



Nitrogen reduction by fill-and-drain wetland receiving high pollution stormwater from impervious road generated by the initial precipitation

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ABSTRACT

Various chemical forms' nitrogen from impervious road stormwater results in challenging nutrient management. This study was performed to monitor and analyze for total nitrogen (TN), organic nitrogen (Org.-N), ammonium nitrogen (NH₄-N) and nitrate-nitrogen (NO₃-N) in stormwater and the effluent a fill-and-drain (FaD) wetland system designed to treat the runoff, generated by the initial 5 mm precipitation, from the impervious road. The wetland system consisted of a sedimentation tank, stormwater collection and coarse particles separation unit, and a FaD wetland, further reduction for dissolved pollutants. It was operated with moderate mass load elimination for TN (67%), Org.-N (49%), NH₄-N (95%) and NO₃-N (99%), respectively. The outflow nitrogen was dominated by Org.-N (94.5%) with an averaged concentration of 3.97 mg/L. Meanwhile, the reduction of nitrogen was limited by the conversion of Org.-N inside FaD wetland. Therefore, it is critical to creating effective ammonification conditions for Org.-N reduction through the FaD system treating stormwater.

Keywords: Fill-and-drain wetland; Impervious road; Nitrogen; Stormwater

1. Introduction

With the rapid urbanization, the stormwater runoff from urban impervious surfaces, for example, highways, roads, parking lots and roofs, has been regarded as one of the increasing important sources of nitrogen to local receiving waters [1–4]. The processes such as the natural nitrogen cycle, fertilizers use, atmospheric deposition and transportation result in rapid nitrogen transport during storm events [5–7]. It has been documented that nitrogen appears with the ranges between 0.6 and 1.4 mg/L for total Kjeldahl nitrogen (TKN) while from 0.14 to 2.2 for NO₃-N plus NO₂-N (nitrite) in the stormwater from urban and highway areas [8,9]. Nitrogen loads in stormwater from urban areas are

greater than those from undisturbed natural lots [6]. Excess N input to water environment causes eutrophication and then results in degradation of habitat quality, alterations in community structure and occurrence of algal blooms [1].

To develop sustainable urban environment, the strategy of low-impact development (LID) was proposed [10,11]. For LID towards the impervious area, the hydrologic and water quality characteristics are expected to occur as close as possible to those before development [12]. Green infrastructures for LID are designed to control stormwater on-site and provide numerous benefits to local areas [13–15]. One of the essential aspects is water resources management [16]. This function can be obtained via the provision of sustainable urban drainage systems with flood alleviation (stormwater

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volume and peak flow), water quality management, recharge of underground water resources and rainwater harvest [17]. And constructed wetland has been commonly used by green infrastructures towards LID resulted from its cost-effectiveness in stormwater management. Constructed wetlands (CWs) are practically divided into free water surface (FWS) ones and subsurface flow (SSF, consisting of horizontal subsurface flow and vertical subsurface flow (VSF)) ones. Nitrogen cycle processes inside wetlands mainly rereferred to particulate settling, plant translocation, ammonia volatilization, sorption of soluble nitrogen on substrates, ammonification (mineralization), nitrification, denitrification, assimilation, and decomposition [6,18,19]. However, nitrification–denitrification is believed to be the most significant pathway for total nitrogen (TN) removal [20]. Substrate mainly supports the plant growth in FWS wetland while can also significantly eliminate pollutants via adsorption in SSF wetland [21,22]. Oxygen and organic carbon are two main chemicals consumed for nitrogen removal in CWs [20]. Compared with the other type of wetland, VSF wetland is found can offer more oxygen to enhance the transformation of TKN. Meanwhile, substrate porosity, oxygen and water quality will influence the microbes and influence the nitrogen removal efficiency [23].

As the nitrogen composition may vary greatly depending on land use type and hydrologic conditions, the nitrogen conversion inside wetlands becomes complex [1,24–26]. For CWs, nitrogen conversion can be improved by the establishment of vegetation and the additional supply of carbon sources [27–29]. Nitrogen removal efficiencies varied greatly and sometimes are unfavorable mainly attributed to the poor conversion of specific nitrogen forms [1,6,9]. In this regard, the information on nitrogen conversion through wetlands still is required to obtain attractive nutrient management. And this study is performed to (1) check the conversion of urban stormwater nitrogen by pumice-woodchip packed fill-and-drain (FaD) wetland, (2) compare the composition variation of nitrogen in impervious road stormwater and effluent of FaD wetland system, and finally (3) based on the result of (1) and (2) estimated the nitrogen reduction capacity of FaD wetland and give suggestions towards the improvement of urban stormwater nitrogen sink.

