#### RESEARCH ARTICLE



# Nitrogen removal efficiency of surface flow constructed wetland for treating slightly polluted river water

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#### Abstract

Restoration and water quality improvement of malodorous as well as slightly polluted rivers have been the global focus for environmental protection research and the development and construction of sponge cities. To date, constructed wetlands have been proven to be one of efficient methods to improve water quality. Nitrogen removal efficiency is a crucial indicator for the performance evaluation in slightly polluted river water treatment. Therefore, current study aimed to investigate the N removal efficiency of 3-stage surface flow constructed wetlands for water treatment. Results show that after a prolonged operation period, constructed wetlands were able to remove  $NH_4^+$ -N,  $NO_3^-$ -N, and TN by 38.4%, 22.3%, and 29.1%, respectively. Further investigations were carried out to investigate the removal efficiency of various N species in the 3-stage wetlands. Findings reveal that NH<sub>4</sub><sup>+</sup>-N was mainly treated in wetland #1 (W1) and wetland #2 (W2), while NO<sub>3</sub><sup>-</sup>-N and TN were in wetland #2 (W2) and wetland #3 (W3). Results also reveal that the influencing factors such as hydraulic retention time (HRT), water temperature (WT), and additional carbon source have significant effect on the removal performance of constructed wetlands.

**Keywords** Constructed wetland  $\cdot$  River pollution  $\cdot$  Nitrogen removal  $\cdot$  HRT  $\cdot$  Carbon source

# Introduction

Along with the rapid urbanization and economic development in China, numerous environmental problems have emerged, among which the river water pollution has become one crucial issue. In 2015, the Chinese government agency has proposed the "Water pollution control action plan," as well as the "Guide for remediation of urban black and odorous water bodies" to address this problem. To restore the slightly polluted river water, physical technologies, e.g., dredging (Chen and Zhang [2015\)](#page-10-0), mechanical algal removal (Pinter et al. [2004;](#page-10-0) Shen et al. [2004\)](#page-10-0), water system connection (Yin and Lu [2008\)](#page-11-0), and diversion dilution (Liang et al. [2004\)](#page-10-0); chemical technologies, e.g., algae removal by chemical agents and dephosphorization by iron salt; microbial technologies, e.g., biological filter (Zhu

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et al. [2015](#page-11-0)) and microbial remediation (Sheng et al. [2013;](#page-10-0) Song et al. [2012\)](#page-10-0); and ecological technologies, e.g., constructed wetlands (Jia et al. [2014](#page-10-0); Shi et al. [2012](#page-10-0); Zhou et al. [2012\)](#page-11-0) and ecological floating island (Xu et al. [2010](#page-10-0); Zhao et al. [2012\)](#page-11-0), can be implemented. Among all these technologies, ecotechnologies became highly popular along with the growing public concern for the ecological environment. In particular, constructed wetlands (CW), which act as an effective buffer between rivers and wastewater treatment plants (Erler et al. [2011](#page-10-0); Vymzal [2018](#page-10-0)), have been widely utilized.

Constructed wetland is an artificial cistern with antiseepage and waterproof layers on the ground, filled with a certain depth of the substrate, planted with aquatic plants, and wastewater is purified by the biological, chemical, and physical synergies of microorganisms, plants, and substrates. It can be categorized into surface flow CW (SF-CW), subsurface flow CW (SSF-CW), tidal flow CW (TF-CW) (Wang et al. [2017\)](#page-10-0), and compound CW (Ávila et al. [2017;](#page-10-0) Tang et al. [2017\)](#page-10-0). Since the first construction and application of CW at Earby England in 1903 (Hiley [1995\)](#page-10-0), abundant research studies on CWs have been carried out. Chavan et al. [\(2007\)](#page-10-0) used a SF-CW to carry out small-scale ecological remediation towards Steamboat Creek at Truckee river, America. Results showed that the TN removal rate was high

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(30~60%) in summer, while no effect on removal efficiency was observed in winter. De Ceballos et al. [\(2001\)](#page-10-0) analyzed a SSF-CW in northeast Brazil, revealed that the 10 days HRT was the most effective HRT for pollutant removal where the removal rate of  $NO_3$ <sup>-</sup>-N was 58~82%. However, due to distinct national situations, these experiences might not be fully applicable in China. Therefore, it is necessary for researchers and engineers to carry out long-term monitoring and systematic evaluation work in China.

