

Effect of filter media and hydraulic retention time on the performance of vertical constructed wetland system treating dairy farm wastewater

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Abstract

This study deals with the dairy wastewater treatment using laboratory scale vertical flow (VF) constructed wetlands with the *Canna indica* plantation over wetland beds due to phytoremediation capabilities. Three laboratory scale VF CWs (CW-A, CW-B and CW-C) each with an area of 0.135 m² filled with gravel (CW-A: 20 mm; CW-B: 10 mm gravel) and sand (CW-C) receiving 0.04 m³ d⁻¹ dairy wastewater were operated for the wastewater purification. Each unit was operated at three hydraulic retention times (HRTs) i.e. 12 h, 24 h and 48 h for assessing its effect on wastewater purification. Among all units, removal rates fluctuated as: total suspended solids (TSS): 64.2–74.5%; biochemical oxygen demand (BOD): 45.3 – 63.1%; ammonium nitrogen (NH₄-N): 29.6 – 56.5% and phosphate phosphorous (PO₄-P): 20.5 – 57.8% at different HRTs. Increase in HRT showed better removal of pollutants in all CWs. Moreover, maximum removal of pollutants excluding TSS and NH₄-N was achieved in CW-B at 48 h HRT. CW-B with similar HRT provided maximum removal of PO₄-P (57.8%), BOD (63.1%) and chemical oxygen demand (COD): 67.4%. Increase in the size of filter media, from sand (0.25 mm) to 20 mm gravel resulted in higher removal of NH₄-N from wastewater.

Keywords: Artificial wetlands, Constructed Wetland (CW) System, Dairy wastewater, Hydraulic Retention Time (HRT), Treatment wetlands, Wastewater Purification



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1. Introduction

Dairy farm practices release a huge volume of wastewater into the surrounding environment [1]. This wastewater is a mixture of animal urine, spilled milk, floor and utensil washings, traces of animal dung etc. It consists of small to medium strength of organic matter, phosphorous and nitrogen which is responsible for the problems of eutrophication in the receiving water bodies and degradation of soil quality as well [2]. In the past, many treatment techniques such as application of organic coagulants [3], advanced oxidation processes, membrane technology, anaerobic digestion [4], activated sludge process, trickling filters, rotating biological contractor, oxidation pond, UV disinfection, etc. have been tested for removing pollutants from dairy wastewater but only limited technologies could be successfully adopted. However long term use of these technologies was impaired due to high treatment cost, skilled labor, problem of sludge disposal, etc. Constructed Wetland (CW) technology is a convenient and eco-friendly alternative which is well suited for treating such kinds of wastewater [5]. Moreover, this technology has added advantages of; low treatment cost, better performance for removal of organic substances and nutrients like N, P along with ability to tolerate load fluctuations in influent [6].

Constructed wetlands (CWs) are engineered systems that have been designed and used to utilize the natural processes involving wetland vegetation, soils, and associated microbial assemblages to assist in wastewater treatment [7]. Among the different designs of CWs, Horizontal, Vertical and Hybrid systems are the most common and had shown effective results in terms of pollutant removal from wastewater [8]. Construction of a CW system, involves consideration of many design parameters such as influent quality, desired effluent quality, organic dosing rate, selection of appropriate filter material and macrophytes, hydraulic retention

time (HRT), BOD/P/N ratio, bed depth, arrangement of beds (parallel or series) and surface partition at bed surfaces [9]. In VF CW system, filter media and plants play an important role in P removal [10]. These potent filter media such as sand and gravels have the capacity to bind phosphate and thus precipitate the phosphate content from wastewater. Along with this, plant such as *Arundo donax* absorb the readily available phosphate which are attached to the filter media surface [11].

The present study focusses on the removal of pollutants i.e. $\text{NH}_4\text{-N}$, TSS, $\text{PO}_4\text{-P}$, BOD and COD using laboratory scale vertical flow CW systems filled with different filter materials and operated at different HRTs for the treatment of dairy farm waste water. The CW system was planted with *Cana indica* owing to previous studies with similar plant, easy availability, price and aesthetic [12].

2. Materials and Methods

2.1. Study Site and Construction of Vertical Flow CW Systems

The present study was conducted at Graphic Era University, Dehradun (30.3165° N , 78.0322° E), Uttarakhand, India. Laboratory scale CW systems were built using round shaped plastic container with a depth of 50 cm (surface area: 0.135 m^2). Three such containers were labelled as CW-A, B, C filled with 20 mm gravel, 10 mm gravel and sand, respectively. On the surface of each CW *Cana indica* was planted. *Canna indica* plant has phytoremediating properties and can easily grow in wet and marshy regions. Arrangement of sampling was made at the bottom of each container using a water tap (Fig. 1).

2.2. Filter Materials

As per previous studies, different filter materials such as gravels of varied sizes, sand, etc. may be used in a CW unit [13]. Sand, being fine in size acts as the best filter material and treats wastewater using physical, biological and chemical processes [14] but is associated with problem of clogging. On the other hand, gravels too have its own advantages of excellent nitrification potential because of greater pore space for air availability and better adhesion surface for microbial films which provides good mineralization of organic nitrogen and oxidation of ammonium ions [15]. This helps in maintaining aerobic conditions on the bed surfaces which supports the pollutant removal processes. Moreover, the gravel filled systems are less susceptible to clogging during their operation.

As the wastewater percolates slowly through the filter material, various physical, biological and chemical processes occur in combination resulting in the removal of pollutants from wastewater. Phosphorous gets attached to sand /gravel surface resulting in adsorption and precipitation processes in the CW unit [15]. These filter materials act like a natural home for variety of microbes such as *Bacillus*, *Micrococcus*, *Pseudomonas* etc. which contribute to organic matter degradation as well as nitrification and denitrification [16]. These filter materials serve as efficient units for removal of pollutants from wastewater (BOD, COD, $\text{NH}_4\text{-N}$, $\text{PO}_4\text{-P}$ and fecal coliforms).

