



Removal of nitrobenzene from wastewater by vertical flow constructed wetland and optimizing substrate composition using Hydrus-1D: optimizing substrate composition of vertical flow constructed wetland for removing nitrobenzene from wastewater

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Abstract

Nitrobenzene is a toxic chemical that is mainly used in industries to produce aniline and many other products, and eventually it is excessively present in industrial wastewaters. In this study, NB removal efficiency data were obtained by experimental scale vertical flow constructed wetlands. Four columns with same size and diameter named as A, B, C and D were used for the experiment and filled with the substrate in three layers having different soil compositions. Synthetic wastewater was prepared in the laboratory and fed to all the wetland columns. The Hydrus-1D model was used to mimic the removal and transport of nitrobenzene by these data with the same boundary conditions. The values of NB removal and the influence of water content and hydraulic conductivity were compared with all the columns, and best composition of substrate was selected on the basis of maximum removal of nitrobenzene. 76.2% was the maximum removal efficiency of NB exhibited by column D, while column A, B and C were having 50.2%, 55.8% and 65.9%, respectively.

Keywords Constructed wetland · Hydrus-1D · Nitrobenzene · Substrate

Introduction

Constructed wetlands (CW) are the water treatment schemes that are being used for many decades to apply the processes that happen in natural wetlands for the treatment of different polluted wastewaters (Aufdenkampe et al. 2001; Haberl et al. 2003; Vymazal 2005). They are designed to take benefit of these processes, but here they happen in a more controlled environment (Kadlec and Knight 1996; Vymazal 2005). Vertical flow constructed wetlands have become a

frequent treatment practice for urban wastewaters from the last decade. They are preferred in some situations, where other treatment systems cannot be applied easily (Paing et al. 2015). They require relatively small surface area and have high purifying efficiency for the treatment of organic matter (Wu et al. 2015). Substrate is the main component in the constructed wetlands. It not only support microbes and the plants growth but also serves significantly in removing pollutants in many ways like filtration, adsorption and sedimentation (Wang 2012). A suitable constructed wetland substrate plays a dynamic part in elevating the sewage treatment efficiency of the constructed wetland (Brix et al. 2001; Mengzhi et al. 2009).

Organic toxins accumulate in the natural environment and cause damage to health and the living organisms. Nitrobenzene (NB) do not exist naturally; it is only manufactured in industries for different purposes. Industrially, it is used in the production of aniline, some medications, dyes and rubbers (Wen et al. 2012) and also found in some polishes and paints as an ingredient (Lv et al. 2013; Pan and Guan 2010). Industrial effluents are often polluted with nitrobenzene, so they should be treated before final disposal into the

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waterbodies. Nitrobenzene is declared as a priority pollutant by many countries. Recent approaches for the treatment of NB in industrial discharges mostly comprise of ozonation, electrochemical reduction, Fenton oxidation and ultrasonic irradiation (Jiang et al. 2011; Subbaramaiah et al. 2014; Zhao et al. 2015). However, the high operating and maintenance costs and difficult handling characteristics of these approaches render their usage in economically concerned regions (Wang et al. 2010). The application of constructed wetlands (CWs) as a widespread, cost-effective and efficient substitute biological treatment technology has rapidly become significant worldwide for the treatment of wastewater (Brix et al. 2001; Vymazal 2005).

It is difficult to identify the nature of the processes that occur within the CW system; there are many multiple physical, chemical and biological processes taking place at the same time, so they are often called black boxes. The best way to understand the hidden processes is to use numerical models (Langergraber et al. 2009). Hydrus-1D is a program that uses finite element method to solve the Richard's equation and numerically solves advection–dispersion type equations for saturated–unsaturated water moment and solute transport, respectively. This software package can be used for various water flow and solutes transport boundary conditions, and it can use different soil hydraulic models. It is used in different studies to simulate solute transport and water moment and solute transport (Jiang et al. 2010; Li et al. 2015; Šimůnek et al. 2006). Many other researchers used Hydrus-1D model for their research like Ma et al. (2010) used it for water infiltration in a large layered soil column, Neumann et al. (2011) used it for the interpolation schemes for solute transport, Rizzo et al. (2014) used it to check the effects of different unsteady organic loads' response of laboratory horizontal flow constructed wetlands to unsteady organic loads, and Tan et al. (2015) used it for field analysis of water and nitrogen transport fate in lowland paddy fields.