In China, persistent work has been done towards CWs. Xie et al. ([2013](#page-10-0)) analyzed the correlation between river water temperature and pollutant removal performance of a laboratory-scale horizontal subsurface CW (HSSF-CW). Yang et al. ([2016](#page-11-0)) investigated the pollutant removal efficiency of a slightly polluted river with a compound CW and found that the  $NH_4^+$ -N and TN removal rates were 41.7% and 25.9%, respectively. Zeng ([2010](#page-11-0)) analyzed the capability of water purification of a SS-CW and found out that the TN removal rate ranged from 5.3 to 38.2%. Chang et al. ([2014\)](#page-10-0) investigated the influence of different flooded/drained time ratios and different outflow rates on the N transformations in three laboratory-scale TF-CW systems. Wang et al. [\(2017](#page-10-0)) explored N transformation and associated microbial characteristics in a modified single-stage TF-CW at five different shunt ratios and found out that the optimal shunt ratio for effective N removal was 1:2. Fu et al. ([2017](#page-10-0)) investigated the effects of supplementing plant-based carbon sources on the nitrogen removal efficiency in a vertical subsurface flow CW (VSSF-CW) and found that the highest removal rates were  $NH_4^+$ -N 91.5%,  $NO_3^-$ -N 94.5%, and TN 92.8%. Zhu et al. ([2017](#page-11-0)) studied the effects of plant compositions on removal rates of pollutants on lab-scale CW, and results indicated that plants with mixed-culture groups improved the removal efficiency of TN. Xu et al. ([2018,](#page-10-0) [2019](#page-10-0)) investigated pollutant removal and microorganism evolution in CW and constructed wetlands combined with microbial fuel cell (CW-MFC) and pointed out that the average removal rate of TN in the CW-MFC was highly significant higher than that in the CW.

Based on the existing knowledge, it is apparent that scientific researches on CW in China are still in laboratory-scale test and lack enough practical engineering work. Moreover, most of the researches focused on SSF-CW, TF-CW, CW-MFC, and compound CW, rather than SF-CW. As a result, it is essential to monitor and investigate the long-term operational performance of a practical engineering project on SF-CW in China, especially in Yangtze River Delta region. This work will serve as a potential guide for the Chinese government agency and engineers making decisions on operational CW. Therefore, major achievements of this study are as follows: (1) design and implementation of SF-CW, (2) optimization of design parameters, (3) analyze hydraulic parameters, (4)

analyze pollutant removal efficiency of SF-CW, and (5) analyze influencing factors.

The primary objective of this study was to quantify the N removal performance in SF-CW at Yangtze River Delta region. For this purpose, the first on-site long-term monitoring project in the region was carried out. In this project, the N removal efficiency of 3-stage SF-CW for water treatment was analyzed, effect of influencing factors such as HRT, WT, and additional carbon source on the removal performance of SF-CW was investigated, and improved operational parameters of SF-CW were proposed.

### Materials and methods

#### Experimental design

Current study was conducted in the Kunshan city, a highly urbanized satellite city at the outskirt of Shanghai. The monitoring experiment was implemented at wetlands in front of the Kunshan Culture & Art Centre. The experimental wetlands were designed and implemented with 3 stage wetlands in series as shown in Fig. [1](#page-2-0). The surface areas of the wetlands were 500, 370, and 270  $m^2$ , respectively, with a water depth ranging from 0.1 to 1.0 m. Four sampling points were selected to collect the water samples for physicochemical analysis. These sampling points were implemented at inlet (W0), and at various locations in experimental wetlands (W1, W2, and W3). These wetlands were connected by DN400 spheroidal tubes, and the wetlands walls were supported with landscape stone cages. Plants used in the wetlands include iris, thalia, reed, lotus, and myriophyllum.

#### Experimental procedure

Current study was carried out with following key steps presented in Fig. [2.](#page-2-0) Long-term operational performance and factors affecting pollutant removal efficiency were analyzed. The effects of HRT, WT, and additional carbon source on pollution removal efficiency were also investigated. Chemical oxygen demand (COD) was measured through manganese III reactor digestion method, and other water quality indicators including ammonical nitrogen (NH<sub>4</sub><sup>+</sup>-N), nitrate nitrogen (NO<sub>3</sub><sup>-</sup>-N), and total nitrogen (TN) were measured following the standard procedures (CEPA, [2002](#page-10-0) and APHA, [2005](#page-9-0)).

#### Water sampling and measurement

Water samples were collected from June 2015 to November 2016, from a total of 19 wetland inlets and outlets, including 6 water samples during precipitation (June–September 2015)

<span id="page-2-0"></span>Fig. 1 Study sites: 3-stage constructed wetlands (W1–W3) implemented at Kunshan Culture & Art Centre, Kunshan, China



and 13 water samples during dry weather (October 2015 to November 2016). Samples were collected once in a month. There were 4 water sampling points (Fig. 1), which were located at the inlet of CW (W1), and water outlets at three CWs (W1, W2, and W3). A Plexiglas water sampler was used to collect water samples. The sampling methods in different climatic conditions were as follows:

- 1. Water sampling in dry condition: Water samples were collected at each sampling point from 9 to 10 am on the same day. Collected water samples were immediately sent to the laboratory for further analysis and storage. Water and air temperature were measured onsite.
- 2. Water sampling after precipitation: Sampling was carried out according to the rainfall intensity. Water samples (200 mL each) were collected in every 2 to 30 min from #0 to W3 sampling points 15 h after rainfall. The mean

concentrations were measured from the mixture of samples obtained at #0, #1, #2, and #3 sampling points.