2. Materials and methods

2.1. Wetland system

The wetland system was located under a bridge (N 36°41'53.6", E 126°34'15.9") of a national road near to Hanseo University in Seosan, South Korea. Usually, the wetland surface area of 1%–2% of the watershed area is recommended for stormwater pollution control. The studied system, including a sedimentation tank (ST) and a FaD wetland (Fig. 1a), was built with an area of 8.5 m² (1.7% of watershed (500 m²)) to capture and treat the runoff with a volume of ~1.25 m³ (the initially 5 mm precipitation) from a typical asphalt paved road. The wetland was packed with cobblestone (20 cm), pumice (15 cm), woodchip (45 cm), cobblestone (10 cm) and quartz stone (2 cm) from bottom to top, respectively (Table 1). As cheap and easily available substrate woodchip has been used commonly by stormwater

management facilities [30–32]. In practice, the leaching of chemical oxygen demand (COD) in the outflow can be observed unsurprisingly. However, significant leaching just is present in the initial operational stage, and the long-term operation of stormwater management facilities can offset this shortfall [29,33]. Otherwise, as woodchip is used for stormwater treatment reactors there are several advantages of supplying void space to minimize the clogging of packing layer, forming anaerobic environment required by specific pollutant reduction, working as mulch to enhance plant growth and providing carbon sources to prompt denitrification as necessary, etc. [3,34–37]. ST worked not only as a runoff capture device but also as a preliminary treatment unit for coarse particle separation to prevent the VSF wetland from clogging. After 24 h, stormwater was transported automatically from ST to wetland with rate of ~5.56 m³/h for further treatment. The FaD had a partial saturated bed with water depth of ~52.5 cm. In order to enhance the treatment performance and the formation of the landscape, the stormwater in the wetland was internally recirculated twice per day with the same rate of feeding. The water was stored in wetland over dry days until the stormwater from the next rainfall event was collected. This study was performed from the end of March to the end of November 2014.

Acorus calamus bought from nursery garden was transplanted in the test-bed wetland on March 26, 2014. The plant roots were directly inserted into the interface between woodchip and cobblestone in the upper layer without using soil. A density of 30 plants/m² was used to provide suitable row spacing and to ensure the initial approximate biomass.

2.2. Rainfall characteristics

During the studied period, the rainfall depth varied from 1.4 to 49.8 mm. The rainfall events during spring had a relatively smaller rainfall depth but longer duration. In addition, 70% of the rainfall events took place with the rainfall depth of less than 10.0 mm. The number of dry days ranged between 1 and 23 d with an average of 8.56 d.

2.3. Water sampling

During rainy days, the stormwater runoff from the road surface was sampled from the road stormwater drain pipe, connecting with ST. Samples were taken before it went into ST. Sampling was conducted based on stormwater hydrograph [38]. Considering the variation of pollutant concentration and/or water flow, event means concentration (EMC) was used to represent the pollutant level of stormwater from inlet and outlet of ST, based on the following equation:

$$EMC = \frac{\int Q_i C_i dt}{\int Q_i dt} \quad (1)$$

where Q_i and C_i are the flow rate and pollutant (mg/L) concentration of the stormwater corresponding to time t .

Over dry days, the sampling was carried out as the stormwater was being transported into wetland and as the water in the wetland was being recirculated, respectively. Considering

