT and COD_{influent}, and K_M had a negative correlation with HRT and COD_{influent}, which is supported by the fitting parameters λ_{3,H} and λ_{5,H} (positive values), φ_{1,H} and φ_{5,H} (negative values), and α_{3,H} and α_{5,H} (positive values). The influence of X_f on the kinetic parameters is softened with values of -0.0007 and 0.0001 for K_M and Y_H, and showing a strongly negative correlation for the μ_{m,H} with a value of -53.0178. In general, this was in accordance with the results shown in Fig. 1. As a whole, the effect of T on heterotrophic kinetic parameters is attenuated, as corroborated through the parameters λ_{3,H}, λ_{4,H}, φ_{3,H}, φ_{4,H}, α_{3,H} and α_{4,H}. This explains the values of angles (of approximately 90°) between T and kinetic parameters observed in Fig. 1.

Concerning the hybrid MBBR-MBR_a, the mathematical models show high values of R^2 for the fitting of the kinetic parameters μ_{m,H}, K_M and Y_H as a function of HRT, X_f, T and COD_{influent} as compared with MBR, with values of 0.9771, 0.9920 and 0.9999, respectively. It should be pointed out that the slight effect of X_f and T on the kinetic parameters, observed in Fig. 1, is supported by the values of λ_{2,H}, λ_{3,H}, λ_{4,H}, φ_{2,H}, φ_{3,H}, φ_{4,H}, α_{2,H}, α_{3,H} and α_{4,H} (Tab. 1). Moreover, the kinetic parameters had usually a positive correlation with COD_{influent} as when COD_{influent} is higher, the gradient of substrate is higher and the μ_{m,H} and Y_H also increase.
The hybrid MBBR-MBR\textsubscript{b} has also high values of $R^2$ (Tab. 1) and the mathematical functions describing its kinetic behavior were similar to those obtained for hybrid MBBR-MBR\textsubscript{a}. It should be noted that $\mu_{\text{m,H}}$ had a strongly positive correlation with $X_T$ (Tab. 1) due to the effect of BD, as explained previously for Fig. 1.

The optimum values of $\mu_{\text{m,H}}$, $K_M$ and $Y_H$ were 0.0172 h\textsuperscript{-1}, 5.2388 mg O\textsubscript{2} L\textsuperscript{-1} and 0.3493 mg VSS mg COD\textsuperscript{-1}, respectively, for the MBR, 0.0032 h\textsuperscript{-1}, 0.5228 mg O\textsubscript{2} L\textsuperscript{-1} and 0.3444 mg VSS mg COD\textsuperscript{-1}, respectively, for the hybrid MBBR-MBR\textsubscript{a}, and 0.0102 h\textsuperscript{-1}, 5.6219 mg O\textsubscript{2} L\textsuperscript{-1} and 0.2855 mg VSS mg COD\textsuperscript{-1}, respectively, for the hybrid MBBR-MBR\textsubscript{b}.

The values of $r_{su,H}$ were determined for each of the operational conditions analyzed in the MBR, hybrid MBBR-MBR\textsubscript{a} and hybrid MBBR-MBR\textsubscript{b}. Furthermore, the value of optimum $r_{su,H}$ was assessed by considering the optimum operational conditions and the optimum response of the different systems, shown in Tab. 1. In the MBR, $r_{su,H}$ was within the range of 20.7146 mg O\textsubscript{2} L\textsuperscript{-1} h\textsuperscript{-1} and 143.6665 mg O\textsubscript{2} L\textsuperscript{-1} h\textsuperscript{-1}, and the optimum value was 143.0274 mg O\textsubscript{2} L\textsuperscript{-1} h\textsuperscript{-1} (Tab. S3, Supporting Information). The value of $r_{su,H}$ varied between 26.4865 mg O\textsubscript{2} L\textsuperscript{-1} h\textsuperscript{-1} and 114.1320 mg O\textsubscript{2} L\textsuperscript{-1} h\textsuperscript{-1} in the hybrid MBBR-MBR\textsubscript{a}, and the optimum value was 27.3662 mg O\textsubscript{2} L\textsuperscript{-1} h\textsuperscript{-1} (Tab. S3, Supporting Information). The hybrid MBBR-MBR\textsubscript{b} showed $r_{su,H}$ values that fluctuated between 8.4726 mg O\textsubscript{2} L\textsuperscript{-1} h\textsuperscript{-1} and 377.9639 mg O\textsubscript{2} L\textsuperscript{-1} h\textsuperscript{-1}, and the optimum value was 75.1161 mg O\textsubscript{2} L\textsuperscript{-1} h\textsuperscript{-1} (Tab. S3, Supporting Information). The optimum $r_{su,H}$ were in the operation ranges of the different systems, which supported the modeling and optimization processes. In light of this, the MBR had the highest rate of organic matter degradation under optimum operational conditions as compared with the hybrid MBBR-MBR systems.