# Hydraulic retention time experiments

Experiments on effect of HRT were conducted in summer (May to June) and winter (December to January) to explore the relationship between water denitrification efficiency and HRT in different seasons, and to determine the optimal HRT. The inflow rate of the wetland was  $28.0 \pm 1.5$  m<sup>3</sup>/h; therefore, the calculated HRT is about 15 h. HRT was designed to be 0.6 day, 1 day, 3 days, 5 days, and 7 days, and the corresponding inflow rates were 28 m<sup>3</sup>/h, 18 m<sup>3</sup>/h, 6 m<sup>3</sup>/h, 3.6 m<sup>3</sup>/h, and  $2.57 \text{ m}^3$ /h, respectively. Based on these values and taking into account the different practices in CW engineering and related





national and international regulations (USEPA report [1988\)](#page-10-0), the experimental procedures were as follows.

- 1. Water pump valve was adjusted according to the electromagnetic flowmeter display data to control the inflow rate at set value.
- 2. The pump flow rate was controlled at the set value by continuously pump water for 5~7 days. Water samples  $(500~1000$  mL) were collected from the inflow  $(40)$  and outflows of CWs (W1, W2, and W3) in summer. In case of winter season, samples were collected only from #0 and W3 because water denitrification test along the CW path was carried out only in summer. All the water samples were immediately sent to laboratory for further analysis and storage at 4 °C if required to analyze at later stage.
- 3. After collecting the water samples at one set level, the valve of the water pump was adjusted to take the inflow rate to the next level, and then repeated step 2.

#### Additional carbon source experiment

Additional carbon source test was carried out in September 2016 to explore the effect of additional carbon source on the N removal performance, particularly the TN. During that period, the wetland vegetation grew vigorously and the substrate microbial activity was high. The preliminary water quality results found that the concentrations of inflow COD and TN were 5.2 mg/L and 2.56 mg/L, respectively. Based on current values and taking into account the monitoring results of the permanganate index and TN in the Taihu lake basin in recent years (2014~2016), four COD concentration levels were designed (multiplied by 1, 1.5, 2, and 2.5). The expansion factor was used to determine the designed values of the inflow COD, which were 5.2 mg/L, 7.8 mg/L, 10.4 mg/L, and 13.0 mg/L. The experimental procedures were as follows:

- 1. Inflow and outlet water samples were collected to measure the COD concentration as baseline concentration.
- 2. 100 g/L concentrated glucose solution was prepared (a total of 20 L).
- 3. Placed the glucose dosing barrel (25 L) at the outlet of storage tank, adjusted the outflow by opening the bottom valve of the barrel (measured by the beaker according to the volume) to make the actual concentration of COD reach the designed value (1.5 times of the baseline concentration). The process of adding the concentrated glucose solution lasted for 36 h.
- 4. Sampling: from the 18th hour, wetland inflow and outflow samples were collected at 6-h interval (i.e., collected at 18th, 24th, 30th, and 36th hours from the start of glucose dosing). The collected water samples were

immediately sent to the laboratory, stored at 4 °C in refrigerator and the measurements were completed within 2 days.

5. After 2 days, steps 2 to 4 were repeated to make actual inflow COD concentration reach to 2.0 and 2.5 times of the baseline concentration.

# Results and discussion

#### Overall N removal performance

Results of the overall monthly N removal performance in CWs are presented in Fig. [3a.](#page-4-0) It can be seen from Fig. [3a](#page-4-0) that the average concentration of inflow TN in the tested wetland was high, and the annual variation range was 1.1–11.8 mg/L, with an average of  $3.53 \pm 2.43$  mg/L. The annual variation range of  $NO_3$ <sup>-</sup>-N was 0.58 to 1.72 mg/L, with an average of  $1.10 \pm 0.31$  mg/L. The annual variation range of NH<sub>4</sub><sup>+</sup>-N was 0.23 to 8.70 mg/L, with an average of  $1.21 \pm 1.86$  mg/L. Compared with the Environmental Quality Standard for Surface Water, TN concentration was in accordance with level V water quality standard of China (> 2 mg/L) but NH<sub>4</sub><sup>+</sup>-N was within the standard IV limit (1.0 to 1.5 mg/L). Results for  $NH_4^+$ -N,  $NO_3^-$ -N, and TN removal are presented in Fig. [3b.](#page-4-0) Findings revealed that the average removal rates of  $NH_4^+$ -N, NO<sub>3</sub><sup>-</sup>-N and TN in CWs were  $38.4 \pm 10.8\%$ ,  $22.3 \pm 6.0\%$ , and 29.1  $\pm$  6.5%, and the removal loads were 0.29  $\pm$  0.46, 0.14  $\pm$ 0.06, and  $0.65 \pm 0.58$  g/m<sup>2</sup> day, respectively. Generally, the removal rate was low and the results obtained in this study were in broad agreement with earlier studies (Gunes et al. [2012;](#page-10-0) Sehar et al. [2016\)](#page-10-0).