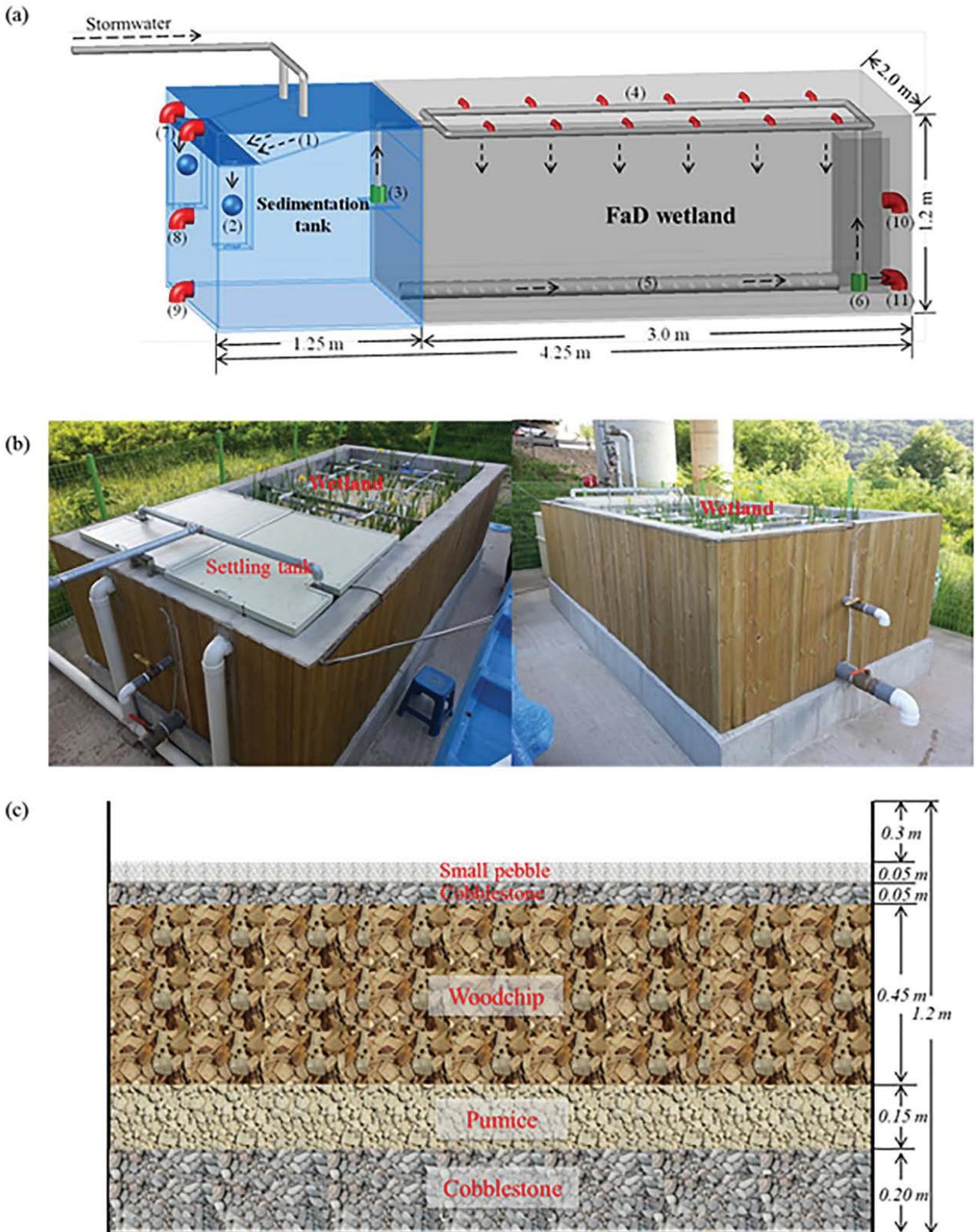


Fig. 1. Configuration of employed wetland system (a) schematic diagram of wetland system, (b) experimental device, and (c) package of wetland substrates. ((1) stormwater sampling site; (2) stormwater capture box; (3) feeding pump; (4) water distributors; (5) drainage pipe; (6) recirculation pump; (7) bypass; (8) sampling hole for settled stormwater (wetland inflow); (9) sediment discharge outlet; (10) overflow exit; (11) water discharge outlet and sampling site).

Table 1
The physical characteristic of the materials used as wetland substrate

Materials	D_{10} (cm)	D_{50} (cm)	D_{60} (cm)	U	Size (cm)	Porosity (%)	PD (kg/m ³)
Woodchip	2.00	3.10	3.40	1.70	1.5–6.5	64	305
Pumice	0.70	0.90	0.90	1.29	0.6–1.3	55	390
Small pebble	–	–	–	–	0.5–1.0	–	1,750
Cobblestone	1.79	2.48	2.58	1.44	0.98–4.17	55	1,611

U – uniformity coefficient; PD – packing density.

the water quality might vary depending on the water depth in settling tank and wetland, we took 5 grab samples to make one composite sample to represent the water quality.

2.4. Analysis

After sampling, temperature, pH, electrical conductivity (EC), dissolved oxygen (DO), turbidity and alkalinity (ALK) were measured immediately. Then the samples were stored in the refrigerator until analysis. All the samples were analyzed within 4 d. Total suspended solids (TSS), total chemical oxygen demand (TCOD), TN, ammonium nitrogen (NH₄-N), nitrate–nitrogen (NO₃-N), total phosphorus (TP) and phosphate (PO₄-P), were measured based on the methods documented by APHA et al. [39].

The differences and relationships among water parameters are detected by one-way ANOVA analysis and Pearson Correlation, respectively. All the statistical analyses were conducted using SPSS software (Version 20.0).

3. Results and discussion

3.1. Nitrogen species and conversion

As to paved-road, the nitrogen appearing in stormwater is mainly sourced from oil, vehicle emissions, dry and wet decomposition. Its concentration can vary greatly with the site due to the traffic condition and rainfall condition. In this study, the average concentrations of TN, NH₄-N, organic nitrogen (Org.-N) and NO₃-N present in stormwater generated by the precipitation within 5 mm were 8.2, 1.68, 5.12 and 1.40 mg/L, respectively. The overall TN content was significantly higher than the average TN concentration (~5.0mg/L) of impervious road stormwater in Korea [40] and the median (2.0 mg/L) of urban areas in the USA [41]. Moreover, nitrogen in stormwater consisted of ~63% Org.-N, ~20% NH₄-N and ~17% NO₃-N, which was consistent with the previous report from the other countries by Collins et al. [6] and Taylor et al. [9] that for urban stormwater organic nitrogen was the predominant nitrogen species.

