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Kinetic modelling and performance evaluation of vertical subsurface flow constructed wetlands in tropics

G.M.P.R. Weerakoon[a], K.B.S.N. Jinadasa[a], Jagath Manatunge[b], Buddhi Wijesiri[c,d,*], Ashantha Goonetilleke[c,d]

[a] Department of Civil Engineering, University of Peradeniya, Peradeniya, 20400, Sri Lanka
[b] Department of Civil Engineering, University of Moratuwa, Moratuwa, 10400, Sri Lanka
[c] Science and Engineering Faculty, Queensland University of Technology (QUT), GPO Box 2434, Brisbane, QLD, 4001, Australia
[d] College of Chemistry and Environmental Engineering, Shenzhen University, Shenzhen, 518060, China

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ABSTRACT

The design of vertical subsurface flow (VSSF) constructed wetlands (CWs) uses kinetic models to calculate the area based on the kinetic reaction rate constant (k) specific to local environmental conditions and target pollutants. Currently, kinetic modelling does not fully account for the impact of the hydraulic loading rate (HLR), which influences the wetland performance. This study used four experimental VSSF CWs operated at HLRs of 5, 10, 20 and 40 cm/day to investigate the applicability of three first order kinetic models combining plug-flow and continuous stirred tank reactor (CSTR) flow patterns. The target pollutants were BOD_5, NH_4^+ and NO_3^-. For each pollutant, estimated k values varied between different HLRs and between plug flow and CSTR models. Assessment of uncertainty in kinetic modelling showed that all three models exhibit a similar trend in predicting the concentrations of BOD_5 and NH_4^+ at 5–20 cm/day HLRs. A substantial removal of BOD_5 (> 88 %) and NH_4^+ (> 70 %) were found for the investigated HLRs, although NO_3- removal was not satisfactory. The HLR had a positive impact on mass removal rates (MRRs) for BOD_5 and NH_4^+. Accordingly, 20 cm/day was deemed as the highest viable HLR for designing effective VSSF wetlands for the removal of BOD_5 and NH_4^+. All three models can be employed to design VSSF wetlands at 20 cm/day HLR to treat BOD_5 using k values of 0.352 (k-C), 0.380 (k-C*) and 0.996 (CSTR) m/day and to treat NH_4^+ using k values of 0.170 (k-C), 0.173 (k-C*) and 0.273 (CSTR) m/day.

1. Introduction

Constructed wetlands (CWs) are a cost effective, robust and environmentally appropriate treatment technology compared to conventional wastewater treatment facilities [1,2]. These systems are engineered in the form of free water surface (FWS), horizontal subsurface flow (HSSF) and vertical subsurface flow (VSSF) within a controlled environment, to mimic the processes of natural wetlands [3–5]. The VSSF wetlands are popular due to their efficiency in treating a wide range of wastewater [6] utilising a relatively small land area compared to FWS and HSSF wetlands. In VSSF wetlands, the intermittent feeding mode provides a greater oxygen transfer potential, resulting in more effective removal of organics [measured as five-day biochemical oxygen demand (BOD_5) and chemical oxygen demand (COD)], suspended solids and ammonium (NH_4^+) [7–9].

The pollutant removal processes in CWs involve a combination of complex physical, chemical and biological processes such as sedimentation, filtration, adsorption, precipitation, volatilisation, biodegradation, nitrification, denitrification and microbial and plant assimilation [10–14]. Generally, these processes occur simultaneously, and microbial degradation, plant uptake and adsorption are the major mechanisms which remove or transform nutrients and organic pollutants from wastewater [15]. However, the rates of removal reactions within a wetland system are related to local climatic conditions, wetland type and its design including hydraulic properties such as hydraulic retention time (HRT) and hydraulic loading rate (HLR) and influent pollutant concentrations, types of vegetation present, type of substrate media and microbial communities [15–17]. The HLR (i.e. the volumetric flow rate divided by the wetland surface area) and HRT (i.e. the ratio of useable wetland water volume to the average flow rate) are the two crucial

* Corresponding author at: Science and Engineering Faculty, Queensland University of Technology (QUT), GPO Box 2434, Brisbane, QLD, 4001, Australia.
E-mail addresses: prabhaw@pdn.ac.lk (G.M.P.R. Weerakoon), shamj@pdn.ac.lk (K.B.S.N. Jinadasa), manatunge@uom.lk (J. Manatunge), b.mahappukankanamalage@qut.edu.au (B. Wijesiri), a.goonetilleke@qut.edu.au (A. Goonetilleke).

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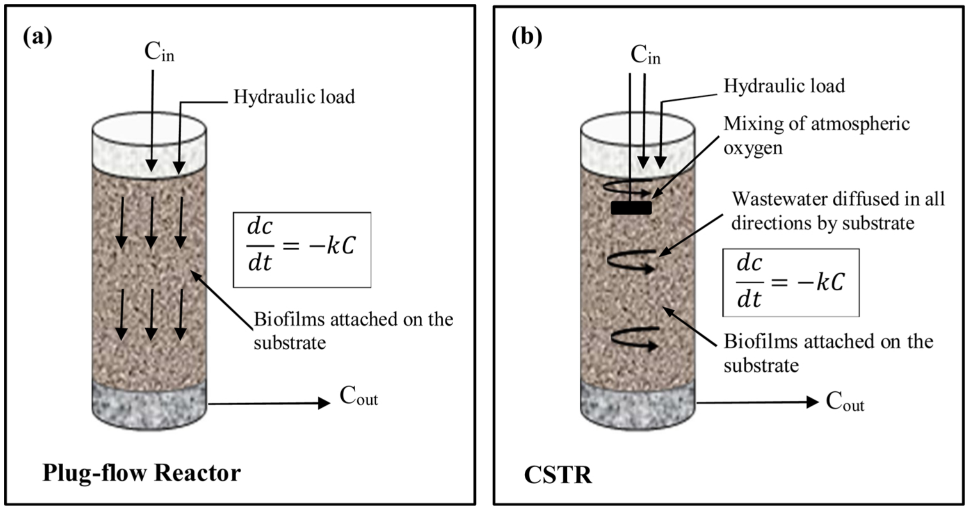

Fig. 1. Kinetic modelling approaches for vertical subsurface flow constructed wetlands: (a) combination of first order kinetics with plug-flow; (b) combination of first order kinetics with continuous stirred tank reactor (CSTR) flow. Note: C_{in} – influent pollutant concentration and C_{out} – effluent pollutant concentration.

parameters that influence the performance of CWs [18].