The monthly variations of NH<sub>4</sub><sup>+</sup>-N concentration in inflow and outflow and removal rates are presented in Fig. [4a.](#page-5-0) Results revealed that the removal rate of  $NH_4^+$ -N in the tested wetland showed at first decreasing trend and then increasing trend from June 2015 to November 2016. From June to September 2015, the  $NH_4^+$ -N removal rate was relatively higher (about 44 to 52%) and gradually decreased from October, reaching to a minimum value of 25.4% in December. From December to March of the following year, due to lower temperature, the NH<sub>4</sub><sup>+</sup>-N removal rate was lower and fluctuated slightly (around 20%). From March 2016, due to the rising temperature and the recovery/growth of wetland plants, the NH<sub>4</sub><sup>+</sup>-N removal rate gradually increased, from 21.7% in March to 35% in May. From June to September 2016, the NH4 + -N removal rate remained at a relatively high level, about 40 to 50%, which was similar to the same period of the previous year. It can be seen that the NH<sub>4</sub><sup>+</sup>-N removal rate was higher in autumn 2016 (46.0%) compared with the same period in 2015 (38.9%). This trend occurred because the NH4 + -N concentration in inflow was higher in autumn 2016.

<span id="page-4-0"></span>Fig. 3 a Monthly variation in inflow nitorgen species. b Removal rates in CWs



The  $NH_4^+$ -N removal rate in wetland improves with the increase of the inflow concentration within a certain range, also the larger substrate concentration improves the  $NH_4^+$ -N removal performance. Similar finding was also observed by Steidl et al. [\(2019\)](#page-10-0) and Wei et al. ([2017\)](#page-10-0) which noted that NH4 + -N removal performance in CW is affected by the inflow NH4 + -N concentration as well as substrate concentration.

The monthly variations of  $NO<sub>3</sub><sup>-</sup>-N$  concentration in inflow, outflow, and removal rate are presented in Fig. [4b.](#page-5-0) It can be seen that the removal rate of  $NO<sub>3</sub><sup>-</sup>-N$  revealed a decreasing trend at first and then an increasing trend. The  $NO_3^-$ -N removal rate was high in June to September 2015, stayed at around 30% with the maximum value of 32.6% appeared in August. From September to December 2015, the  $\overline{NO_3}^-$ -N removal rate gradually decreased to 16.2%. From December to March of the following year, the  $NO<sub>3</sub><sup>-</sup>-N$  removal rate remained at a low level (11~15%) due to low temperature and plant dormancy. From March 2016, with the rising temperature and the recovery of wetland plants, the  $NO<sub>3</sub><sup>-</sup>-N$  removal rate gradually increased from 11.5% in March to 24% in June.  $NO_3$ <sup>-</sup>N removal rate remained relatively high from June to September 2016, which was similar to the same period of the previous year (June–September 2015). However, the temperature gradually decreased from October 2016, but the reduction of  $NO<sub>3</sub><sup>-</sup>-N$  removal rate was not significant. At the end of the experiment in November 2016, the  $NO<sub>3</sub><sup>-</sup>-N$  removal rate was

still 22.7%. Moreover, the average removal rate in autumn 2016 (25.1%) was close to the summer removal rate (26.5%), which was higher than the same period of the previous year (21%). It was mainly due to the higher permanganate index of wetland inflow during this period. The removal of NO<sub>3</sub><sup>-</sup>-N mainly depends on the denitrification by heterotrophic denitrifying microorganisms which have higher requirements on the carbon to nitrogen ratio (C/N ratio) in the substrate. This observation agrees with the results observed by Katyal et al. ([1988\)](#page-10-0), Blecken et al. ([2009\)](#page-10-0) and Chen et al. [\(2013\)](#page-10-0) which showed that higher organic matter content also improves the denitrification process.

Results on the monthly variations of the TN concentration in inflow and outflow and removal rate are presented in Fig. [4c.](#page-5-0) The TN removal rate indicated a decreasing trend at first and then increasing trend from June 2015 to November 2016. The TN removal rate was high, stayed around 30% with the maximum of 34.4% in June to September 2015. From October, the removal rate was gradually decreased to 21.8% in December. Whereas, from December to March of the following year, the TN removal rate remained at a relatively low level of about 20% with slight fluctuation (minimum value 18.3%) due to lower temperature and plant dormancy. From April 2016, with the rising temperature and the recovery of wetland plants, the TN removal rate gradually increased

