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Numerical simulation of phosphorus removal in horizontal subsurface flow constructed wetlands

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ABSTRACT

The removal of total phosphorus (TP) in horizontal subsurface flow constructed wetlands (HSF CW) is numerically investigated using the Freundlich linear isotherm for adsorption. For the numerical simulation, the Visual MODFLOW code family, based on the finite difference method, was used. This model is applied in the simulation of five pilot-scale HSF CWs. Influent–effluent TP concentration experimental data from these facilities were used to first calibrate this model and derive appropriate values of the distribution coefficient K_d and the first-order removal coefficient λ. Then, the model was verified using the values of λ and K_d from calibration and independent experimental data from these facilities. The calibrated model was applied to comparatively simulate differences in removal efficiency under various design and operational parameters.

Keywords: Wastewater treatment; Constructed wetlands; Total phosphorus removal; Adsorption; Freundlich isotherm; Numerical modeling; MODFLOW

1. Introduction

The use of horizontal subsurface flow constructed wetlands (HSF CWs) for the treatment of wastewater is considered to be an attractive ecological and economic solution, especially for small settlements [1–5]. Among the various wastewater constituents, phosphorus removal presents some challenge, governed by the phenomenon of adsorption. The removal of total phosphorus (TP) in HSF CWs is still under study [6–10]. Several studies, mostly experimental ones, propose the use of alternative filter media materials to enhance phosphorus removal [11–17].

A literature search revealed limited use of computer codes for the simulation of phosphorus removal in HSF CWs [18]. Recently, artificial neural networks have been successfully used to develop models predicting the performance of HSF CWs in phosphorus removal [19].

Regarding adsorption, the type of porous medium has a principal role. For the evaluation of adsorption

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characteristics, available experimental data have to be analyzed. In some cases, the Langmuir isotherm is used [20–22], while other studies consider both the Freundlich and the Langmuir isotherms [23,24].

In this study, the removal of TP in HSF CWs is numerically investigated using the Freundlich linear isotherm for adsorption. For the numerical simulation, the Visual MODFLOW (Modular Finite-difference Groundwater Flow Model) code family [25], based on the finite difference method (FDM), was used. Experimental data from five pilot-scale HSF CWs, which were constructed and operated in our laboratory [26], were used in model validation. The study provides values for the TP first-order removal coefficient λ and the distribution coefficient K_d for various experimental conditions.

2. Materials and methods

2.1. Mathematical formulation

Phosphorus is a constituent whose fate in soils and porous media is, generally, dominated by the process of adsorption. The relevant partial differential equation describing the phenomenon is derived from mass balance of the pollutant. In the three-dimensional (3-D) space, considering convection, dispersion, sources/sinks, equilibrated adsorption, and first-order irreversible kinetic reactions, this equation reads [27]:

$$\theta R_d \frac{\partial C}{\partial t} = \frac{\partial}{\partial x_i}\left(\theta D_{ij}\frac{\partial C}{\partial x_j}\right) - \frac{\partial}{\partial x_i}(q_i C) + q_s C_s - \lambda_1 \theta C - \lambda_2 \rho_b S \quad (1)$$

where C is the solute concentration [ML^{-3}], S is the adsorbed concentration [M pollutant/M solid], q_i is the Darcy velocity [$L\,T^{-1}$], D_{ij} is the hydrodynamic dispersion coefficient tensor [$L^2 T^{-1}$], q_s is the volumetric flow rate per unit volume of aquifer representing fluid sources (positive) or sinks (negative) [T^{-1}], C_s is the concentration of the source or sink flux [ML^{-3}], θ is the porosity [–], ρ_b is the dry bulk density of the soil [ML^{-3}], λ_1 is the removal coefficient for the dissolved phase [T^{-1}], and λ_2 is the removal coefficient for the adsorbed phase [T^{-1}]. Usually, it is assumed $\lambda_1 = \lambda_2 = \lambda$.