Generally, HLR and HRT are correlated; hence for a given HLR, HRT can be adjusted by the active depth of the wetland cell [14]. Past studies have revealed that lower HLRs and longer HRTs result in better removal of pollutants [15,19,20]. Chang et al. [19] reported that percentage removal of NH_4^+, COD and BOD_5 show a negative response to an increase in HLR from 20 to 120 cm/day. At higher HLRs, CWs require larger land areas for wastewater treatment [21]. According to Metcalf and Eddy [22], efficient removal of pollutants occurs within 4–15 days of HRT with corresponding nominal HLR range of 1.3–5 cm/day. Therefore, it is important to investigate the feasibility of wetland usage with high HLRs to achieve possible land area reductions and to optimise treatment performance.

However, due to the complex pollutant removal processes and the unpredictability of the changes in the local environmental conditions, the design of CW systems has been based mainly on experience and rule-of-thumb without accepted standard procedures [14]. Several modelling approaches have been employed considering the relationships between treatment efficiency and influential factors in order to replicate the performance of CWs [16,23]. The primary empirical design approach can be identified as the first order kinetic model (k–C model/Kickuth Equation), integrated with plug-flow assumptions [24–26]. This model simplifies the CW system to a 'black box', accounting for the exponential reduction of pollutants from inlet to outlet. However, the k–C model does not consider the interactions between soil, plants, water and microorganisms [27]. Further, k–C model fails to account for the influence of environmental factors (e.g. precipitation and evapotranspiration) which may produce a secondary hydraulic regime within the wetland cell, invalidating steady-state theoretical models [25,28].

Therefore, more sophisticated models such as plug-flow dispersion (PFD) model and tanks in series (TIS) model have been developed, which simulate non-ideal hydraulic conditions describing the detention time distribution in a wetland [3]. However, the use of such models is limited due to the complexity and extensive data requirements. Therefore, first order kinetic models and simple regression approaches are commonly used to investigate the performance of CWs. The k–C model has been modified by Kadlec and Knight [29] to form the k–C* model, which considers pollutant degradation allowing a non-zero background concentration (C*) to represent the remaining effluent pollutant concentrations after reaching a plateau towards the end of the treatment process [10,30,31]. The k–C* model has been widely used for the design of CWs in recent years [25,32].

In the case of VSSF wetlands, the assumption of ideal plug-flow pattern may not be appropriate with the feeding mode of wastewater, and likely to deviate through the unsaturated packed media [33]. Therefore, continuous stirred tank reactor (CSTR) behaviour was considered in earlier studies [34]. In fact, the flow regime in subsurface flow wetlands varies between plug-flow and CSTR flow patterns [35].

Modelling CWs is difficult due to the intrinsic complexity in biological wastewater treatment and associated environmental processes which are subject to inherent variability [36,37]. As such, various simplifying assumptions underpin kinetic models, which may lead to uncertainty arising from measured data, model parameters and model structure [38]. In fact, parameters of CW models are considered to be constants, limiting the variability associated with these parameters being taken into account [39,40]. This could influence the accuracy of model predictions. Therefore, assessing uncertainty in model predictions is necessary to inform accurate interpretation of modelling outcomes, and thereby to improve the design of CWs [37,41].

The research study discussed in this paper evaluated three empirical models for their applicability in the design of VSSF CWs. The study used experimental data from VSSF CWs operated at four different HLRs in tropical climatic conditions, and employed a robust methodology for assessing modelling uncertainty. The research outcomes are expected to contribute to the design of more efficient VSSF CWs suitable for tropical areas.

2. Materials and methods

2.1. Kinetic modelling

Three kinetic models, combining first order biological degradation kinetics with plug-flow and CSTR flow patterns were used to relate concentrations of BOD_5, NH_4^+ and NO_3^- in the influent and effluent in a VSSF wetland system. Fig. 1 illustrates these modelling approaches for a separate VSSF wetland bed (modified from [42]).

Model 1: First order kinetics with plug-flow assumption (k–C model)

This model considers the exponential degradation of pollutants from the inlet to the outlet assuming idealized plug-flow conditions, correlating the influent and the effluent pollutant concentrations as expressed by Eqs. (1) and (2).

$$\frac{C_{out}}{C_{in}} = exp^{-\left(\frac{k_1}{HLR}\right)} \tag{1}$$

$$k_1 = \frac{\ln C_{in} - \ln C_{out}}{1/_{HLR}} \quad (2)$$

Where, C_{in}, C_{out}, HLR and k_1 represents influent pollutant concentration (mg/L), effluent pollutant concentration (mg/L), hydraulic loading rate (m/day) and first order area-based removal rate constant (m/day) for Model 1, respectively.

Model 2: Modified first order kinetics with plug-flow assumption (k–C* model)

The k–C model has been modified by Kadlec [29], assuming exponential removal of pollutants to include non-zero background wetland concentrations (C*) as shown in Eqs. (3) and (4). C* is an irreducible effluent concentration that results from internal biogeochemical cycling and life cycle of biota, which produce some residual material within wetland cells and can be measured as BOD, TSS (total suspended solids), nitrogen, phosphorus and fecal coliforms. There will always be some residual background concentrations of these pollutants regardless of the size of the wetland or the characteristics of the influent wastewater. Thus, C* effectively sets a lower limit to the effluent concentration of a treatment wetland [43,44].

$$\frac{C_{out} - C^*}{C_{in} - C^*} = e^{-\left(\frac{k_2}{HLR}\right)} \quad (3)$$

$$k_2 = \frac{ln(C_{in} - C^*) - ln(C_{out} - C^*)}{1/_{HLR}} \quad (4)$$

Where, C* and k_2 represents irreducible background concentration (mg/L) and first order area based removal rate constant (m/day) for Model 2, respectively.

Model 3: First order kinetics with CSTR flow assumption

This model was developed considering the CSTR flow behaviour in CWs. The first order kinetics in a wetland reactor is expressed by Eq. (5) and CSTR flow pattern in the reactor can be expressed by Eq. (6).

$$\frac{dC}{Dt} = -k_v C_{out} \quad (5)$$

$$\frac{dC}{Dt} + \frac{1}{\tau}C_{in} = \frac{1}{\tau}C_{out} \quad (6)$$

Where, C, k_v and τ represents pollutant concentration (mg/L), volumetric reaction rate constant (per day) and hydraulic retention time (days).