Core objective of this study was to optimize the VFCW substrate by trying different compositions of sand, silt, clay and ferric powder to get the best composition to evaluate the NB removal capacity of media in all columns. Furthermore, the objective was also to evaluate the nitrobenzene transport in the subsurface flow using Hydrus-1D model and observe the effects of water content and hydraulic conductivity on each substrate and to ensure the maximum removal of nitrobenzene.

Materials and methods

Four laboratory-scale identical columns (diameter of 35 cm and 65 cm in length) were used to test the simulation tool under defined conditions. These columns were labeled as A, B, C and D. Soil samples for substrate were made having

different compositions of sand, silt, clay and ferric powder with varying bulk densities. All the vertical flow constructed wetland columns were filled with these substrate compositions in three layers as shown in Fig. 1. Layer 1 is 30 cm high, and layers 2 and 3 are both 15 cm high. Compositions of the substrate for all the columns are given in Table 1. The main filter layers mainly consist of the mixture of sand, silt, clay and ferric powder in different constitutions, while the 5-cm gravel layer below the filter substrate serves as the drainage system. Synthetic wastewater was supplied to all the wetland columns due to health and safety reasons and also for the comparison of all columns. Same sample of the synthetic wastewater was fed to all the columns so that the substrate efficiency can be determined. The synthetic sewage was prepared in the laboratory before feeding each column by mixing the following different constituents: 200 mg/L of chemical oxygen demand (COD), 40 mg/L of ammonium as N ($\text{NH}_4\text{-N}$), 5 mg/L of nitrate nitrogen ($\text{NO}_3\text{-N}$), 20 mg/L of nitrobenzene and 6.30 mg/L of total phosphorus (TP) and 20 mg/L of NB.

The experiment was set for 40 days, and observations were made every 5 days to feed the new wastewater to the columns and to collect the data from four different observation points in each column. This data collected was simulated in the Hydrus-1D for the removal and transport of

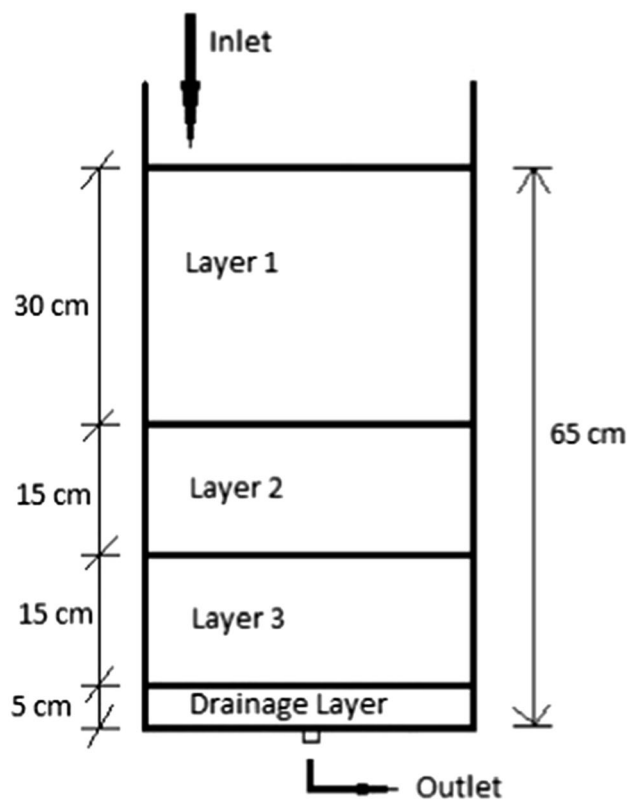


Fig. 1 Layout of the VFCW column



Table 1 Different compositions of substrates for all the columns

Column	Height (cm)	Soil texture	Sand %	Silt %	Clay %	Ferric powder ratio	Bulk density BD (g/cm ³)
A	30	Sand	92	6	2	20:01	1.39
	15	Loamy sand	80	12	8	–	1.45
	15	Sandy loam	74	16	10	20:03	1.47
B	30	Sandy loam	70	20	10	–	1.46
	15	Loam	45	40	15	–	1.43
	15	Loam	35	45	20	–	1.42
C	30	Sand	88	8	4	–	1.42
	15	Loamy sand	80	14	6	20:03	1.44
	15	Sandy loam	72	20	8	–	1.45
D	30	Sandy loam	64	23	13	20:01	1.47
	15	Loam	50	32	18	20:04	1.46
	15	Sandy loam	70	22	8	–	1.45

nitrobenzene. Water content, hydraulic conductivity (K) and the NB removal efficiency are discussed. The investigation about coupled water flow movement and solute transport was completed in Hydrus-1D for all laboratory-scale constructed wetland columns with soils of different physical properties and composition. For this purpose, van Genuchten-Mualem soil hydraulic model was selected with no hysteresis.