The values of ρ_b are calculated by the equation:

$$\rho_b = (1-\theta)\rho_r \quad (2)$$

where ρ_r is the density of solid grains of the porous material (usually 2.65 g cm^{-3}).

In Eq. (1), R_d is the retardation factor given by the following equation:

$$R_d = 1 + \frac{\rho_b}{\theta} K_d \quad (3)$$

where K_d is the distribution coefficient [$L^3 M^{-1}$], which is determined by the equation:

$$K_d = \frac{\partial S}{\partial C} \quad (4)$$

K_d expresses the distribution of the pollutant concentrations between solid and liquid phases, S and C, respectively. Regarding the dependence of S on C, the frequently used adsorption isotherms are the Freundlich and Langmuir ones [27].

In our study, the linear Freundlich isotherm was used, which is expressed by the equation:

$$S = K_d \cdot C \quad (5)$$

2.2. Available experimental data

The removal of TP was numerically investigated. Available experimental data from five pilot-scale HSF CWs were used. These pilot-scale units were operated in parallel experiments continuously for two years (2004–2006), in the Laboratory of Ecological Engineering and Technology. The experimental layout, procedures, obtained data, and their statistical analyses are presented in detail by Akratos and Tsihrintzis [26], therefore, are only briefly presented here. The pilot-scale CW units were rectangular and contained various local porous media (i.e. medium gravel-MG, fine gravel-FG, and cobbles-CO) and two vegetation species (reed-R and cattails-C). One CW unit was unplanted (Z). Each tank was made of steel and had dimensions $L = 3.0$ m in length, $W = 0.75$ m in width, and $H = 1.0$ m in depth, while the thickness of the porous material was $d = 0.45$ m. Synthetic wastewater was used as feedwater, which was produced and introduced three times daily (every eight hours) in the five pilot-scale HSF CWs. The ratio BOD/COD was about 0.6. TP concentrations in the influent, effluent, and at distances of 1/3 and 2/3 of the unit length were measured during the experiments using vertical in-planted perforated plastic pipes. For more details, refer to Akratos and Tsihrintzis [26]. Operational characteristics of all CW tanks (i.e. ambient temperature and other meteorological parameters, hydraulic residence time [HRT], etc.) were the same [26].

2.3. Simulation by MODFLOW computer code

For the numerical simulation of TP removal in the pilot-scale HSF CWs, the MODFLOW family code was used [25]. It is based on the FDM and is one of the most widely used computer codes for ground-water flow investigations. MODFLOW was combined with the MT3DMS package [28], which offers the possibility to simulate the removal of TP using the Freundlich linear isotherm for adsorption. More details on these two models are given in [25,29].

First-order decay for TP removal and linear Freundlich isotherm for adsorption were assumed. MODFLOW was calibrated to obtain optimum values of the removal coefficient λ in Eq. (1) by adopting a trial-and-error procedure, similar to that described in Liolios et al. [30,31]. In this procedure, TP concentration values computed by the model at the CW outlet were matched with the corresponding experimental values. Proper values for the distribution coefficient K_d were obtained which varied within the proposed range in the literature. For verification, the model was run using λ and K_d values obtained in calibration, and a comparison was made between the computed and the experimental results of TP concentration at distances 1/3 and 2/3 of the CW unit length.

3. Results and discussion

3.1. Phenomenological coefficients and other parameters of the porous media for each tank

The main phenomenological coefficients and other parameters, which were introduced into the MODFLOW computer code for the simulation of the experiments, were the porosity, the hydraulic conductivity, and the inflow rate for each unit.

The hydraulic conductivity K [L T^{-1}] was computed using the following equation [1]:

$$K = \frac{\rho g \theta^{3.7} D_{50}^2}{255(1-\theta)\mu} \tag{6}$$

where ρ is the water density [ML^{-3}], g is the gravity acceleration [L T^{-2}], D_{50} is the experimentally estimated diameter of each porous media [L], μ is the dynamic water viscosity [ML^{-1} T^{-1}], and θ [–] is the porosity. The experimentally measured values of porosity in the five CW units varied between 0.28 and 0.37, affected by porous media size and vegetation type and density. D_{50} varied between 6 and 90 mm [26].