The probability plots of TN, Org.-N, NH₄-N and NO₃-N from stormwater and outflow are provided in Fig. 2 to assess the difference between input and output nitrogen. Pollutant duration curves in FaD wetland are created to focus on FaD wetland performance (Fig. 3).

The input TN concentration ranged between 2.1 and 11.7 mg/L with an average of 8.2 mg/L while the output ranged from 2.3 to 12.0 mg/L with a mean of 4.2 mg/L. This

result indicates that the FaD wetland system was capable of reducing stormwater TN discharge even though there were several occasions that the output TN concentration was higher than its input. Nonetheless, comparable to the criteria for Lakes and Reservoirs (0.36 mg/L) and for Rivers and Streams (0.69 mg/L) towards the Aggregate Ecoregion IX [42], the discharges still exceed the 0.69 mg/L criterion. In addition, the concentration ranges of Org.-N, NH₄-N and NO₃-N were 0.01–8.33, 0.09–3.51 and 0.55–3.56 mg/L in the stormwater and 0.94–11.21, 0.02–0.49 and 0.01–0.25 mg/L in the outflow. As a result, the outflow nitrogen happened with 95% Org.-N, 3% NH₄-N and 2% NO₃-N, respectively. Moreover, the levels of TN, Org.-N in effluent changed as their inflow concentrations varied during the operational stage while NH₄-N and NO₃-N had stable outflow concentrations ($p = 0.017, 0.037, 0.967$ and 0.157 for TN, Org.-N, NH₄-N and NO₃-N, respectively). And the outflow NH₄-N and NO₃-N appeared with a concentration of less than 0.50 mg/L.

As demonstrated in Fig. 3, the reduction of nitrogen significantly took place during the initial first day in FaD wetland ($p < 0.05$), and then just except for several occasions with an observed variation the levels of TN, Org.-N, NH₄-N and NO₃-N in the effluent were relatively stable. This result also suggests that excessive treatment time plays no obvious role in nitrogen reduction by FaD wetland.

Overall, the studied FaD wetland was capable of removing nitrogen from the inflow with an efficiency of 36%. It was notable that overall FaD wetland gave 88.2% and 88.0% reduction for NH₄-N and NO₃-N, respectively. Comparatively, the reduction efficiencies of TKN and NH₄-N are higher than those wetlands treating wastewater [43] and urban or agricultural stormwater or runoff-impacted surface waters [44]. The possible reason is that the internal recirculation in this study supplied sufficient oxygen to make the nitrification to perform more effectively.

During the study stage, 35.95 m³ stormwater was fed into FaD. Due to the combined effect of both precipitation and evaporation, the total water volume did not show a significant change. The behavior and fate of nitrogen are pursued and given in Fig. 4. The input nitrogen load was 5.54 kg/ha-y while the output and reduction were 1.82 and 3.72 kg/ha-y, respectively. Meanwhile, the stormwater nitrogen entering the treatment system with the form of Org.-N, NH₄-N, and NO₃-N were 3.46, 1.14 and 0.94 kg/ha-y, respectively. And their corresponding output loads were 1.76, 0.05 and 0.01 kg/ha-y, respectively. Based on the input and output, the FaD system achieved an attractive

