Desalination and Water Treatment
www.deswater.com

doi: 10.1080/19443994.2013.877850

54 (2015) 103–112
April

Taylor & Francis
Taylor & Francis Group

Pollutant removal efficiency of mesocosm HSSF-constructed wetlands treating highway runoff with different filter materials and HRT

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Received 16 November 2012; Accepted 10 December 2013

ABSTRACT

In this study, cobble-stone (CS) and broke-stone materials were used as filter media in horizontal sub-surface flow constructed wetlands (HSSF-CWs) for highway runoff treatment. These wetlands were planted with *Phragmites* spp. plants. The Lukou viaduct section of the Nanjing airport expressway was selected for collection of highway runoff. The HSSF-CWs were subjected to two hydraulic retention times (HRT) of 12 and 24 h with different hydraulic loads and influent flow rate. Based on the results, the CW with broken-stone (BS) substrate achieved significantly higher removal efficiency for SS (75 and 55%), chemical oxygen demand (45 and 50%), NH_3-N (78 and 88%), and total phosphorus (77 and 34%) in 12 and 24 h HRT compared with CS substrate wetlands. The CW with CS substrate showed lower removal efficiencies than the BS substrate wetland. The removal efficiencies for Cu, Zn, and Pb for the 12 h HRT were 62, 80, and 9%, respectively, and the concentrations of Cu, Zn, Pb, and Cr were generally under the standard limits. This work will aid design and improvement of constructed wetlands for small scale treatment of highway runoff.

Keywords: Highway runoff; HRT; HSSF-CWs; Pollutants; Filter substrates

1. Introduction

Highway runoff pollution has become a crucial environmental issue in recent years. Urban surface runoff particularly highway runoff generates heavy pollutant loads [1]. It has been identified as one of the various potential pollutant sources that are detrimental to the surface water quality. Even though highways may occupy only 5–8% of the catchment area, they can contribute 50% of suspended solids, 16% of hydrocarbons, and 35–75% of heavy metals [1]. Research on highway runoff in China started late, toward the end of the last century. Even then, very little attention focused on pollution caused by road runoff, especially its treatment by small-scale constructed wetlands (CWs) under local environmental conditions [2,3]. CWs are considered more reliable and effective control measures compared to other systems and are suitable because they are passive systems that reduce pollutants from various wastewaters [4]. Many previous studies reveal the importance and pollutant removal efficiency of various filter substrates, e.g. gravel, zeolite, shale, ceramic filter, peat, fly-ash bricks, as well as cobble-stone (CS)

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and broken-stone (BS) filter substrates in CWs [5–10]. The removal efficiency of these filter materials ranged from 45 to 95% for various pollutants such as total suspended solids (TSS), chemical oxygen demand (COD), biological oxygen demand, total nitrogen (TN), ammoniacal-nitrogen (NH_3-N), total phosphorus (TP), and heavy metals [4,6,11-13]. The pollutant removal efficiency of various filter substrates depends on many factors such as design of the CWs, physical and chemical properties of the filter materials (such as porosity and surface area, etc.), planted or unplanted CWs, hydraulic loadings, and hydraulic retention times (HRT) [6,12,14,15]. A sedimentation tank is also an important means of pre-processing and controlling road runoff pollution [11] and could effectively remove suspended particles and parts of other complex pollutants present in surface runoff. The purification mechanism in CW is very complicated, where physical, chemical, and biological processes play important roles in pollutants removal [4,14,16].

Treatment efficiency of CWs relies on various aspects such as design, HRT, flow pattern, water depth, pollutant loads, and wetland plants [5,17]. The treatment efficiency of CWs is strongly influenced by the flow pattern, hydraulic loading, and HRT conditions [18]. Previous studies suggested that CW performance in treating highway runoff was generally a function of hydraulic loading rate and HRT, also affected by storm intensity, runoff volume, size of the CW (area and volume), and type of filter media [14]. Horizontal sub-surface flow constructed wetlands (HSSF-CWs) used for treatment of various stormwater runoff have also been designed to achieve a specific HRT based on the maximum expected amount of stormwater volume received [19]. However, in case of highway runoff, it is very difficult to estimate the stormwater volume due to its direct relation to the amount of rainfall and weather conditions [16]. Achieving a specific or minimum HRT and its importance in highway treatment process has been discussed repeatedly [11]. Highway runoff influent to the HSSF-CWs continues its way under the surface of the filter bed in a more or less horizontal path until it reaches the outlet zone. During this passage, wastewater comes into contact with a network of aerobic, anoxic, and anaerobic zones. The aerobic zones occur around roots and rhizomes that provide oxygen to the substrate [3,12,20]. The organic removal increases with long run time mainly due to the increase in microbial populations in CWs [13,21]. Various literature reviews on the transformation and removal processes of nutrients in HSSF-CWs has been reported in previous studies [12].

