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# Biokinetic model for nitrogen removal in free water surface constructed wetlands

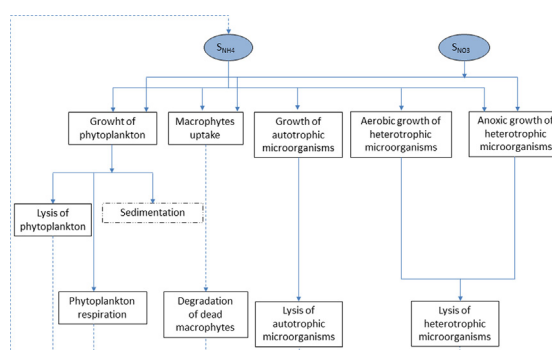
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## HIGHLIGHTS

- A mechanistic biokinetic model for nitrogen removal was developed.
- Immobility of microorganisms was supposed in order to simulate observed data.
- Oscillations in oxygen concentration were essential for simulating denitrification.
- Contribution of each process in nitrogen removal was quantified.
- Most influential parameters were related to growth and lysis of microorganisms.

## GRAPHICAL ABSTRACT



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## ABSTRACT

In this article, a mechanistic biokinetic model for nitrogen removal in free water surface constructed wetlands treating eutrophic water was developed, including organic matter performance due to its importance in nitrogen removal by denitrification. Ten components and fourteen processes were introduced in order to simulate the forms of nitrogen and organic matter, the mechanisms of autotrophic and heterotrophic microorganisms in both aerobic and anoxic conditions, as well as macrophytes nitrogen uptake and release. Dissolved oxygen was introduced as an input variable with a time step of 0.5 days for mimicking eutrophic environments: aerobic conditions were assigned during daylight hours and anoxic conditions during the night. The sensitivity analysis showed that the most influential parameters were those related to the growth of heterotrophic and autotrophic microorganisms. The model was properly calibrated and validated in two full scale systems working in real conditions for treating eutrophic water from Lake L'Albufera (València). In the studied systems, ammonium was mainly removed by the growth of autotrophic microorganisms (nitrification) whereas nitrate was removed by the anoxic growth of heterotrophic microorganisms (denitrification). Macrophyte uptake removed between 9 and 19% of the ammonium entering to the systems, although degradation of dead standing macrophytes returned a significant part to water column.

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## 1. Introduction

During last decades, multiple efforts have been done in order to preserve natural water bodies from eutrophication. Reducing nutrient

loads (mainly, nitrogen and phosphorus) by treating urban wastewater was an important step, but in some cases it was demonstrated to be insufficient for recovering water quality in eutrophicated systems (Martín et al., 2013). For example, in Sweden, where one of the major problems for surface water is eutrophication caused by the diffuse pollution from agricultural sources, 1574 constructed wetlands (CWs) were built between 1996 and 2006 with the aim of reducing agricultural runoff and

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restoring the Baltic Sea good ecological status (Arheimer and Pers, 2016). Furthermore, some experiences carried out worldwide have demonstrated the usefulness of CWs, both free water surface (FWSCWs) and subsurface flow (SSFCWs) configurations, for treating eutrophic water in order to remove nutrients and phytoplankton biomass (He et al., 2007; Li et al., 2008; Martín et al., 2013; Tang et al., 2009).

CWs efficiency removal depends on a large amount of factors (e.g. inlet concentrations, loading rates, hydraulic configuration, vegetation cover, temperature and pH) whose influence can be different for each process. Given the complex task of taking into account the numerous interdependencies, modelling has been demonstrated to be a useful tool for simulating the performance of these systems and many typologies of models have been recently developed. Among them all, mechanistic or process-based models are considered to be the most useful for understanding systems performance (Langergraber, 2008). The vast majority of process-based models have been developed for simulating SSFCWs treating wastewater, being CWM1 (Langergraber et al., 2009), CW2D (Langergraber and Šimůnek, 2005) and BIO-PORE (Samsó and García, 2013) some of the most robust and widely accepted. However, this kind of models are less abundant for simulating FWSCWs and some of the available ones, such as Galanopoulos and Lyberatos (2016), include a limited number of components and interactions. Specifically, seven components and five processes are used in this model for simulating eutrophic water treatment in FWSCWs including nitrogen and organic matter forms, whereas phosphorus influence and interactions between water column and sediment layer were not considered.