<span id="page-5-0"></span>

Fig. 4 Concentration of a  $NH_4^+$ -N, b  $NO_3^-$ -N, and c TN in inflow and outflow and their removal rates

from 20% in April to 35% in July. From June to September 2016, it remained relatively high, similar with the same period of the previous year. However, temperature was decreased gradually from October to the end of the experiment, but the reduction of the TN removal rate was not substantial and remained at a high level (30%), close to the summer removal rate, higher than the same period of previous year. It is assumed that it is mainly due to the higher inflow concentration of  $NH_4^+$ -N and organic matter (permanganate index) in this period. The NH<sub>4</sub><sup>+</sup>-N removal rate was high and significantly contributed to the TN removal. Meanwhile, the increase in organic matter could also promote the denitrification reaction to a certain extent and improve the removal of  $NO<sub>3</sub><sup>-</sup>-N$  (Vymzal [2018](#page-10-0)). Therefore, the adverse effect of temperature was





Fig. 5 Monthly removal rates of a  $NH_4^+$ -N, b  $NO_3^-$ -N, and c TN in different water temperature

minimum and the TN removal rate was high. Previous studies also revealed the similar trend that  $NH_4^+$ -N removal contribute more to TN removal performance, instead of  $NO<sub>3</sub><sup>-</sup>-N$  (Steidl et al. [2019\)](#page-10-0).

# Denitrification efficiency in 3-stage wetlands  $(NH_4^{\text{+}}-N, NO_3^{\text{-}}-N,$  and TN)

The denitrification efficiency along the path of 3-stage wetlands was investigated from June 2015 to September 2016. The investigation period covered winter (December to February), spring (March to May), summer (June to August), and autumn (September to November). Figure 5a shows the result of NH<sub>4</sub><sup>+</sup>-N removal performance along the 3-stage wetlands. It can be seen from Fig. 5a that the average NH<sub>4</sub><sup>+</sup>-N removals in W1, W2, and W3 CWs were 0.16, 0.11,

and 0.07  $g/m<sup>2</sup>$  day, respectively. The first two wetlands removed 80% of NH<sub>4</sub><sup>+</sup>-N. Additionally, in higher temperature, the NH<sub>4</sub><sup>+</sup>-N removal proportion of the first two wetlands was relatively higher. In winter and early spring, when the temperature was relatively low, the NH<sub>4</sub><sup>+</sup>-N removal amount was more evenly distributed (seen from January to March 2016). This trend indicated that the assimilation of plants and microorganisms contributed significantly to the NH<sub>4</sub><sup>+</sup>-N removal. The microbial activities were weak in winter and spring due to low temperature. Wetland plants were harvested or still in the germination stage and were unable to remove the large amount of  $NH_4^+$ -N. Therefore, the  $NH_4^+$ -N removal amount of each wetland were more consistent. In their previous study, Luo et al. ([2020\)](#page-10-0) noted similar findings that in a 3-stage SF-CW, NH4 + -N removal proportion can reach up to 43.0–99.0% in the first two wetlands.

The result of  $NO_3$ <sup>-</sup>-N removal performance along the 3stage wetlands pathway is presented in Fig.  $5b. NO<sub>3</sub><sup>-</sup>-N$  $5b. NO<sub>3</sub><sup>-</sup>-N$  was mainly removed in the second and third wetlands. The average  $NO<sub>3</sub><sup>-</sup>-N$  removal amounts of W1, W2, and W3 were 0.09, 0.13, and 0.25  $g/m^2$  day, respectively. The last two wetlands removed  $80\%$  NO<sub>3</sub><sup>-</sup>-N of the total removal amount, especially third wetland, removed more than  $50\%$   $NO<sub>3</sub><sup>-</sup>-N$ . The removal of NO<sub>3</sub><sup>-</sup>-N mainly relies on denitrifying bacteria (facultative aerobic microorganisms) which requires higher amount of carbon source  $(BOD<sub>5</sub>/TN > 3~5)$  and dissolved oxygen (DO  $< 0.5$  mg/L). The biodegradability was poor due to the low organic matter in the wetland inflow, which was not conducive to the denitrification reaction. Similar trend was observed in earlier studies (Wei et al. [2017;](#page-10-0) Chen et al. [2013](#page-10-0); Katyal et al. [1988](#page-10-0); Blecken et al. [2009](#page-10-0)). Therefore, the  $NO<sub>3</sub><sup>-</sup>-N$  removal load of all wetlands was low. However, due to the low DO concentration in W3 (average DO in W1 and W2 was  $>$ 3.5 mg/L and < 1.8 mg/L in W3), the  $NO<sub>3</sub><sup>-</sup>-N$  removal was high in W3.