The combination of Eqs. (5) and (6) gives a simplified first order kinetics combined with CSTR flow pattern in terms of first order area-based removal rate constant (k_3, m/day) as expressed by Eq. (7), correlating concentrations in the influent and effluent [27, 34, 46].

$$k_3 = \frac{HLR(C_{in} - C_{out})}{C_{out}} \quad (7)$$

2.2. Experimental wetland setup

The data for BOD_5, NH_4^+ and NO_3^- was collected from four experimental VSSF wetland units of 1.4 m × 0.5 m × 0.6 m (length × width × height) operated at four different HLRs of 5, 10, 20, and 40 cm/day. The experimental setup (Fig. 2) was maintained in the open air premises at the University of Peradeniya, Sri Lanka (80° 35′ 59″ E, 7° 16′ 00″ N). The mean temperature, average relative humidity and the average annual rainfall in the region were 24.6 °C, 84 % and 2132 mm, respectively.

All wetland units were prepared as presented in Weerakoon et al., [21], using 10–20 mm gravel as the wetland media and 30–50 mm gravel for the drain field. Eight rhizomes (each approximately 30 cm high), of *Typha angustifolia* (narrow leaf cattail) were planted in each wetland unit. Soon after planting, the wetland beds were kept wet using tap water for four weeks to facilitate plant growth. Thereafter, the units were fed with tap water for another two weeks at a nominal rate to flush-out and remove pollutants from the system. Then, the synthetic wastewater was applied.

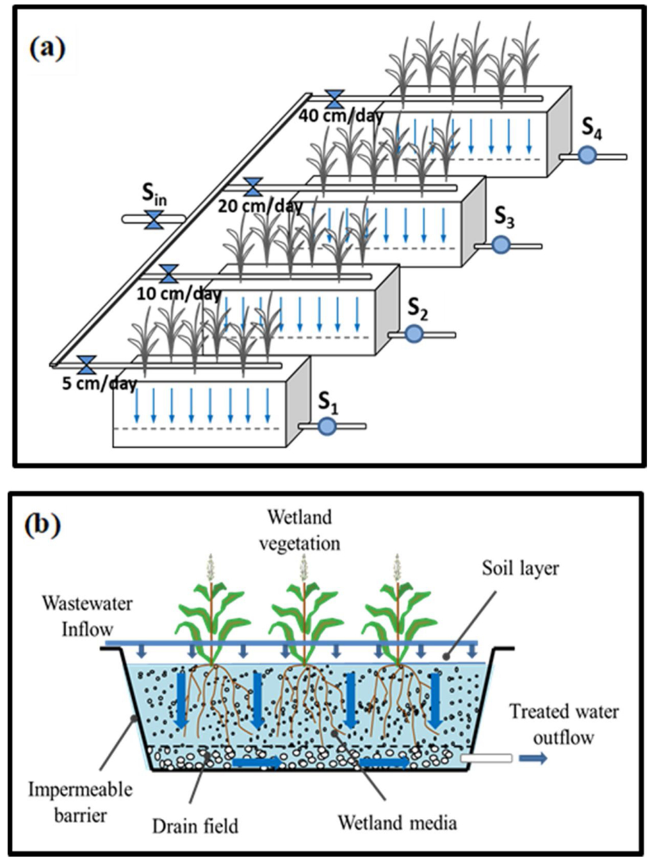

Fig. 2. (a) Experimental wetland setup (S_{in}, S_1, S_2, S_3 and S_4: Sample points), (b) Schematic diagram of a vertical susbsurace flow (VSSF) wetland unit.

2.3. Synthetic wastewater

The study investigated the applicability of VSSF wetlands for secondary or tertiary treatment of domestic wastewater. As such, synthetic wastewater similar to septic tank effluent was prepared, since actual wastewater of septic tank effluent quality fluctuate over time. The compounds used in the synthetic wastewater included, urea (30 g), granular sugar (75 g), $(NH_4)_2SO_4$ (10 g), $MgSO_4$ (10 g), $FeCl_3$ (0.25 g), $MnSO_4.H_2O$ (1 g), phosphate buffer solution (250 mL) and septage sludge (500 mL) in 500 L of tap water [21]. The composition of phosphate buffer solution had KH_2PO_4 (8.5 g), K_2HPO_4 (21.75 g), $Na_2HPO_4.7H_2O$ (33.4 g) and NH_4Cl (1.7 g) in 1 L of tap water [46]. Septage sludge was collected from municipal gulley bowsers and kept below 4 °C until used. The flow rates relating to each HLR was determined considering the surface area of the wetland unit. The synthetic wastewater was applied to the four different VSSF wetland units to achieve the desired HLRs of 5, 10, 20 and 40 cm/day in each wetland unit as shown in Fig. 2(a), through a constant head arrangement. The flow rates related to each HLR was determined considering the surface area of the wetland unit. A valve arrangement was used to control the wastewater flow into each wetland unit and the inflows were monitored frequently to minimize variations.

2.4. Sample collection and laboratory testing

Samples were collected from influent (S_{in}) and effluent (S_1, S_2, S_3, S_4) from each wetland unit (Fig. 2(a)) into 500 mL pre-cleaned PET (Polyethylene terephthalate) bottles at two-week intervals for water quality testing, after an acclimatizing period of two weeks. BOD_5, NH_4^+ and NO_3^- concentrations were determined in accordance with the Standard Methods for Examination of Water and Wastewater [47]. The volumes of influent and effluent wastewater flow in relation to each VSSF unit were measured volumetrically at 2–3 day intervals in order to obtain the daily mean average discharge.

2.5. Data analysis

2.5.1. Estimation of kinetic reaction rate constant and uncertainty assessment

Kinetic reaction rate constants (k) for BOD_5, NH_4^+ and NO_3^- in relation to the three models at each HLR were estimated employing non-linear regression using MATLAB in-built function *nlinfit* [48]. The function *nlinfit* uses 'non-linear least squares regression' as the optimization technique. Given the estimated k, the variations in the concentrations in the effluent against the concentrations in the influent in each water quality parameter could be predicted.

In the prediction of concentrations in the effluent, an error term was specified for the *nlinfit* function in the form of proportional error model ($y = f + \theta f\varepsilon$, where function value 'f' and error parameter 'θ' with initial estimate set to default value (= 1) are independent components; $\varepsilon \sim N(0, 1)$). This accounted for the variance of 'y' in the form of $var[y \mid k, C_{in}] = \theta^2 f(C_{in}, k)^2$ given the estimated 'k' and C_{in}, and standard error of the estimated 'k' given by the observed Fisher Information Matrix [48], enabling the quantification of uncertainty associated with the predictions by each kinetic model. As such, a large number of mathematical simulations of the predicted concentrations in the effluent of each water quality parameter were undertaken by accounting for residual errors and parameter (k) estimation errors for each model, enabling the quantification of uncertainty limits at 95 % interval. The upper and lower uncertainty limits would define the range at which the predicted values vary according to the accounted errors.