Plenty of work has been carried out on highway runoff treatment by CWs in developed countries, but there is still a lack of sufficient research work in China, especially on small-scale treatment of highway runoff by HSSF-CWs. Hence, the current study was carried out using a lab-scale evaluation of removal efficiency for HSSF-CWs. The main objectives were to examine the efficacy and capacity of CS and BS substrates in HSSF-CW for highway runoff treatment and to evaluate the optimal design factors under different HRT and hydraulic loads. The removal efficiency combined with information of HRT and various filter substrates may be useful in the design and improvement of small-scale HSSF-CWs for future applications in densely populated areas where land for stormwater treatment is limited.

2. Methodology

2.1. Sampling and analysis

The Lukou viaduct section of the Airport Expressway of Nanjing, China was selected as the sampling area for collection of highway runoff. Asphalt pavement material was used for construction of this expressway and the average hourly traffic load on this section was 270 vehicles/h. The catchment area was 960 m^2, and the surrounding land uses included residential, agriculture, and transportation. Highway runoff was collected in a sedimentation tank and raw highway runoff samples were collected to analyze for its physicochemical characteristics. The experiment was conducted in year 2008–2009. Highway runoff samples were collected during various rainfall events and sampling interval times were set according to rainfall duration and runoff rate. Each sample volume was set at 1 L. Simultaneously, rainfall characteristics were also investigated with the help of data recorded by a JS-2 siphon rain gauge (not included in this paper). Influent and effluent highway runoff samples from CWs were studied in the laboratory to assess the changes in the concentrations of TSS, COD, NH_3-N, TN, TP, and heavy metals. Highway runoff samples were analyzed immediately (within 1–2 h) after sampling according to the methods described by the APHA manual [22]. Statistical analysis and graphical work were completed with Origin 7.0 (OriginLab) and data were analyzed through one-way ANOVA to compare the performance of all parameters studied from both CWs. Particle size analysis was also carried out in the current study as particle size interval is one of the most important factors in removal process of heavy metals. The volume mean diameter, which reflects the particle size, was measured by laser diffraction (Malvern Mastersizer 2000).

2.2. Experimental CW units

The design for CW units was based on previous studies, where the removal efficiency of CWs under 12 and 24 h HRT conditions was investigated [11]. The design of both experimental CW units was similar except the type of filter material in the CW substrates (Fig. 1(a–b)). Two sets of experimental CW units were designed with dimensions of 80 cm in length, 40 cm in width, and 65 cm in height. The CW units were designed with 12 and 24 h HRT conditions with two different flow rates in each unit and were built with Plexiglas and PVC sheets. In order to separate the perforated plate and treatment area in the HSSF-CWs, inlet valves were setup at a height of 10 cm in each unit. The first CW unit (CS-CW) was filled with CS substrate as a filter media (matrix size 6–8 cm) with short flow inhibition to prevent the clogging problem. The BS material was used as the filter media in the second CW unit (BS-CW) with a matrix composition consisting of a 15 cm thick top soil layer, a 15 cm layer of fine particles (1–2 cm in diameter), followed by a 10 cm layer with medium size particles (2–4 cm in diameter), and a 10 cm layer with larger particles (6–8 cm in diameter). The substrate layering scheme implemented in CS-CW unit was similar to the BS-CW unit. The CS and BS filter materials are very different in their physical and chemical properties such as porosity, surface area, and absorption capacity. The BS filter material has higher absorption capacity as well as larger surface area. The porosity of BS and CS substrates was 0.37 and 0.36, respectively.

The different sized filter materials were used in the substrate to provide better plant growth, avoid clogging, and to improve aeration conditions in the CWs. A valve was setup at a height of 20 cm to regulate the water level. The CW units, storage tank, and metering pumps were placed in an indoor environment in order to have a controlled experiment. Highway runoff was collected in a storage tank for 2 h to simulate the pre-sedimentation process prior to intermittent feeding in the CWs. Another regulatory valve was also setup at a height of 30 cm to control the HRT and water level in CW units. These wetland units were planted with *Phragmites* spp. plants with 8–10/m^2 density but the plant growth was not satisfactory due to the inadequate indoor lighting time. A total of 23 highway runoff samples were collected for investigation during the current study. The influent flow rates were set to 2.96 and 2.88 L/h, whereas hydraulic loads were set to 29.6 cm d^{-1} in CS-CW and 28.8 cm d^{-1} in BS-CWs for the 12 h HRT. The influent flow rates were set to 1.48 and 1.44 L/h, whereas hydraulic loads were set to 14.8 cm d^{-1} in CS-CW and 14.4 cm d^{-1} in BS-CW for the 24 h HRT (Table 1). The HRT was calculated by the following formula:

$$t = \frac{n \times L \times W \times D}{Q} \qquad (1)$$

where

n effective porosity of media (% as a decimal)