The pore volume V_p [L^3] was estimated by the relation:

$$V_p = L\, W\, d\, \theta \tag{7}$$

Finally, the inflow rate Q_{in} [L^3 T^{-1}] was computed for each tank using the corresponding HRT:

$$Q_{in} = \frac{V_p}{\text{HRT}} \tag{8}$$

The following Table 1 provides, for each HSF CW and for HRTs of 6, 8, 14, and 20 d, the values of the above phenomenological parameters.

3.2. Model calibration: determination of K_d and λ

The inflowing TP concentration in the experiments was about 9 mg L^{-1}, a typical value for domestic wastewater. According to the literature [1,6,32–34], this value is considered as a low one and so, the use of the Freundlich linear isotherm is suggested. Based on the CW literature for similar porous media [21,24], the following four representative values of the distribution coefficient were tested: K_d = 0.1, 0.25, 1.0, and 2.0 cm^3 g^{-1}. Next, using the above values of K_d, optimal values of the first-order removal coefficient λ for the simulation of TP removal were determined. For this purpose, the measured concentration values at the

Table 1
Phenomenological coefficients and other parameters for each pilot-scale HSF CW [30]

Tank	HRT (d)	θ (–)	K (m s^{-1})	V_p (m^3)	Q_{in} (L d^{-1})
MG-R	6	0.35	0.7196	0.3544	59.1
	8	0.36	0.7640	0.3594	44.9
	14	0.38	0.9743	0.3848	27.5
	20	0.35	0.7196	0.3544	17.7
MG-C	6	0.33	0.5620	0.3341	55.7
	8	0.34	0.5980	0.3392	42.4
	14	0.34	0.6366	0.3443	24.6
	20	0.33	0.5620	0.3341	16.7
MG-Z	6	0.37	0.9120	0.3746	62.4
	8	0.37	0.9120	0.3746	46.8
	14	0.37	0.9120	0.3746	26.8
	20	0.37	0.9120	0.3746	18.7
FG-R	6	0.29	0.0199	0.2936	48.9
	8	0.29	0.0199	0.2936	36.7
	14	0.31	0.0262	0.3139	22.4
	20	0.29	0.0199	0.2936	14.7
CO-R	6	0.28	3.8710	0.2835	47.3
	8	0.28	3.8710	0.2835	35.4
	14	0.28	3.8710	0.2835	20.3
	20	0.28	3.8710	0.2835	14.2

outlet of the five CW units in the afore-mentioned trial-and-error procedure were used.

For each CW unit and for HRTs of 6, 8, 14, and 20 d, Table 2 provides representative values of the average temperature T_{av} [°C], the inlet C_{in} and outlet C_{out} TP concentrations [mg L^{-1}], the bulk density ρ_b [g cm^{-3}], and the values of λ [d^{-1}] for $K_d = 0.1$, 0.25, 1.0, and 2.0 $cm^3 g^{-1}$, respectively. Regarding the variation of ρ_b in the planted tanks, this was due to changes of porosity caused by plant growing during the two-year experiments (2004–2006).

3.3. Model verification

As mentioned, in order to verify the model, a comparison was made between the computed and the experimental values measured at distances 1/3 and 2/3 of the CW length. The λ and K_d values estimated in the previous section were used. To assess the goodness of the verification, linear regression lines of the form $y = \gamma x$ were fitted between computed (y) and experimental (x) values.