2.5.2. Comparison of model performance

The accuracy of estimated k values for each water quality parameter using each model was compared using Normalized Objective Function (NOF) and Model Efficiency (ME) [5].

The NOF can be computed using Eqs. (8) and (9). The NOF value, ranging from 0 – ∞, measures the differences between predicted and the measured values. The ideal value of NOF is 0.0. However, a validated model is acceptable to test alternatives for NOF values ranging from 0.0 – 1.0.

$$Root\ Mean\ Squre\ Error\ (RMSE) = \sqrt{\sum_{i=1}^{N} \frac{(P_i - M_i)^2}{N}} \tag{8}$$

$$NOF = \frac{RMSE}{M_{mean}} \tag{9}$$

Where, M_{mean} is the mean of the measured values and N is the total number of measurements.

The ME is a normalized statistic that determines the relative magnitude of the residual variance compared to the variance of the measured data and is computed based on Eq. (10).

$$ME = 1 - \frac{\sum_{i=1}^{N}(M_i - P_i)^2}{\sum_{i=1}^{N}(M_i - M_{mean})^2} \tag{10}$$

Generally, ME ranges between -∞ and 1, and measures the variations accounted by the model. A higher ME value corresponds to a closer match between predicted and measured values and the best value of ME is 1.0. However, values between 0.0 and 1.0 are generally considered as acceptable levels of model performance. The values < 0.0 indicate that the mean measured value is a better predictor than the simulated value, implying unacceptable model performance. Simulation results are considered to be good for values of ME > 0.75, whereas the ME values between 0.36 − 0.75 are also considered as satisfactory [45].

2.5.3. Evaluation of wetland treatment performance

The effect of HLR on wetland treatment performance was evaluated on the basis of removal efficiency (RE), mass loading rate (MLR) and mass removal rate (MRR). The RE was calculated as the percentage change in pollutant concentration from inlet to outlet, using Eq. (11). The MLR and MRR in g/m^2/day were calculated using Eqs. (12) and (13), respectively.

$$RE = \frac{C_{in} - C_{out}}{C_{in}} \times 100\% \tag{11}$$

$$MLR = C_{in} \times HLR \tag{12}$$

$$MRR = (C_{in} - C_{out}) \times HLR \tag{13}$$

Where, C_{in} and C_{out} represent the pollutant concentrations at the inlet and outletof each wetland system, respectively.

Moreover, Normality of influent and effluent wastewater characteristics was determined using Anderson Darling test. One way ANOVA test was used to determine the significance of the treatment differences between wetland systems operated at different HLRs for removal of BOD_5, NH_4^+ and NO_3^- at 95 % confidence level ($p < 0.05$). All statistical analyses were conducted using 'MINITAB 16' software.

3. Results and discussion

3.1. Kinetic reaction rate constant (k)

Model Eqs. (2), (4) and (7) corresponding to Model 1, Model 2 and Model 3 as discussed in Section 2.1 were used to estimate the kinetic reaction rate constants k_1, k_2 and k_3 for BOD_5, NH_4^+ and NO_3^-, respectively. For Model 2, the background concentrations (C*) were obtained from the measured lowest concentrations of BOD_5 (1.2 mg/L), NH_4^+ (0.4 mg/L) with NH_4^+ as N (0.3 mg/L), and NO_3^- (2 mg/L) with NO_3^- as N (0.4 mg/L) in the effluent from the experimental CWs in this study, similar to Trang et al. [49] and Babatunde et al. [25]. When compared to the C* values for subsurface flow constructed wetlands reported by US EPA [44]: BOD_5 (1–10 mg/L), NH_4^+ as N (< 0.1 mg/L) and NO_3^- as N (< 0.4 mg/L), it was noted that the C* values used in this study were within this range only for BOD_5. The estimated k values for BOD_5, NH_4^+ and NO_3^- corresponding to each model at different HLRs are given in Table 1.

According to Table 1, it was noted that the estimated k values for BOD_5, NH_4^+ and NO_3^- are different between HLRs and could be due to the increase in Mass Loading Rates (MLRs) of BOD_5 (2.4–19.4 g/

Table 1
Estimated kinetic reaction rate constants (k).

Parameter/ Model	Kinetic reaction rate constants (m/day) at different hydraulic loading rates (HLRs)			
	5 cm/day	10 cm/day	20 cm/day	40 cm/day
BOD_5				
Model 1	0.1267	0.2193	0.3517	0.4835
Model 2	0.1444	0.2423	0.3804	0.5102
Model 3	0.6086	0.8032	0.9961	0.944
NH_4^+				
Model 1	0.0936	0.1505	0.1702	0.1423
Model 2	0.0965	0.1543	0.1732	0.1441
Model 3	0.2841	0.3522	0.2726	0.1710
NO_3^-				
Model 1	−0.0240	−0.0656	−0.0898	0.0844
Model 2	−0.0282	−0.0765	−0.1049	0.1156
Model 3	−0.0189	−0.0480	−0.0718	0.0940

m^2.day), NH_4^+ (1.9–15.5 g/m^2.day), NO_3^- (0.5–4.4 g/m^2.day) (from Table 5) and different biochemical degradation rates of pollutants within the wetland cells. According to Babatunde et al. [25], k value depends on HLR and influent quality, and Tran et al. [50] reported that k could increase slightly as HLRs increase for BOD_5 and COD, but not for TN and TP. In this study, the estimated k values of BOD_5 for Model 1 and Model 2 tended to increase with increasing HLRs from 5 to 40 cm/day, and the statistical analysis showed a linear relationship between k values and HLRs for BOD_5 at 95 % confidence level ($R^2 \approx 0.9$). However, Model 3 did not exhibit a good linear relationship between k value and HLRs for BOD_5 ($R^2 \approx 0.54$). On the other hand, for NH_4^+, k value first increased linearly from 5 to 20 cm/day HLRs, and decreased at 40 cm/day HLR, for Models 1 and 2. For NO_3^-, k value negatively increased from 5 to 20 cm/day HLRs for all three models, and conversely, a positive k value was obtained for 40 cm/day HLR. This confirms an increment of NO_3^- within the wetland bed, potentially associated with the enhanced nitrification and lower de-nitrification rates in VSSF units. Furthermore, the statistical analysis showed a linear relationship between k value and HLR for both, NH_4^+ and NO_3^- up to HLR of 20 cm/day at 95 % confidence level ($R^2 \approx 0.9$) for Models 1 and 2. However, a distinctive trend in k values over HLR was not observed for NH_4^+ and NO_3^- for Model 3. Despite this, it was found that for Model 3, k values for BOD_5 and NH_4^+ were higher than those for Model 1 and Model 2 at all four HLRs. However, for NO_3^-, k values of Model 3 were lower than that of the other two models. These variations could be due to the portrayal of the hydrologic behaviour of the models. As per Grismer et al. [51], many studies agree that the flow pattern in a subsurface flow CW cannot be simply described as a plug flow reactor or CSTR reactor, but uncertainties remain about which model to apply for a specific CW design.