L length of bed (ft)

W width of bed (ft)

D average depth of liquid in bed (ft)

Q average flow through bed (ft^3/day)

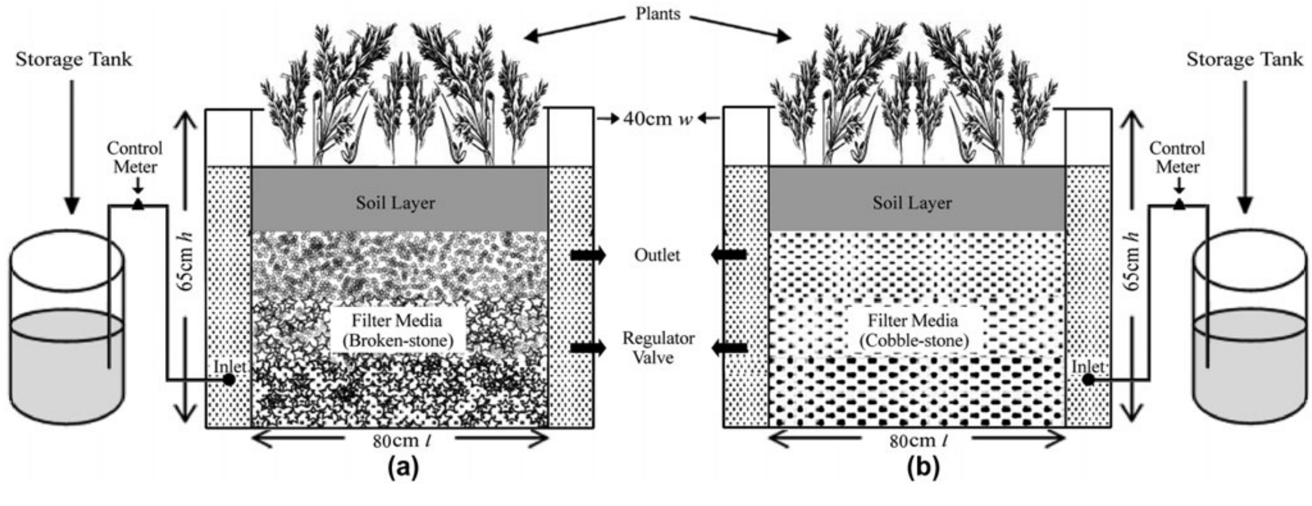

Fig. 1. Schematic diagram of HSSF-CW units (a) BS-CW unit and (b) CS-CW unit.

Table 1
Hydraulic parameters for HSSF-CW units

Parameters		CS-CW	BS-CW
Porosity		0.37	0.36
12 h HRT	Water flow rate (L/h)	2.96	2.88
	Hydraulic loading (cm d^{-1})	29.6	28.8
24 h HRT	Water flow rate (L/h)	1.48	1.44
	Hydraulic loading (cm d^{-1})	14.8	14.4

Pollutant removal efficiency of the HSSF-CWs was calculated by following formula:

$$\eta = \frac{C_o - C_e}{C_o} \times 100\% \qquad (2)$$

where

n	removal efficiency
C_o	influent concentration
C_e	effluent concentration

3. Results and discussion

The influent and effluent concentrations were studied for suspended solids, COD, ammonia nitrogen, TN, TP, and heavy metals to determine the performance of HSSF-CWs with different HRT, hydraulic loads, and flow rates. Results are presented in Table 2 with the mean values of the pollutants in the influent and effluent. Figs. 2–4 represent the removal efficiency of the CW units for major pollutants presented in highway runoff. Nitrogen was presented mainly in the forms of ammoniacal nitrogen (dissolved ammoniacal nitrogen and ammonium ion) and TN. The pH and temperature of raw highway runoff ranged between 6.97–7.82 and 11.9–21.6℃, respectively. Data analysis showed a significant difference between influent and effluent values for most of the parameters. Overall, results for removal efficiency of both wetlands operated under 12 and 24 h HRTs showed that there was no significant difference ($p > 0.05$) between the two HRTs for most of the parameters studied. The particle size interval of primary highway runoff ranged from 20 to 75 μm in current study. The BS-CW showed higher removal capability than CS-CW for particles in this particle size interval. The main removal path in the studied CWs was screening and settling of the pollutants, which are both physical processes. In the low HRT, the level of the biological decomposition of the substances presented in the effluent compared to their physical retention must