The results are presented in Table 3. For best fit, both the slope γ of these equations and the coefficient of determination R^2 should be as close to 1.0 as possible. As Table 3 shows, the values of slope γ are slightly higher than 1.0 for units MG-R and MG-Z, which means that the computational simulation overestimates slightly the TP concentrations for these units. For the other units, the values are slightly less than 1.0. Concerning the coefficient of determination R^2, its values are better for the units MG-C, FG-R, and CO-R in comparison to units MG-R and MG-Z. R^2 values are generally low, particularly for the above-mentioned units MG-R and MG-Z. However, the values for the other three units range at an acceptable level (~0.6) for similar studies; however, this also indicates that several other parameters are needed to describe these complex systems and phosphorus removal processes, and improve model performance.

As the results of Table 3 show, the optimum value of K_d is 0.1 $cm^3 g^{-1}$, because the corresponding γ values are closer to 1.0, and the R^2 values are greater and closer to 1.0 than those for the other values of K_d. Nevertheless, the difference between the γ and R^2 values between $K_d = 0.1$ $cm^3 g^{-1}$ and 0.25 $cm^3 g^{-1}$ is small, indicating that most appropriate values for distribution coefficient K_d may be in the range between 0.10 and 0.25 $cm^3 g^{-1}$. This value is also included in the range of relevant K_d values (0.03–0.28 $cm^3 g^{-1}$) suggested by Molle et al. [21], concerning phosphorus removal in HSF CWs with calcareous gravel porous

Table 2
Values of the removal coefficient λ for TP removal in the pilot-scale HSF CWs

HRT	Tank	T_{av} (°C)	C_{in} (mg L^{-1})	C_{out} (mg L^{-1})	ρ_b (g cm^{-3})	K_d ($cm^3 g^{-1}$) 0.1	0.25	1.0	2.0
						λ (d^{-1})			
6	MG-R	15.7	9.2	9.2	1.723	0.000	0.000	0.000	0.000
	MG-C			7.0	1.776	0.026	0.019	0.006	0.003
	MG-Z			5.8	1.670	0.047	0.037	0.012	0.004
	FG-R			2.8	1.882	0.107	0.074	0.024	0.013
	CO-R			6.9	1.908	0.025	0.016	0.005	0.003
8	MG-R	12.1	9.2	7.5	1.709	0.015	0.010	0.004	0.002
	MG-C			3.9	1.762	0.062	0.044	0.015	0.008
	MG-Z			5.9	1.670	0.034	0.027	0.009	0.005
	FG-R			1.4	1.882	0.128	0.082	0.028	0.017
	CO-R			5.5	1.908	0.025	0.020	0.007	0.003
14	MG-R	16.2	9.7	5.8	1.643	0.023	0.018	0.006	0.004
	MG-C			2.3	1.749	0.061	0.044	0.015	0.008
	MG-Z			5.5	1.670	0.025	0.019	0.007	0.004
	FG-R			1.1	1.829	0.090	0.063	0.021	0.011
	CO-R			1.6	1.908	0.069	0.044	0.015	0.006
20	MG-R	15.2	8.8	5.7	1.723	0.012	0.007	0.003	0.002
	MG-C			1.8	1.776	0.046	0.035	0.011	0.006
	MG-Z			4.5	1.670	0.020	0.017	0.005	0.003
	FG-R			1.1	1.882	0.056	0.037	0.012	0.007
	CO-R			2.5	1.908	0.034	0.022	0.007	0.004

Table 3
Correlation of experimental and computed results

Tank	$K_d = 0.1\,cm^3\,g^{-1}$		$K_d = 0.25\,cm^3\,g^{-1}$		$K_d = 1\,cm^3\,g^{-1}$		$K_d = 2\,cm^3\,g^{-1}$	
	γ	R^2	γ	R^2	γ	R^2	γ	R^2
MG-R	1.0562	0.2166	1.0640	0.2120	1.0664	0.2017	1.0711	0.1596
MG-C	0.9024	0.6435	0.8989	0.6429	0.8977	0.6425	0.8972	0.6293
MG-Z	1.1188	0.2612	1.1198	0.2601	1.1200	0.2511	1.1272	0.2299
FG-R	0.7494	0.5592	0.7482	0.5528	0.7476	0.5514	0.7462	0.5488
CO-R	0.9369	0.6597	0.9293	0.6232	0.9248	0.6173	0.9205	0.6065

media, and the value $0.28\,cm^3\,g^{-1}$ suggested in the HYDRUS manual by Langergraber and Simunek for vertical SF CWs [35].

