Desalination and Water Treatment
www.deswater.com

doi: 10.1080/19443994.2014.922315

54 (2015) 3534–3543
June

Taylor & Francis
Taylor & Francis Group

Nitrogen reduction in stormwater from livestock lots by vertical subsurface flow wetlands packed with woodchips

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Received 15 January 2014; Accepted 14 March 2014

ABSTRACT

This study was done to investigate the performance of vertical subsurface flow (VSF) wetlands with additional carbon sources in reducing the nitrogen in stormwater from livestock lots. Three experimental lab-scale VSF wetlands packed with woodchips were constructed. The wetlands were operated with number of dry days (NDD) of 2, 4, and 8, respectively, for a duration of 136 d. The average removal efficiencies of total nitrogen (TN) were 26.2, 34.1, and 50.0% at NDD as 2, 4, and 8, respectively. The average nitrogen removal rate based on woodchips volume was 3.6 g N m^{-3} d^{-1} for NDD of 2, 2.1 g N m^{-3} d^{-1} for NDD of 4, and 1.7 g N m^{-3} d^{-1} for NDD of 8. Nitrification and denitrification were the major mechanisms of nitrogen reduction in the studied wetlands. The contribution of other pathways to nitrogen removal was small. The removal of total Kjeldahl nitrogen (TKN) and TN was not affected by the influent pH, whereas the removal of TKN was enhanced by the increase in influent alkalinity. Nitrification was promoted by the abundant oxygen supplied during the course of recirculation, whereas denitrification might be suppressed to some degree by the oxygen.

Keywords: Stormwater; Livestock lots; Vertical subsurface flow wetlands; Woodchips; Carbon sources; Nitrogen removal

1. Introduction

Usually, storm run-off from livestock zones is rich with nitrogen [1], which might be carried into the surface water in the form of ammonium nitrogen or nitrate [2]. The cumulative nitrogen in a discharged water body can lead to adverse ecological effects such as eutrophication, hypoxia, and algal bloom. Thus, the reduction in nitrogen transport from livestock watersheds is increasingly taken seriously.

Vertical subsurface flow (VSF) wetlands are capable of transporting a greater amount of oxygen than horizontal flow wetlands [3]. Hence, they are more effective in reducing organic matters and ammonia [4]. As a kind of typical small on-site decentralized wastewater treatment system, VSF constructed wetlands have been acknowledged as an effective and economical technology to eliminate the pollutants from wastewater and stormwater worldwide [5–7], especially in the countries with limited available land.

In order to further improve the performance of VSF wetlands, some innovative operational measures such as tidal flow and recirculation have been applied. Prost-Boucle and Molle [8] demonstrated that nitrification is strongly dependent on the recirculation rate.

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Presented at the 5th IWA-ASPIRE Conference, 8–12 September 2013, Daejeon, Korea

Sun et al. [9] showed that the ammonia nitrogen (NH_4–N) removal efficiency was increased by 51% after effluent recirculation was used in VSF wetlands. In this study, the VSF wetland with an intermittent inflow was adopted, allowing the appearance of unsaturated and saturated zones in the wetland, where nitrification and denitrification can occur.

In addition, several materials such as sucrose, methanol, and glucose have been used as carbon sources to enhance denitrification. However, the relatively inexpensive approach to relieve the limitation of carbon in denitrification is to employ solid carbon sources (e.g. wheat saw, sawdust, bark, and woodchips) [10]. Also, the employment of woodchips in a denitrification bed has been well documented for the past decade, because they are cheap, highly permeable, and readily available [11–13]. However, few studies have been conducted on woodchips-packed wetlands, especially small on-site decentralized VSF wetlands.

The objective of our research is to investigate the performance of woodchips-packed recirculated VSF wetlands operated with different dry days, on eliminating nitrogen in stormwater from livestock lots. The specific aims include: (i) to access the removal of total nitrogen (TN), total Kjeldahl nitrogen (TKN), and NH_4–N; (ii) to identify the effect of recirculation on nitrogen removal; and (iii) to investigate the environmental factors affecting nitrogen conversion.

2. Materials and methods

2.1. Wetlands design

Opaque acryl columns with a height of 1.0 m and an internal diameter of 0.10 m were used to construct the VSF wetlands. Besides woodchips, small amounts of quartz, and small vermiculite are employed to pack the wetlands (Table 1). Three column VSF wetlands with the same configuration were constructed for this study (Fig. 1). Acorus Calamus was planted on the top layer of the wetland, where sand and soil were not employed to reduce the risk of clogging.

2.2. Experimental procedures

As natural livestock stormwater was not available, synthetic stormwater was used, which was achieved by diluting piggery slurry from a local livestock farm with tap water. The TCOD concentration was controlled to around 100 mg/L.