Table 2
Average and statistical deviation values of highway runoff quality and pollutant removal efficiency of CS-CW and BS-CW units

Water quality parameters	Raw highway runoff quality	HRT (12 h)					HRT (24 h)				
			CS-CW		BS-CW			CS-CW		BS-CW	
		Influent•	Effluent	Removal %	Effluent	Removal %	Influent	Effluent	Removal %	Effluent	Removal %
TSS#	42–290	64* ± 26.08	22ˇ ± 14.68	63	15ˇ ± 8.33	75	87* ± 47.62	25ˇ ± 17.74	46	25ˇ ± 19.40	55
COD#	67–310	132* ± 41.12	80ˇ ± 38.39	40	67ˇ ± 15.64	45	106* ± 57.23	69ˇ ± 40.47	37	49ˇ ± 18.34	50
NH_3-N#	1.58–4.8	3.76* ± 1.83	1.49ˇ ± 1.24	67	0.81ˇ ± 0.62	78	2.02* ± 1.75	0.63ˇ ± 0.74	71	0.28ˇ ± 0.35	88
TN#	5.9–11.2	8.2* ± 3.49	7.1* ± 3.18	11	7.9* ± 3.44	−6	3.7* ± 2.85	2.5* ± 1.77	18	3.8* ± 2.42	−28
TP#	0.2–0.5	0.35* ± 0.12	0.17ˇ ± 0.09	48	0.08ˇ ± 0.08	77	0.14* ± 0.13	0.12* ± 0.09	−7	0.07* ± 0.05	34
pH	6.97–7.82	7.63	7.48	–	7.85	–	7.22	7.19	–	7.61	–
Temp. °C	11.9–21.6	10–22					22–30				

• Average (mean) values of influent and effluent concentrations;

#Values are in mg L^{-1};

*, ˇ Values of each parameter marked by different symbol are significantly different ($p < 0.05$).

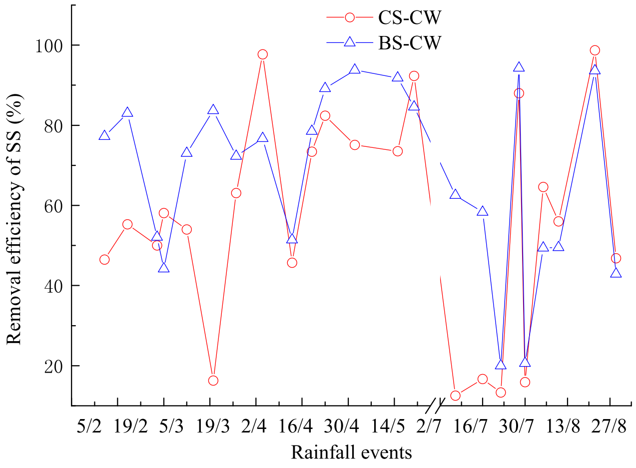

Fig. 2a. Suspended solids removal efficiency of CS-CW and BS-CW units.

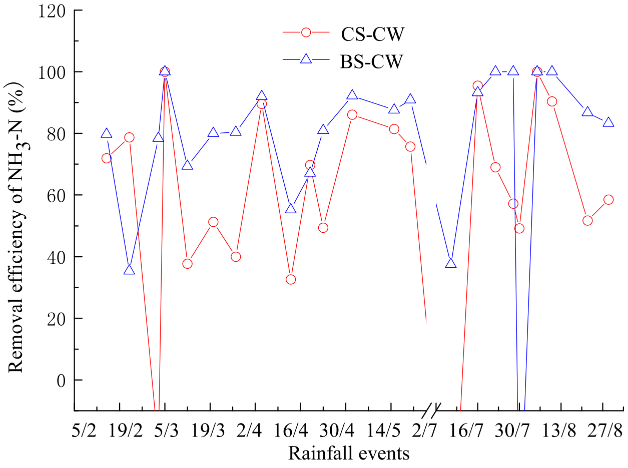

Fig. 3a. NH_3-N removal efficiency of CS-CW and BS-CW units.

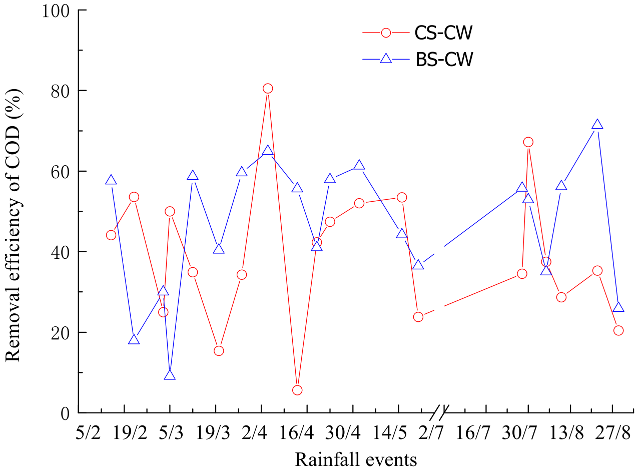

Fig. 2b. COD removal efficiency of CS-CW and BS-CW units.