3.4. Effect of design and operational parameters on TP removal

Using the calibrated and verified model, the influence of the main CW design and operational parameters (i.e. vegetation, temperature, porous media, and HRT) on TP concentration prediction could be tested. For this purpose, the calibrated model was run with the values of λ for $K_d = 0.1\,cm^3\,g^{-1}$ estimated in Section 3.2 (Table 2) and the values of the main phenomenological coefficients and other parameters reported in Section 3.1 (Table 1). The results include TP concentration along the centerline of each HSF CW, and are presented in Figs. 1–4. It was assumed that the inlet TP concentration of the wastewater which was injected in the CW units was the same for all the runs, i.e. $9\,mg\,L^{-1}$.

Regarding the influence of vegetation, Fig. 1 compares predicted TP concentrations in three units, MG-Z, MG-C, and MG-R. These were similar in all design parameters except the plant. The temperature was 15°C and HRT 8 d. Cattails (MG-C) were more

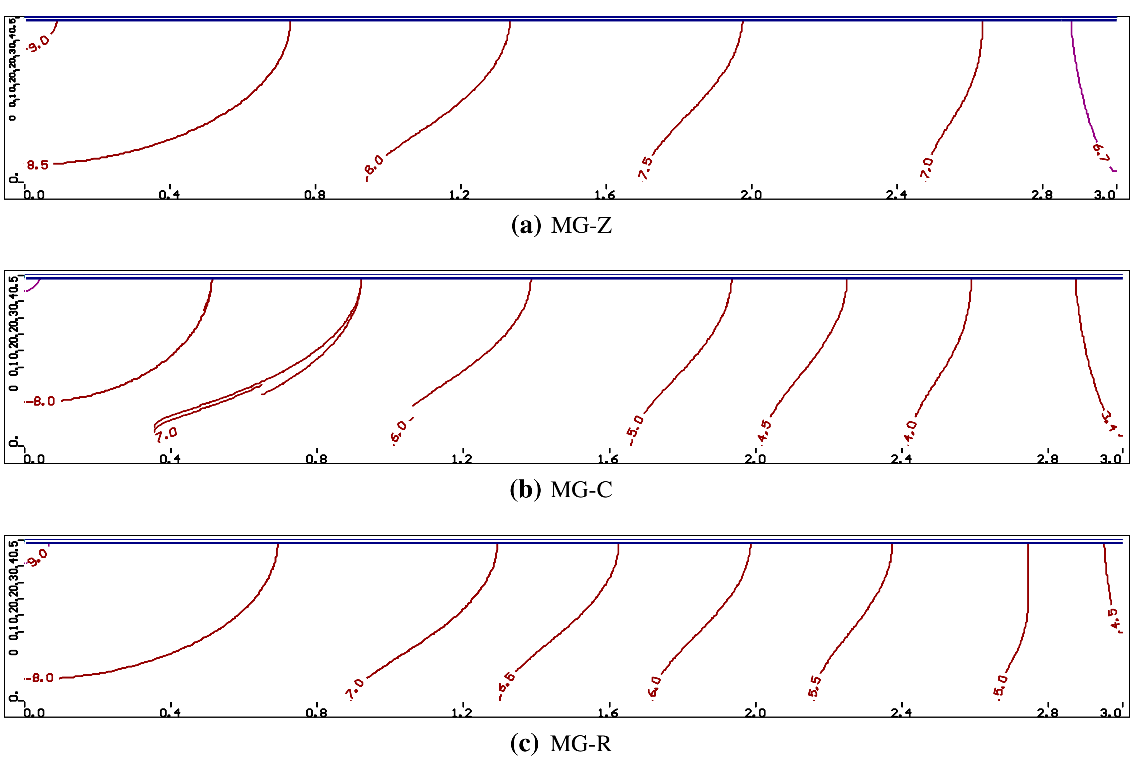
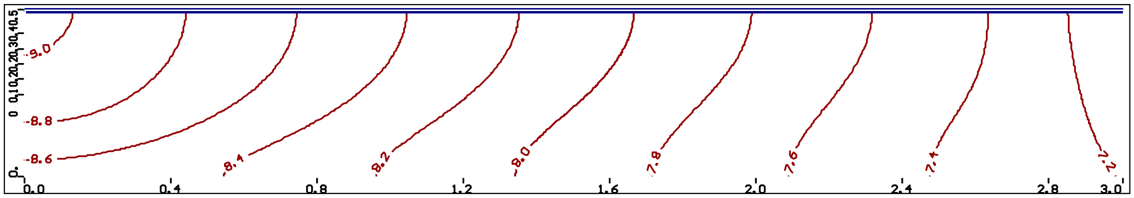
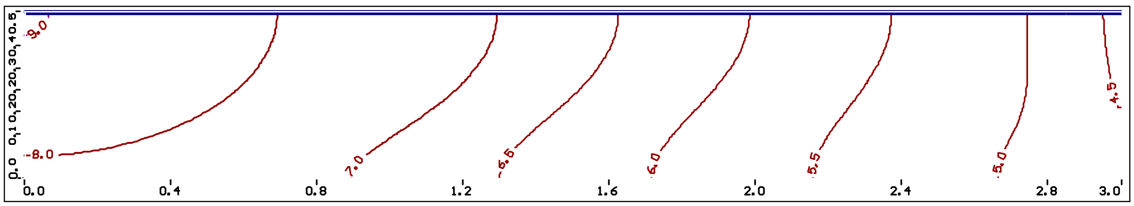