TN removal performance along the 3-stage wetland pathway is presented in Fig. [5c.](#page-5-0) It can be seen from the results that the TN was mainly removed in the second and third wetlands. The average TN removal amounts in W1, W2, and W3 CWs were 0.33 g/m<sup>2</sup> day, 0.49 g/m<sup>2</sup> day, and 0.59 g/m<sup>2</sup> day, respectively. Results revealed the last two wetlands were more effective and removed 76.4% TN. This was mainly related to the proportion of  $NH_4^+$ -N and  $NO_3^-$ -N in TN. The  $NH_4^+$ -N was removed in the first and second wetlands, while  $NO<sub>3</sub><sup>-</sup>-N$  was removed in the second and third wetlands. In most cases, the inflow concentration of  $NH_4^+$ -N is smaller than that of NO<sub>3</sub><sup>-</sup>-N. Therefore, the removal of TN was mainly conducted in the last two wetlands. These results are consistent with the findings of Guo et al. [\(2017](#page-10-0)). In a previous study, researchers noted that the single-stage CWs cannot achieve high removal rate for the TN due to their inability to provide both aerobic and anoxic conditions at the same time (Vymazal [2007](#page-10-0); Vymzal [2018\)](#page-10-0).

## Effect of HRT and temperature on the N removal efficiency

The N pollutant removal rates under different HRT in summer season are presented in Fig. [6a.](#page-7-0) Results revealed that the NO<sub>3</sub><sup>-</sup>-N removal was increased with the HRT and reached to a higher value (23.1%) under 3-day HRT. It was changed slightly (25.8%) when HRT was 7 days. The TN removal showed similar trend with the  $NO<sub>3</sub><sup>-</sup>-N$  removal but had a higher removal (31.8%) when HRT was 5 days, and only increased 1.9% when HRT increased from 5 to 7 days. Trend for NH4 + -N removal was different from the TN and  $NO<sub>3</sub><sup>-</sup>-N$  where removal rates were almost similar and remained at about 40%. Earlier studies also revealed that the HRT is closely related to the degradation and removal efficiency of these pollutants. It is an important parameter to maintain the operation of wetlands and fully exert the purification effect (Wu et al. [2012](#page-10-0); Vymzal [2018\)](#page-10-0).

It is well understood that the removal of  $NO<sub>3</sub><sup>-</sup>-N$  depends on denitrifying bacteria, and the denitrifying bacteria is facultative aerobic microorganism (Katyal et al. [1988](#page-10-0); Chen et al. [2013;](#page-10-0) Wei et al. [2017](#page-10-0)). Cross-sectional flow rate and DO in the CWs were decreased with the increase in HRT, which is beneficial to the denitrification process. As a result, the removal rate of  $NO_3$ <sup>-</sup>-N was gradually increased and reached to the maximum level (38.3%) when HRT was 5 days. However, C/ N was relatively low since the inflow water was slightly polluted river water; therefore, the overall removal rate of  $NO<sub>3</sub><sup>-</sup>$ N was also very low. In addition,  $NO<sub>3</sub><sup>-</sup>-N$  removal accounts with high proportion ( $> 60\%$ ) and plays an important role in TN removal. Hence, it can be concluded that there was an obvious similarity between  $NO<sub>3</sub><sup>-</sup>-N$  and TN removal rates. Earlier studies also revealed that NH<sub>4</sub><sup>+</sup>-N removal mainly depends on the nitrification by aerobic nitrifying bacterial species (Wu et al. [2015](#page-10-0); Vymzal [2018](#page-10-0)). The higher oxygen level in CWs was beneficial to the nitrification process. Therefore, the removal rate of NH<sub>4</sub><sup>+</sup>-N was much higher compared to  $NO<sub>3</sub>$ <sup>-</sup>-N. The change in  $NH<sub>4</sub>$ <sup>+</sup>-N removal rate with the HRT was not apparent due to the low concentration in the substrates.

The N pollutant removal rates under different HRT in winter are shown in Fig. [6b.](#page-7-0) It can be seen that in winter with lower temperature, the removal rates of NH<sub>4</sub><sup>+</sup>-N were 21.8%, 24.2%, 25.4%, 28.9%, and 25.0%; NO<sub>3</sub><sup>-</sup>-N were 17.6%, 15.0%, 13.0%, 16.4%, and 16.1%; and TN were 20.1%, 21.2%, 22.1%, 20.3%, and 18.9%, respectively, in 0.6-, 1.0-, 3.0-, 5.0-, and 7.0-day HRT. The change in removal rate of each N pollutant was less affected by HRT in winter. Pearson correlation analysis (Table [1](#page-7-0)) also revealed that the correlation between each N pollutant and HRT was greater than 0.05, indicating that the HRT had negligible effect on the removal rate of N pollutant in winter. This trend could be happened due to factors such as low inflow concentrations of the N

<span id="page-7-0"></span>Fig. 6 Pollutants removal efficiencies under different HRT in a summer and b winter



pollutants resulted in the less microbial reactions. Seasonal factor (temperature) might be another reason as water temperature and the microbial activity were low which affect the growth and microbial activities. Meanwhile, in winter, the wetland plants had been harvested; therefore, the stems and leaves of the plants were not able to intercept the pollutants effectively. The above analysis shows that HRT should be 5 days in summer in order to achieve best N pollutant removal performance. Nevertheless, the effect of HRT on N removal in winter is negligible. Earlier studies also reported that 5-day HRT was suitable for the optimal pollutants removal performance in CWs (Wu et al. [2015](#page-10-0)).