Nitrobenzene degradation

From different physical, chemical and biological processes for the treatment of nitrobenzene-polluted wastewater, the biological treatment is the best and cost-effective option (Lv et al. 2013). In anaerobic conditions, sewage effluent transforms nitrobenzene to aniline but under aerobic condition no aromatic amine was detected. The degradation of NB could happen in oxidative and reductive paths simultaneously; NB could be reduced to aniline in anaerobic conditions and then followed by aerobic process; it could further be degraded and mineralized. NB degrades by the cleavage of its ring in aerobic conditions, and ammonia is released. Then, the altered compounds could fully be mineralized to CO₂ and H₂O by the microbial activities. In the wetland substrate bed, there exist a huge number of definite micro- and macro-gradients of redox circumstances that assist the growth of highly varied microbial groups capable of various redox reactions. These processes are described thoroughly in Fig. 2.

In our experiment, NB was used as a contaminant in the synthetic wastewater to treat it using wetland technology. NB

itself is not soluble in the water; it was mixed with different chemicals to get synthetic wastewater which was fed to the substrate soil. About above 90% of the NB was removed in average from all the columns. From that 90%, some amount was volatilized and the rest of it was removed from the wastewater and absorbed by the substrate of the column in different layers. Because the NB was absorbed from the different soil layers across the column at different nodes, it was difficult to know the exact amount of NB volatilized and the amount of NB absorbed, but the total amount of the NB removal was determined.

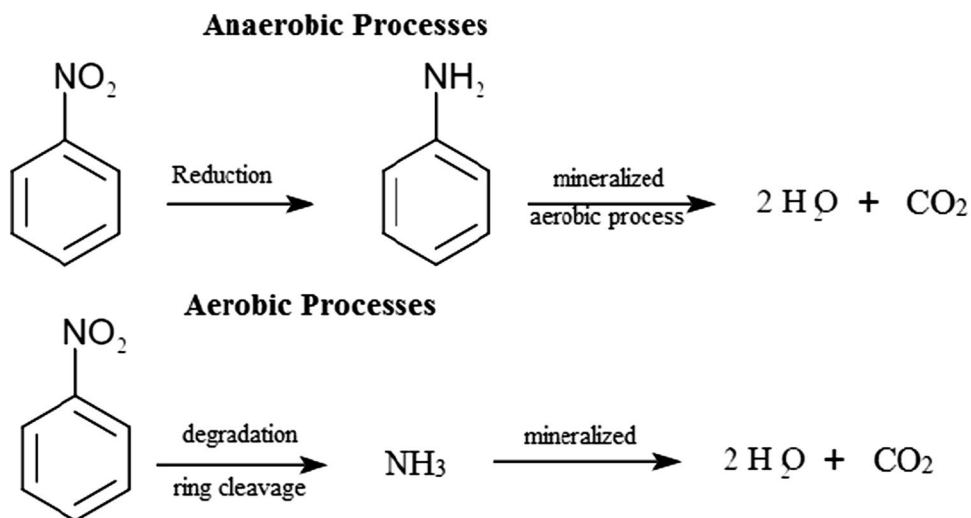
Hydrus-1D

Hydrus-1D uses finite element method to resolve the Richard's equation (Eq. 1) that is used for simulating the one-dimensional water flow movement in variably saturated medium.

$$\frac{\partial \theta}{\partial t} = \frac{\partial}{\partial z} \left[K(\theta) \left(\frac{\partial h}{\partial z} - \cos \alpha \right) \right] - S \quad (1)$$

where “ θ ” is the volumetric water content [L^3L^{-3}], “ t ” is the time [T], z is the spatial longitudinal coordinate axis [L] (positive in upward direction), “ h ” is described as the water pressure head [L], “ α ” is the inclination angle between the vertical axis and the flow direction of water flow (i.e., for the vertical flow, $\alpha = 0$, and when flow direction is horizontal, $\alpha = 90$, and $0 < \alpha < 90$ for inclined water flow), “ S ” represents the sink term [$L^3L^{-3}T^{-1}$], and K is represents the unsaturated hydraulic conductivity function [LT^{-1}] that is further explained by the following relationship (Simunek et al. 2005).