### 3.2. Modeling and optimization of the process of total nitrogen removal

The efficiency of TN removal generally decreased when the HRT was lower for each WWTP as the ammonium loading rate was higher. Moreover, the higher the biomass concentration was, the higher the efficiency of TN removal was (Tab. S2, Supporting Information).

The results of the multivariable statistical analysis for nitrogen removal are shown in Fig. 2. This analysis shows three triplot diagrams for the MBR (Fig. 2a), hybrid MBBR-MBR\textsubscript{a} (Fig. 2b) and hybrid MBBR-MBR\textsubscript{b} (Fig. 2c).

Figure 2

The HRT had usually a positive correlation with the TN removal and the autotrophic kinetic parameters. It was in accordance with the fact that the efficiency of TN removal increased when the HRT was higher.

Regarding the biomass concentration, MLSS did not have any correlation (angle of 90° approximately) with the TN removal and the autotrophic kinetic parameters for the MBR (Fig. 2a). In the case of the hybrid MBBR-MBR\textsubscript{a}, BD presented a strongly negative correlation with the TN removal and the autotrophic kinetic parameters, although MLSS showed a positive correlation (Fig. 2b). MLSS and BD showed a positive correlation with the TN removal and the autotrophic kinetic parameters for the hybrid MBBR-MBR\textsubscript{b} (Fig. 2c), which implies that the attached biomass had an influence on the nitrogen removal and the autotrophic kinetics higher than that obtained for the hybrid MBBR-MBR\textsubscript{a}. This supported the higher TN removal efficiency under the HRTs analyzed for the hybrid MBBR-MBR\textsubscript{b}.

The T did not affect the different systems studied regarding the TN removal as its influence was attenuated due to the fact that the WWTPs were not out in the open, but they were inside a laboratory. It also occurred in the process of COD removal, as explained previously.

In general, the TN removal and the kinetic parameters for autotrophic biomass had a positive correlation in the three biological systems, so the autotrophic kinetics supported the results of TN removal.
The mathematical models, which describe \( Y_{TN} \) as a linear function of the variables \( HRT, X_T, T \) and \( TN_{influent} \), are defined in Tab. 2 through the coefficients \( \beta_{0,A}, \beta_{1,A}, \beta_{2,A}, \beta_{3,A} \) and \( \beta_{4,A} \) for the MBR, hybrid MBBR-MBR, and hybrid MBBR-MBR.

**Table 2**

The mathematical models for the process of nitrogen removal have a high goodness of fit due to the values of \( R^2 \) obtained for the MBR, hybrid MBBR-MBR, and hybrid MBBR-MBR (Tab. 2). Regarding the MBR, \( TN \) removal shows a positive correlation with \( HRT \), and a negative correlation with \( T \) and \( TN_{influent} \) (Fig. 2). This is supported by the values of \( \beta_{1,A} \) (positive), \( \beta_{3,A} \) (negative) and \( \beta_{4,A} \) (negative) (Tab. 2). Moreover, \( \beta_{2,A} \) (-0.0005) reveals the slight influence of MLSS on the TN removal as the angle between these vectors is of approximately 90° (Fig. 2).

In relation to the hybrid MBBR-MBR, \( TN \) removal had a positive correlation with \( HRT, MLSS \) and \( T \), and a negative correlation with \( BD \) and \( TN_{influent} \). This is confirmed by the positive values of \( \beta_{1,A} \) for \( HRT \) and \( \beta_{3,A} \) for \( T \); the negative value of \( \beta_{2,A} \) could demonstrate that the influence of \( BD \) on the TN removal is higher than that exerted by MLSS (Tab. 2) due to the growth of attached biomass on the carriers moving inside the bioreactor.

The coefficients \( \beta_{1,A}, \beta_{2,A}, \beta_{3,A} \) and \( \beta_{4,A} \) for the hybrid MBBR-MBR corroborate that TN removal is directly proportional to \( HRT, MLSS \) and \( BD \), and is inversely proportional to \( T \) and \( TN_{influent} \) (Tab. 2).