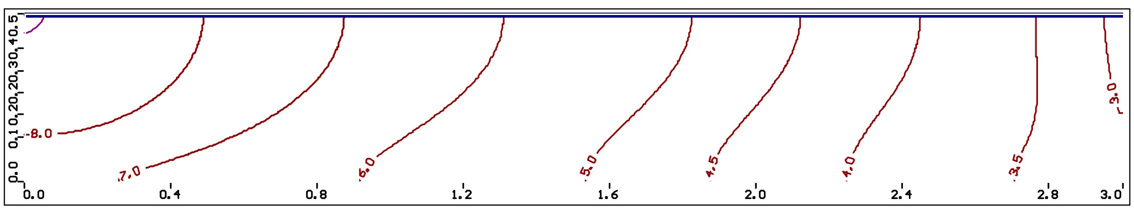
Fig. 1. Effect of vegetation on TP removal: profiles of TP concentration ($mg\,L^{-1}$) in cross sections of three HSF CW units, filled with medium gravel (MG), operating at temperature 15°C, and HRT of 8 d: (a) unplanted (MG-Z); (b) cattails (MG-C); (c) reeds (MG-R).



(a) T = 8 °C

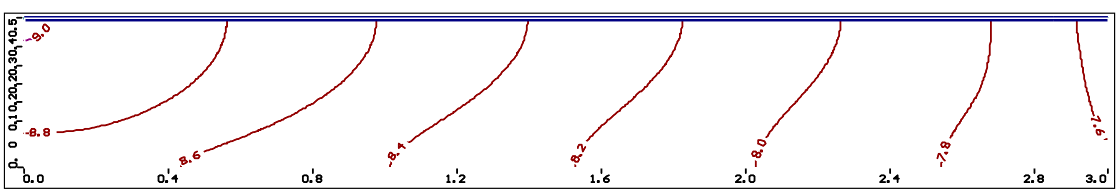


(b) T = 15 °C

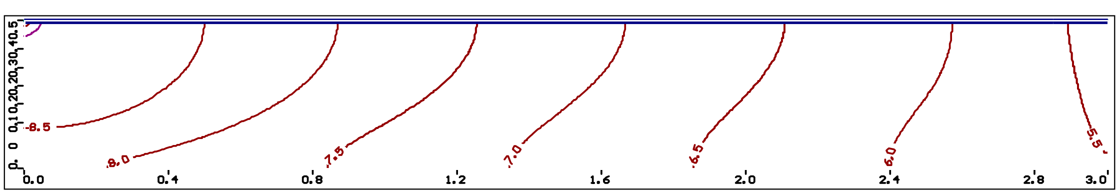


(c) T = 20 °C

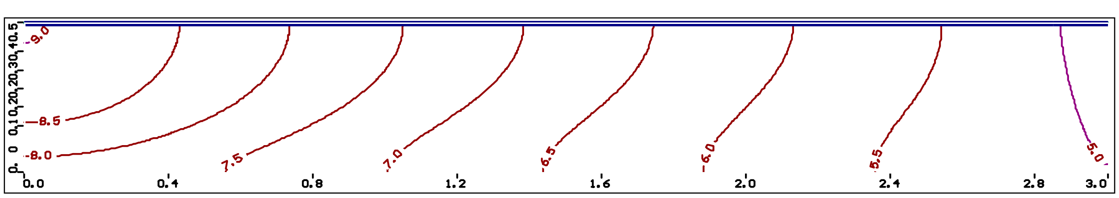
Fig. 2. Effect of temperature (*T*) on TP removal: profiles of TP concentration (mg L^{-1}) in cross sections of one HSF CW, filled with medium gravel and planted with reeds (MG-R), operating at a HRT of 8 d under three different temperatures: (a) 8°C; (b) 15°C; (c) 20°C.