For stormwater wetlands, inflow occurs only during wet days, and thus, the stormwater may be allowed to rest in the column during dry days until the next rainfall event. The number of dry days (NDD) is similar to hydraulic retention time (HRT), but does not constantly depend on the persistent days without rain activity. In this study, NDD rather than HRT was used to describe the residence time in the column wetlands.

Three VSF wetlands were operated with NDD of 2, 4, and 8, with corresponding HRT of about 2, 4, and 8 d, respectively. About 2.1 L of stormwater as influent was fed into each wetland within 1.5 min. The instant hydraulic loading rate was about 257 m/day, which is two times faster than that of a rapid sand filter. In stormwater wetlands, the heights of the saturated zone and unsaturated zone were 0.33 m and 0.42 m, respectively (Fig. 1). In order to make maximum use of the wetlands during dry days, the stored stormwater was recirculated after 24 h of retention in the columns. The recirculation frequency of NDD of 2, 4, and 8 was 1, 3, and 7, respectively. After the specified retention time for each wetland, the effluent was collected for laboratory tests, and another batch of stormwater was fed to the columns. The wetland system was operated for 136 d in this study.

2.3. Measurements of physical and chemical parameters

Temperature, pH, and electrical conductivity were measured *in situ* using a YSI 556 portable water quality monitor. Turbidity was measured by a turbidimeter (HACH, 2100N). Samples were kept in a refrigerator immediately after collection, and were tested within a period of four days. The other water quality parameters were measured using the *Standard Methods for the Examination of Water and Wastewater*, 19th edition [14].

2.4. Statistical analyses

All statistical tests were performed using IBM SPSS version 11.5 for Windows. One-way ANOVA was used to determine significances of the treatment effect on the performance. Bivariate correlations were adopted to test the relationships between the loading rates and effluent concentrations, and between the loading rates and removal rate.

3. Results and discussion

3.1. Characteristics of nitrogen removal

3.1.1. Nitrogen removal

The concentrations of different species of nitrogen in the influent and effluent are given in Table 2.

Table 1
Packing order of the lab-scale constructed VSF wetland

Layer height (cm)	Media					
	Substrate	Width (cm)	Height (cm)	Length (cm)	Volume (cm^3)	Porosity (%)
90–95 (top)	Vermiculite	4.8–5.5 mm in diameter				45
85–90	Quartz	1.9–3.1	2.0–5.0	0.8–1.9	5.8–15.6	41
30–85	Woodchips	1.0–3.1	2.6–11.5	0.1–1.1	0.4–16.8	66
20–30	Quartz	1.4–2.4	1.7–3.6	0.6–1.7	1.7–11.0	40
10–20	Quartz	1.9–3.1	2.0–5.0	0.8–1.9	5.8–15.6	41
0–10 (bottom)	–	–	–	–	–	100

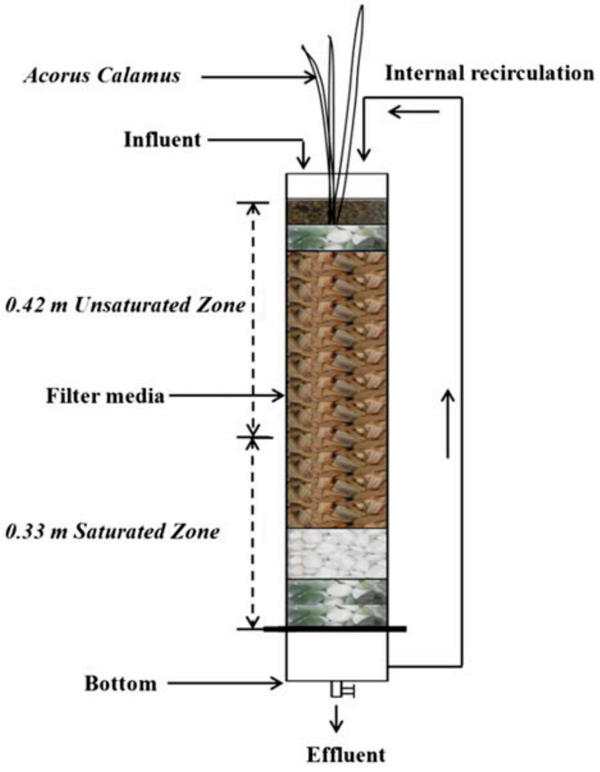

Fig. 1. Schematic diagram of the VSF wetland.

Generally, except on several occasions, the VSF wetlands are efficient in removing nitrogen (Fig. 2).