Fig. 2 Common degradation process of NB



$$K(h, z) = K_s(z) \cdot K_r(h, z) \quad (2)$$

where K_r and K_s are the relative and the saturated hydraulic conductivities, respectively [LT^{-1}].

Soil hydraulic properties include residual and saturated water contents, saturated hydraulic conductivity and empirical coefficients (α and n); all these parameters for all layers of soils were predicted in Hydrus-1D that uses Rosetta Dynamically Linked Library (DLL) for this purpose. Soil hydraulic properties for all the soil layers of four columns are given in Table 2.

Hydrus-1D uses advection–dispersion type and diffusion type equations for the solute transport in the liquid and gaseous phases, respectively. Convection type equations describe the movement of dissolved components with the flowing water, and dispersive transport is due to the differential

velocities of water flow at the pore scale. Transport of non-adsorbent and inert solute in one-dimensional vertically saturated medium is described as follows:

$$\frac{\partial(\theta C)}{\partial t} = \frac{\partial}{\partial z} \left(\theta D \frac{\partial C}{\partial z} \right) - \frac{\partial(v\theta C)}{\partial z} \quad (3)$$

where “ C ” represents the solute concentration of the solute, t is the time, θ is the volumetric water content, D is the hydrodynamic dispersion distribution coefficient, z is the positive depth in downward, and v is the average pore water velocity.

For simulation purposes, concerns in Hydrus-1D, for equilibrium conditions of solute transport model, Crank–Nicholson conditions were selected as time weight scheme and Galerkin finite elements were selected as space weight scheme. For physical and chemical

Table 2 Soil hydraulic properties for all the columns

Column	Q_r (cm ³ /cm ³)	Q_s (cm ³ /cm ³)	Alpha (α) (1/cm)	N	K_s (cm/day)	I
A	0.049	0.418	0.037	2.848	636.62	0.5
	0.047	0.404	0.036	1.744	125.17	0.5
	0.046	0.397	0.034	1.558	71.53	0.5
B	0.044	0.39	0.032	1.496	59.81	0.5
	0.051	0.380	0.011	1.505	17.88	0.5
	0.062	0.396	0.008	1.559	12.7	0.5
C	0.048	0.411	0.037	2.339	349.31	0.5
	0.044	0.402	0.039	1.761	140.68	0.5
	0.042	0.39	0.036	1.528	74.67	0.5
D	0.048	0.392	0.026	1.445	38.71	0.5
	0.056	0.392	0.015	1.454	17.39	0.5
	0.041	0.392	0.034	1.499	67.57	0.5

Where Q_r and Q_s are residual water content and saturated water contents, respectively, alpha (α) and n are the parameter in the soil water retention functions, K_s is the saturated hydraulic conductivity, and “ I ” is the tortuosity parameter in the conductivity function

non-equilibrium solute transport, dual porosity model with two site sorption in the mobile zone was selected as solute transport model which represents the physical and chemical non-equilibrium. “Concentration” boundary condition and “zero concentration gradient” were considered as upper boundary conditions and lower boundary conditions, respectively.

Solute transport parameters include bulk density, longitudinal dispersivity (Disp.), dimensionless fraction of adsorption sites and immobile water content which depend on the solute transport models. Set equals to zero when physical non-equilibrium was not considered. Some solute-specific parameters for solute were also needed like molecular diffusion coefficient in free water (Diffus. W.) and molecular diffusion coefficient in soil–air (Diffus. G.). The values for longitudinal dispersivity, Diffus. W. and Diffus. G. were taken from the literature (Huang et al. 2015; Schulze-Makuch 2005).