According to the mathematical models proposed, the optimum value of \( HRT \) is 30.4 h for the three systems analyzed. Regarding the biomass concentration, the optimum performance of the hybrid MBBR-MBR is achieved at high values of \( X_T \), and the optimum behavior of the MBR and hybrid MBBR-MBR is obtained at low values of \( X_T \). Moreover, the MBR and hybrid MBBR-MBR have the optimum performance for low values of \( T \) and \( TN_{influent} \), and the hybrid MBBR-MBR has its best efficiency for high \( T \) and \( TN_{influent} \) (Tab. 2).

In general, the values of \( R^2 \) obtained for the mathematical models fitting the autotrophic kinetics of the hybrid MBBR-MBR systems as a function of \( HRT, X_T, T \) and \( TN_{influent} \) are higher than that corresponding to the MBR (Tab. 2). Thus, the goodness of fit is better in the hybrid MBBR-MBR systems, probably due to the coexistence of suspended and attached biomass, as discussed for the heterotrophic kinetics.

The effect of \( T \) on the autotrophic kinetic parameters is softened due to the interior installation of the WWTPs (Fig. 2), which is confirmed by the values of \( \lambda_{3,A}, \lambda_{4,A}, \phi_{3,A}, \phi_{4,A}, \alpha_{3,A} \) and \( \alpha_{4,A} \) for the three systems (Tab. 2). HRT has a higher influence on the \( \mu_m,A \) than on the \( K_{NH} \) and \( Y_A \), as confirmed by \( \lambda_{1,A}, \phi_{1,A} \) and \( \alpha_{1,A} \) (Tab. 2) as the ammonium loading rate is directly related to the \( \mu \) corresponding to the biomass. In general, the effect of \( X_T \) on the autotrophic kinetics is attenuated, as indicated by \( \phi_{2,A} \) and \( \alpha_{2,A} \) (Tab. 2), probably due to competitive phenomena between suspended and attached biomass. It should be pointed out that \( X_T \) has the highest influence on the \( \mu_m,A \) for the hybrid MBBR-MBR as \( \lambda_{2,A} \) shows the highest value (Tab. 2). This is in accordance with the greatest influence of attached biomass present in the hybrid MBBR-MBR, which allows to obtain better TN removal efficiency.

In general, it should be noted that \( K_{NH} \) and \( TN_{influent} \) are directly proportional as \( K_{NH} \) reaches a constant value at high substrate concentrations according to Monod model.

Regarding the optimum values of \( \mu_m,A, K_{NH} \) and \( Y_A \), they were 0.0304 h\(^{-1}\), 1.0574 mg N L\(^{-1}\) and 0.4067 mg O\(_2\) mg N\(^{-1}\), respectively, for the MBR, 0.0208 h\(^{-1}\), 5.2873 mg N L\(^{-1}\) and 1.5912 mg O\(_2\) mg N\(^{-1}\), respectively, for the hybrid MBBR-MBR, and 0.0172 h\(^{-1}\), 0.7384 mg N L\(^{-1}\) and 0.8506 mg O\(_2\) mg N\(^{-1}\), respectively, for the hybrid MBBR-MBR.

The values of \( r_{su,A} \) were also determined for the operational conditions and optimum operational conditions (optimum \( r_{su,A} \)) analyzed in the three systems (Tab. 2). In the MBR, \( r_{su,A} \) was within the range of 40.7476 mg N L\(^{-1}\) h\(^{-1}\) and 368.1191 mg N L\(^{-1}\) h\(^{-1}\), and the optimum value was 64.6223 mg N L\(^{-1}\).
The value of $r_{su,A}$ varied between 36.6366 mg N L$^{-1}$ h$^{-1}$ and 59.7009 mg N L$^{-1}$ h$^{-1}$ in the hybrid MBBR-MBR$_A$, and the optimum value was 37.1061 mg N L$^{-1}$ h$^{-1}$ (Tab. S3, Supporting Information). The hybrid MBBR-MBR$_B$ showed $r_{su,A}$ values that fluctuated between 31.7557 mg N L$^{-1}$ h$^{-1}$ and 98.5505 mg N L$^{-1}$ h$^{-1}$, and the optimum value was 42.9504 mg N L$^{-1}$ h$^{-1}$ (Tab. S3, Supporting Information). The optimum values of $r_{su,A}$ were within the operation ranges of the different systems, which supported the modeling and optimization processes. In this regard, the MBR had also the highest rate of total nitrogen removal under optimum operational conditions.

To finish this section, Fig. 3 shows that the type of system, HRT and BD are the main variables which have influence on the performance for the biological processes of COD and TN removal as they had a positive correlation with the organic matter and nitrogen removal and the heterotrophic and autotrophic kinetics of the different bioreactors. Nevertheless, T and MLSS have practically no effect on the response of the system, as discussed previously.