During the operation stage, the TN concentrations in the influent and effluent averaged 17.7 ± 3.1 and 13.0 ± 2.4 mg L^{-1} at NDD of 2, 18.2 ± 2.9 and 11.9 ± 2.8 mg L^{-1} at NDD of 4, and 19.4 ± 2.5 and 11.5 ± 6.6 mg L^{-1} at NDD of 8. The percentage removals of TN at NDD of 2, 4, and 8 were 26, 35, and 41%, respectively, which shows that the influent nitrogen removal increased significantly as NDD increased ($p < 0.05$). This is because of the longer retention time which benefits nitrogen removal by increasing the contact time between nitrogen and biofilm. It should be noted that most of the TN in the influent was dissolved: DTN accounted for 82% at NDD 2, 84% at NDD 4, and 88% at NDD 8. Consequently, physical filtration did not account for the majority of the nitrogen sink. On average, the woodchips wetlands achieved dissolved nitrogen removal of 23% for NDD 2, 35% for NDD 4, and 54% for NDD 8. However, this result was different from that of the same woodchips wetlands treating stormwater from an urban zone [15], in which the nitrogen was reduced by 46, 52, and 26% for NDD 2, 4, and 8, respectively. This difference was likely due to the stormwater characteristics.

The average influent TKN concentration was reduced by woodchips wetlands from 16.4 to 2.3 mg L^{-1}, from 16.1 to 8.4 mg L^{-1}, and from 16.7 to 7.0 mg L^{-1} for NDD 2, 4, and 8, respectively. However, the removal of NH_4–N and org.-N at different NDDs was different. The influent NH_4–N was greatly removed at NDD 8 (83%) rather than 2 (28%), while the influent org.-N was greatly removed at NDD 2 (42%) rather than 8 (23%), suggesting that the TKN removal was closely related to NH_4–N removal ($p < 0.001$) especially at a longer retention time. These results also imply that the release of org.-N obviously occurred during longer retention time. In addition, the effluent org.-N at NDD 8 was significantly related to PTN ($r = 0.963$; $p < 0.001$), which means that this system has a high potential for producing particulate org.-N.

The effluent NO_3–N concentration averaged 2.08 ± 1.04 mg L^{-1} at NDD 2, 3.57 ± 2.00 mg L^{-1} at NDD 4, and 4.54 ± 2.94 mg L^{-1} at NDD 8. The corresponding net cumulative concentration was 0.70, 1.54, and 1.90 mg L^{-1}, respectively. It is concluded that the nitrification rate was higher than the denitrification rate. The results confirmed the study by Connolly

Table 2
Conversion of nitrogen in different operational conditions

Items	NDD = 2			NDD = 4			NDD = 8		
	Influent (mg L^{-1})	Effluent (mg L^{-1})	Decrease (%)	Influent (mg L^{-1})	Effluent (mg L^{-1})	Decrease (%)	Influent (mg L^{-1})	Effluent (mg L^{-1})	Decrease (%)
TN	17.7(3.1)	13.0(2.4)	26	18.2(2.9)	11.9(2.8)	35	19.4(2.5)	11.5(6.6)	41
TKN	16.4(3.0)	10.8(2.3)	34	16.1(3.0)	8.4(2.8)	48	16.7(2.7)	7.0(6.8)	58
DTN	14.6(2.8)	11.2(2.1)	23	15.3(2.8)	10.0(2.0)	35	17.0(2.5)	7.9(2.9)	54
PTN	3.09(1.49)	1.83(1.24)	41	2.89(1.39)	1.98(1.74)	31	2.35(0.72)	3.58(5.48)	−52
NH_4–N	9.47(1.70)	6.85(1.51)	28	9.46(2.03)	4.61(1.98)	51	9.82(2.40)	1.63(0.90)	83
org.–N	6.89(3.25)	3.98(2.90)	42	6.68(3.51)	3.76(3.35)	44	6.93(3.85)	5.33(7.07)	23
NO_3–N	1.38(1.04)	2.08(1.06)	−51	2.03(1.14)	3.57(2.00)	−76	2.64(1.07)	4.54(2.94)	−72

Note: PTN = TN—DTN; org.N = TKN—NH_4–N.