(a) MG-R

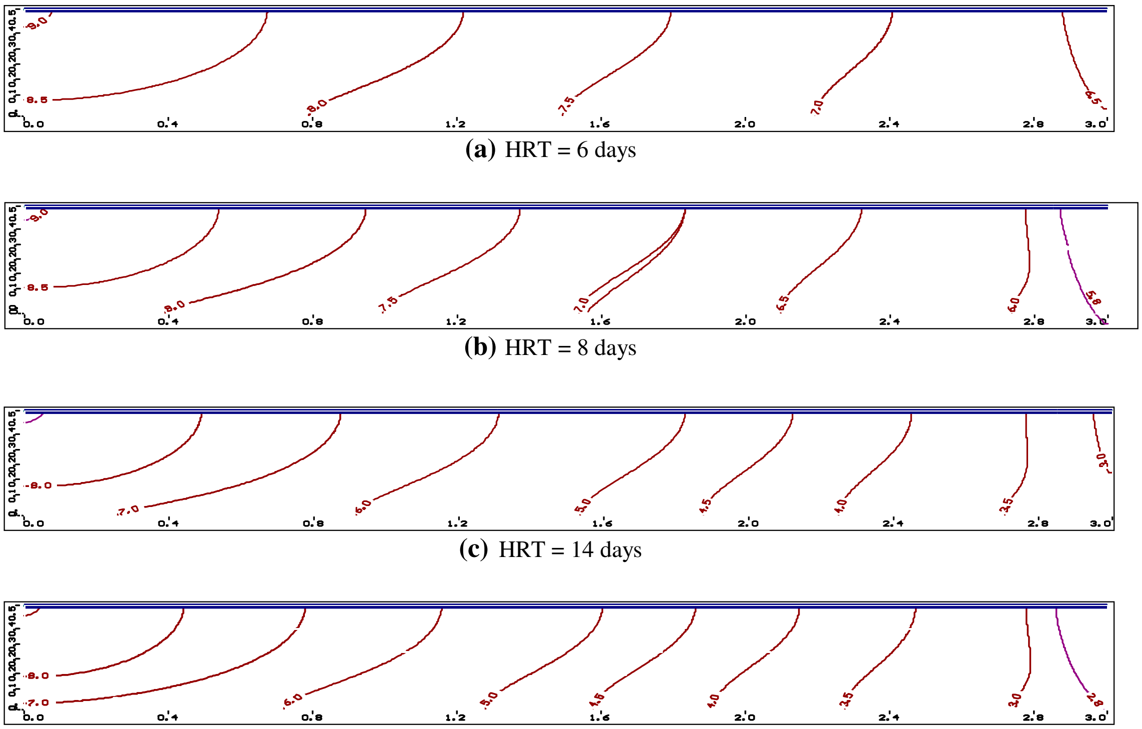


(b) FG-R



(c) CO-R

Fig. 3. Effect of porous media on TP removal: profiles of TP concentration (mg L^{-1}) in cross sections of three HSF CW units planted with reeds, operating at a temperature of 15°C, and HRT of 8 d: (a) MG-R; (b) FG-R; (c) CO-R.



(a) HRT = 6 days

(b) HRT = 8 days

(c) HRT = 14 days

(d) HRT = 20 days

Fig. 4. Effect of HRT on TP removal: profiles of TP concentration (mg L^{-1}) in cross sections of MG-R unit, planted with reeds, at a temperature of 20˚C, and HRT of: (a) 6 d; (b) 8 d; (c) 14 d; (d) 20 d.

effective in TP removal, compared to reeds (MG-R), as seen by the lower TP concentration at the downstream end of the MG-C unit. This may have to do with the fact that cattails have a more vigorous root system [26,36]. The TP removal was the lowest for the unplanted tank (MG-Z).

The effect of the temperature on TP removal is presented in Fig. 2. The performance of one unit with porous material medium gravel, planted with reeds (MG-R), operating at a HRT of 8 d was simulated for three different values of temperature (i.e. 8, 15, and 20˚C). The highest removal efficiency is seen for the highest temperature, apparently due to enhanced bacterial performance.

The effect of porous media on TP removal is presented in Fig. 3. Three CW units (FG-R, MG-R, and CO-R) are simulated, operating under the same conditions (temperature 15˚C; HRT 8 d) and planted with the same plant (reeds), but containing different porous media (fine gravel, medium gravel, and cobbles). Best performance was recorded for the tanks filled with cobbles (CO-R) and fine gravel (FG-R), while the poorest for the bed filled with medium gravel. The most possible