Table 1 Correlation between pollutant removal rates and HRT (Pearson, 2-way)

<b>Seasons</b>		$NH_4^+$ - N	$NO3$ - N	TN
Summer	Pearson correlation	0.539	$0.938*$	$0.976***$
	Saliency (bilateral)	0.349	0.019	0.004
Winter	Pearson correlation	0.646	$-0.084$	$-0.504$
	Saliency (bilateral)	0.239	0.893	0.386

The results presented in Table 2 and Fig. [7](#page-8-0) revealed the effect of temperature on N removal efficiency. Results showed significant correlations between water temperature and N pollutants removal  $(p < 0.01)$ . In case when water temperature was higher than 16 °C, the average removal rates of  $NH_4^+$ -N,  $NO<sub>3</sub><sup>-</sup>-N$ , and TN were increased to 89%, 69%, and 61%, respectively, compared with lower than 16 °C. Moreover, significant correlations were also found between various seasons and N pollutant removal rates ( $p < 0.01$ ). The N removal rates in different season were as follows: summer > autumn > spring > winter. This trend is similar with the earlier findings

Table 2 Correlation between pollutant removal rates and water temperature

Parameters		$NH_4^+$ - N	$NO3$ - N	TN
Removal rate	W.T > 16 °C	$45.1\%$	25.6%	33.1%
	W.T < 16 °C	23.9%	$15.1\%$	20.5%
Correlation-W.T		$0.904*$	$0.906*$	$0.778*$
Difference-season $(F$ value)		25.958*	$16.941*$	14.884*

 $*p < 0.01$ 

<span id="page-8-0"></span>

Fig. 7 Monthly variations in water temperature and pollutant removal rates

(Beutel et al. [2009](#page-10-0); Wu et al. [2015](#page-10-0); Vymzal [2018\)](#page-10-0), which indicated that the rate of nitrate loss in wetland was highly seasonal and generally enhance in summer.

# Effect of additional carbon source on  $NH_4^+$ -N, NO<sub>3</sub><sup>-</sup>-N, and TN removal efficiency

In current study, glucose was used as additional carbon source to explore its effect on nitrogen removal in CW. The improved water quality parameters are presented in Table 3. The removal rates of NH4 + -N under different COD inflow values are presented in Fig. 8a. Results revealed that the change of  $NH_4^+$ -N removal rate was low (40%) when the inflow concentration was 0.22 mg/L. With the increase of inflow COD, the carbon source was added 1.5 times, 2 times, and 2.5 times as the baseline value, but the  $NH_4^+$ -N removal rates only increased to 6.1%, 5.7%, and 7.6%, respectively. Correlation analysis (Table [4\)](#page-9-0) also revealed that there was no significant correlation between inflow COD and NH<sub>4</sub><sup>+</sup>-N removal rate  $(p > 0.05)$ . In conclusion, for slightly polluted river water with low NH<sub>4</sub><sup>+</sup>-N, the addition of carbon source has negligible effect on its removal efficiency in SF-CW. It might be mainly related to the low inflow  $NH_4^+$ -N and the characteristics of the SF-CW. Since the inflow  $NH_4^+$ -N concentration was very low, it had reached or was approaching its outflow threshold. This makes nearly impossible to further reduce its concentration by relying solely on SF-CW. On the other hand, the NH4 + -N removal depends on the autotrophic bacteria, rather



Fig. 8 Inflow and outflow concentrations and removal rates of a  $NH_4$ <sup>+</sup>-N,  $\mathbf{b}$  NO<sub>3</sub><sup>-</sup>-N, and **c** TN under additional carbon source

than the carbon source. Therefore, the increase of organic matter content had negligible effect on NH<sub>4</sub><sup>+</sup>-N removal (Wei et al. [2017;](#page-10-0) Chen et al. [2013](#page-10-0); Katyal et al. [1988;](#page-10-0) Blecken et al. [2009](#page-10-0)). This observation is consistent with the results observed by Huang et al. ([2019](#page-10-0)) which pointed out that



Table 3

<span id="page-9-0"></span>**Table 4** Correlation between inflow  $\mathrm{COD}_{\mathrm{Mn}}$  concentration and N pollutant removal efficiencies

	Pollutants				
	$\text{COD}_{\text{Mn}}$	$NH_4^+$ -N	$NO3 - N$	TN	
Pearson correlation Saliency (bilateral)	$0.954**$ 0.000	0.277 0.360	$0.946**$ 0.000	$0.911**$ 0.000	

 $*$  $p$  < 0.01

COD concentration hardly affected activities of nitrifying bacteria. Earlier study also noted that the carbon source, as an electron acceptor of microbial denitrification process, has an important influence on the denitrification efficiency of CW (Wang et al. [2016\)](#page-10-0).