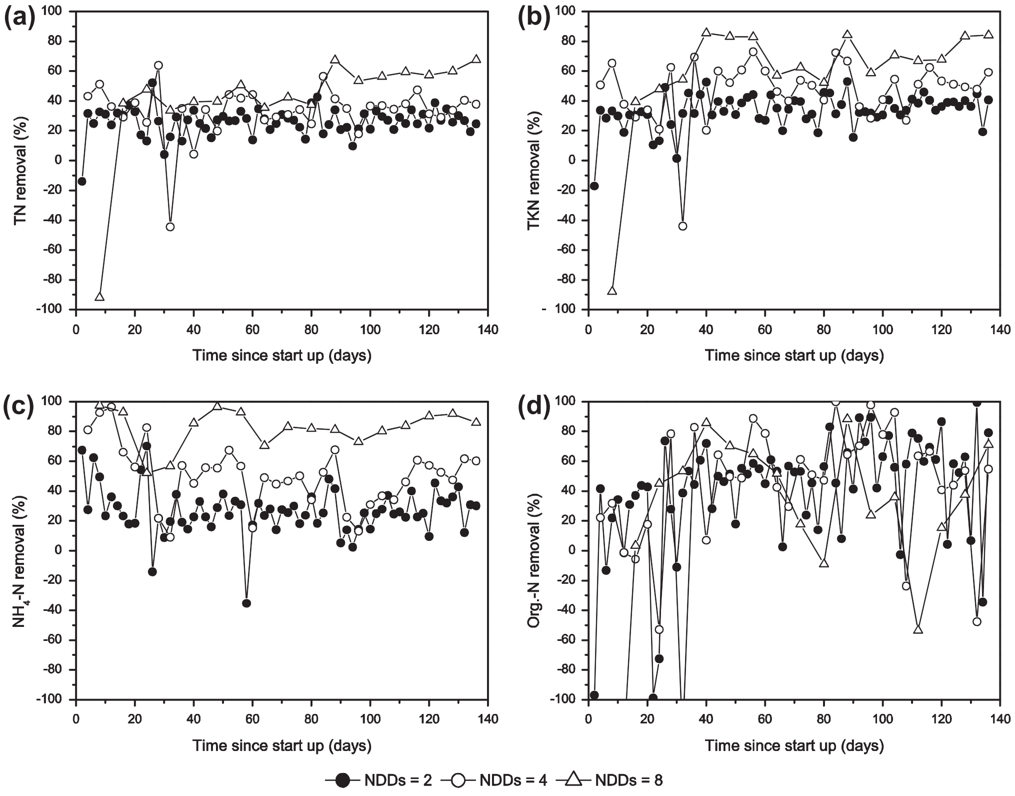

Fig. 2. Removal efficiency of different species of nitrogen with respect to time.

et al. [16] that recirculation could lead to higher NH_4–N percentage removal and more accumulative NO_3–N in the water. Still, amounts of NO_3–N were not converted to N_2, which was attributed to the inhibition of denitrification and the encouragement of nitrification by the relatively high DO concentration in the water [17]. Also, Ruane et al. [18] have reported that the nitrification was stronger in the upper part than in the deeper part of woodchip beds, whereas denitrification was stronger in the deeper part.

3.1.2. *Effect of loading rate on treatment performance*

The effect of loading rate on the outflow concentration was pursued (Fig. 3). As shown in Fig. 3(a), the Pearson correlation coefficient (*R*) between TN loading rate and effluent concentration was 0.355. Still, effluent concentrations increased with an increase in loading rates ($p < 0.001$). In addition, the effluent concentrations of TKN, NH_4–N, and org.-N were also somewhat enhanced by loading rates ($p < 0.001$) (Fig. 3(b)–(d)), especially for NH_4–N.

The performance of VSF wetlands was also investigated in terms of the effect of loading rate on the removal rate. The relationships between the loading and removal rates of TN, TKN, NH_4–N, and org.-N were pursued for the three wetlands (Fig. 4).

As shown, there was a good linear relationship between the loading and removal rates for TN and TKN, and a poor linear relationship between the loading and removal rates for NH_4–N and org.-N. However, statistic correlations ($p < 0.001$) were observed between the loading and removal rates for TN, TKN, NH_4–N, and org.-N, indicating that higher loading resulted in higher removal.

3.2. Nitrogen removal rates based on woodchip volume

Usually, the nitrogen removal rate can be calculated based on woodchip volume and effective porosity of woodchip. In this study, nitrogen removal rates considering the volume of the media were calculated to check the performance of VSF wetlands with additional carbon sources in eliminating nitrogen. As shown in Fig. 5, except for one occasion for each wetland without nitrogen reduction, the nitrogen removal rates varied between 0.35 and 7.59 $g\,N\,m^{-3}\,d^{-1}$, 0.33 and 4.65 $g\,N\,m^{-3}\,d^{-1}$, and 1.22 and 2.65 $g\,N\,m^{-3}\,d^{-1}$ with NDD 2, 4, and 8, respectively. The nitrogen removal rate averaged at 3.6 $g\,N\,m^{-3}\,d^{-1}$ for NDD 2, 2.1 $g\,N\,m^{-3}\,d^{-1}$ for NDD 4, and 1.7 $g\,N\,m^{-3}\,d^{-1}$ for NDD 8. It was also documented by Healy et al. [19] that the removal rate decreased as HRT increased. The removal rate for NDD 2 was not stable because the micro-organisms could not acclimate well to the new environment after new stormwater was fed.

