Using numerical simulation of a one stage vertical flow wetland to optimize the depth of a zeolite layer
Bernhard Pucher, Hernán Ruiz, Joëlle Paing, Florent Chazarenc, Pascal Molle and Guenter Langergraber

ABSTRACT
This simulation study investigates the treatment performance of a compact French vertical flow wetland using a zeolite layer in order to increase ammonium nitrogen removal. For the modelling exercise, the biokinetic model CW2D of the HYDRUS Wetland Module is used. The calibrated model is able to predict the effect of different depths of the zeolite layer on ammonium nitrogen removal in order to optimize the design of the system. For the model calibration, the hydraulic effluent flow rates as well as influent and effluent concentrations of chemical oxygen demand (COD) and NH₄-N have been measured. To model the adsorption capacity of zeolite, Freundlich isotherms have been used. The results present the simulated treatment performance with three different depths of the zeolite layer, 10 cm (default), 15 cm and 20 cm, respectively. The increase of the zeolite layer leads to a significant decrease of the simulated NH₄-N effluent concentration.

Key words | French vertical flow wetland, HYDRUS Wetland Module, zeolite

INTRODUCTION
In the field of wastewater treatment, treatment wetlands (TWs) are a fast growing alternative to conventional intensive technical treatment technologies. TWs are used for many purposes due to their simplicity of operation and reliable treatment performance without the requirement of complex infrastructure and high investment costs. When it comes to the evaluation of their performance, TWs are still looked at as being black box systems where contaminated water enters and treated water leaves the system. This thinking also reflects in the process of the systems design, where a certain amount of surface area per person equivalent is requested (ÖNORM B 2505 2009; DWA-A 262 2006) or simple first-order decay models are used (Rousseau et al. 2004). Kadlec (2000) showed that such models are not suitable to deal with the complex treatment processes occurring in TWs. With the introduction of new modelling approaches trying to describe these complex and simultaneously active physical, chemical, and biological processes that mutually influence each other, a better understanding is gained and new design tools can be developed (Kadlec & Wallace 2000; Langergraber 2011).

This study is carried out to support the design development of compact French vertical flow (VF) wetland systems treating domestic wastewater. Compact French VF wetlands aim to achieve the treatment efficiencies of the classical two-stage French VF wetland with a single stage. Ruiz et al. (2016) investigated several different compact systems; one particular system uses a zeolite layer as reactive filter media to improve the ammonium nitrogen (NH₄-N) removal. The optimal depth of the zeolite filter layer is of interest as zeolite is very costly compared to gravel or sand.
MATERIALS AND METHODS

Experimental setup

The pilot of the compact French VF wetland has a footprint of 2.3 m². The setup of the filter includes four main layers: first, 20 cm pea gravel (2–4 mm), second, 10 cm zeolite (2–5 mm), third, 20 cm crushed gravel (4–10 mm), and fourth, 20 cm Mayennite (0.5–4 mm) and a 25 cm free drainage layer. The system is fed intermittently with raw domestic wastewater over 3.5 days followed by a resting period of 7 days. The main data collection was carried out on the third day of loading providing nine measurements during the main measurement campaign. For the simulation study, a new measurement campaign, namely the second campaign, was carried out providing data for all 3 days of loading. For detailed information on the experimental setup readers are referred to Ruiz et al. (2016).

Simulation study

HYDRUS Wetland Module

HYDRUS numerically solves variably saturated water flow and solute transport equations. The water flow equation incorporates a sink term to account for water uptake by plant roots. The solute transport equations consider convective-dispersive transport in the liquid phase, diffusion in the gaseous phase, as well as non-linear non-equilibrium reactions between the solid and liquid phases (Šimůnek et al. 2012). Version 2 of the HYDRUS Wetland Module includes two biokinetic model formulations simulating reactive transport in TWs: CW2D and CWM1 (Langergraber & Šimůnek 2012). For the simulation of VF wetlands providing aerobic conditions, CW2D is commonly applied (e.g. Morvannou et al. 2014; Karlsson et al. 2015; Pálfy et al. 2016) and therefore also chosen for this work. The mathematical formulation of CW2D is based on those of the IWA Active Sludge Models (Henze et al. 2000) and includes 12 components and nine processes. The components are dissolved oxygen (O₂), organic matter (OM) divided into readily available (CR), slowly available (CS) and inert organic matter (CI), heterotrophic microorganisms (XH), autotrophic microorganisms (XANs, XANb), nitrogen compounds as ammonium (NH₄-N), nitrite (NO₂⁻N), and nitrate (NO₃⁻N) nitrogen, and nitrogen gas (N₂), as well as inorganic phosphorus (PO₄-P). The nine processes described in CW2D are related to the activities of the heterotrophic and autotrophic bacteria groups, respectively (Table 1).