The removal rates of  $NO<sub>3</sub><sup>-</sup>-N$  under different inflow COD concentration are presented in Fig. [8b](#page-8-0). Results revealed that the change in  $NO_3$ <sup>-</sup>-N removal rate was low when the inflow concentration was 1.43 mg/L. Results also indicated that with the increase of inflow COD concentration (1.5, 2, and 2.5 times of initial value), the  $NO_3^-$ -N removal rate was increased from initial 15.2% to 19.0%, 21.9%, and 25.3%. The  $NO_3$ <sup>-</sup>-N removal rate in additional carbon source group was increased by 24.6%, 44.1%, and 66.1% compared with initial results. Correlation analysis (Table 4) indicated that there was significant correlation between inflow COD concentration and NO<sub>3</sub><sup>-</sup>-N removal rate ( $p < 0.01$ ). It can be seen that for the slightly-polluted river water with low  $NO<sub>3</sub><sup>-</sup>-N$  content, the addition of carbon source had an obvious effect on  $NO_3$ <sup>-</sup>N removal efficiency in SF-CWs. Denitrifying bacteria belonging to facultative aerobic microorganisms required higher DO and carbon source for effective denitrification process. Under anoxic conditions, the organic carbon source in water generally used as the electron donor, and the nitrification products  $(NO<sub>2</sub>-N$  and  $NO<sub>3</sub>-N)$  used as electron acceptors to reduce NO<sub>3</sub><sup>-</sup>-N to nitrogen (Blecken et al. [2009;](#page-10-0) Chen et al. [2013](#page-10-0); Wu et al. [2015\)](#page-10-0). Therefore, it can be concluded that the abundant organic carbon source is a necessary condition for the effective denitrification in CWs. With the gradual increase of COD in inflow, the organic carbon source which is used as the electron donor gradually accumulated and enhanced denitrification intensity and  $NO_3^-$ -N removal (Blecken et al. [2009\)](#page-10-0).

The removal rates of TN under different inflow COD concentration are presented in Fig. [8c](#page-8-0). Results revealed that the change of TN removal rate was similar with  $NO<sub>3</sub><sup>-</sup>-N$  removal when inflow concentration was 2.63 mg/L. With the increase in inflow COD concentration (1.5, 2, and 2.5 times of initial value), the TN removal rate was increased from initial 18.4% to 23.7%, 28.4%, and 32.4%. The TN removal rate of additional carbon source group increased by 28.9%, 54.7%, and 76.6%, compared with baseline result. Correlation analysis (Table 4) indicated that there was a significant correlation

between inflow COD concentration and TN removal rates  $(p < 0.01)$ . Hence, it can be concluded that under low TN concentration, the addition of the carbon source has an obvious effect on TN removal efficiency in SF-CWs (Blecken et al. [2009](#page-10-0); Liu et al. [2018\)](#page-10-0). The removal of TN mainly depends on the coupling of amination, nitrification, and denitrification process (Blecken et al. [2009](#page-10-0); Vymzal [2018](#page-10-0)). As  $NO<sub>3</sub><sup>-</sup>-N$  was accounted for a larger proportion of TN, the TN removal rate and  $NO_3^-$ -N removal rate showed an obvious correlation. Similar results were also observed by Wang et al. [\(2016\)](#page-10-0), which stated that an increase in COD/N ratio led to increasing reduction in NO<sub>3</sub><sup>-</sup>-N; meanwhile, efficient nitrification and denitrification promoted TN removal in CWs.

### Conclusion

Current study was conducted to evaluate the performance of 3-stage CWs, herein referred as W1, W2, and W3 for treating slightly polluted river water in Kunshan city. Results revealed that average removal rates of  $NH_4^+$ -N,  $NO_3^-$ -N, and TN were 38.4%, 22.3%, and 29.1%, respectively. The NH<sub>4</sub><sup>+</sup>-N was mainly treated in the first and the second CWs (W1, W2), while  $NO<sub>3</sub><sup>-</sup>-N$  and TN were in the second and the third CWs (W2, W3). Results on influencing factors indicated that HRT should be fixed between 3- and 7-day duration to ensure the outflow compliance rates and effective  $NO<sub>3</sub><sup>-</sup>-N$  and TN removal. A significant positive correlation was found between water temperature and N removal rates with the fact that N removal rates were higher in summer and autumn, but lower in spring and winter. Addition of external carbon source also promoted the removal of NO<sub>3</sub><sup>-</sup>-N and TN, but it had negligible effect on NH<sub>4</sub><sup>+</sup>-N removal. Results of this pilot study revealed that the N removal efficiency of CWs was stable and reliable. Therefore, it is feasible to transform the traditional impervious surfaces in city centre and squares into low impact development techniques such as constructed wetlands. For future research, authors intend to investigate the N removal performance of SF-CWs for moderately polluted rivers, instead of slightly polluted rivers.

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