Dispersivity was calculated using the following general equation given by Schulze-Makuch (2005)

$$\text{Disp.} = c(L)^m \quad (4)$$

where “ c ” is a parameter characteristic specific for a given geological medium, “ m ” is the scaling exponent factor and “ L ” is the flow distance for unconsolidated media (e.g., a

sandy aquifer). Here c and m were identified by Schulze-Makuch (2005).

$$\text{Disp.} = c(L)^m = 0.20(L)^{0.44} \quad (5)$$

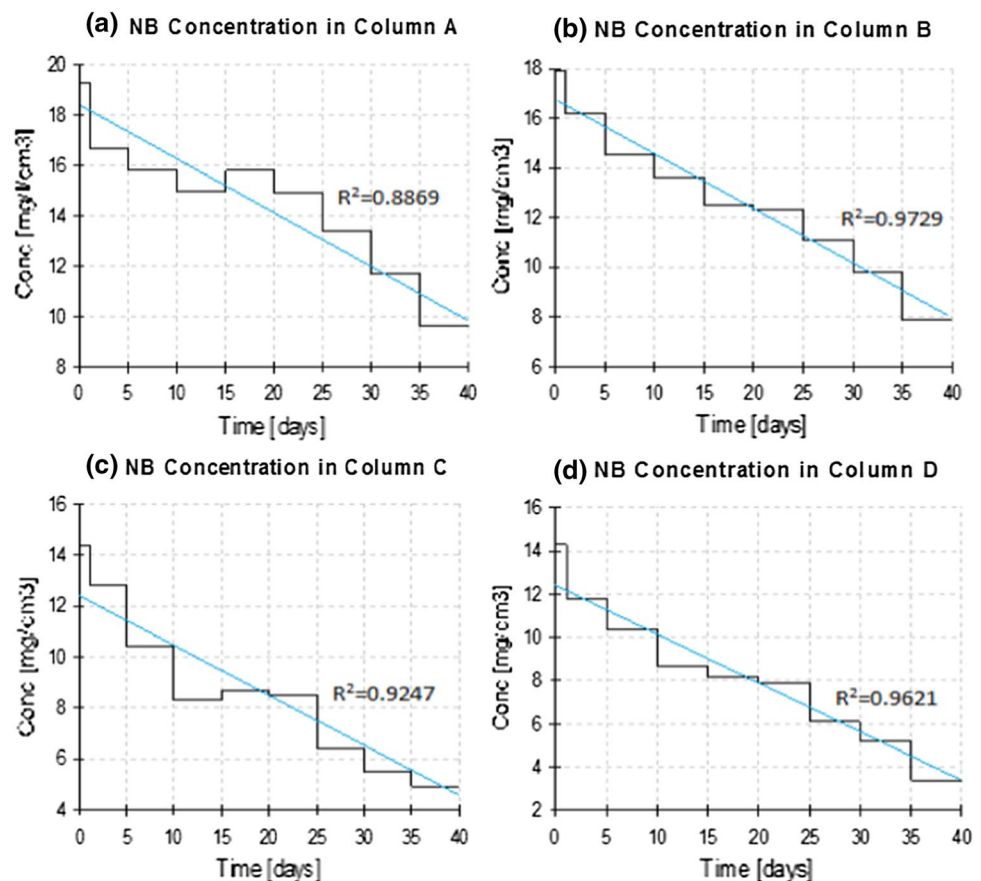
The molecular diffusion coefficient of nitrobenzene in water at room temperature and pressure was $1.0333 \text{ cm}^2/\text{day}$ given by Huang et al. (2015), and its molecular diffusion coefficient in free air was $6556.4 \text{ cm}^2/\text{day}$ (Van der Perk 2013).

Results and discussion

Nitrobenzene removal was considered as solute transport for all the soil composition samples in all the wetland columns. Hydraulic conductivity and water content were also investigated for every column. Results for these parameters are discussed here in detail.

Trend of NB removal in all the columns is shown in Fig. 3. Figure 3a–d represents the NB removal in the columns A, B, C and D, respectively. In the column A, initial concentration of NB was 19.3 mg/L and the final concentration was 6.9 mg/L , so the removal efficiency was 50.26% with R^2 value of 0.8869 . Column A was the least efficient column because most of the substrate composition was only sand although the ferric powder was also used in the first