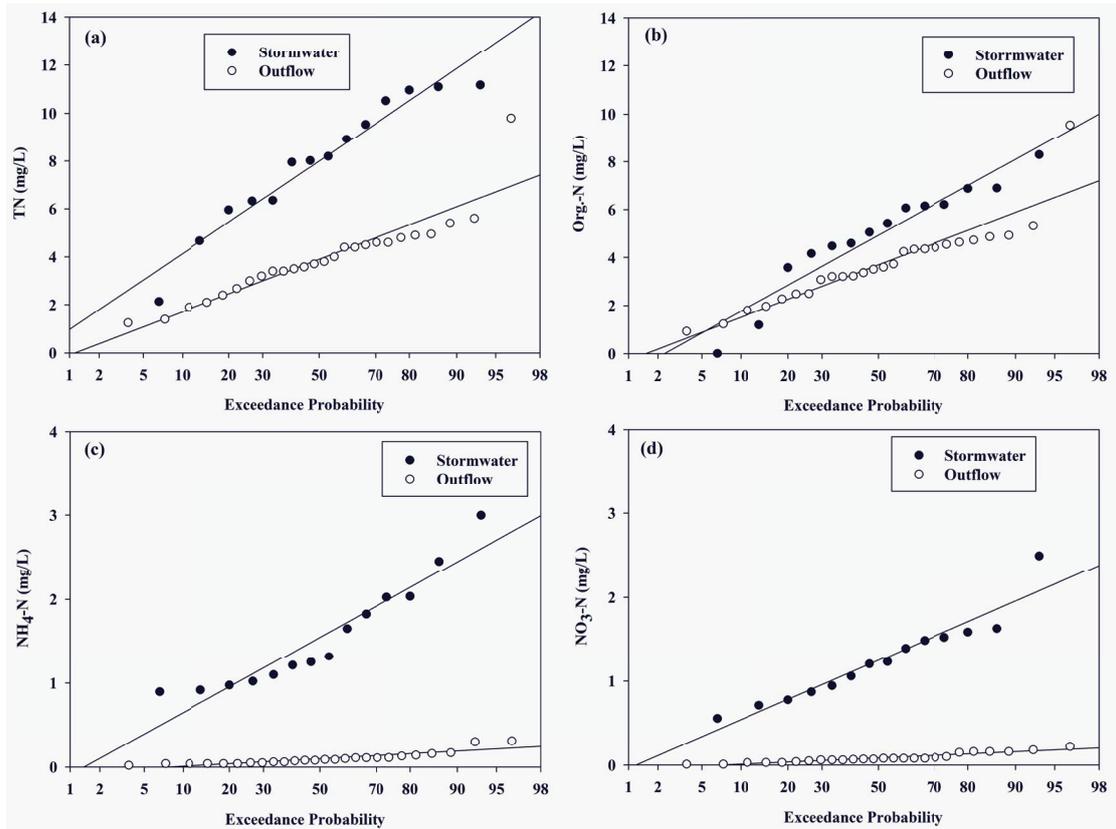


Fig. 2. Comparison of the TN, Org.-N, $\text{NH}_4\text{-N}$ and $\text{NO}_3\text{-N}$ in road stormwater and wetland outflow (a) TN, (b) Org.-N, (c) $\text{NH}_4\text{-N}$, and (d) $\text{NO}_3\text{-N}$.

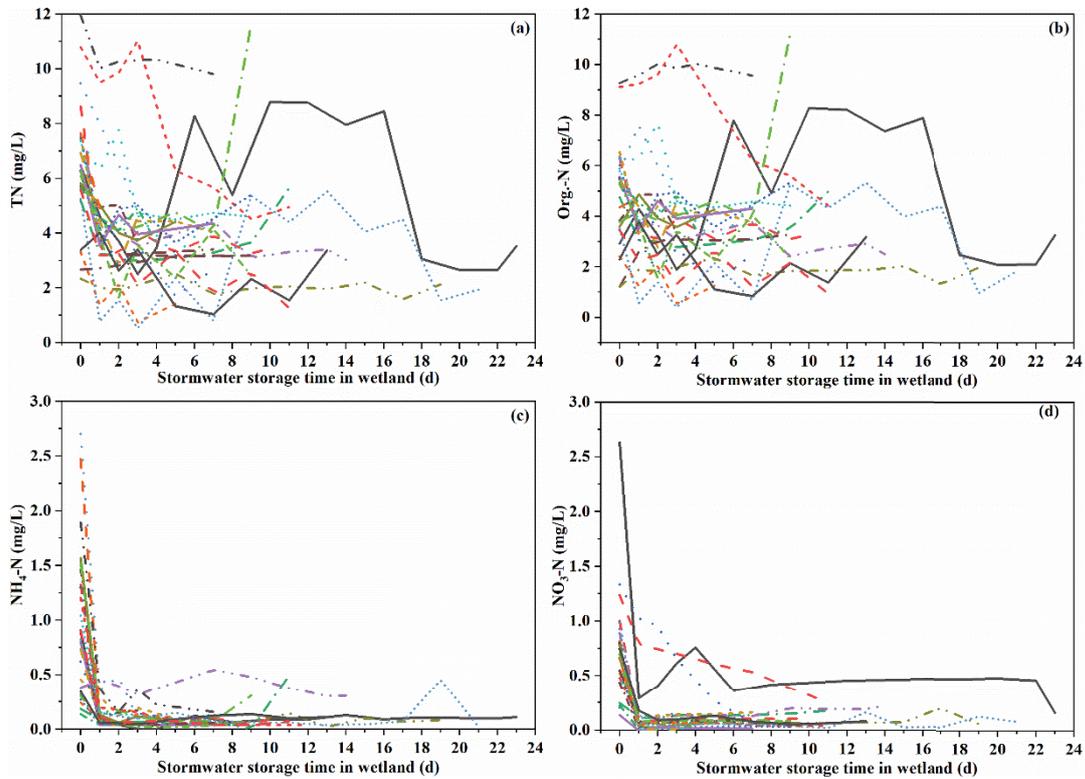


Fig. 3. Change of nitrogen concentration in FaD wetland over time (a) TN, (b) Org.-N, (c) $\text{NH}_4\text{-N}$, and (d) $\text{NO}_3\text{-N}$.