**Figure 3**

**4. Conclusions**

The organic matter removal ($Y_{OM}$) and total nitrogen removal ($Y_{TN}$) could be modeled depending on hydraulic retention time (HRT), total biomass concentration ($X_T$), temperature ($T$), and COD of the influent ($\text{COD}_{influent}$) for organic matter removal or TN of the influent ($\text{TN}_{influent}$) for total nitrogen removal according to the following equation:

$$Y_{OM/TN} = \beta_{0,H/A} + \beta_{1,H/A} \cdot \text{HRT} + \beta_{2,H/A} \cdot X_T + \beta_{3,H/A} \cdot T + \beta_{4,H/A} \left( \frac{\text{COD}_{influent}}{\text{TN}_{influent}} \right)$$

Heterotrophic and autotrophic kinetic performance in terms of maximum specific growth rate ($\mu_m$), half-saturation coefficient ($K_s$) and yield coefficient ($Y$) could be modeled as a function of HRT, $X_T$, T, and COD$_{influent}$ or TN$_{influent}$ according to the following equations:

$$\mu_m = \frac{\lambda_{1,H/A}}{\text{HRT}} + \frac{\lambda_{2,H/A}}{X_T} + \frac{\lambda_{3,H/A}}{T} + \frac{\lambda_{4,H/A}}{T} + \lambda_{5,H/A} \left( \frac{\text{COD}_{influent}}{\text{TN}_{influent}} \right)$$

$$K_s = \frac{\phi_{1,H/A}}{\text{HRT}} + \frac{\phi_{2,H/A}}{X_T} + \frac{\phi_{3,H/A}}{T} + \frac{\phi_{4,H/A}}{T} + \frac{\phi_{5,H/A}}{\left( \frac{\text{COD}_{influent}}{\text{TN}_{influent}} \right)}$$

$$Y = \frac{\alpha_{1,H/A}}{\text{HRT}} + \frac{\alpha_{2,H/A}}{X_T} + \frac{\alpha_{3,H/A}}{T} + \frac{\alpha_{4,H/A}}{T} + \frac{\alpha_{5,H/A}}{\left( \frac{\text{COD}_{influent}}{\text{TN}_{influent}} \right)}$$

(iii) In general, the values of coefficient of determination ($R^2$) for the different models regarding COD and TN removal and kinetic behavior supported the mathematical modeling and were higher for the hybrid MBBR-MBR systems than those obtained for the MBR. This was probably due to the coexistence of suspended and attached biomass in the hybrid MBBR-MBR systems.

**Acknowledgment**

The authors wish to thank the Ministry of Education, Culture and Sport of Spain for the FPU grant no. AP2010-1552 awarded to J.C. Leyva-Díaz, as well as the University of Granada in the training plan “Programa de Ayudas Puente” and the program “Convocatoria de ayudas a Proyectos de I+D Excelencia 2013” (CTM2013-48154-P).

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Symbols used

- **BD** \([\text{mg} \, \text{L}^{-1}]\) biofilm density
- **COD** \([\text{mg} \, \text{O}_2 \, \text{L}^{-1}]\) chemical oxygen demand
- **COD\text{influent}** \([\text{mg} \, \text{O}_2 \, \text{L}^{-1}]\) chemical oxygen demand of the influent
- **HRT** \([\text{h}]\) hydraulic retention time
- **K_M** \([\text{mg} \, \text{O}_2 \, \text{L}^{-1}]\) half-saturation coefficient for organic matter
- **K_NH** \([\text{mg} \, \text{N} \, \text{L}^{-1}]\) half-saturation coefficient for ammonia nitrogen
- **Q_o** \([\text{L} \, \text{h}^{-1}]\) volumetric flow rate of influent
- **Q_s** \([\text{L} \, \text{h}^{-1}]\) volumetric flow rate of effluent
- **r_{su}** \([\text{mg} \, \text{O}_2 \, \text{L}^{-1} \, \text{h}^{-1}]\) substrate degradation rate
- **r_x** \([\text{mg} \, \text{VSS} \, \text{L}^{-1} \, \text{h}^{-1}]\) cell growth rate
- **S_o** \([\text{mg} \, \text{L}^{-1}]\) substrate concentration of influent
- **S_s** \([\text{mg} \, \text{L}^{-1}]\) substrate concentration of effluent
- **T** \([\circ\text{C}]\) temperature
- **V** \([\text{L}]\) bioreactor volume
- **X_T** \([\text{mg} \, \text{L}^{-1}]\) total biomass concentration
- **Y_A** \([\text{mg} \, \text{O}_2 \, \text{mg} \, \text{N}^{-1}]\) yield coefficient for autotrophic biomass
- **Y_H** \([\text{mg} \, \text{VSS} \, \text{mg} \, \text{COD}^{-1}]\) yield coefficient for heterotrophic biomass
- **\mu_{m,A}** \([\text{h}^{-1}]\) maximum specific growth rate for autotrophic biomass
- **\mu_{m,H}** \([\text{h}^{-1}]\) maximum specific growth rate for heterotrophic biomass