One of the most complete models for FWSCWs was developed by Gargallo et al. (2016), where total suspended solids, phytoplankton and phosphorus in eutrophic water were simulated taking into account the effects of avifauna and wind in resuspension processes, as well as vegetation cover (VC) in resuspension and sedimentation mechanisms. In this model, Gargallo et al. considered both organic and total inorganic phosphorus (OP and TIP, respectively,  $\text{mg P L}^{-1}$ ) and the last one was divided into soluble (DIP, dissolved inorganic phosphorus) and particulate (PIP, particulate inorganic phosphorus) fractions. Furthermore, phosphorus accumulated inside the phytoplankton cells was simulated by means of the component  $P_{\text{int}}$  ( $\text{mg P mg Chl a}^{-1}$ ).

However, nitrogen forms and nutrient uptake by plants were not included in this model.

The aim of this paper is (1) to develop a process-based model for nitrogen performance in free water surface constructed wetlands treating eutrophic water, (2) to calibrate and validate it in two full-scale systems working in real conditions and (3) to look into the main processes in relation to nitrogen removal, focusing mainly in nitrate and ammonium.

## 2. Methods

### 2.1. Site description and experimental data

Experimental data for calibrating and validating the model was collected in a set of two FWSCWs located in the natural reserve area known as *Tancat de la Pipa*, in València (Spain). Calibration was carried out in the unit named as FG1 ( $13,509 \text{ m}^2$ ) and validation in FG2 ( $18,240 \text{ m}^2$ ). These FWSCWs, which were planted with cattails (*Thypha* spp.), operated in series in order to treat hypertrophic water from Lake L'Albufera.

These systems were monitored from April 2009 to April 2012. During these three years they worked continuously, except in three periods that water input was stopped due to maintenance tasks. More details about CWs configuration and functioning can be found in Martín et al. (2013).

Three points were studied for water quality: P0 at the inlet to FG1 and P1 and P2 at the outlet of FG1 and FG2, respectively. They were monitored every two weeks from April 2009 to October 2011, and monthly from November 2011 to April 2012 ( $n = 64$ ). Water samples were collected in 2 L bottles, transported and preserved at  $4^\circ\text{C}$  until they were analysed in laboratory, no later than 24 h. Ammonium, nitrate, nitrite, total nitrogen (TN), soluble and total chemical oxygen

demand ( $\text{COD}_s$  and  $\text{COD}_T$ , respectively) were measured using the Spectroquant® Analysis System by Merck, while dissolved oxygen (DO), pH and temperature were measured in situ using portable field measurement equipment (WTW-Multi 340i). Phosphates, total phosphorus, phytoplankton and total suspended solids, as well as inflow, outflow, meteorological data and VC estimations obtained in Gargallo et al. (2016) and Martín et al. (2013) were used as input data for calibrating and validating the model.

In order to establish the fractioning of the organic matter measurements into the organic components of the model ( $S_s$ ,  $S_i$ ,  $X_s$  and  $X_i$ , see Table 1 in next section) one test was carried out in point P0 in February 2010. Particulate and soluble COD and  $\text{BOD}_5$  were measured in P0 ( $\text{COD}_p = 18.2$ ,  $\text{COD}_s = 19.8$ , particulate  $\text{BOD}_5 = 16.0$ , soluble  $\text{BOD}_5 = 3.9 \text{ mg O}_2 \text{ L}^{-1}$ , respectively). Assuming that soluble  $\text{BOD}_5$  corresponds to  $S_s$  and total  $\text{BOD}_5$  to the sum of  $S_s$  and  $X_s$ , it was obtained that  $X_s$  and  $X_i$  correspond to 88% and 12% of  $\text{COD}_p$ , respectively, whereas  $S_s$  and  $S_i$  to 20% and 80% of  $\text{COD}_s$ . These percentages were applied to  $\text{COD}_T$  and  $\text{COD}_s$  measured in the FWSCWs during the studied period.