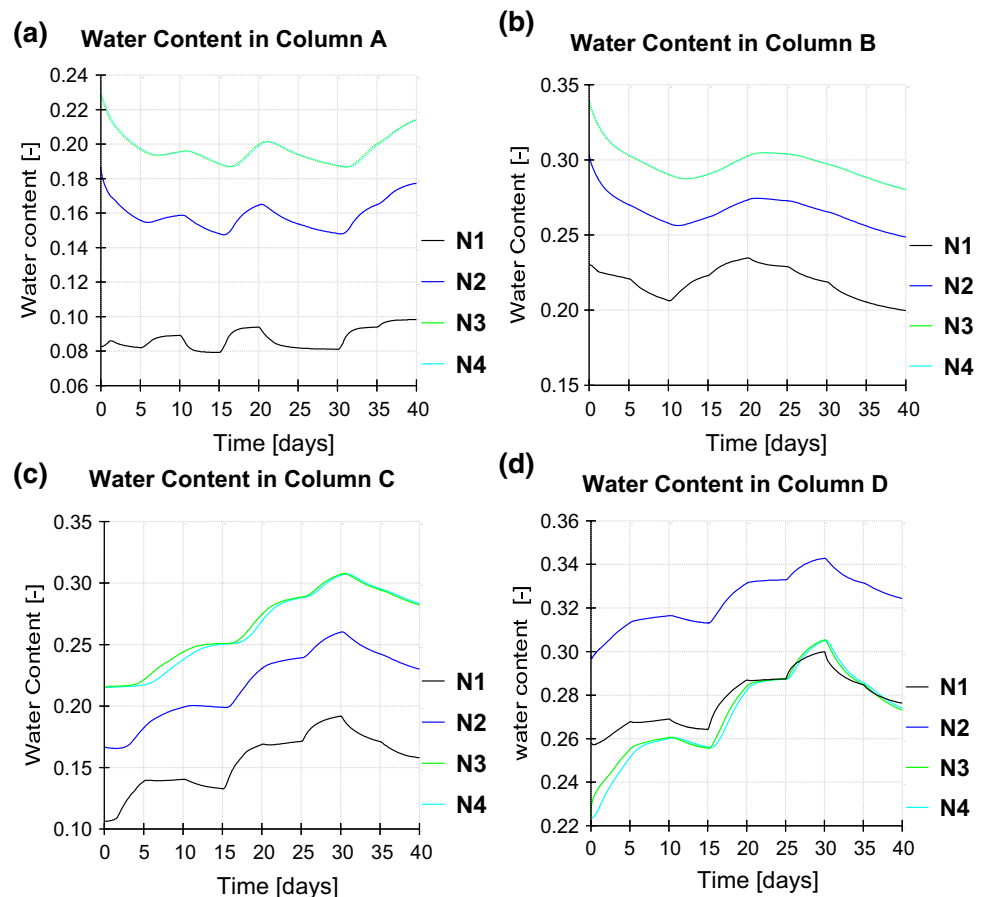
Fig. 3 Nitrobenzene concentration in all the columns A, B, C and D



30-cm layer and the third 15-cm layer of the soil, but its ratio was only 20:1 and 20:3, respectively. Compared with the column A, column B has better results for the NB removal. Column B consists of sandy loam and loamy soils that have relatively more constitution of silt and clay. On the first day, the NB concentration was 17.9 mg/L, and at the end of final time it was 7.9 mg/L having with the removal efficiency of 55.86% and R^2 value of 0.9729. Trend of removal was consistent because the substrate used in this column was a mixture of only sand, silt and clay. Column C gave better results as compared to A and B with the removal efficiency of 65.97%. The initial concentration of NB was 14.4 mg/L and the final concentration was 4.9 mg/L with R^2 value of 0.9247 that represents the irregular trend because of the composition of the substrate. Most of the composition was sand with lesser amount of both silt and clay and the ferric powder compositions of 20:1, 20:3 and 20:5 in the first, second and third layers, respectively. Figure 3d represents the trend of NB removal in column D with the initial concentration of

14.3 mg/L and the final concentration of 3.4 mg/L and R^2 value of 0.9621; the trend was rather smoother as compared with column C. Column D was the most efficient column with the removal efficiency of 76.22%. The substrate composition was sandy loam and loam with more of the silt and clay as compared to all other columns. Its second layer also contained ferric powder mixed with the mixture of sand, silt and clay in 20:4 ratio. In columns A, B and C, the ratio of silt to clay was less as compared to column D. It was well recognized that the downward descending movement of solutes in finer soils was slower as compared to the coarse soils because fine soils have the lower values of hydraulic conductivities. Therefore, the extent of solute leaching percolating downward in the column A, column B and column C was less than that in column D. Similar results have been exhibited by Jiang et al. (2010) and Tan et al. for the transport of bromide (Br) and nitrogen transport, respectively (Jiang et al. 2010; Tan et al. 2015).