3.2. Uncertainty analysis

Fig. 3 shows the upper and lower uncertainty limits encompassing a 95 % uncertainty interval for each predicted value of BOD_5, $NH_4^+$and NO_3^- concentrations in the effluent at HLR of 5 cm/day. Figs. S1–S3 in the Supplementary information show upper and lower uncertainty limits for each predicted value of BOD_5, $NH_4^+$and NO_3^- concentrations in the effluent at HLRs of 10, 20 and 40 cm/day. Accordingly, the concentrations of BOD_5 and NH_4^+ in the effluent for all four HLRs predicted by the three models lie within the uncertainty limits. However, for NO_3^-, uncertainty limits encompass only the concentrations predicted at HLR of 40 cm/day. The predicted NO_3^- concentrations vary drastically around upper uncertainty limit corresponding to HLRs of 5, 10, and 20 cm/day. This signifies the complexity in nitrate removal mechanisms prevailing in VSSF wetlands.

Moreover, as uncertainty varies proportionately to the predicted values, relative uncertainty bandwidth (RUB), which is the ratio between the difference in uncertainty limits and corresponding predicted value, was determined. This enabled the comparison of the uncertainty associated with the predictions of different models at different HLRs. Fig. 4 shows RUBs for BOD_5 for Models 1–3, and Figs. S4 and S5 in the Supplementary information show RUBs for NH_4^+ and NO_3^- for Models 1–3.

As evident from Fig. 4, variability in predicted concentrations of BOD_5 in effluent at a given point in time decreases at higher HLRs. Similarly, according to Fig. S4, the predicted NH_4^+ concentrations in the effluent exhibit relatively low variability at high HLRs and relatively high variability at low HLRs for all three models. On the other hand, according to Fig. S5, NO_3^- concentrations in the effluent at a given point in time show low variability at low HLRs for all three models. These results match closely with the observations derived from Figs. 3 and S1–S3.

3.3. Model evaluation

Fig. 5 illustrates the measured and predicted concentrations of BOD_5, NH_4^+ and NO_3^- in the effluent corresponding to each model. It is evident that predicted values of each model are similar and within the measured concentration range at each HLR. Further, the NOF and ME values given in Table 2 also confirm that the predictions for BOD_5, NH_4^+ and NO_3^- closely match with the measured values.

3.4. Wetland treatment performance

3.4.1. Wastewater characteristics

Table 3 shows the average concentrations of BOD_5, NH_4^+ and NO_3^- in the influent and effluent in the VSSF units over the study period. The normality test conducted showed that the data are normally distributed.

It was noted that the mean influent BOD_5 concentration of 48.9 mg/L reduced to 3.7–14.2 mg/L, at 5–40 cm/day HLRs. Similarly, the mean influent NH_4^+ concentration of 39.1 mg/L reduced to 6.1, 8.5 and 15.8 mg/L at 5, 10 and 20 HLRs, respectively. However, NH_4^+ reduction was relatively low at 40 cm/day HLR (26.9 mg/L). On the other hand, the mean influent NO_3^- concentration of 11.1 mg/L increased to 17.6, 20.9 and 17.2 mg/L at 5, 10 and 20 cm/day HLRs, respectively. However, the mean influent NO_3^- concentration reduced from 11.1 mg/L to 8.6 mg/L at 40 cm/day HLR. This is attributed to the lower nitrification rates at higher HLRs due to reduced contact time, limiting the accumulation of NO_3^- in the effluent.

3.4.2. Pollutant removal efficiency

Table 4 shows the average pollutant REs for BOD_5, NH_4^+, and NO_3^- in the four VSSF wetland units operated at HLRs of 5, 10, 20, and 40 cm/day, respectively. The data confirm that the increase in HLR has a negative impact on RE for BOD_5 and NH_4^+. However, negative REs for NO_3^- were obtained for HLRs of 5, 10, and 20 cm/day due to the observed increase in NO_3^- concentrations in the effluent (Table 3).

Removal mechanisms for BOD_5 in a CW system include adsorption, sedimentation, and microbial metabolism [52]. The increase in HLR was found to exert a negative impact on BOD_5 removal. This is attributed to the fact that the effectiveness of removal mechanisms is influenced by contact time [53]. However, REs for BOD_5 were considerably high at HLRs of 5, 10, and 20 cm/day (more than 88 %) and consistent with past studies [21,49]. The one way ANOVA test also confirmed this observation, giving no significant treatment difference in BOD_5 removal between HLRs of 5, 10, and 20 cm/day.

The removal of NH_4^+ and NO_3^- in CW systems is achieved through several interrelated mechanisms including ammonification, ammonia volatilization, nitrification, de-nitrification, plant and microbial assimilation and adsorption. Nitrification, which is the main NH_4^+ removal mechanism, requires an aerobic environment [54]. In fact, VSSF wetlands allow higher oxygenation due to the manner in which wastewater is fed [55]. In the current experiments, the negative impact on NH_4^+ RE at higher HLRs could have been influenced by the lower contact time and reduced oxic environment. From the results obtained, it was observed that NH_4^+ removal was substantial up to HLR of 20 cm/day (more than 70 %), compared to RE at HLR of 40 cm/day (52.1 %). However, the one way ANOVA test confirmed that there is no significant treatment difference between HLRs of 5 and 10 cm/day, although it was observed that there is a treatment difference between 10 and 20 cm/day HLRs.