Abbreviations

- **DCA** detrended correspondence analysis
- **K_s** substrate half-saturation coefficient
- **MBBR** moving bed biofilm reactor
- **MBR** membrane bioreactor
- **MBBR-MBR** moving bed biofilm reactor-membrane bioreactor
- **MLSS** mixed liquor suspended solids
- **RDA** redundancy analysis
- **S** substrate concentration
- **TN** total nitrogen
- **TN_{influent}** total nitrogen of the influent
- **VSS** volatile suspended solids
- **WWTP** wastewater treatment plant
- **Y** yield coefficient
**Y\textsubscript{OM}** removal of organic matter

**Y\textsubscript{TN}** removal of total nitrogen

**µ** specific growth rate

**µ\textsubscript{m}** maximum specific growth rate

**References**


### Tables

Table 1. Mathematical modeling of the organic matter removal and heterotrophic kinetics and optimization of the operational conditions, organic matter removal and heterotrophic kinetic parameters for the MBR, hybrid MBBR-MBR<sub>a</sub> and hybrid MBBR-MBR<sub>b</sub>

<table>
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<tr>
<th>Parameter</th>
<th>Wastewater treatment plant</th>
<th>MBR</th>
<th>Hybrid MBBR-MBR&lt;sub&gt;a&lt;/sub&gt;</th>
<th>Hybrid MBBR-MBR&lt;sub&gt;b&lt;/sub&gt;</th>
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Table 2. Mathematical modeling of the total nitrogen removal and autotrophic kinetics and optimization of the operational conditions, total nitrogen removal and autotrophic kinetic parameters for the MBR, hybrid MBBR-MBR$_a$, and hybrid MBBR-MBR$_b$.

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<th>Parameter</th>
<th>Wastewater treatment plant</th>
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<th>Hybrid MBBR-MBR$_b$</th>
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</tr>
<tr>
<td>Y$_{TN}$ (%)</td>
<td>80.46</td>
<td>75.94</td>
<td>74.35</td>
<td></td>
</tr>
<tr>
<td>$\mu_{max}$ (h$^{-1}$)</td>
<td>0.0304</td>
<td>0.0208</td>
<td>0.0172</td>
<td></td>
</tr>
<tr>
<td>$K_{S}$ (mg N L$^{-1}$)</td>
<td>1.0574</td>
<td>5.2873</td>
<td>0.7384</td>
<td></td>
</tr>
<tr>
<td>$Y_{a}$ (mg O$_2$ mg N$^{-1}$)</td>
<td>0.4067</td>
<td>1.5912</td>
<td>0.8506</td>
<td></td>
</tr>
</tbody>
</table>
Figure 1. Triplot diagram for RDA of the kinetic parameters for heterotrophic biomass, $\mu_{mHr}$, $K_M$, $Y_{Hr}$ and COD removal (Tab. S2, Supporting Information) in relation to the variables HRT, MLSS, BD, T and COD$_{influent}$ (Tab. S1, Supporting Information) in the MBR (a), hybrid MBBR-MBR$_a$ (b) and hybrid MBBR-MBR$_b$ (c).
Figure 2. Triplot diagram for RDA of the kinetic parameters for autotrophic biomass, $\mu_{m,a}$, $K_{NH}$, $Y_a$, and TN removal (Tab. S2, Supporting Information) in relation to the variables HRT, MLSS, BD, T and TN influent (Tab. S1, Supporting Information) in the MBR (a), hybrid MBBR-MBR$_a$ (b) and hybrid MBBR-MBR$_b$ (c).
Figure 3. Triplot diagram for RDA of the COD and TN removal, heterotrophic and autotrophic kinetic parameters, $\mu_{m,H}$, $K_M$, $Y_H$, $\mu_{m,A}$, $K_{NH}$, $Y_A$, in relation to the variables: wastewater treatment technology (system), HRT, MLSS, BD and T.
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The aim of this study was to develop mathematical models in terms of organic matter and nitrogen removal and kinetic performance to optimize the operation conditions. The operation conditions were the hydraulic retention time, total biomass concentration, temperature and chemical oxygen demand of the influent or total nitrogen of the influent.