2.3. Hydraulic Retention Time (HRT)

HRT is the most easily changed operational variable in the design of CW system [17]. The performance of a Constructed Wetland system largely depends on the selected HRT because it reflects a contact period between wastewater and the wetland system [18].

Previous studies by DeBusk and Reddy [19] and Kadlec and Knight [20], recommended that longer the contact period of water with the filter material inside the wetland, more removal of pollutants occurs. In the past many studies were conducted at different HRTs (8 h to 20 days) (Table 1) However, most of these studies were focussed on the treatment of domestic wastewater. To the best of our knowledge, none of the study has been conducted for treatment of dairy farm wastewater at shorter HRTs.

Based on previous studies related to filter materials and HRT in CWs, this research work was designed to analyze the performance of laboratory scale vertical sub-surface flow Constructed Wetland systems operated with different filter material and HRTs and their combined effect on removal of organic and inorganic pollutants from dairy wastewater.

2.4. Experimental Design and Collection of Samples

Dairy wastewater samples were collected from dairy farm located nearby Graphic Era University, Dehradun Uttarakhand, India. In this study, different filter materials i.e. gravel of 10 mm and 20 mm sizes and sand were used in separate CW units (A, B and C). *Canna indica* plant was selected for surface plantation and was collected from a sewage drain located near Graphic Era University. Filter materials and the plants were brought to the laboratory and washed properly before starting the experiment. Plastic containers of depth 50 cm and diameter 41.5 cm each were taken for the experiment. 20 mm and 10 mm gravels were filled throughout the container A

(CW-A) and container B (CW-B), respectively while container C (CW-C) was filled from top to bottom with washed sand (0.25 mm). Control systems for all the three CW systems were also considered for the study. The control CW systems had the same materials and other specifications as CW-A, CW-B and CW-C, but were operated at zero HRT unlike CW-A, CW-B and CW-C.

On the basis of retention time, flow rates, bed surface area and concentrations of organic matter and nitrogen, the decomposition constant coefficients (k) for wastewater treated in VF beds were calculated. First order equation form presented in (Eq. (1)), which uses k_v rate and the HRT for VF beds:

$$C_{out}/C_{in} = \exp.(-k_A/q) \quad (1)$$

Here, q = hydraulic loading rate in m d^{-1} i.e. calculated as the ratio of flow rate (Q) in $\text{m}^3 \text{d}^{-1}$ and surface area (A) in m^2 whereas k_A is the decomposition constant in m d^{-1} [21].

Forty litres of dairy farm wastewater was dosed intermittently (batch feeding mode) everyday to each container. Dairy influent was analyzed for pH, ORP (oxidation reduction potential), EC (electrical conductivity), TDS (total dissolved solids), TSS (total suspended solids), salinity, DO (dissolved oxygen), temperature, BOD (biochemical oxygen demand), COD (chemical oxygen demand), $\text{NH}_4\text{-N}$ (ammonium-nitrogen), and $\text{PO}_4\text{-P}$ (phosphate phosphorous) before dosing to the CW systems. Treated samples from control beds and all the three systems were collected in 1 litre plastic bottles. These treated water samples were collected at specific HRTs (0 h, 12 h, 24 h and 48 h) through the knob at the base of each container after HRT of 12 h, 24 h and 48 h, respectively. The study was carried out for a period of three months (from March to May 2019).

2.5. Analytical Procedures

Regular samples were collected from the outlets of all the three containers after HRT of 0 h, 12 h, 24 h and 48 h, respectively and stored in refrigerator for analysis. Samples from the control beds of all the three CW systems were collected at zero h HRT i.e. no HRT was set in these beds. All the untreated and treated water samples were analysed in laboratory as per the guidelines of standard methods for examination of water and wastewater [22] and Hach Manual. pH, ORP, EC, TDS, salinity and temperature were analyzed during the study period by using multiparameter system (Hach SensION+ MM 150). DO was measured using DO meter (Hach SensION). Similarly, other parameters were measured using methods such as TSS (colorimetric method), BOD (3-days incubation method), COD (reactor digestion method), NH₄-N (salicylate method) and PO₄-P (molybdovanadate method).

2.6. Determination of Removal Rate

The efficiency of the vertical sub-surface constructed wetland system for removal of pollutants was calculated in terms of removal rate using the following equation [5].

$$\text{Removal rate (\%)} = (C_i - C_o) * 100 / C_i \quad (2)$$

C_i = Influent concentration (mg L^{-1}); C_o = Effluent concentration (mg L^{-1})

3. Results and Discussion

3.1. pH, DO, NO₃-N and NH₄-N

1 Average pH in the influent was recorded as 7.2 ± 0.4 which changed between 7.23 ± 0.2 in
 2 control, 7.3 ± 0.3 to 7.5 ± 0.3 in CW-A, 7.2 ± 0.4 to 7.5 ± 0.2 in CW-B and 7.4 ± 0.3 to 7.7 ± 0.2
 3 in CW-C unit (Table S1). Although no significant change in pH was observed at different HRTs
 4 yet maximum pH change was observed at 48 h HRT in all CW systems. Average DO
 5 concentration in the influent was found to be $4.3 \pm 2.0 \text{ mg L}^{-1}$. The control bed of CW-A showed
 6 a fluctuation of 2.9 to 4.9 mg L^{-1} during the study period while in CW-B control bed, it ranged
 7 between 3.0 to 5.0 mg L^{-1} . The control bed of CW-C showed minimum and maximum DO
 8 values of 2.9 and 4.9 mg L^{-1} , respectively. In the treated water, DO was recorded in the range of
 9 4.9 ± 0.35 to $5.8 \pm 0.8 \text{ mg L}^{-1}$ among all CW systems. CW B (10 mm gravel filled unit) showed
 10 4.9 ± 0.35 , 5.0 ± 0.6 and $5.2 \pm 0.2 \text{ mg L}^{-1}$ average DO concentrations at HRTs 0, 12, 24 and 48 h,
 11 respectively (Fig. 2). A slightly higher DO increase (5.1 ± 0.5 to $5.8 \pm 0.8 \text{ mg L}^{-1}$) was observed
 12 in CW-A (20 mm gravel filled) and CW-C i.e. sand filled unit (5.1 ± 0.6 to $5.4 \pm 0.7 \text{ mg L}^{-1}$). The
 13 percent DO increase at outlets w.r.t influent DO concentration was observed as 34.9 and 25.6%
 14 in CW-A and CW-C respectively at 24 h HRT and 20.9% in CW-B at 48 h HRT. Maximum DO
 15 increase of 5.8 mg L^{-1} was recorded in 20 mm gravel filled system at an HRT of 24 h.