explanation for this is that the origin of the porous materials prevails over size in TP removal: FG and CO were collected from a river bed (igneous material containing Si, Al, and Fe, which contribute significantly to the absorption of phosphorus). Conversely, the MG was obtained from a quarry, containing mostly Ca and very low quantities of Al and Fe [26].

Finally, regarding the influence of the HRT, the results are presented in Fig. 4. The MG-R unit is simulated for $T = 20$˚C and four residence times, i.e. 6, 8, 14 and 20 d. The highest removal occurs for the highest HRT.

4. Conclusions

A numerical simulation of flow and TP transport and removal in porous media has been presented with emphasis on modeling HSF CW performance. The Visual MODFLOW code, combined with the MT3DMS module, was used for the computational simulation of two-years experiments realized in five rectangular pilot-scale HSF CW units. The Freundlich linear isotherm for adsorption was employed using four estimated values of the distribution coefficient K_d. The computational results were in acceptable agreement with the experimental ones.

Optimal values for the first-order reaction rate λ for TP removal were estimated for each typical value of K_d using inverse problem procedures. Based on the experimental results, values in the range 0.1–0.25 $cm^3 g^{-1}$ for K_d were the optimum among the tested values for the pilot-scale units. The proposed values of first-order reaction rate λ and K_d can be used effectively for the prediction of performance and optimum design of full-scale CWs, having similar design and operational characteristics with the investigated HSF CWs herein.

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