Fig. 4 Water content for all columns with respect to time

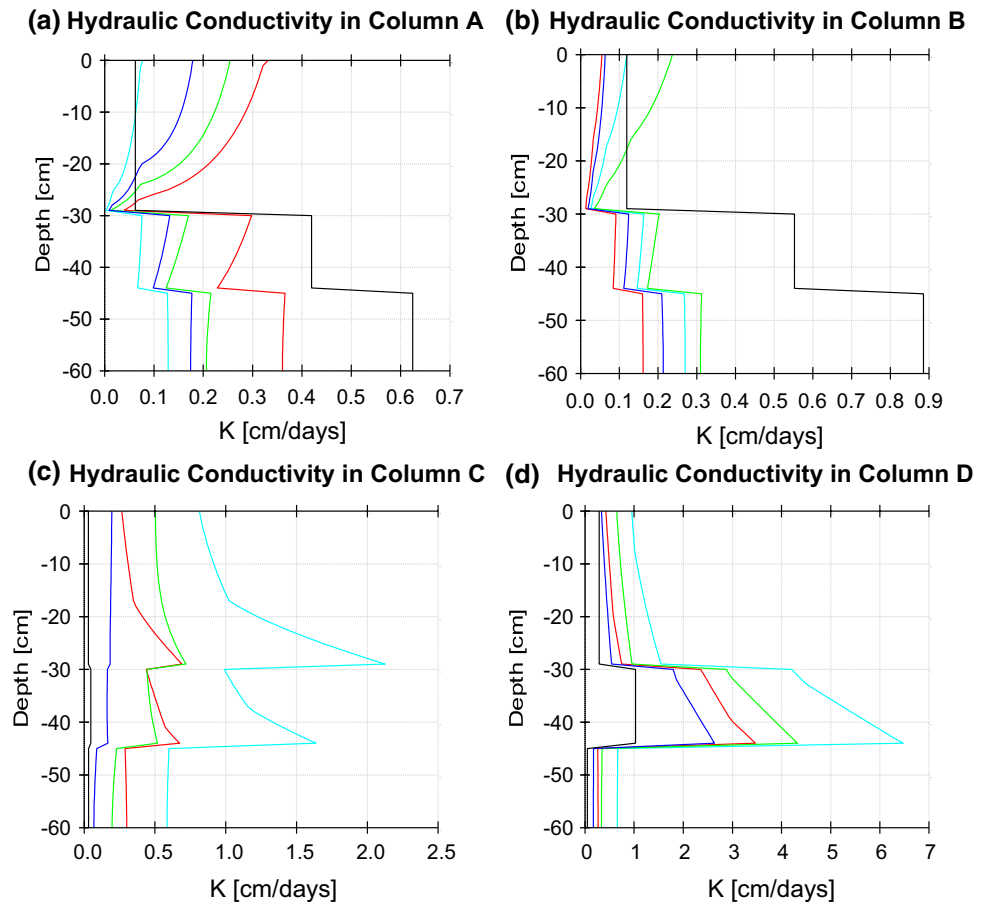


Water content is an important parameter in the constructed wetlands. Figure 4 represents the soil volumetric water content of all the columns obtained from Hydrus-1D. Figure 4a–d represents the soil water content for column A, column B, column C and column D, respectively. The results are described with time on *x*-axis and water content on *y*-axis. Figure 4a represents the water content in column A, which exhibits just small variation in the water content of the soil layers, and the trend was almost the same for all the layers because the water inflow was the same for all the layers, and the small variation was just because of the different substrate compositions. Figure 4b also shows the similar behavior for column B, but it shows the decreasing trend of water content with time. The similarity in the results was due to the fact that same effluent was applied to all the layers that exhibit the same flow conditions and water content shows nearly identical trend for all the observation points. The fluctuations in the lines can be better explained on the basis of different substrate compositions.

The behavior of column C as shown in Fig. 4c displays the increasing trend of water content and also shows the increasing values with respect to every observation points. In the first observation point, its initial value was 0.11 and the final value was 0.16; for the second point the initial value was 0.17 and the final was 0.23; third and fourth points show similar values for water content, and the lines are almost merged into each other and it is because these points lie on the same soil layer. The column D represents the same behavior as column C, but the last two points show relatively decreased water content with respect to the first two points although it is increasing from the initial to the final time.

Figure 4d shows the water content in the column D where initial value of the first line is 0.258 and the final value is 0.278; the second line starts from 0.298 and ends at 0.327. Third and the fourth lines show very small difference because of the same soil layer: Third line stars at 0.233 and ends at 0.273 and the fourth line starts at 0.225 and ends very near to the ending point of line three that is 0.275.