The removal process for NO_3^- in CWs is mainly driven by the denitrification process which requires the wetland bed to be anoxic [54]. However, NO_3^- removal could also be influenced by nitrification rates. Results of the current experiments indicated an increase in NO_3^- in the effluent at HLRs of 5, 10 and 20 cm/day, thus, resulting in negative REs (Table 3). Further, it was noted that this increment is high at 5 and 10

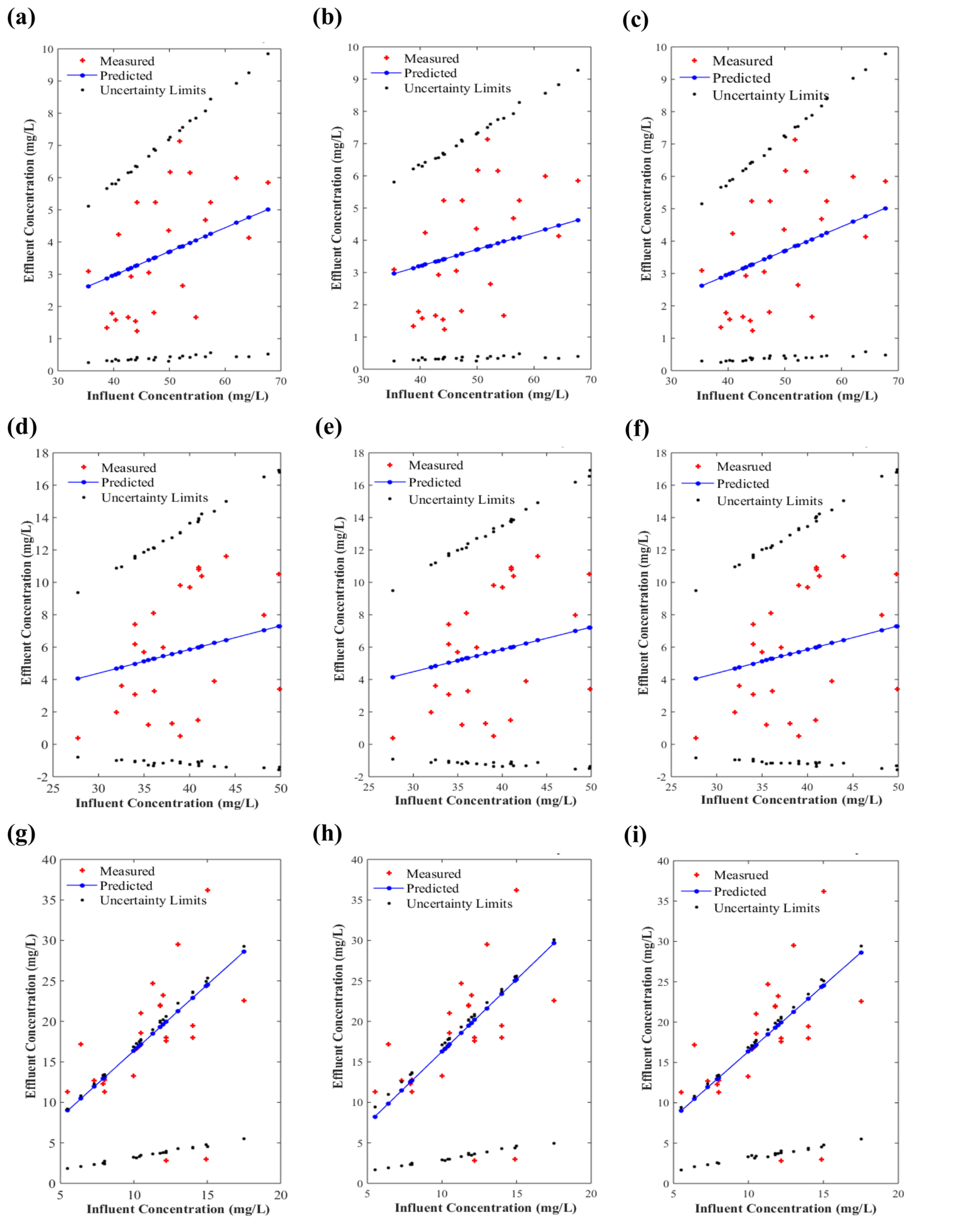

Fig. 3. Uncertainties associated with the predictions of concentrations of BOD_5, NH_4^+, and NO_3^- in the effluent at a hydraulic lording rate (HLR) of 5 cm/day: (a) BOD_5 from Model 1; (b) BOD_5 from Model 2; (c) BOD_5 from Model 3; (d) NH_4^+ from Model 1; (e) NH_4^+ from Model 2; (f) NH_4^+ from Model 3; (g) NO_3^- from Model 1; (h) NO_3^- from Model 2; (i) NO_3^- from Model 3.

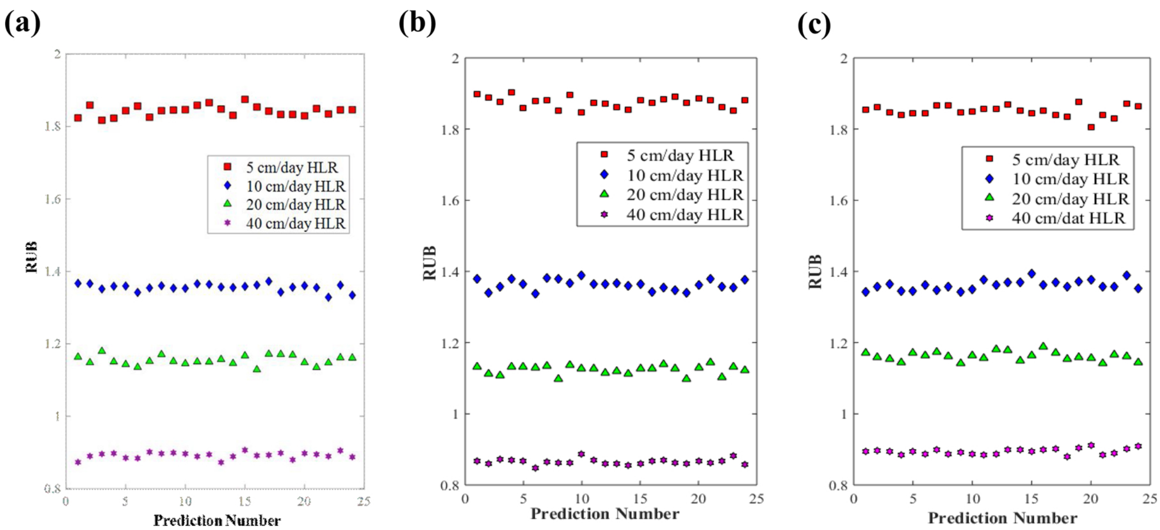

Fig. 4. Variation of relative uncertainty bandwidth (RUB) for BOD_5: (a) Model 1; (b) Model 2; (c) Model 3. *Note: Prediction Number corresponds to each predicted value of effluent concentration.*