16 Nitrification is a key characteristic of vertical flow systems [23]. The influent showed an
 17 average $\text{NO}_3\text{-N}$ concentration of $1.5 \pm 0.8 \text{ mg L}^{-1}$. Effluents of control and all the CW systems
 18 showed higher $\text{NO}_3\text{-N}$ concentrations; 2.1 ± 0.5 - $3.0 \pm 0.3 \text{ mg L}^{-1}$ in CW-A, 1.8 ± 0.3 - 3.7 ± 0.4
 19 mg L^{-1} in CW-B and 1.7 ± 0.3 - $1.98 \pm 0.5 \text{ mg L}^{-1}$ in CW-C. In all the CW systems $\text{NO}_3\text{-N}$
 20 concentration showed an increase (42.9% in CW-A, 32.1% in CW-B and 13.1% in CW-C) when
 21 HRT was increased from 12 h to 24 h however, when HRT was further extended to 48 h, the
 22 $\text{NO}_3\text{-N}$ concentration showed a decrease in effluent (Table S1). This might be due to

denitrification process which occurs at longer HRTs. It has also been reported that at longer HRT a well-developed microbial community in CWs gets suitable contact time to remove pollutants [24, 25]. In a study by Toet et al. [25] positive nitrogen removal was observed in CWs with an HRT of 0.8 days in comparison to 0.3 day HRT. Further longer HRT led to anoxic conditions in the bed which facilitates denitrification process [26]. In addition, the effect of HRT may differ between CWs depending on the dominant plant species, temperature, type of wastewater and feeding mode as those factors can affect the hydraulic efficiency of wetlands [24, 25].

Maximum nitrate concentration (3.0 mg L^{-1}) i.e. 100% increase w.r.t influent concentration was observed in CW-A (20 mm gravel filled) at a HRT of 24 h. while minimum $\text{NO}_3\text{-N}$ concentration of 1.7 mg L^{-1} was observed in CW-C (sand filled) at an HRT of 48 h.

Average concentration of $\text{NH}_4\text{-N}$ in the dairy influent was recorded as $48.0 \pm 3.5 \text{ mg L}^{-1}$. Average $\text{NH}_4\text{-N}$ concentrations at CW-A outlet (20 mm gravel filled unit) was found as $27 \pm 3.5 \text{ mg L}^{-1}$ (12 h), $21 \pm 4.1 \text{ mg L}^{-1}$ (24 h) and $20.9 \pm 3.2 \text{ mg L}^{-1}$ (48 h HRT) whereas at CW-B outlet, average $\text{NH}_4\text{-N}$ concentration was recorded as 29.1 ± 3.7 , 24.3 ± 4.0 and $25 \pm 2.5 \text{ mg L}^{-1}$ at HRTs 12, 24 and 48 h (Table S1). The $\text{NH}_4\text{-N}$ concentrations in the control beds of CW-A, CW-B and CW-C fluctuated between 29.1 to 38.1 mg L^{-1} , 33.1 to 41.4 mg L^{-1} and 39.6 to 45.2 mg L^{-1} , respectively (Fig. 4). During its retention insided the wetland bed, water undergoes certain physical, chemical and biological processes which leads to removal of toxic pollutants like ammonium, nitrogen, suspended solids and phosphate. The major processes involved in removal of ammonium nitrogen in a CW are nitrification-denitrification [27]. Nitrogen removal in constructed wetland depends upon environmental parameters (e.g. pH, oxygen, temperature etc.), operating conditions (hydraulic and pollutant loading, detention time, influent feed mode,

recirculation, organic carbon addition etc.) and presence of microorganisms [24]. Among these parameters HRT plays an important role as it affects the contact duration between microbes and pollutants [28]. In this study, CW-A unit provided 43.8% removal of $\text{NH}_4\text{-N}$ at 12 h HRT which was subsequently increased to 56.3 and 56.5% at 24 h and 48 h HRTs. In CW-B unit (10 mm gravel filled), average removal percentage was recorded as 39.4% at 12 h HRT, further showed an increase of nearly 10% on extending HRT to 24 h. However no further increased was observed in $\text{NH}_4\text{-N}$ removal when HRT was taken to 48 h (Fig. 5). More or less similar trend was observed in sand filled unit, CW-C which provided showed 29.6% $\text{NH}_4\text{-N}$ removal at 12 h HRT. This was further increased to 43.1% and 43.8% at 24 h and 48 h HRTs, respectively. In a similar study [29], with field scale VF CW system (20 mm gravels), the removal of $\text{NH}_4\text{-N}$ was more (75% removal in 24 h HRT) as compared to lab-scale VF CW system filled with 20 mm gravels (56.3% in 24 h). Our results are not in an agreement with a previous study carried out by Akratos and Tsihrintzis [30] in which the overall removal of $\text{NH}_4\text{-N}$ was achieved as 74.9 and 87.5% at HRTs of 14 and 20 days, respectively. The differences in result may be due to variation in HRTs of both studies which were 0-48 hrs in our study against 14-20 days in the compared study.