Model set-up

The transport domain is chosen as a 2D vertical plane with a width of 10 cm and an overall depth of 70 cm, not including the drainage layer. The finite element mesh consists of 102 nodes and 100 elements. At the top of the domain, an atmospheric boundary condition (BC) is applied, while on the bottom, the constant head BC is set to −2 cm to respect the free flowing drainage property (Langergraber 2001).

Hydraulic model

For the simulation of the water flow within the system the van Genuchten Mualem model implemented in HYDRUS is chosen (Mualem 1976; van Genuchten 1980; Šimůnek et al. 2012). This model is described by the following equations:

\[ \theta(h) = \begin{cases} \theta_s + \frac{\theta_k - \theta_s}{1 + \alpha \cdot h^{1/m}} & h < 0 \\ \theta_k & h \geq 0 \end{cases} \quad (1) \]

\[ K(h) = K_s \cdot S_e^h \cdot [1 - (1 - S_e^{1/m})^m]^2 \quad (2) \]

\[ S_e = \frac{\theta - \theta_s}{\theta_k - \theta_s} \quad (3) \]

Table 1 | CW2D processes (Langergraber & Šimůnek 2012)

<table>
<thead>
<tr>
<th>Heterotrophic bacteria</th>
<th>Autotrophic bacteria</th>
</tr>
</thead>
<tbody>
<tr>
<td>Hydrolysis</td>
<td>Conversion of CS into CR</td>
</tr>
<tr>
<td>Aerobic growth of XH on CR</td>
<td>Mineralization of OM</td>
</tr>
<tr>
<td>Anoxic growth of XH on CR</td>
<td>Denitrification on NO₂⁻N</td>
</tr>
<tr>
<td>Anoxic growth of XH on CR</td>
<td>Denitrification on NO₃⁻N</td>
</tr>
<tr>
<td>Lysis of XH</td>
<td>Production of OM</td>
</tr>
<tr>
<td>Aerobic growth of XANs on NH₄⁻N</td>
<td>First step of nitrification</td>
</tr>
<tr>
<td>Lysis of XANs</td>
<td>Production of OM</td>
</tr>
<tr>
<td>Aerobic growth of XANb on NO₂⁻N</td>
<td>Second step of nitrification</td>
</tr>
<tr>
<td>Lysis of XANb</td>
<td>Production of OM</td>
</tr>
</tbody>
</table>
where $\Theta_r$ = residual and $\Theta_s$ = saturated water content \([L^2/L^3]\), $K_s$ = saturated hydraulic conductivity \([L^2/T] \cdot (L^2/h)^{-1}\), the empirical parameters $\alpha$ \([L^{-1}]\) and $n$ \([-\)] influence the shape of the functions $\Theta(h)$ and $K(h)$, and $l$ \([-\)] is defined as the pore-connectivity parameter. $S_e$ equals the effective water content as shown in Equation (3) and $m$ is defined as $m = 1 - 1/n$ where $n > 1$. $L$ = length unit after preference.

Fitting these parameters for all four layers to the measured outflow of the system (Ruiz et al. 2016) using the Marquardt-Levenberg type parameter estimation technique would lead to a high number of possible solutions. Thus, in this study only the material parameters of the fourth layer are fitted using the inverse simulation in HYDRUS (Šimůnek et al. 2012). This step will respect the measured water flow of the system and provide a good fit for the water flow model.

For each filter media six parameters have to be determined. $K_s$ is determined by the falling head method applied in a laboratory setup (Dane & Topp 2002). $\Theta_s$ is assumed to be equal to the total porosity for each filter media (Morvannou et al. 2013). Values of $\Theta_s$ for the gravel layers (layers 1 and 3) are taken from Morvannou et al. (2013) as similar materials were used in a first stage of a French VF wetland. For zeolite the parameters were not measured, i.e. $\Theta_r$, $\alpha$, $n$, are taken form the work of Pucher (2013). The pore connectivity parameter $l$ was set to 0.5 for all filter media (Morvannou et al. 2013).