Once per season, from December 2011 to September 2012, DO and pH were measured every 15 min for 24 h in points P0, P1 and P2. In these point, alkalinity was measured every three weeks from July to December 2015 ( $n = 8$ ) using the methodology by APHA (1991).

### 2.2. Model development

The Activated Sludge Model series structure (Henze et al., 2000) was used for representing the processes involved in nitrogen removal. Matrix notation was used to represent the effect of each process on each component by means of the specific stoichiometric coefficients and processes kinetic rates. Components included in the model are listed in Table 1. The capital letter S was used for denoting soluble components and X for particulate ones. Following CWM1 (Langergraber et al., 2009),  $S_{\text{NO}_3}$  was assumed to include the sum of nitrite and nitrate concentrations and for stoichiometric calculations it was considered to be nitrate. Given the relation between nitrogen and organic matter performance, especially in the denitrification process, the later one was included in the model.

Organic nitrogen (ON), TN,  $\text{COD}_s$  and  $\text{COD}_T$  were obtained by adding the nitrogen or the organic matter content of the corresponding components (Eqs. (1)–(4)), where  $i_{\text{NX}_s}$ ,  $i_{\text{NX}_i}$ ,  $i_{\text{NS}_s}$  and  $i_{\text{NS}_i}$  ( $\text{mg N mg COD}^{-1}$ ) are nitrogen content in  $X_s$ ,  $X_i$ ,  $S_s$  and  $S_i$ , respectively,  $X_p$  ( $\text{mg Chl a L}^{-1}$ ) is phytoplankton concentration,  $i_{\text{NX}_p}$  ( $\text{mg N mg Chl a}^{-1}$ ) is nitrogen content in phytoplankton ( $X_p$ ) and  $i_{\text{CODX}_p}$  ( $\text{mg COD mg Chl a}^{-1}$ ) refers to organic matter content in phytoplankton.

$$\text{ON} = i_{\text{NX}_s} \cdot X_s + i_{\text{NX}_i} \cdot X_i + i_{\text{NS}_s} \cdot S_s + i_{\text{NS}_i} \cdot S_i \quad (1)$$

$$\text{TN} = S_{\text{NH}_4} + S_{\text{NO}_3} + \text{ON} + i_{\text{NX}_p} \cdot X_p \quad (2)$$

$$\text{COD}_s = S_s + S_i + 4.57 \cdot S_{\text{NO}_3} \quad (3)$$

$$\text{COD}_T = \text{COD}_s + X_s + X_i + X_p \cdot i_{\text{CODX}_p} \quad (4)$$

**Table 1**  
Description of the components included in the model.

| Components           | Description                                        | Units                  |
|----------------------|----------------------------------------------------|------------------------|
| 1. $S_{\text{NH}_4}$ | Ammonium concentration.                            | $\text{mg N L}^{-1}$   |
| 2. $S_{\text{NO}_3}$ | Nitrate concentration.                             | $\text{mg N L}^{-1}$   |
| 3. $S_s$             | Reactive soluble organic matter concentration.     | $\text{mg COD L}^{-1}$ |
| 4. $S_i$             | Inert soluble organic matter concentration.        | $\text{mg COD L}^{-1}$ |
| 5. $X_s$             | Reactive particulate organic matter concentration. | $\text{mg COD L}^{-1}$ |
| 6. $X_i$             | Inert particulate organic matter concentration.    | $\text{mg COD L}^{-1}$ |
| 7. $X_H$             | Heterotrophic microorganisms concentration.        | $\text{mg COD L}^{-1}$ |
| 8. $X_A$             | Autotrophic microorganisms concentration.          | $\text{mg COD L}^{-1}$ |
| 9. $X_{\text{ml}}$   | Living macrophyte biomass.                         | $\text{g COD m}^{-2}$  |
| 10. $X_{\text{md}}$  | Dead standing macrophyte biomass.                  | $\text{g COD m}^{-2}$  |

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