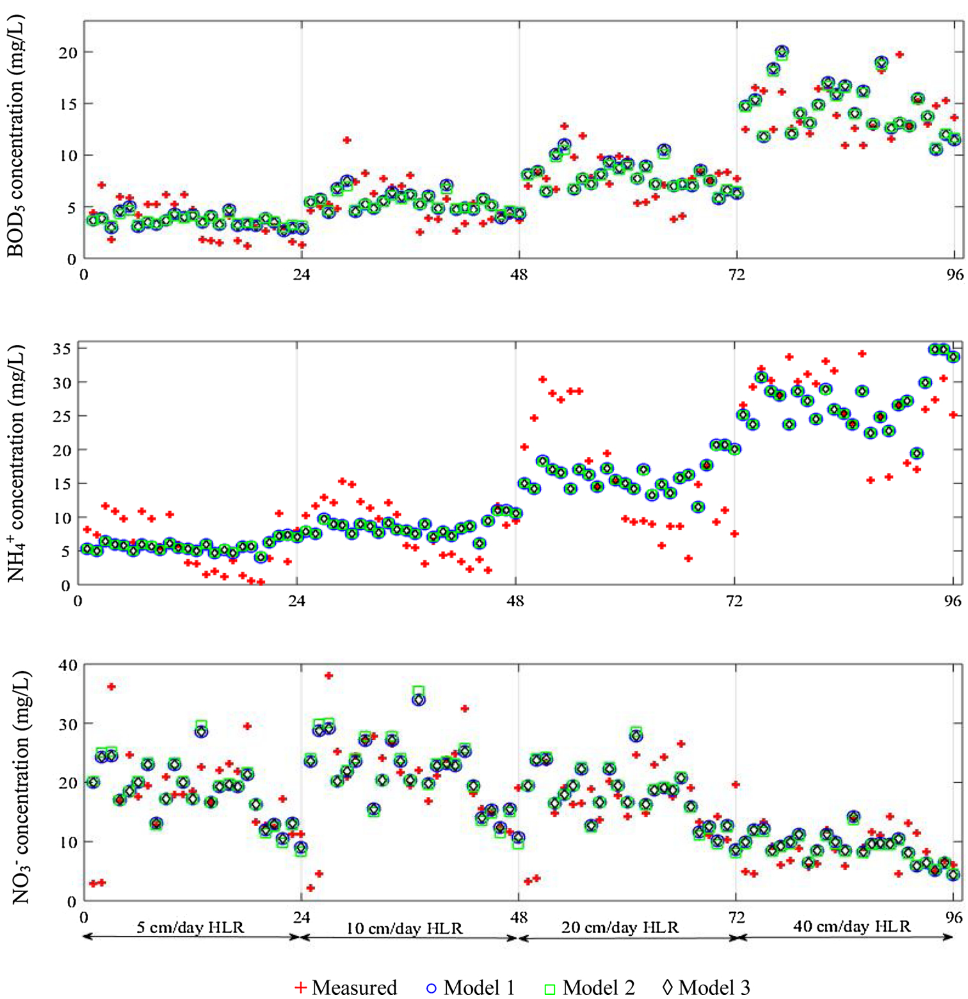

Fig. 5. Measured and predicted concentrations of BOD_5, NH_4^+ and NO_3^- in the effluent.

cm/day HLRs, on average -40.7 % and -53.3 % respectively, and could be attributed to NO_3^- formation through nitrification [56] and insufficient de-nitrification with stronger oxic environment. However, a positive RE of 42.4 % was obtained at the highest HLR (40 cm/day). This could be attributed to the reduced oxic environment and the lower nitrification rate influenced by higher hydraulic flow and reduced contact time. The statistical analysis showed that there is no significant treatment difference for the REs for NO_3^- between HLRs of 5, 10, and 20 cm/day.

Table 2
Results of wetland treatment performance evaluation criteria.

Model/Parameter	Normalised Objetive Function (NOF)	Model Efficiency (ME)
Model 1		
BOD_5	0.29	0.25
NH_4^+	0.42	0.36
NO_3^-	0.41	0.72
Model 2		
BOD_5	0.29	0.25
NH_4^+	0.43	0.37
NO_3^-	0.42	0.75
Model 3		
BOD_5	0.29	0.26
NH_4^+	0.46	0.43
NO_3^-	0.41	0.72

Table 3
Average concentrations of BOD_5, NH_4^+ and NO_3^- in the influent and effluent.

Parameter	Influent (mg/L)	Effluent (mg/L)			
		5 cm/day	10 cm/day	20 cm/day	40 cm/day
BOD_5	48.9 ± 8.4	3.7 ± 1.9	5.4 ± 2.1	7.8 ± 2.1	14.2 ± 2.3
NH_4^+	39.1 ± 6.6	6.1 ± 4.1	8.5 ± 4.1	15.8 ± 8.3	26.9 ± 5.5
NO_3^-	11.1 ± 2.9	17.6 ± 7.4	20.9 ± 8.7	17.2 ± 6.3	8.6 ± 3.2

Table 4
Average removal efficiencies of BOD_5, NH_4^+ and NO_3^-.

Parameter	Removal Efficiency (%)			
	5 cm/day	10 cm/day	20 cm/day	40 cm/day
BOD_5	93.6 ± 3.0	91.4 ± 2.9	88.2 ± 3.0	79.2 ± 7.5
NH_4^+	87.1 ± 8.1	82.5 ± 8.7	70.3 ± 14.7	52.1 ± 14.1
NO_3^-	−40.7 ± 50.5	−53.3 ± 65.0	−20.1 ± 55.3	42.4 ± 29.3

Table 5
Average mass loading rate (MLR) and mass removal rate (MRR) of BOD_5, NH_4^+ and NO_3^-.