In the study by Cameron and Schipper [20], the mean nitrogen removal rates for 10–23 months were 3.0 and 4.9 $g\,N\,m^{-3}\,d^{-1}$ (softwood) and 3.3 and 4.4 $g\,N\,m^{-3}\,d^{-1}$ (hardwood) at the temperatures of 14 and 23℃, respectively, in denitrification beds treating municipal potable water dosed with KNO_3. Van Driel et al. [21] observed that nitrogen removal rates ranged from 2.1 to 3.7 $g\,N\,m^{-3}\,d^{-1}$ in up-flow denitrification beds for the treatment of ground water. The differences of performance between treatments packed with

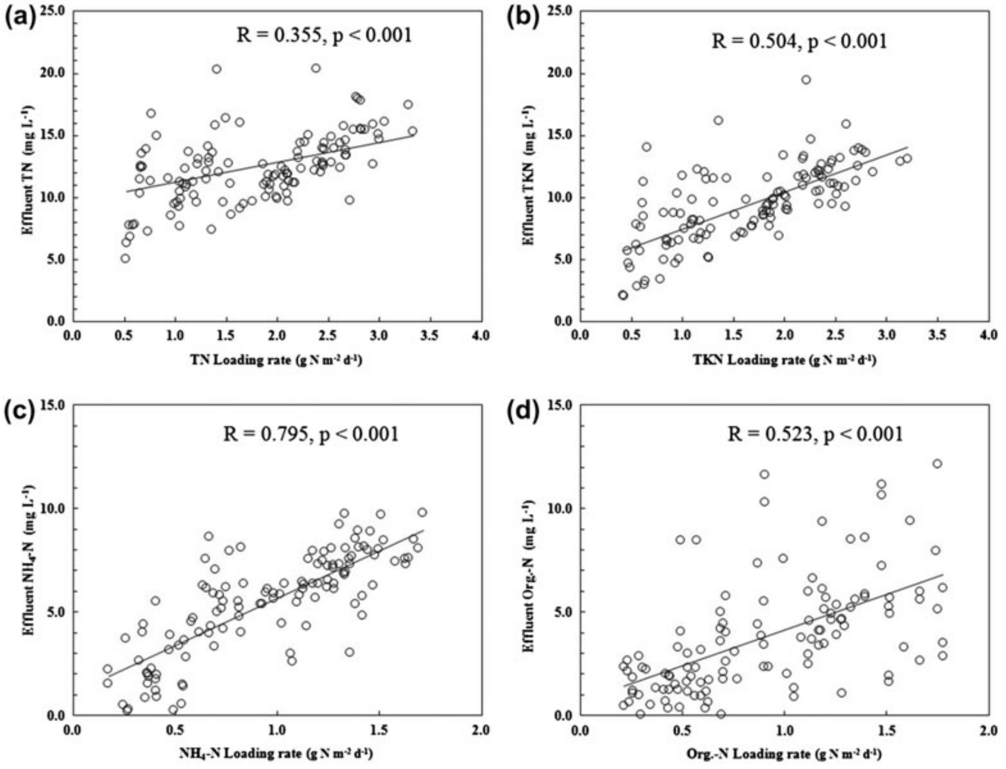

Fig. 3. The relationship between loading rate and effluent concentration.