3.2. TSS and $\text{PO}_4\text{-P}$

Total suspended solids (TSS) are the substances of size > 2 microns and are present in the form of inorganic materials, though dust, bacteria, algae etc. may also contribute to total solid concentration in water [31, 32]. The average concentration of suspended solids in dairy influent was recorded as $628.0 \pm 53.0 \text{ mg L}^{-1}$ (Table S1). The control beds of CW-A and CW-B showed

1 fluctuations in TSS concentration from 288.0 to 515.0 mg L⁻¹ and 365.0 to 615.0 mg L⁻¹
2 respectively whereas in sand control beds, fluctuation of 250.0 to 500.0 mg L⁻¹ was recorded (Fig.
3 2). Gravel (20 mm) in CW-A showed a removal of 64.2% of TSS at 12 h HRT which showed
4 more or less similar removal rate when HRT was increased to 24 h HRT. Likewise, no further
5 increase in removal rate was observed at 48 h HRT (Fig. 6). Gravel (10 mm) in CW-B unit also
6 showed similar pattern with 67.9% removal at 12 h and 68.4% at 24 h HRT. No further increase
7 in the removal rate was seen at 48 h HRT. Sand showed maximum removal of suspended solids
8 (73.1–74.5%) at all HRTs in CW-C because sand acts as a fine filter medium and is capable to
9 remove small to large size suspended particles from the wastewater (Fig. 5). Vymazal [33] in one
10 of his study recorded 75% removal of suspended solids using gravel media as substrate. Another
11 studies suggested that TSS removal is mainly a physical process and is also affected by retention
12 time [34–36] but in our study, HRT did not produce a significant change in the TSS removal
13 capacity of all three CW units. However, substrates play a very vital role in TSS removal as
14 recorded in the study conducted by Manios et al. [37] in which removal percentage of TSS was
15 found to be 85 and 90% in gravel and sand respectively. In our study, sand filter with 48 h HRT
16 arrangement was found to be most suitable for achieving maximum (74.5%) TSS removal from
17 the waste water.

18 Phosphorous in wetlands occurs as phosphate in organic and inorganic compounds. In a
19 wetland system, P removal occurs by adsorption and precipitation mechanisms. The P present in
20 the wastewater usually gets bind to the filter substrates of the CW beds as a result of adsorption
21 and precipitation reactions by combining with the Ca, Al and Fe present in the gravel or sand
22 substrates [31]. Thus the ability of VF CW beds to reduce P is dependent on the contents of these

minerals in the filter media. Other factors like plant uptake and biological reactions do not play major role in P removal [31]. Thus, filter media has an important role in removing phosphorus from wastewater. $\text{PO}_4\text{-P}$ in influent has an average concentration of $41.0 \pm 4.7 \text{ mg L}^{-1}$. In the control system of CW-A and CW-B, the $\text{PO}_4\text{-P}$ concentration was found to be fluctuating between 35.5 to 41.5 and 31.2 to 41.8 mg L^{-1} , respectively while in sand (CW-C) control bed, it was recorded to range from 30.8 to 40.1 mg L^{-1} (Fig. 3). Average $\text{PO}_4\text{-P}$ concentration at the outlet of CW-A was recorded as; $32.6 \pm 4.8 \text{ mg L}^{-1}$ at 12 h, $26.4 \pm 3.6 \text{ mg L}^{-1}$ at 24 h and $22.8 \pm 2.9 \text{ mg L}^{-1}$ at 48 h HRT (Table S1). In CW-A (20 mm gravel filled unit), removal rate of $\text{PO}_4\text{-P}$ was observed as 20.5% at 12 h HRT. An increment of 15.1% was recorded at 24 HRT resulting in 36.6% removal. On further increasing HRT to 48 h, $\text{PO}_4\text{-P}$ removal rate was increased upto 44.4%. In CW-B (10 mm gravel filled unit), average $\text{PO}_4\text{-P}$ concentration in treated water was observed as; 27.3 ± 4.4 , 19.4 ± 3.6 and $17.3 \pm 3.3 \text{ mg L}^{-1}$ at 12, 24 and 48 h HRTs, respectively. The average removal rate was recorded as 33.4% at 12 h HRT; 52.7% at 24 h HRT and further an increase of 5.1% at 48 h (Fig. 2(b)). In CW-C (Sand filled unit), $\text{PO}_4\text{-P}$ removal rates was recorded as 25.9, 44.1 and 46.1% at 12 h, 24 h and 48 h HRTs respectively (Fig. 7). Among all the three CW units, CW-B (10 mm gravel filled unit) showed maximum removal of $\text{PO}_4\text{-P}$ at 48 h HRT. Phosphorous removal mainly takes place by absorption by plants and sorption processes on the media used, and this phosphorous can be stored in the accumulated sediments [32]. Seo et al. [32], in his study, explained that P may remain bound to the media components as a result of precipitation and adsorption reactions with ions (Ca, Fe or Al) present in sand or gravels. The particle size of substrates which are suitable for P removal may vary significantly.

Our results showed a higher $\text{PO}_4\text{-P}$ removal as compared to a study conducted by Arias et al. [35] in which the maximum removal (45.6%) of $\text{PO}_4\text{-P}$ was achieved at HRT of 7 days. In another study conducted by Stearman et al. [36], maximum $\text{PO}_4\text{-P}$ removal was recorded as 10% at 0.7 day HRT.

3.3 . BOD_3 and COD

Biochemical Oxygen Demand (BOD) determines the amount of oxygen required by the biological organisms to degrade organic pollutants. It is a pollution indicating parameter. The average BOD_3 value found in the dairy influent was $815.0 \pm 33 \text{ mg L}^{-1}$ (Table S1). The BOD value in control beds of CW-A and CW-B fluctuated from 402.0 to 558.0 and 400.0 to 510.0 mg L^{-1} , respectively (Fig. 3) while the average BOD values were observed as 466.4 and 475.4 mg L^{-1} in CW-A and CW-B, respectively. CW-C control bed showed BOD range between 410.0 to 550.0 mg L^{-1} with an average value of 498.1 mg L^{-1} .