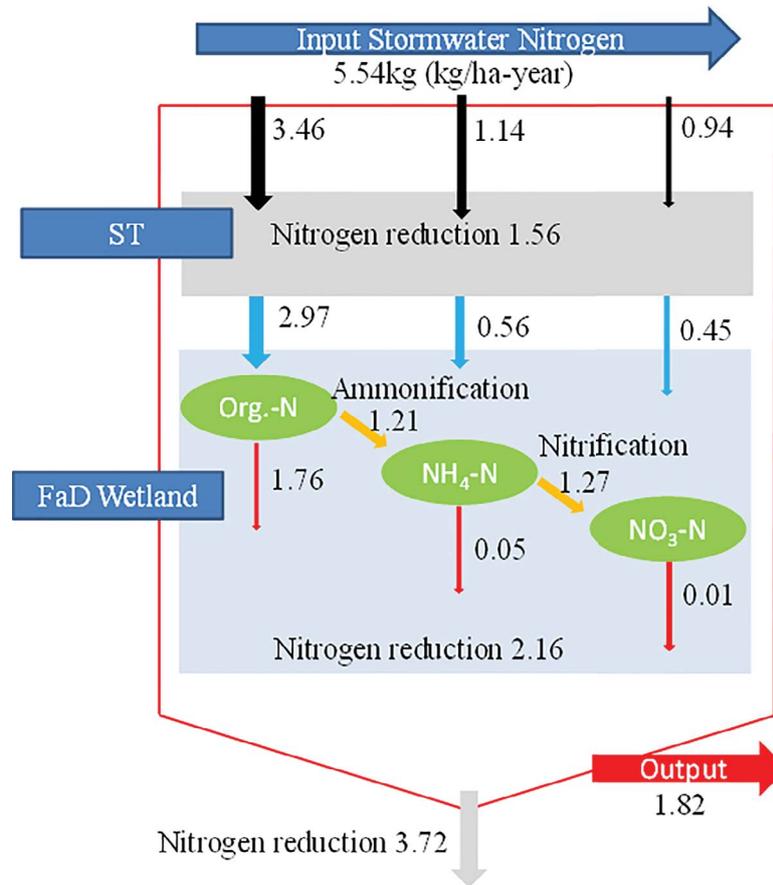


Fig. 4. Behavior and fate of stormwater nitrogen.

mass load reduction of 67.1% (27.1% in ST while 40% in FaD wetland). Specifically, the mass load of Org.-N, NH₄-N, and NO₃-N happened with 14.2%, 50.9% and 52.1% in ST while 34.9%, 44.7% and 46.8% in FaD wetland, respectively.

The nitrogen sink in wetland systems can result from the processes including sedimentation, filtration, mineralization, NH₄-N fixation (ion exchange), microbial assimilation, nitrification, denitrification and plant uptake [1]. The result indicates that the nitrogen sink performance was mixed and greatly governed by nitrogen species. Usually, the nitrogen appeared predominantly in dissolved form in stormwater and a low reduction via sedimentation is expected [1,45]. Also, in this study, the nitrogen present in stormwater did not appear with a great amount of particulate forms ($R^2 = 0.0092$, $p > 0.05$ between TN and TSS) and got a lower removal rate in a sedimentation basin. Even though both NH₄-N and NO₃-N were reduced with a high rate of around 90%, the low inflow concentrations of NH₄-N (1.68 ± 0.77 mg/L) and NO₃-N (1.40 ± 0.74 mg/L) coupled with small scale conversion of Org.-N indicate that nitrification-denitrification did not remarkably arise inside the wetland. Therefore, the ammonification, via which organic nitrogen is biologically converted into ammonia, was a limiting factor of the removal of nitrogen by nitrification and followed by denitrification. This result was consistent with the observation from the stormwater wetlands operated with other types that Org.-N could not be removed with a high

extent or even can be increased in the outflow [46–48]. Even though the reason has not been fully identified, it is believed that this result is related to the system design, substrates and operational conditions [1]. The present result shows plant uptake was not a significant pathway for nitrogen sink with a mass fraction of 3.7% to the total stormwater nitrogen. In fact, plants generally need several growing seasons to reach their maximum aboveground biomass. During the study period, the vegetation did not develop fully resulting in a small amount of nitrogen assimilated. Hence, the amount of sequestered in plants is expected to increase with operational years. The nutrient percentage sequestered in plants depends mostly on the inflow load. When inflow loading is high, the uptake is almost the same as compared to low loading but the percentage as compared to inflow is lower [49]. The load in stormwater wetlands is much lower as compared to municipal wastewater or tile drainage so in this case, the removal potential of plants is higher. The proportion of nitrogen removal by plant uptake has been reported within the common range of 0.5%–40.0% of the TN removal in comparison to typically 60%–95% removed by denitrification [50,51].