Reactive transport

The influent concentrations of the model system are derived from the measurements of Ruiz et al. (2016). In CW2D, chemical oxygen demand (COD) is divided into three fractions, readily available organic matter (CR), slowly available organic matter (CS) and inert organic matter (CI) respectively. The CR:CS ratio is 2:1 and CI is chosen roughly at the measured COD effluent concentration (Toscano et al. 2009).

To model adsorption of NH$_4$-N within the zeolite layer a measured Freundlich adsorption isotherm is used (Dal Santo 2009). Equation (4) describes the chemical non-equilibrium two-site adsorption model implemented in HYDRUS. The time dependency of adsorption is described by the concept of two-side sorption, where one part sorption is instantaneous on one part of the exchange sides and considered to be time-dependent on the other side (Šimůnek et al. 2012).

$$ \frac{d s_k^h}{dt} = \alpha \cdot \left[ (1-f) \cdot \frac{k_{s,k} \cdot C_k^{\infty} - s_k^h}{\frac{1}{1+\eta_k \cdot C_k^{\infty}} - s_k^h} \right] $$

where $s_k^h$ = adsorbed concentration of sites assumed to be time dependent \([mg \cdot kg^{-1}]\), $C_k^{\infty}$ = concentration in the aqueous phase \([mg \cdot dm^{-3}]\), $\alpha$ = first order exchange rate coefficient \([d^{-1}]\), $k_{s,k}$ and $\eta_k$ are the adsorption isotherm coefficient for the Freundlich equation when $\eta_k = 0$. The fractions of exchange sites assumed to be in equilibrium with the solution phase is defined as $f$ \([-\)] . In the HYDRUS Wetland Module it is assumed that all adsorption sites are time-dependent by setting $f = 0$.

Calibration procedure

The calibration procedure was as follows:

1. Calibration of the water flow model by determining the missing soil hydraulic parameters using the inverse simulation provided in HYDRUS as described above.

2. Reactive transport simulations:

(a) At first, the model was run using the standard parameter set for the CW2D biokinetic model (Langergraber & Šimůnek 2005).

(b) The first adjustment addresses the unlimited growth of the two bacterial groups namely heterotrophic (XH) and two groups of autotrophic bacteria (Nitrosomonas XANs and Nitrobacter XANb). In the HYDRUS Wetland Module, no growth limiting function for the bacterial growth is implemented. To eliminate the limitless growth (Samsó & García 2013; Pálfy et al. 2016) the lysis rate ($b_h$) of both bacterial groups is increased by changing the growth and die-off ratio.

(c) The most interesting parameter, namely the NH$_4$-N effluent concentration, is addressed by two parameters: the maximum growth rate of XANs ($\mu_{XANs}$) and the first order exchange rate coefficient $\alpha$ of the adsorption model (Equation (4)) which is used to simulate the availability of adsorbed NH$_4$-N for XANs. The adsorption model is part of the regular HYDRUS software. In a first step, several simulations are carried out using the standard parameter for $\mu_{XANs}$ and different values for $\alpha$.

(d) These results are then compared to measured data from pilot scale plots of the same experiment without reactive material to generate realistic behaviour for the adsorption model. The fine-tuning is done by changing the maximum growth rate of the nitrifiers, i.e. $\mu_{XANs}$ For NO$_3$-N, no fitting is performed within this study.

All parameter adjustments are carried out using a trial and error approach. To reach the final results, $b_h$ had to be adjusted again in order to ensure biomass equilibrium.
RESULTS AND DISCUSSION

Measured data

Table 2 presents the results of the first and second measurement campaign for the most important wastewater constituents COD, NH$_4$-N, NO$_2$-N and NO$_3$-N (Ruiz et al. 2016). The influent parameters are used as input for the model.

Calibration of water flow

The determination of the soil hydraulic parameters for the four different filter media and the final simulation of the hydraulic flow within the system sums up the first step of the simulation procedure. Figure 1 illustrates the results of the water flow simulation. Table 3 lists the final set of soil hydraulic parameters. The parameters of the Mayennite layer were fitted using the inverse simulation procedure. The non-marked values in the following table are estimated from different authors (Langergraber 2001; Morvannou et al. 2013; Pucher 2015).