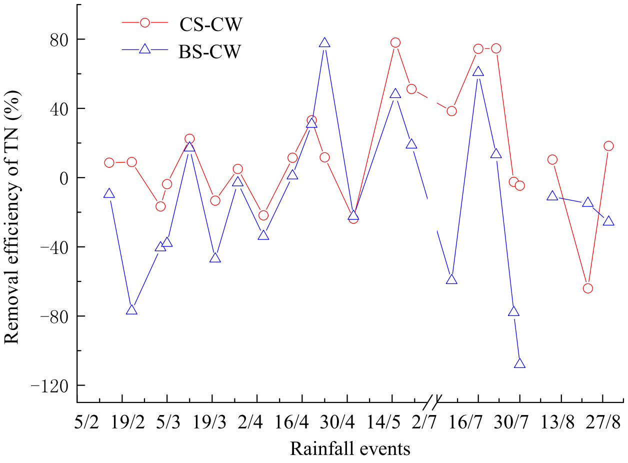

Fig. 3b. TN removal efficiency of CS-CW and BS-CW units.

be low. It is also reported in previous studies that suspended solids play a major role in the fate of the contaminants and nutrients in the highway runoff [11]. This is further supported by the results from the current study since the HRT differentiation did not result in significant removal efficiency performance differentiations.

3.1. pH and temperature

The average pH values of influent to and effluent from the CS-CW and BS-CW units were 7.63 and 7.48 in 12 h HRT and 7.22 and 7.19 in 24 h HRT, respectively. The pH values decreased in the initial stage of CS-CW but increased slightly in BS-CW. The possible reason for the increase in pH could be a decrease in the oxygen requirement in the CW system. Kadlec and Knight reported the buffer states of CWs, where reduction of ferric-ion occurs if the system overflows [4]. Effluent values were within the range of standard limits which may create favorable conditions for metal precipitation of inorganic compounds. The influent temperature in both CW units ranged between 10 and 30°C. Bulc and Slak reported that low temperatures can have a considerable effect on water quality, as a result of a decrease in biological decomposition [23]. Overall results show that removal efficiency was lower at lower temperature and higher at higher temperature. The possible reason could be the increase in biofilm thickness and lower diffusion of

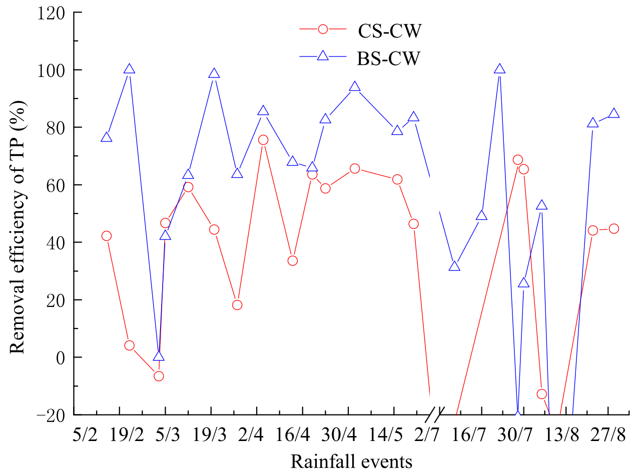

Fig. 4. TP removal efficiency of CS-CW and BS-CW units.

dissolved oxygen to micro-organisms. Previous studies also suggest that the actual effect of temperature on removal efficiency could be determined by longer operation of CWs [24,25].