Parameter	Hydraulic Loading Rate (HLR) (cm/day)			
	5	10	20	40
BOD_5 (g/m^2/day) (n = 24)				
MLR	2.4 ± 0.4	4.9 ± 0.9	9.5 ± 1.5	19.4 ± 3.2
MRR	2.3 ± 0.4	4.5 ± 0.8	9.2 ± 1.5	14.5 ± 3.3
R^2	0.978	0.978	0.989	0.912
NH_4^+ (g/m^2/day) (n = 24)				
MLR	1.9 ± 0.3	3.9 ± 0.6	7.6 ± 1.3	15.5 ± 2.6
MRR	1.7 ± 0.3	3.3 ± 0.7	6.9 ± 1.6	6.2 ± 2.7
R^2	0.836	0.846	0.912	0.459
NO_3^-(g/m^2/day) (n = 24)				
MLR	0.5 ± 0.1	1.1 ± 0.3	2.2 ± 0.6	4.4 ± 1.2
MRR	−0.1 ± 0.3	−0.4 ± 0.6	1.4 ± 0.5	1.4 ± 1.4
R^2	0.003	0.033	0.431	0.655

3.4.3. *Mass loading rates and mass removal rates*

Table 5 shows average MLRs, MRRs and correlation coefficients between MLRs and MRRs for BOD_5, NH_4^+ and NO_3^- at different HLRs. It is evident that MRRs are positively influenced by the increasing HLRs. The MRRs for BOD_5 show a very strong linear relationship with the incoming mass loads ($R^2 > 0.9$) for all four HLRs. On the other hand, MRRs for NH_4^+ show a fairly strong relationship with the incoming mass loads in VSSF wetland units operated at HLS of 5, 10 and 20 cm/day HLRs (R^2 = 0.83, 0.85 and 0.91, respectively). However, the VSSF wetland unit operated at 40 cm/day HLR did not show a strong relationship (R^2 = 0.46). On the other hand, MRRs of NO_3^- do not show a relationship with incoming mass loads in the VSSF wetland units operated at 5 and 10 cm/day HLRs (R^2 = 0.005 and 0.033), and show a weak relationship at 20 cm/day (R^2 = 0.43) HLR. Thus, it can be concluded that low anoxic environments are not favourable for NO_3^- removal in VSSF wetlands. However, the VSSF wetland unit operated at 40 cm/day HLR, showed a moderate relationship between MLRs and MRRs (R^2 = 0.65). This could be attributed to low nitrification rates due to reduced contact time and favourable de-nitrification due to the anoxic environment associated with the higher HLR.

In summary, the mass removal of BOD_5 was efficient for all four HLRs, while mass removal of NH_4^+ was efficient for 5–20 cm/day HLRs. However, the mass removal of NO_3^- was not satisfactory for 5–20 cm/day HLRs.

Table 6
Recommended design parameters for respective models for designing and performance monitoring of vertical subsurface flow constructed wetlands.

Parameter & HLR (cm/day)	Kinetic reaction rate constants (m/day)		
	Model 1 (k–C model)	Model 2 (k–C* model)	Model 3 (First order CSTR model)
BOD_5			
5	0.1267	0.1444	0.6086
10	0.2193	0.2423	0.8032
20	0.3517	0.3804	0.9961
40	0.4835	0.5102	0.944
NH_4^+			
5	0.0936	0.0965	0.2841
10	0.1505	0.1543	0.3522
20	0.1702	0.1723	0.2726

3.5. *Recommendations for wetland design*

Results of model evaluation revealed that HLR is a significant parameter in designing VSSF wetlands using empirical kinetic models. Also, it was noted that all three models (first order k–C, k–C* and first order CSTR) have performed satisfactorily in estimating kinetic reaction rate constants with respect to BOD_5 and NH_4^+ removal in VSSF wetlands, with very strong correlations for BOD_5 for 5–40 cm/day HLRs and NH_4^+ for 5–20 cm/day HLRs. This shows the difficulty of describing the reactor type (plug-flow or CSTR) for VSSF wetlands. According to the uncertainty analysis, all three models can be recommended as satisfactory predictive tools for design and performance monitoring of VSSF wetlands in tropical environments for BOD_5 and NH_4^+ removal. Table 6 gives recommended design parameters ('k' values and HLR). However, since 20 cm/day HLR was found to be the highest viable HLR, corresponding k values of 0.3517, 0.3804 and 0.9961 m/day for BOD_5 removal and 0.1702, 0.1732 and 0.2726 m/day for NH_4^+ removal can be used for k–C, k–C* or first order CSTR model, respectively, for the design of VSSF wetlands in tropical climatic regions.

4. Conclusions

The applicability of three first order kinetic models (k–C, k–C* and first order CSTR) for the design of VSSF CWs in tropics was evaluated using four laboratory scale experimental VSSF wetland configurations operated at four different HLRs of 5, 10, 20 and 40 cm/day. Results revealed that HLR is a key parameter in designing VSSF wetlands using empirical models. Also, results confirmed that all three models perform satisfactorily in the estimation of kinetic reaction rate constant (k) for treating BOD_5 at HLRs of 5, 10, 20 and 40 cm/day, and NH_4^+ at 5, 10 and 20 cm/day HLRs. This highlights the difficulty in describing hydrologic behaviour (plug-flow or CSTR) of VSSF wetlands. The estimated k values for BOD_5 varied as 0.1267 – 0.4835 m/day for the first order k–C model, 0.1444 – 0.5102 m/day for the k–C* model and 0.6086 – 0.944 m/day for the first order CSTR model, for 5–40 cm/day HLRs. The estimated k values for NH_4^+ varied as 0.0936 – 0.1702 m/day for the k–C model, 0.0965 – 0.1732 m/day for the k–C* model and 0.2841 – 0.2726 m/day for the first order CSTR model, for 5–20 HLRs. Evaluation of treatment performance of VSSF wetlands showed more than 88 % removal of BOD_5 and more than 70 % removal of NH_4^+ at 5, 10 and 20 cm/day HLRs, while NO_3^- removal was not satisfactory at these HLRs. Further, it was evident that HLR has a positive impact on MRRs for BOD_5 and NH_4^+ at 5, 10 and 20 cm/day HLRs with a strong correlation between MLRs and MRRs. Therefore, 20 cm/day was deemed as the highest viable HLR for VSSF wetland designs for tropical climatic conditions. Therefore, in designing VSSF wetlands, corresponding k values of 0.3517, 0.3804 and 0.9961 m/day for BOD_5 removal and 0.1702, 0.1732 and 0.2726 m/day for NH_4^+ removal can be used for first order k–C, k–C* and CSTR models, respectively.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary material related to this article can be found, in the online version, at doi:https://doi.org/10.1016/j.jwpe.2020.101539.

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