The prediction of the soil hydraulic parameters is an important first step. Within an ongoing experimental setup, the determination of these parameters can be challenging. Morvannou et al. (2013) present a good overview on future experiments required when modelling is also considered at a certain point.

Reactive transport simulation

The final input parameters used for the reactive transport simulations are presented in Table 4. Changes made to the

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**Table 2** Measured influent and effluent parameters (1st campaign: median values on the third day of loading, $n = 9$; 2nd campaign: measured values, $n = 2$)

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Unit</th>
<th>COD [mg L$^{-1}$]</th>
<th>NH$_4$-N [mg L$^{-1}$]</th>
<th>NO$_2$-N [mg L$^{-1}$]</th>
<th>NO$_3$-N [mg L$^{-1}$]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Influent concentrations</td>
<td></td>
<td>850</td>
<td>94</td>
<td>0.008</td>
<td>2.0</td>
</tr>
<tr>
<td>Effluent concentrations</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>1st campaign</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>3rd day</td>
<td></td>
<td>144</td>
<td>12.0</td>
<td>0.05</td>
<td>51.0</td>
</tr>
<tr>
<td>2nd campaign</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>1st day</td>
<td></td>
<td>225</td>
<td>21.7</td>
<td>1.65</td>
<td>104.5</td>
</tr>
<tr>
<td></td>
<td></td>
<td>368</td>
<td>26.8</td>
<td>1.95</td>
<td>127.5</td>
</tr>
<tr>
<td>2nd day</td>
<td></td>
<td>198</td>
<td>12.9</td>
<td>5.10</td>
<td>76.5</td>
</tr>
<tr>
<td></td>
<td></td>
<td>187</td>
<td>17.0</td>
<td>3.25</td>
<td>70.0</td>
</tr>
<tr>
<td>3rd day</td>
<td></td>
<td>124</td>
<td>11.4</td>
<td>3.25</td>
<td>43.0</td>
</tr>
<tr>
<td></td>
<td></td>
<td>134</td>
<td>15.0</td>
<td>3.10</td>
<td>69.5</td>
</tr>
</tbody>
</table>

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**Table 3** Final soil hydraulic parameters for all layers of the TW

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Unit</th>
<th>$\Theta_r$ [cm$^3$ cm$^{-1}$]</th>
<th>$\Theta_s$ [cm$^3$ cm$^{-1}$]</th>
<th>$\alpha$ [cm$^{-1}$]</th>
<th>$n$ [I–I]</th>
<th>$k_s$ [cm h$^{-1}$]</th>
<th>$l$ [I–I]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Gravel 2–4</td>
<td></td>
<td>0.31$^a$</td>
<td>0.37$^b$</td>
<td>0.14</td>
<td>2.68</td>
<td>252$^b$</td>
<td>0.5$^c$</td>
</tr>
<tr>
<td>Zeolite 2–5</td>
<td></td>
<td>0.047$^d$</td>
<td>0.55$^b$</td>
<td>0.141$^d$</td>
<td>3.67$^d$</td>
<td>288$^b$</td>
<td>0.5$^c$</td>
</tr>
<tr>
<td>Gravel 4–10</td>
<td></td>
<td>0.22$^a$</td>
<td>0.415$^b$</td>
<td>0.145</td>
<td>2.68</td>
<td>324$^b$</td>
<td>0.5$^c$</td>
</tr>
<tr>
<td>Mayennite 0.5–4</td>
<td></td>
<td>0.0472$^e$</td>
<td>0.42$^b$</td>
<td>0.119$^e$</td>
<td>3.91$^e$</td>
<td>247$^e$</td>
<td>0.5$^c$</td>
</tr>
</tbody>
</table>

$^a$Morvannou et al. (2013).
$^b$Measured.
$^c$Mualem (1976).
$^e$Fitted (inverse solution).
standard parameter set (Langergraber & Šimůnek 2005), in order to gain biomass equilibrium and fit the simulated and measured NH$_4$-N effluent concentrations, are presented in Table 5.