Fig. 5 Hydraulic conductivity for all the columns A, B, C and D



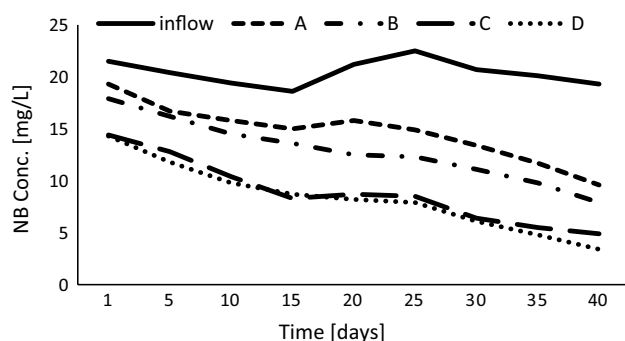


Fig. 6 Comparison of NB removal in all the VFCW columns

These results obtained from Hydrus-1D show very good representation of the water contents in the soil columns. They help to understand the nature of the soil layers for the water moment in the substrate. Similar results for water content have been represented by Erfani Agah and Wyseure (2013), Jiang et al. (2010) and Tan et al. (2015).

Hydraulic conductivity is different for every soil layer in each column. Hydraulic conductivity is taken with respect to depth that demonstrates its variation in the different layers. Figure 5 shows the hydraulic conductivity of all the experimental columns where Fig. 5a represents the hydraulic conductivity in column A, whereas Fig. 5b–d represents the hydraulic conductivity in column B, column C and column D, respectively. Five different lines show its value at five different points in the soil column. First black line shows the hydraulic conductivity at the initial point while blue, green, cyan and red lines represent the hydraulic conductivity at 15, 30, 45 and 60 heights from the surface, respectively.

For column A as shown in Fig. 5a, hydraulic conductivity significantly decreases in the first layer like in the last node its initial value was 0.33 cm/d and at the ending of this layer its value was 0.05 cm/d; in the second layer, the decreasing trend was lesser than the first layer. At the depth of 30 cm and 45 cm, there was an abrupt change in the values that was because of the change in the soil layers. In the third 15-cm layer, the hydraulic conductivity become almost constant till end that was because of the addition of ferric powder and the increased value of silt and clay in the soil composition. In the column B as represented in Fig. 5b, the trend of hydraulic conductivity was almost similar with the column A, but here its value was lesser than that in column A. In the third column represented in Fig. 5c, the third cyan line is much different

from the others because the third layer had more amount of soil and less composition of silt and clay and it also contained the varying amounts of ferric powder. Figure 5d shows the hydraulic conductivity trend for column D, and it represents the normal behavior for the first and the third layers, but there was an abrupt increase in the hydraulic conductivity values for the second layer.

Correlation between NB removal results of all the columns and the effluent concentration was made that represent the linear relationship among them. The most common correlation coefficient, the Pearson product-moment correlation coefficient, and the most widely recognized correlation were utilized used in this study. Figure 6 can be used to compare the NB removal mechanism in all the columns which indicates that column D is the column that gives best results with R^2 value of 0.9854. Column C also gives the satisfactory results and is very likely to have a similar trend with the column D with R^2 value of 0.9646; columns A and column B both have similar trends with R^2 values of 0.9759 and 0.9820, respectively, and the NB removal is more in column B as compared to A; all these results are due to the different substrate compositions. Similar results have been exhibited by Erfani Agah and Wyseure (2013), Jiang et al. (2010) and Tan et al. (2015) for the transport of bromide (Br) and nitrogen, respectively.

Conclusion

In this work, different compositions of the substrate were used in four vertical flow constructed wetland columns to treat nitrobenzene from the laboratory-synthesized wastewater. It was concluded that the substrate compositions for all the columns showed different potential for the removal of nitrobenzene although the column D was demonstrated to have maximum removal efficiency of 76% while column A, B and C were having 50.25%, 55.86% and 65.97% removal efficiencies, respectively. A paired-samples *t* test was performed to determine the effect of variations between all the columns. It was performed as column D with columns A, B and C, and the results were at significant level of 0.05, evaluating the best efficiency of column D. The results showed that the removal of nitrobenzene depends on the substrate composition in the vertical flow constructed wetlands. It was evident that different substrate compositions may affect the removal



capability of the constructed wetland. Thus, constructed wetland technology was accessed as the best and economical wastewater treatment option.

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