3.2. Suspended solids and COD

Most of the large particles and other solids from highway runoff were removed in a pretreatment tank. Performance efficiency for suspended solids is presented in Fig. 2(a). The main removal processes in the CWs studied were physical processes, such as screening and settling. Previous studies showed that suspended solids play a major role in the fate of contaminants and nutrients in highway runoff [14]. This phenomenon is further supported by the fact that HRT variations did not result in significant performance variations in this current research, where removal efficiencies in CWs show no significant difference. Bulc and Slak reported that accumulation of solid particles depends on average traffic conditions [23]. They reported that fractions of heavy metals and other pollutants depend on particle size and that concentrations are generally higher on smaller particles (10 μm). The average particle size of solids in primary highway runoff ranged from 20 to 75 μm in current study. The average concentration of SS in the influent was 63 mg/L, whereas the effluent concentrations of CS and BS-CWs were 22 and 15 mg/L for the 12 h HRT conditions. The average effluent concentrations of SS in 24 h HRT conditions in both CW units was similar (25 mg/L). The BS-CW shows good SS removal efficiency in low HRT conditions, where average removal rates in both wetlands were 63 and 75%, and treatment effect in BS-CW was slightly better than CS-CW. he BS filter material has larger surface area as well as more porous compared to CS substrate. Sedimentation and adhesion are also main factor for matrix and suspended solids removal [14]. Therefore, removal efficiency was lower in higher HRT than at lower HRT conditions as it decreased to 46 and 55% removal, probably due to the effects of initiation of several other chemical and biological processes. The possible biological and chemical processes include dilution, decomposition, microbial oxidation, ion exchange, precipitation, and adsorption [25]. The removal of SS was more stable under 12 h HRT conditions and there was a substantial change in SS removal in BS-CW and CS-CW which may be due to the different HRT and substrate properties. The surface property of filter media is the main factor that affects sedimentation process [14].

Results show that the removal efficiencies of SS from both CW units were comparatively higher (63 and 75%) in lower HRT (12 h) with slightly higher flow rate and hydraulic loadings. The effluent values of SS in the current study fluctuated parallel to the changes in influent values. Higher removal rates in BS-CW unit show that the suspended solids may be accumulated and retained with BS substrate. Lower SS removal from CS-CW shows that CS filter material is not as efficient as BS filter material. It also proves that physicochemical properties of BS filter are comparatively better than CS material. The removal of SS in the current study was lower than several other studies in which 70–75% [5], >84% [23], and 57–82% [24] removal were observed. In the context of the results from this study, it may be presumed that both CS and BS filter media are efficient for removal of suspended solids from highway runoff. However, lower removal efficiencies suggest further long term monitoring and operation of CWs.

The findings for COD removal efficiency are presented in Fig. 2(b). The average concentration of COD in the influent was 132 mg/L, whereas the effluent concentrations of CS and BS CWs were 80 and 67 mg/L for 12 h HRT, respectively. The average removal rates of the COD in the CS-CW and the BS-CW for a 12 h HRT were 40 and 45%, and; in general, the BS-CW showed slightly better performance than the CS-CW. The dissolved and particulate COD removal rates were 26 and 55%, respectively, in the CS-CW, whereas the removal rates were 41 and 69% in the BS-CW for 12 h and 24 h HRTs. The removal efficiency of dissolved COD and total COD was poor in general because removal of dissolved COD mainly depends on the matrix and microbial adsorption. These results suggest that the growth of micro-organisms in the wetland was not ideal, and it is more difficult to grow micro-organisms with the chemical and biological quality of highway runoff water. Indeed, one possible reason that has been suggested in the literature for low COD removal is the lower microbial activities and poor plant growth that can decrease the oxygen availability for microbial growth in CWs [5,14].

The average concentration of COD in the influent was 106 mg/L, whereas effluent concentrations of CS and BS CWs were 69 and 49 mg/L for a 24 h HRT. The total COD removals in the CS-CW and the BS-CW were 37 and 50%, respectively, when the HRT increased to 24 h. Results reveal that the impact of HRT, flow rates, and hydraulic loading was minimal on COD removal as there was little difference in COD removal efficiencies in both CWs under 12 and 24 h HRT conditions. COD removal relies mainly on microbiological degradation by the attached matrix and plant roots. The results are consistent with the findings of Terzakis et al., who reported 49–54% COD removal under similar HRT conditions [11], which were very low compared to other findings, where COD removal varied between 64–82% [4,21]. Differences between these studies could be due to limited oxygen in the HSSF-CWs and it could be avoided to improve COD removal but may affect performance for other pollutants [4,6]. It can be concluded from the present investigation that COD removal efficiency in the BS-CW was better than in the CS-CW. The higher surface area and chemical properties of the BS filter media could be the reason for this trend that enhances various chemical processes such as nitrification–denitrification [24].