3.2. Overall treatment performance for other pollutants

As shown in Table 2, TSS, TCOD and TP in stormwater were reduced from 257, 188 and 0.70 to 24, 75 and 6.6 mg/L in

Table 2
Variation of selected water quality parameters (average \pm standard deviation) (mg/L)

Item	TSS	TCOD	TN	Org.-N	NH ₄ -N	NO ₃ -N	TP	PO ₄ -P
SW	257 \pm 174	188 \pm 97	8.2 \pm 2.7	5.12 \pm 2.42	1.68 \pm 0.77	1.40 \pm 0.74	0.70 \pm 0.55	0.05 \pm 0.05
SW	24 \pm 28	75 \pm 31	6.6 \pm 2.2	4.89 \pm 1.95	0.93 \pm 0.67	0.75 \pm 0.47	0.16 \pm 0.05	0.05 \pm 0.03
Outflow	9 \pm 8	149 \pm 131	4.2 \pm 2.2	3.97 \pm 2.15	0.11 \pm 0.10	0.09 \pm 0.06	0.11 \pm 0.07	0.02 \pm 0.01

SW – stormwater after sedimentation in ST for 24 h.

ST, and then to 9, 149 and 0.11 mg/L in wetland, respectively, on average. In addition, ST removed 91% TSS, 60% TCOD, and 77% TP from stormwater. It indicates that solids were greatly reduced in ST; therefore, the employment of the sedimentation basin could significantly reduce the probability of clogging in FaD wetland. Because, considerable organic matters and phosphorus were particle-associated ($R^2 = 0.5935$, $p < 0.05$ for TCOD and TSS; $R^2 = 0.6632$, $p < 0.05$ for TP and TSS), TCOD and TP were also reduced hugely. And there is no surprise to detect a COD export from the wetland because woodchip was allowed to release organic matters supporting denitrification by providing carbon sources and creating low oxygen condition due to the biodegradation of leached organics. However, it should be noted that the increase of COD concentration mainly occurred in the initial stage, and the treatment facility would exhibit favorable COD elimination over time [30,34].

3.3. Variation of temperature, EC, pH, ALK and DO in FaD wetland

The statistical analysis on temperature, EC, pH, ALK and DO in the inflow and outflow of the wetland is shown in Table 3.

Temperature plays a very important role in landscape formation and pollutant reduction. As to FaD wetland, the highest water temperature appeared in summer with 26°C while the lowest in spring and autumn with around 10°C. There was a slight difference between water and air: the water had slightly higher temperatures in spring and autumn while the slightly lower temperatures in summer in comparison to air. Usually, the temperature is important in terms of the activities of nitrifying bacteria and the denitrification potential in treatment wetlands because it affects both the microbial activity and oxygen diffusion [52]. Nitrification and denitrification rates can be decreased remarkably at water temperatures $< 15^\circ\text{C}$ or $> 30^\circ\text{C}$ [53]. However, in contrast to the general trend of nitrogen conversion with respect to temperature, several studies have

documented that there is no significant difference in treatment performance for landfill leachate by wetlands between high and low temperatures [54]. In this study, it was also found that temperature was not a significant factor influencing nitrogen removal ($p > 0.05$).

It was detected that pH was decreased while ALK was increased in the studied wetland. The significant pH decrease ($p < 0.05$) observed might result from the hydrolysis and acidogenesis of complex polymers, originating from organic bed medium or inoculum, under locally anaerobic conditions [55]. In the wetland, pH varied between 6.44 and 7.33 depending on the rainfall event. Nitrifiers perform better as $\text{pH} > 7.2$ and are depressed as $\text{pH} < 6.0$ [56]; while denitrifiers work optimally in the range of 6.5–7.5 [57]. Hence, no significant depression to nitrification and denitrification took place in the wetland. In the woodchip-packed wetland, the increased ALK might be related to several biological processes, including denitrification, manganese(IV) reduction, iron(III) reduction, sulfate reduction and methane fermentation [58]. Nitrification is a process requiring ALK, which means that the increase in ALK in wetland would promote the nitrification process.

DO concentration in water varied greatly depending on the water temperature and oxygen consumption and supply. The averaged concentrations are 7.45 mg/L for inflow while 6.47 for outflow. In the treatment facility due to the aerobic biodegradation of organic matter from woodchip DO concentration used to be decreased [35,37,59]. In our wetland, DO was consumed both by the biodegradation and conversion of pollutants from stormwater and the materials, especially organic matter, from woodchip. As a result, DO concentration inside the wetland was expected to decrease significantly. However, only a slight decrease was observed. This is because the internal recirculation operation over dry days supplied abundant oxygen to compensate for the DO depletion inside the FaD wetland. Generally, a DO level higher than 2.0 mg/L does not affect nitrification while the level higher than 0.09 mg/L can give a significant inhibition to denitrification [56]. This study

Table 3
Variation of basic water quality (average \pm standard deviation)

Item	Temperature ($^\circ\text{C}$)	DO (mg/L)	pH (–)	ALK (mg/L)	EC ($\mu\text{s}/\text{cm}$)
SW	–	–	7.16 \pm 0.26	36 \pm 16	805 \pm 836
Inflow	20.6 \pm 3.8	7.42 \pm 1.24	7.82 \pm 0.90	67 \pm 19	502 \pm 323
Outflow	20.7 \pm 4.0	6.47 \pm 1.31	7.09 \pm 0.17	89 \pm 15	505 \pm 265

SW – stormwater; inflow: stormwater after sedimentation in ST for 24 h.