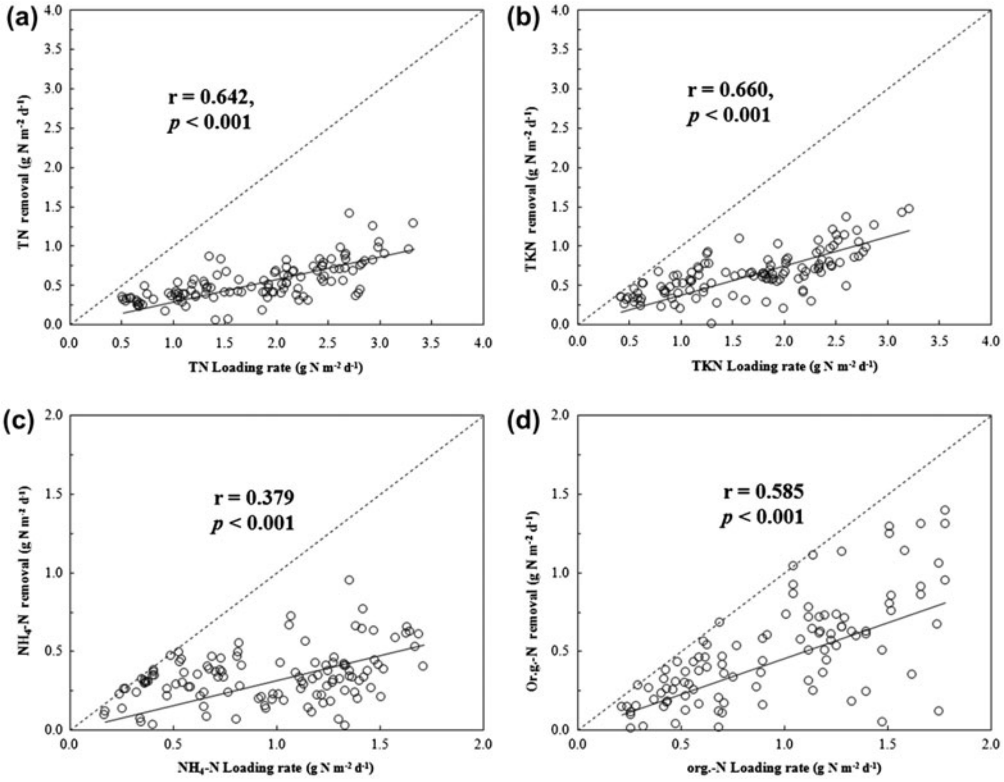

Fig. 4. Relationship between loading and removal rates.

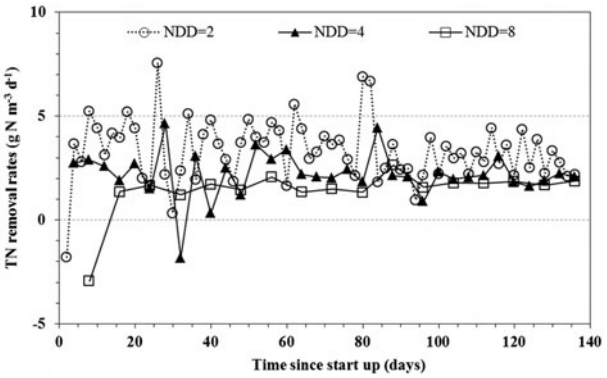

Fig. 5. Nitrogen removal rates based on woodchip volume.

woodchips might be related to the depth of the saturated zone and the specific characteristics of inflow.

3.3. Environmental factors influencing nitrification and denitrification

3.3.1. Effect of water temperature

Although denitrification rate does not increase as the same extent as the nitrification rate, both will increase with an increase in temperature within the range of 10–25℃ [22]. The effect of temperature on nitrification and denitrification based on the change of TKN and TN removal efficiencies with respect to average water temperature during dry days was investigated. As shown in Fig. 6(a), when the temperature was below 21℃, the removal efficiency of TKN was lower, and the lowest removal efficiency occurred. In the case of TN (Fig. 6(b)), when the water temperature was below 23℃, the overall TN removal performance was poor. Pearson bivariate correlation analysis shows that the removal of TKN was promoted by the water temperature at NDD 2 ($p = 0.001$) and 8 ($p = 0.018$), while the removal of TN was only enhanced at NDD 8 ($p = 0.015$). However, it should be noted that the performance of VSF wetland systems was adversely affected by higher temperature via promoting TKN release. The decrease of TKN and TN removal efficiencies when temperature was above 24℃ was due to the higher leaching of org.-N at higher temperature [20].

3.3.2. Effect of pH and alkalinity in the influent

Nitrification and denitrification might be affected by the environmental pH and alkalinity, and