The average BOD_3 decrease at outlet amongst the three setup was most prominent in CW-B outlet (10 mm gravel filled). The BOD_3 removal rate was observed as: 46.3, 60.9 and 62% in CW-A at 12 h, 24 h and 48 h HRTs respectively (Fig. 6(a)). In CW-B (10 mm), the removal rate was recorded as 48.1 and 62.2% at 12 h and 24 h HRTs, respectively while no further change was observed at 48 h HRT (Fig. S1). In CW-C, the BOD_3 removal rate was found to be 45.2, 57.1 and 58.5% at 12 h, 24 h and 48 h HRTs, respectively. Using a similar setup comprising of a 2-stage SSVF CW and a SSHF CW, Job [38] obtained BOD_5 removal rate of approximately 65%. The BOD_3 removal rate in our study showed an increase in all CW units

(14.6 % in CW-A, 14.1 % in CW-B and 11.8% in CW-C) when HRT was increased from 12 to 24 h.

Chemical Oxygen Demand (COD) indirectly measures the amount of organic compounds in wastewater similar to BOD. It is also another pollution indicating parameter like BOD. During the study period, average concentration of COD recorded in dairy influent was $1,230.0 \pm 48 \text{ mg L}^{-1}$ (Table S1). The average concentration of COD was found to be decreased by 56.5% at 12 h, 64.6% at 24 h and 64.9% at 48 h HRT in CW-A. An increase of approximately 8% was observed in COD removal rate when HRT was increased from 12 h to 48 h in CW-A unit (Fig. 6(a)). In CW-B (10 mm gravel) removal was observed as 58.1, 66.5 and 67.4% at HRTs 12, 24 and 48 h. In CW-C, the percent removal was minimum among all the three filter materials and was recorded as 54.6, 62.3 and 63.0% at 12 h, 24 h and 48 h HRT, respectively. Plant activities have essential role in organic matter degradation [39, 40]. This occurs due to increase in plant biomass, leading to high amount of microbial development among the roots. Thus microbial degradation as well as biofilm formation is favoured leading to decrease in organic matter content of the wastewater.

From Table S1, it can be observed that maximum degradation of organic substances occurred in CW-B (10 mm gravel filled) at 24 h HRT. In a similar study [41] removal efficiency and removal rate of COD in gravel filled CW units were found in the range of 32.9 - 50.4% and $7.4 - 20.6 \text{ g m}^{-2} \text{ d}^{-1}$ in VFCW, respectively in 4.9, 2.4 and 1.2 days of HRT. A study conducted by Abed [42] stated that hydraulic retention time (HRT) of 24 h is enough for removal of pollutants from wastewater by the activities of filter media as well as plants.

4. Conclusions

The constructed wetland involves diverse processes (physical, chemical & biological) for removal of pollutants. Further, optimal operating parameters (i.e., water depth, hydraulic retention time and pollutant load) and wetland design also influence treatment performance of CW. The major findings of study are summarized as follows:

- No significant change in pH was observed with variation in HRT.
- The study showed higher removal of $\text{NH}_4\text{-N}$ and BOD_3 with 48 h HRT in comparison to 12 h and 24 h HRTs. Removal rates of $\text{NH}_4\text{-N}$ and BOD_3 were higher in gravel filled units compared to sand. The gravel has more porosity and provides more surface area for attachment of microbial community hence results in more pollutant removal.
- Sand filled CW unit showed 10% higher removal of TSS compared to gravel filled units. Results indicate that change in HRT has least effect on TSS removal rate in all CW units.
- Maximum DO increase of 5.8 mg L^{-1} was recorded in 20 mm gravel filled system at an HRT of 24 h. Results showed that change in HRT did not influence DO however change in DO was affected due to filter media size.
- The maximum $\text{PO}_4\text{-P}$ removal (57.8%) was achieved by 10 mm gravel filled unit at 48 h HRT. For better $\text{PO}_4\text{-P}$ removal synthetic and industrial products with high phosphorus sorption capacity and hydraulic conductivity can be explored as alternative substrates in CWs in case of poor absorptive media.
- During the study, change in removal rate of pollutants ($\text{NH}_4\text{-N}$, BOD_3 , COD) was significantly higher (12.2-18.6%) when HRT was changed from 12 h to 24 h. A slight

1 increase ($< 3\%$) in the removal rate of pollutants was observed when HRT was increased
2 from 24 h to 48 h. However this trend was not observed in case of TSS removal.

3 The study showed good removal of pollutants in all filter materials, however further study is
4 recommended to validate findings on real-scale CW systems. We recommend assessment of
5 planted vegetation for phytoremediation potential and biofilm formation on filter media in future
6 studies for better understanding on pollutant removal mechanisms in CW.

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17 **Author Contributions**

18 D.M (PhD student) conducted all the experiments and wrote the manuscript. P.K.S (Professor)
19 revised the manuscript. A.R (Associate Professor) wrote and revised the manuscript.

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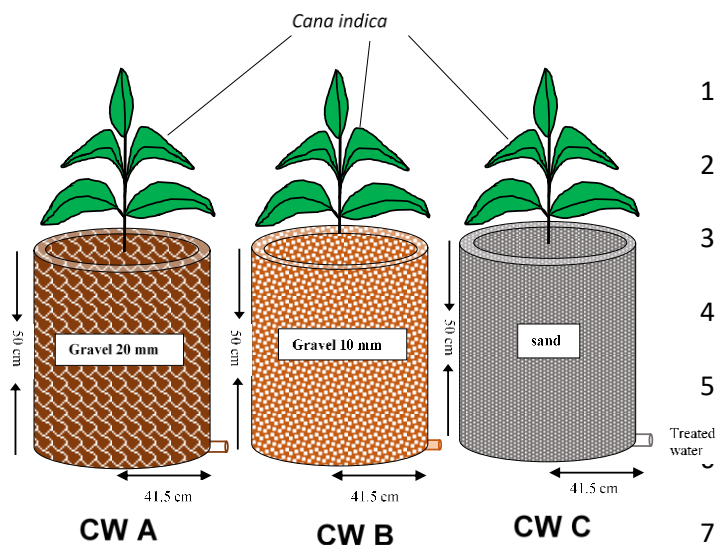


Fig. 1. Layout of CW-A, CW-B and CW-C for treatment of dairy farm wastewater.