Figures 2 and 3 illustrate the concentrations of the heterotrophic and autotrophic bacterial biomass (XH and XANs, respectively). Steady state conditions were reached after about 200 days' simulation time. The different behaviour during loading and resting times can be observed. The concentration of autotrophic bacteria mass is highest in layer 2 at 25 cm, which represents the zeolite layer.

In Figure 4, the simulated sorption of NH$_4$-N with the zeolite layer is shown. Steady state is reached from the beginning, when the initial condition for adsorbed NH$_4$-N is set to 2,000 mg kg$^{-1}$. The Freundlich isotherm coefficients $k_{s,k}$ and $\beta_k$, presented in Equation (4), are set to 660 mL $\mu$g$^{-1}$ and 0.76, respectively (Dal Santo 2009).

The results in Figures 3 and 4 show the growth of autotrophic MO on NH$_4$-N is happening during the loading time while in the resting period there is die-off leading to a decrease in the nitrification. This is also true for the zeolite layer. The greater amount of autotrophic MO in the second layer relates also to measurements done by Morvannou et al. (2011).

Simulation results for the most important effluent parameters COD and NH$_4$-N are presented in Figures 5 and 6, for the last loading period of 3.5 days of the overall simulation time. For the first measurement campaign, the mean value plus/minus standard deviation is shown while the second campaign is illustrated by the values from two measurements.

Figure 6 shows simulated effluent concentration for NH$_4$-N at different values of $\mu$ and $\alpha$. Slower growth of XANs (decreasing values of $\mu$) at constant $\alpha$ increases the NH$_4$-N effluent concentration. Faster sorption, i.e. a higher value of $\alpha$ at constant $\mu$, further increases the NH$_4$-N effluent concentration. The best fit could be achieved for $\mu = 0.8$ d$^{-1}$ and $\alpha = 0.008$ d$^{-1}$, respectively. These values have been used for the further simulations.

Although the sources of zeolite used by Dal Santo et al. (2010) and Ruiz et al. (2016) have been different, the results showed that assuming both types of zeolite are similar (in terms of hydraulic and sorption behaviour, respectively) and that the parameters measured by Dal Santo et al. (2010) can be used for the simulation study. As the experimental system was quite young, an increased NH$_4$-N adsorption capacity of an established sludge layer was not considered.
Figure 3 | Simulated autotrophic bacteria concentrations (XANs) in three different filter depths.

Figure 4 | Simulated adsorption and desorption of NH₄-N within the zeolite layer.

Figure 5 | Simulated COD effluent concentration over the last 3.5 days of loading.
Using the calibrated model, the depth of the zeolite layer is increased to 15 cm (+5 cm) and 20 cm (+10 cm), respectively. The results, presented in Figure 7, show a decrease in the simulated NH₄-N effluent concentration with an increase of the zeolite layer depth. A summary of the simulated effluent concentrations for different depth of the zeolite layer, namely COD, NH₄-N and NO₃-N, is listed in Table 6.

**CONCLUSION**

The following conclusions can be drawn:

- The water flow model could be calibrated well using literature data for the gravel and zeolite layers and applying the inverse simulation provided by HYDRUS for determining the soil hydraulic properties of the Mayenne layer. As a good calibration of the water flow model is required, several measurements should be
carried out when modelling is also considered at a certain point (see e.g. Morvannou et al. 2013).

- To reach equilibrium biomass concentrations, adjustment of lysis rates \( b \) for each bacterial group was needed. Although the implementation of a growth limiting function based on the pore space within the media, would be a better solution, especially for systems without prolonged resting periods.
- A good overall match of measured and simulated data for the \( \text{NH}_4\text{-N} \) and COD effluent concentration was observed.
- The expected behaviour of the zeolite layer, i.e. during loadings \( \text{NH}_4\text{-N} \) is adsorbed on the media and is available for autotrophic bacteria during non-loading periods, can be simulated. However, no calibration was carried out for the regeneration process and further studies are required.
- Simulation results show that increasing the zeolite layer decreases the \( \text{NH}_4\text{-N} \) effluent concentrations significantly.
- In general, further validation is needed in order to proof the simulated results when increasing the depth of the zeolite layer. This may be the next step in order to establish this model as a design support tool to help upcoming experimental designs.

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First received 1 September 2016; accepted in revised form 15 November 2016. Available online 28 November 2016