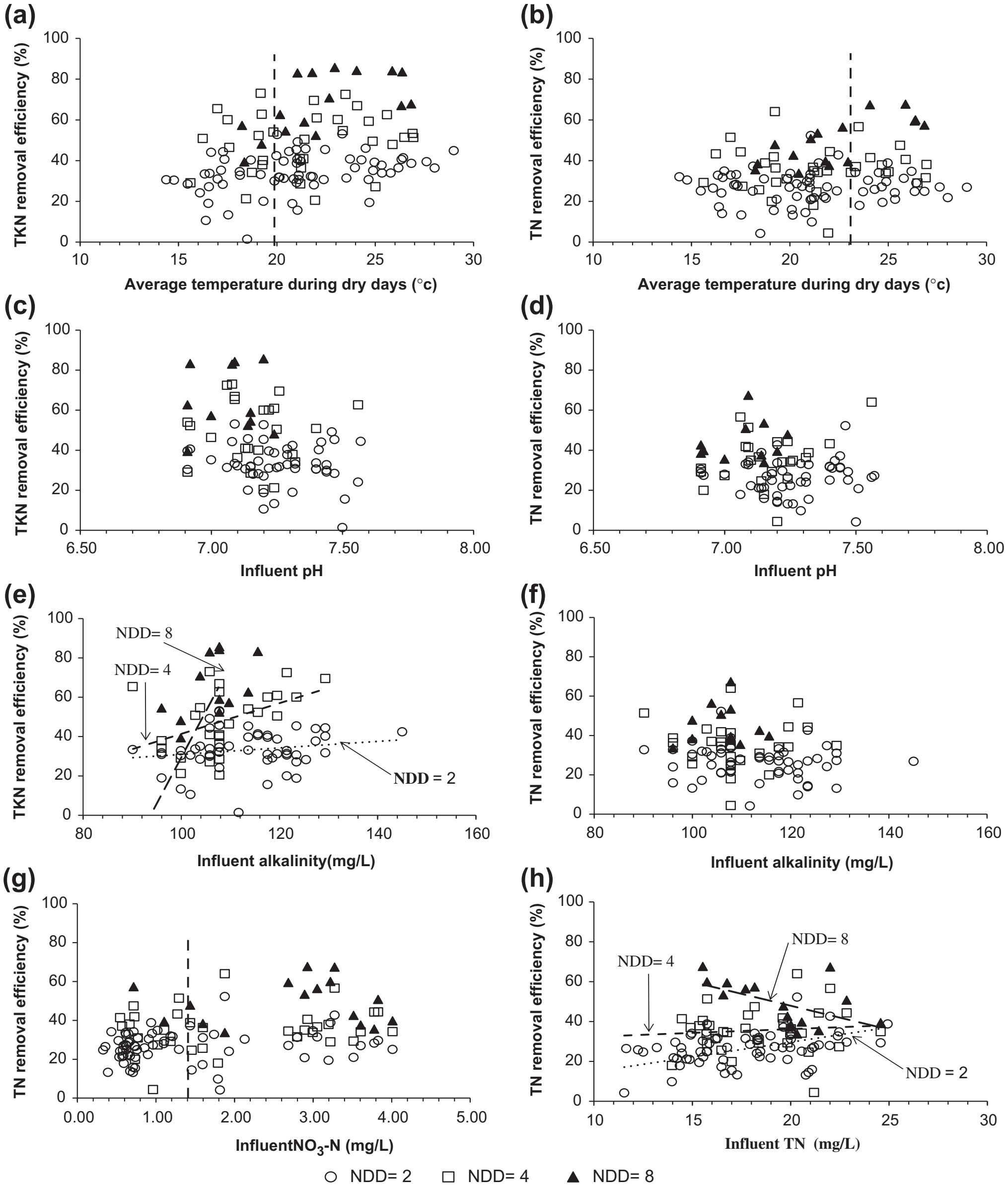

Fig. 6. Effect of water temperature, influent pH, influent alkalinity, influent NO_3–N, and influent TN on nitrogen removal.

nitrification will be much more sensitive to pH than denitrification [23]. Denitrifiers operate best in the pH range of 6.5–7.5, and nitrification prefers pH 7.2 or higher [3]. The effects of the pH and alkalinity on nitrification and denitrification are shown in Fig. 6(c) and (d).

The influent pH ranged from 6.91 to 7.57, which was close to the optimum for denitrification, and also was a minor influence on nitrification in wetland systems [3]. Therefore, it is natural that both TN and TKN removal were not suppressed by influent pH in this study.

However, the effects of alkalinity on nitrification and denitrification were different (Fig. 6(e) and (f)). Although the removal of TKN appeared to be enhanced by influent alkalinity, the removal TN was not affected by it. This is because the nitrification directly consumes alkalinity, while it is not required for denitrification.

3.3.3. *Effect of NO_3–N concentrations in the influent*

As a terminal electron acceptor, the concentration of NO_3–N might be a limitation for denitrification.

The effect of input NO_3–N on nitrogen removal is shown in Fig. 6(g). The influence of the influent NO_3–N concentration on TN elimination depends on the retention time. The removal of TN was constrained by the input NO_3–N at NDD 2 and 4 when the influent NO_3–N concentration was lower than around 1.2 mg L^{-1}, but this constrained effect was not observed at NDD 8. This is because the stronger nitrifying reaction could offset the restrictions from low NO_3–N concentration at a longer retention time.

3.3.4. Effect of TN concentrations in the influent

The effect of influent TN concentration on TN removal was investigated. As shown in Fig. 7(h), the nitrogen removal efficiency increased with the inflow TN concentration for NDD 2 and 4, while the removal efficiency decreased as the inflow TN concentration increased at NDD 8. The decrease was attributed to the greater release of nitrogen from woodchips at longer retention time.

3.3.5. Effect of DO concentrations

The influent DO concentration was relatively high with an average of 5.87 ± 2.57 mg L^{-1}. The recirculation operation also provided more oxygen for wetlands. The higher amount of available oxygen could promote nitrification. In this regards, the removal efficiencies of TKN and NH_4–N were not related to influent DO concentration ($p > 0.05$).