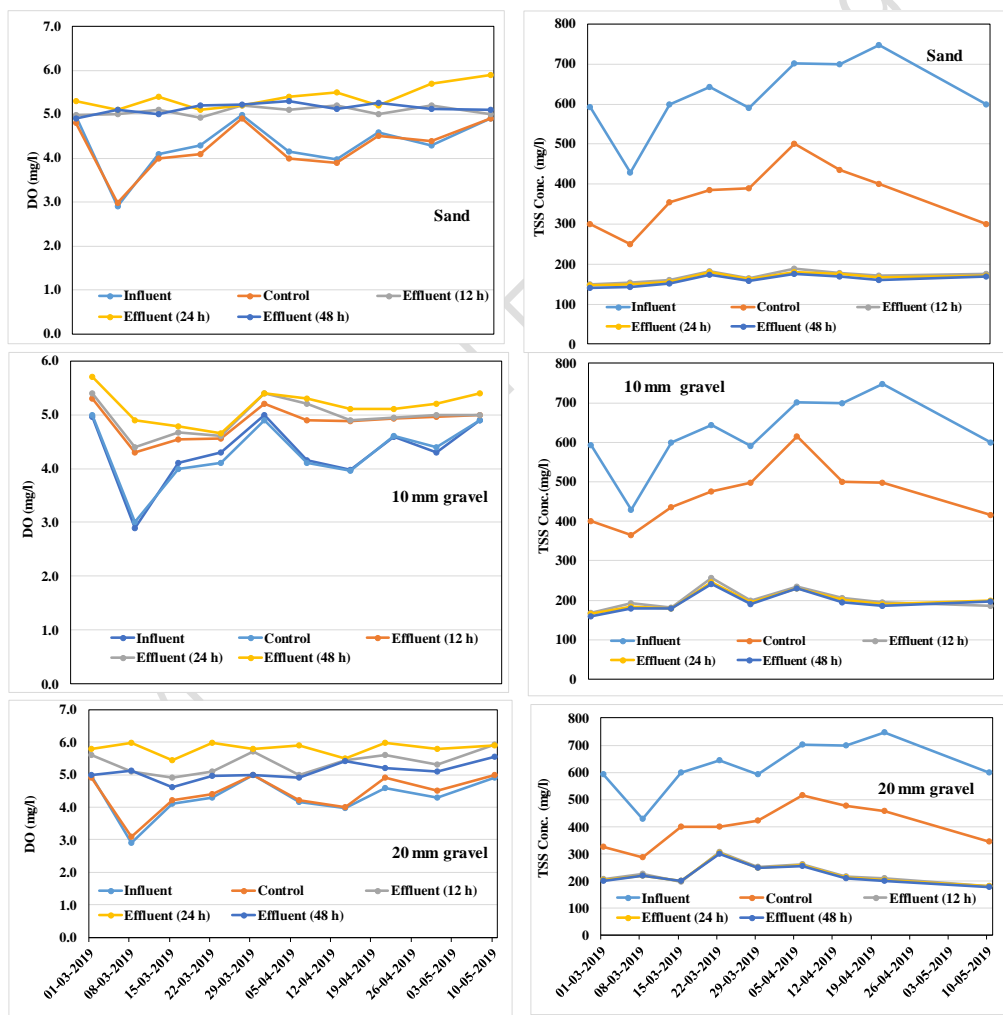


Fig. 2. Continuous DO and TSS concentrations in influent and effluent in control, CW-A, CW-B and CW-C during the study period.