3.3. NH_3-N and TN

The removal of ammoniacal nitrogen in CWs depends on many processes and previous studies show that the removal of nitrogen is a complicated process [19,21–23]. Nitrification followed by denitrification, assimilation, mineralization of organic nitrogen, ammonia volatilization, and adsorption of ammonia onto substrates are the key pathways for nitrogen removal [20,21,23,24]. The major problem for nitrogen removal in various CWs is the availability of oxygen for nitrification and subsequent availability of carbon sources for biological denitrification [26]. Generally, nitrification is more efficient in free surface water flow CWs than in subsurface flow CWs. Poor plant growth and porosity of the filter substrate are also major factors for low oxygen availability in CWs [24]. NH_3-N removal efficiencies of the HSSF-CWs are presented in Fig. 3(a). The average influent concentrations of NH_3-N for 12 and 24 h HRTs were 3.76 and 2.02 mg/L, respectively. The effluent concentrations of CS and BS-CWs were 1.49 and 0.81 mg/L at the 12 h HRT and 0.63 and 0.28 mg/L, respectively, at the 24 h HRT. The removal efficiencies for NH_3-N in HSSF-CWs for the 12 h HRT using CS and BS filter substrates were 67 and 78%, respectively. However, statistical tests show that the NH_3-N removal efficiency was not significantly better in the BS-CW. The majority of the NH_3-N was present in dissolved form (3.37 mg/L), and particulate organic nitrogen (1.37 mg/L) in the highway runoff waters and removal rates were 62 and 64% in the CS-CW, while 80 and 72% in the BS-CW. NH_3-N removal increased to 71 and 88%, respectively, in both CS-CW and BS-CW with an increase in HRT (24 h). The removal of NH_3-N in the CS-CW increased by only 4%, while BS substrate showed higher removal efficiency with a 10% increase. These changes indicate that the adsorption of ammonia with BS-CW changed markedly, whereas microbial activities were minimal in the CS-CW with changes in HRT. These results show that microbial nitrification and chemical stability of the substrates were the leading factors. It is important to note that pH values were not high. The average pH values ranged between 7.63 and 7.48 in both CWs; therefore, NH_3-N loss through vitalization would be limited since it generally requires a pH of 9.3 or greater and the fact that the HSSF-CWs have no free water surface [4]. Hence, algal activity is negligible in these systems and therefore, pH values do not increase [26,27].

TN effluent concentrations of the CS-CW and BS-CWs were 7.1 and 7.9 mg/L for the 12 h HRT and 2.5 and 3.8 mg/L for the 24 h HRT (Table 2). Results (Fig. 3(b)) show that the CS-CW was slightly better than the BS-CW for TN removal. The average removal rates in both wetlands were 11 and −6% for the 12 h HRT. The removal of TN shows very different trends (increasing and decreasing) for these two different substrates with both higher and lower hydraulic conditions. The lower amount of COD and nitrogen in highway runoff may be the main reason for lower concentrations of TN as the limited carbon source and slow microbial degradation process affects denitrification [21,26]. The macrophytes have a limited role in nitrogen removal in HSSF-CWs that have shorter retention time [21]. The CS-CW shows better TN removal due to its effectiveness in the nitrification process. These trends indicate that TN increases as COD gradually degrades, whereas ammoniacal nitrogen decreases due to nitrification. The results for a 24 h HRT show similar trends to the 12 h HRT conditions, where removal efficiency changed to 18 and −28%, respectively, in both the CS and BS-CWs. This trend in TN concentration was probably due to the increase in ammonia adsorption and nitrification, which increases the concentration of residual nitrogen. The TN removal was very low in this study compared with other studies [12,20,24,25], that reported 38–70% removal efficiencies for CWs. The HSSF-CWs could achieve better nitrogen reduction if wetlands can operate with higher denitrification capacity [21,24].

3.4. TP

Results for TP removal are presented in Table 2 and overall findings showed that phosphorus removal was not so high in the current study. The influent concentrations in lower and higher HRT experimental conditions were 0.35 ± 0.12 and 0.14 ± 0.13 mg/L, respectively. The effluent concentrations of TP in the CS-CW and the BS-CW were 0.17 and 0.08 mg/L (12 h HRT) and 0.12 and 0.07 mg/L (24 h HRT), respectively. The phosphorus removal efficiency trends are presented in Fig. 4. The effluent concentrations of dissolved and particulate TP were 0.06 and 0.28 mg/L, respectively. Results showed that average TP removal rates in CS-CW and BS-CW were 48 and 77% for the 12 h HRT; where BS-CW showed statistically not, though better TP removal than the CS-CW. The dissolved and particulate phosphorus removal rates were 28 and 64% in the CS-CW, and 64 and 73% in the BS-CW. The adsorption capacity of the particles in the BS substrate led to good removal efficiency of particulate phosphorus. This result reveals that comparatively high pH values are conducive to dephosphorization. Many previous studies reveal that low phosphorus removals may be due to the capacity of the filter media to absorb or precipitate the incoming phosphorus [20].