However, oxygen is also one of the most important factors to influence denitrification. Korom [24] and Healy et al. [17] reported that the specific DO contents resulted in a facultative change in the denitrifier's electron acceptor from O_2 to NO_3^-. Saliling et al. [13] reported that the nitrogen removal efficiency increased over the depth of the woodchips media. Ruane et al. [18] showed that the nitrogen removal efficiency increased and the NO_3–N concentration decreased with the increase of depth of the woodchip bed.

In this study, it was observed that NO_3–N accumulated in the effluent, and the effluent DO concentrations were around 3 mg/L for each wetland. In addition, the saturated zone of the media was only 0.33 m. Consequently, denitrification in VSF wetlands, especially in the shallow part of the bed, was likely to be negatively affected by oxygen. However, TN removal efficiency also was not observed to be affected by the DO in the influent ($p > 0.05$). This is because recirculation can provide favorable amount of oxygen for the wetland beds [9].

3.3.6. Effect of available carbon sources

As shown in Fig. 7, organic matter, mainly in dissolved form, was significantly released over time, especially during the initial 60 d, and the effluent COD concentrations became stable after that. However, the nitrogen removal rates did not show any significant change after 60 d ($p > 0.05$) (Fig. 5). This means that denitrification in VSF wetlands packed with woodchips was not limited by carbon sources. However, there was more cumulative NO_3–N at longer retention time. The reason might be that denitrification was constrained by oxygen or matter produced during the degradation of woodchips. This needs to be further investigated in later studies.

One thing of concern is the duration that woodchip can supply carbon. As mentioned previously the carbon was not lack in the duration of 136 d. And a number of studies have documented that woodchip can be for a long time. The study by Robertson [25] indicates that woodchips can deliver stable NO_3 removal rates over decadal timeframes. The investigation by Roberson et al. [26] into the treatments which have

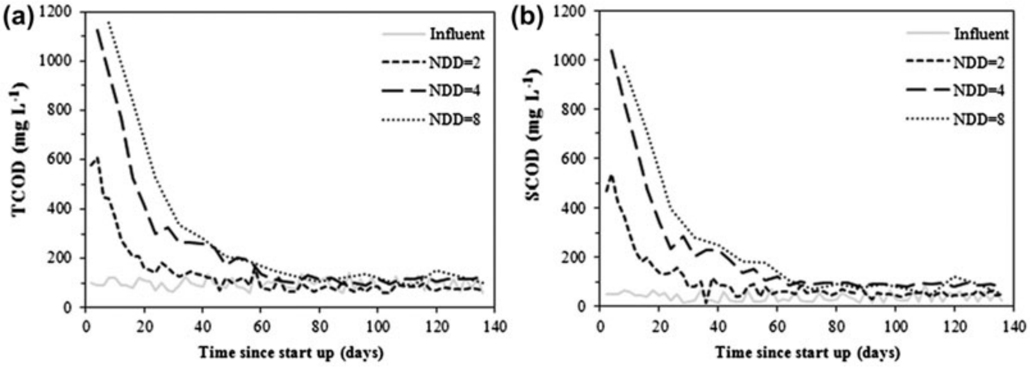

Fig. 7. TCOD and SCOD concentrations in the influent and the effluent.

been operated for 3–5 years showed that the treatment system still has favorable nitrogen removal. In addition, Eimear et al. [27] estimated that woodchip employed in their denitrification beds will possibly be replaced after 2–3 years due to the clogging rather than the lack of carbon.

4. Conclusions

Woodchip-packed VSF wetlands operated with recirculation were tested in reducing nitrogen in livestock stormwater during dry days. Comparisons were made for the conversion of different species of nitrogen in different operational conditions. In addition, the factors that influence nitrogen removal were investigated.

The VSF wetlands were capable of reducing nitrogen, especially with large NDD, via nitrification and denitrification. TN and TKN removals greatly increased with the increase in NDD. However, the nitrification rate was higher than the denitrification rate, and caused NO_3–N to accumulate in the effluent. The selection of NDD can be carried out based on the required removal efficiencies or the discharged effluent pollutants concentrations. Both the removal of TKN and TN were promoted by the increased water temperature below 24˚C, but the effect of temperature on TKN was more significant than on TN. TKN and TN removals were not affected by influent pH, whereas the removal of TKN was enhanced by an increase of influent alkalinity. Recirculation promoted nitrification by providing more oxygen; however, high oxygen concentration likely suppressed the denitrification to some degree. The systems achieved stable nitrogen removal due to the sufficient carbon sources in the wetlands.

Acknowledgment

This research was supported by the Korea Ministry of Environment as part of “The Eco-Innovation Project: Nonpoint source pollution control research group.” The authors are grateful for the support.

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