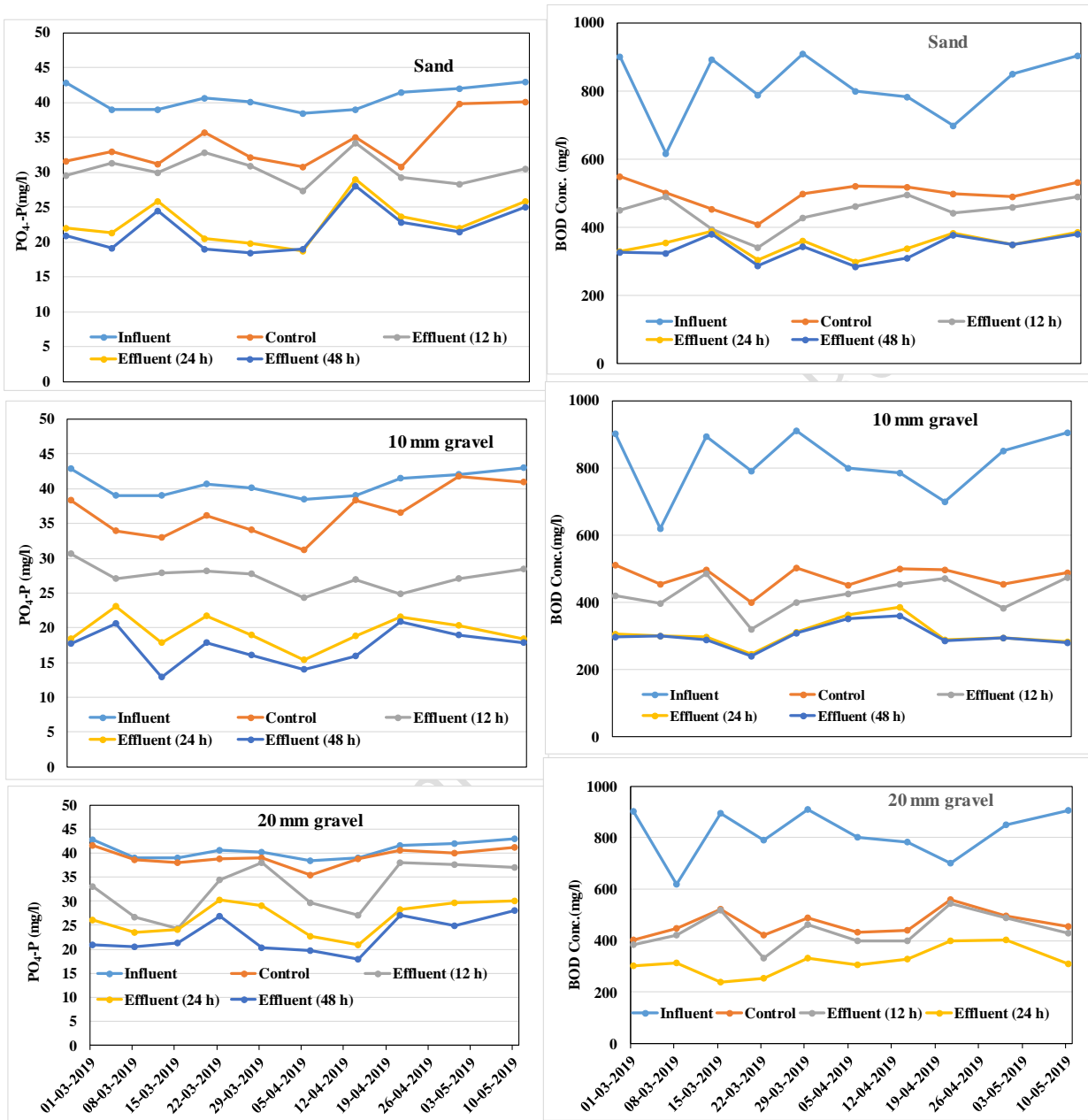


Fig. 3. Continuous $PO_4\text{-P}$ and BOD concentrations in influent and effluent in control, CW-A, CW-B and CW-C during the study period.

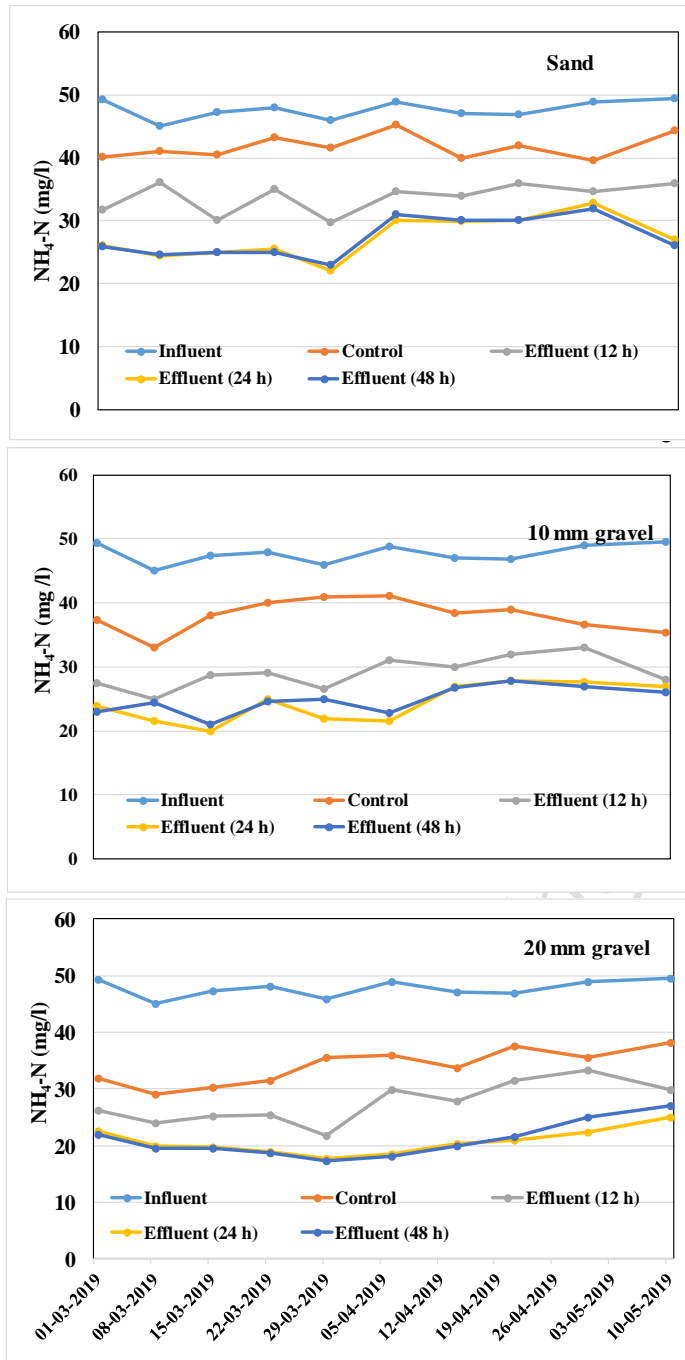


Fig. 4. Continuous $\text{NH}_4\text{-N}$ concentrations in influent and effluent in control, CW-A, CW-B and CW-C during the study period.

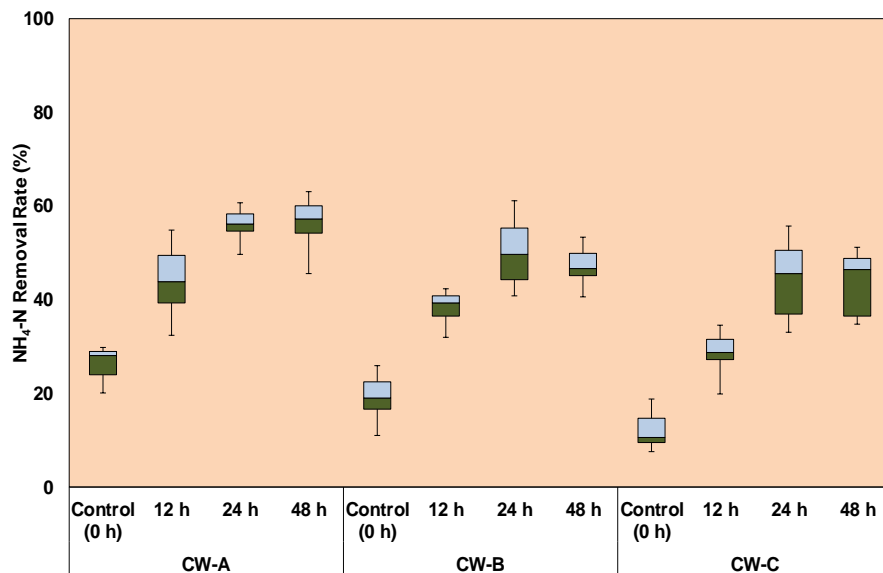


Fig. 5. $\text{NH}_4\text{-N}$ removal rates at different HRTs in VF CW system.