The TP removal shows a different trend under longer HRT and lower flow rates and hydraulic loadings in both tested CW units, where the concentration of TP was increased in the CS-CW (−7%), while the BS-CW shows low TP removal (34%) compared with 12 h HRT. This trend is consistent with other research findings where studies have shown that different matrices have different result for phosphorus release, generally varying between 40 and 80% [11,15,20]. This phenomenon shows that CS may have a different phosphorus adsorption mechanism than a BS substrate. CS substrate has a double-layer adsorption capacity (i.e. potential for physical adsorption, ion exchange, or deposition of phosphorus) that continues to increase the concentration of phosphorus in a saturated solution while the amount of phosphorus adsorption is still increasing to a different degree. It is known that concentrations of phosphorus, generally present as orthophosphate, are very low in highway runoff. Therefore, the adsorption capacity and adsorption characteristics are important for the choice of matrix, but one must also consider the matrix analytical rate [4,20,25]. It can be concluded from the results that phosphorus removal was higher in lower HRT and slightly higher flow rates and hydraulic loadings compared with higher HRT and lower flow rates and hydraulic loading conditions.

3.5. Heavy metals

Results from current study indicate clearly the presence of heavy metals in the atmosphere close to the studied highway, probably including resuspended particles from the road. The concentrations of heavy metals in highway runoff may depend on various factors such as wind, rainfall characteristics, and volume sampled [26]. Many parameters play a very crucial role in heavy metal removal mechanisms, with pH affecting the chemical properties of various substrates [27,28]. Highway runoff in the present study had higher pH (7.22–7.63) values, which affects metal speciation. Particle size intervals also play an important role in heavy metal removal from CWs [23]. The average particle size of solids in current study ranged from 20 to 75 μm. Results reveal that the removal rates of Cu in CS and BS-CWs were 63 and 62%, with treatment being very similar for the two different substrates. The removal rates of Zn in the CS and the BS-CWs were 63 and 82% (12 h HRT) and 81 and 60% (24 h HRT). Results reveal that Zn removal in the CS-CW was increased with larger retention time, whereas they decreased in the BS-CW. This reverse trend probably occurred due to the pH of the runoff water and the retention time. Both substrates showed better performance at higher pH values as Zn is present in particulate form, but HRT and pH affects their particle adsorption properties [13,27–29]. The CS-CW shows better Pb removal under the 12 h HRT, whereas BS-CW shows very weak performance. Results were quite different under the 24 h HRT, where the concentration of Pb increased in the effluent water. The removal efficiency of Pb was 40 and 9% for the 12 h HRT in the CW-CW and the BS-CW, respectively. Lower removal efficiency possibly occurred due to the dissolution of Pb [29]. Analysis showed that the matrix absorption was balanced at lower Pb concentration and adsorption and desorption reactions were the possible reason for the trend. This trend also occurred for Cd.

Previous studies reveal that heavy metal removal is correlated with SS removal and the heavy metals removed by CWs were probably due to those attached with suspended solids. These were either screened or settled in the wetland systems. However, results from the current study do not fully support this phenomenon as removal efficiency of heavy metals was comparatively low [28]. Studies examining correlation between heavy metal removal and TSS removal suggest that higher concentrations of some heavy metals in effluent compared to influent highway runoff could be due to the surface absorption capacity of filter substrates and solids present in highway runoff [28–30]. Many previous studies show significant reduction of heavy metals from CWs [30]. The removal of lead and cadmium by clinoptilolite in solutions at various pH values had better removal at acidic pHs where the metal species were dominantly cationic [31]. Hence, pH has a significant impact on metal removal from highway runoff. Relatively, lower heavy metal removal efficiency in both experimented CWs shows that there is a need for further research to optimize Pb and Cd removal.

4. Conclusion

Results indicated that the performance of the CW with BS substrate was better than the CW with CS substrate at treating highway runoff. The BS-CW achieved mean removal efficiencies for SS, COD, NH_3-N, and TP of 75, 45, 78, and 77% for a 12 h HRT and higher HLR, whereas 55, 50, 88, and 34%, respectively, for the 24 h HRT and lower HLR conditions. CS-CW and BS-CW showed low removal efficiency for TN and COD. Removal efficiencies for heavy metals were similar in both CS-CW and BS-CWs. Removal efficiencies were high for Zn and low for Pb in the BS-CW. Overall, the findings in the current study show that the CWs perform similarly in lower and higher HRT conditions. Thus, the 12 h HRT CW is recommended since it can treat higher amounts of highway runoff with similar pollutant removal efficiencies. Additionally, the 12 h HRT CW is preferred since it requires less land and lower construction costs. The findings from this study could be useful for implementation of HSSF-CWs for small-scale treatment of highway runoff in fast growing infrastructure and densely populated areas, where availability of land is a big challenge.

Acknowledgments

This study was supported by National Natural Science Foundation of China (NSFC, No. L50909019). The authors would like to thank Prof. Katz Lynn, University of Texas, Austin, USA and Dr Ryan Winston, NC States University, USA for their valuable suggestions and helping us to improve the manuscript. We would also like to thank anonymous reviewers for their insightful comments for improving our manuscript.

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