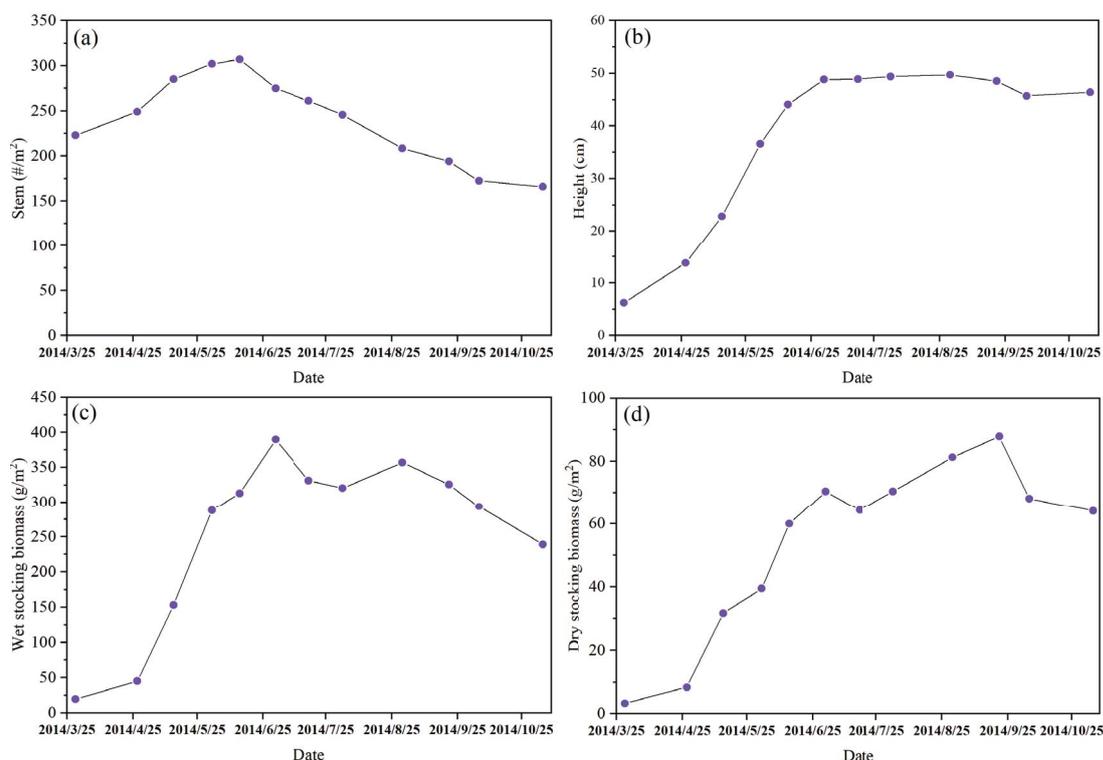


Fig. 5. Vegetation of FaD wetland studied during the operational stage (a) stem, (b) height, (c) wet stocking biomass, and (d) dry stocking biomass.

demonstrates that the denitrification processes seemed to be stressed by DO. In fact, however, denitrification is associated with reaction rims that penetrate into the woodchip rather than being restricted to the grain surfaces [60,61]. Therefore, DO may not be a significant factor affecting the denitrification in woodchip-packed FaD wetlands.

3.4. Vegetation

For the present study, the vegetation did not develop fully. Based on Fig. 5, the landscape of plants mainly occurred from May to September with the highest height of 50 cm. The change in height over time suggests that the optimum growth time is May to August. Due to the self-thinning effect shoot density declined since the middle of June. Meanwhile, the maximum wet biomass took place in June while the dry during September. Overall, the variation of height and stocking biomass follows the S-curve growth equation. According to the result, the growth rate constant is 0.06 d^{-1} for height, 0.11 d^{-1} for wet stocking biomass and 0.05 d^{-1} for dry stocking biomass.

4. Conclusion

As to the studied wetland system, the nitrogen from paved-road stormwater can be effectively removed. However, the outflow TN concentration was still higher than the criteria to protect the Lakes & Reservoirs and Rivers & Streams recommended. The conversion of nitrogen from

organic nitrogen to $\text{NH}_4\text{-N}$ and following $\text{NO}_3\text{-N}$ in the FaD wetland was the limiting factor of the further reduction for nitrogen. An investigation with long time operation is expected to check whether the unfavorable organic nitrogen reduction is temporary. Also, the efforts should be taken to find out the reason for low organic nitrogen reduction and the effective media and/or configuration/or operation to achieve the high organic nitrogen reduction in the FaD wetland system.

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