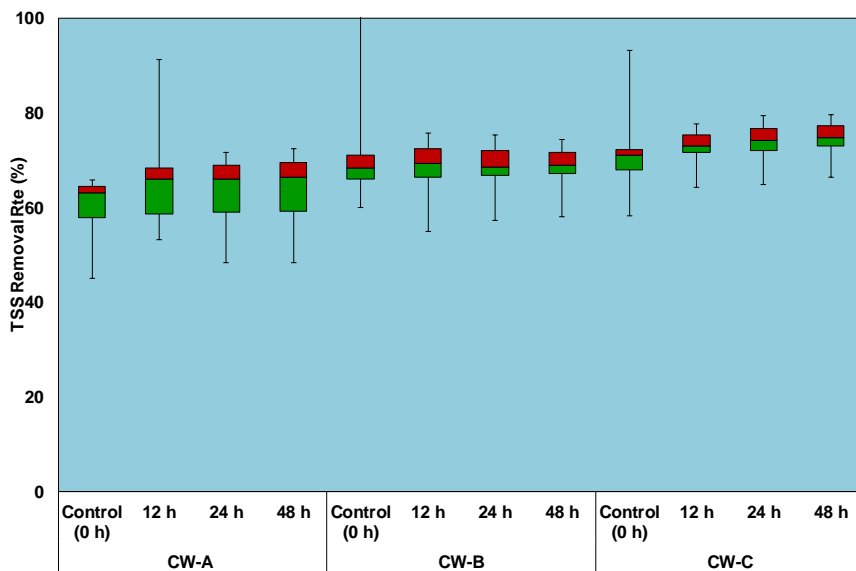


Fig. 6. TSS removal rates at different HRTs in VF CW system.

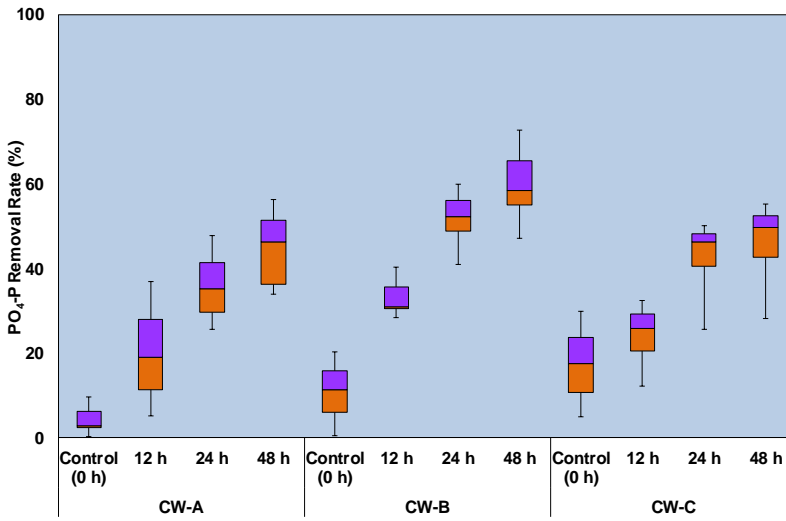


Fig. 7. $\text{PO}_4\text{-P}$ removal rates at different HRTs in VF CW system.

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2 **Table 1.** Comparative Table Representing HRT Studies Carried Out by Various Researchers

Reference	Design	Type of wastewater	HRT	Removal rate (%)						
				BOD	COD	PO ₄ -P	TN	NH ₄ -N	NO ₃ -N	TSS
Thalla et al. [47]	VF CW	Domestic wastewater	12 h	68	50	75	-	67.2	47.5	-
			24 h	83	65	90	-	84.4	66.8	-
Zhu et al. [49]	VFCW	Domestic wastewater	48 h		85	77.9	89.9	-	-	81.2
Raphael et al. [48]	VFCW	Raw gray water	3 d	35.4	56.7	65	92	-	-	59.6
Stearman et al. [36]	SSF CW	Nursery irrigation runoff	0.7 d	-	-	10.0	70.8	-	-	-
			1.2 d	-	-	7.7	71.8	-	-	-
			1.9	-	-	-1.7	69.9	-	-	-
Arias et al. [35]	SSF CW	Domestic sewage	7 d	-	-	45.6	-	-	-	-
Ghosh and Gopal [45]	VF CW	Dairy processing industry wastewater	1 d	30.4	30.8	26.1	35.4	81.0	40.8	80.2
			2 d	69.6	72.2	55.6	80.5	92.9	85.8	90.2
			3 d	82.2	85.0	63.3	84.9	99.4	95.9	95.6
			4 d	93.9	97.1	92.7	94.6	92.7	98.6	98.8
Akratos and Tsihrintzis [30]	HF CW	Synthetic wastewater	6 d	87.9	88.7	29.3	44.8	19.1	-	-
			8 d	94.0	92.6	81.8	80.2	77.9	-	-
			14 d	91.2	92.8	90.3	78.9	74.9	-	-
			20 d	90.2	91.8	98.5	85.9	87.5	-	-
Sirianuntapiboon et al. [46]	VF (UF) CW	Domestic wastewater	0.75 d	88.0	84.0	84.0	63.0	30.0	-	42.0
			1.5 d	91.0	82.0	89.0	67.0	64.0	-	89.0
			3 d	91.0	90.0	93.0	84.0	62.0	-	76.0
Toet et al. [18]	SF CW	STP effluent	0.3 d	-	3.5	-4.5	14.3	16.9	21.2	-
			0.8 d	-	0.5	-10.1	24.3	29.2	29.8	-
			2.3 d	-	0.9	5.4	41.4	65.1	44.0	-
			9.3 d	-	-12.8	1.6	63.1	87.0	85.6	-

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