

## Advanced treatment of sewage plant effluent by horizontal subsurface flow constructed wetlands: effect of coupling with cellulosic carbon sources

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### ABSTRACT

In order to construct an ecological treatment system for sewage plant effluent, horizontal subsurface flow constructed wetlands (HSCWs) coupled with cellulosic carbon sources were investigated. Cellulosic carbon sources including synthetic cellulose carbon (composed of cellulose, hemicellulose, and lignin) and alkali-modified corn straw had been tested for enhancing the removal of typical pollutants such as nitrogen and phosphorus in HSCWs. The results showed that HSCWs coupled with cellulosic carbon sources could intensify the denitrification process significantly, and the systems with alkali-modified corn straw performed much better than the systems with synthetic cellulose carbon. Constructed wetlands (CWs) packed with zeolite, fly ash ceramsite, sandstone, and fine sand had a better overall operation after adding 40 g alkali-modified corn straw. The best removals in HSCWs packed with zeolite, fly ash ceramsite (TN 76.89% and TP 78.90%) and HSCWs packed with fly ash ceramsite (TN 88.69% and TP 77.68%) were obtained with 40 g alkali-modified corn straw. However, much higher TN and TP removal efficiency up to 92.63% and 88.21% was realized in CWs packed with polyethylene plastic filler after adding 20 g alkali-modified corn straw. Most of the effluent chemical oxygen demand was below 30 mg/L after adding alkali-modified corn straw. These results indicated that the cellulosic carbon source such as corn straw could be used to improve the ecological treatment effect of tailwater at low cost.

*Keywords:* Constructed wetlands; Cellulosic carbon sources; Sewage plant effluent; Advanced treatment; COD/N ratio

### 1. Introduction

There is still a large amount of pollutants such as nitrogen, phosphorus and organic matter in the tailwater treated by secondary treatment in sewage treatment plants. Direct discharge of tailwater will increase the eutrophication of water bodies. More and more attention has been paid to the deep treatment of the sewage plant effluent. Tailwater generally has problems such as large volume, low organic matter concentrations, complex composition and high concentrations of nitrogen and phosphorus. Therefore,

it is especially important to seek an energy-saving and low-consumption ecological treatment technology. As an important ecological engineering technology, constructed wetlands (CWs) are effective and suitable for the treatment of micro-polluted water such as sewage plant effluent, agricultural non-point source polluted water and eutrophic lake water [1–3]. Typically, CWs can be classified into three types: surface flow CWs, horizontal subsurface CWs and vertical flow CWs according to the flow mode of water [4]. Among various types of CWs, horizontal subsurface flow constructed wetlands (HSCWs) can be of great potential

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because of high hydraulic load capacity, small installation area, great oxygen transport capacity and high decontamination performance [5]. It has been widely applied worldwide in the past decades [6,7].

Substrate in CWs has a great influence on the growth of microorganisms, adsorption, and deposition of pollutants [8,9]. Except for traditional substrates such as soil, sand, and gravel, the emerged substrates can be classified into ion-exchange substrates, sorption substrates and electron donor substrates [10]. Considering the construction cost and decontamination effect, substrates such as gravel, sandstone, blast furnace slag, alum slag, pelleted clay, etc. have been widely used in CWs nowadays [11]. However, some materials which have limited sorption capacity may not be used for a long time without considerate maintenance such as backwashing. Therefore, the appropriate selection of substrates in CWs is crucial. Zeolite has high potentiality to exchange cations such as  $\text{NH}_4^+$  and toxic metals from wastewater, so it has been considered as effective substrates in removing nitrogenous compounds and biodegradable organics [12–14]. Fly ash ceramsite is produced by high temperature burning of fly ash, clay, and admixture. Fly ash ceramsite not only is good for the adsorption of phosphate in sewage but also has good regeneration performance since fly ash contains more Si, Al active sites and a larger specific surface area [15]. Polyethylene plastic filler is a new type of microbial carrier with large specific surface area, good impact load resistance, and good hydrophilicity, and is suitable for the growth of biological membranes. It generally does not require reverse cleaning and can be used repeatedly. The suitable specific gravity allows it to be in a suspended flow, which enhances the impact of cutting of bubbles in the water and increases the oxygen content of the water [16]. However, research on the application of polyethylene plastic filler to CWs has rarely been carried out [10]. In this study, three groups of HSCWs were constructed with polyethylene plastic filler, zeolite, fly ash ceramsite, sandstone, and fine sand.

The nitrogen and phosphorus removal in CWs mainly depend on sedimentation, matrix adsorption, plant absorption and microbial transformation [17,18]. Studies have shown that the lack of carbon sources is a key factor limiting the denitrification performance of CWs. The carbon source is considered to be the controlling factor in the denitrification process because organic carbon acts as the electron donor to guarantee the complete denitrification while nitrate acts as the electron acceptor [19,20]. Also, the main ways to remove phosphorus from CWs are matrix adsorption and plant uptake, but microbes such as polyphosphate bacteria also have a certain effect on phosphorus removal [21]. The polyphosphate bacteria will compete with denitrifying bacteria for carbon sources, which will intensify the shortage of carbon sources. The addition of carbon source is an important way to ensure the normal life activities of ecosystem microorganisms and promote the removal of nitrogen and phosphorus by microorganisms. Insufficient carbon source in sewage plant effluent is a problem that must be faced in nitrogen and phosphorus reduction with CWs. Therefore, it is necessary to strengthen the research on external carbon source utilization technology. The traditional external carbon sources are mainly low molecular organic substances

such as methanol, acetic acid, glucose and sucrose [22]. They have been widely used in the denitrification of wastewater because of easy biodegradation. Nevertheless, they also have disadvantages such as high-cost and storage hazards [23]. Agricultural biomass such as corn straw can be an abundant, effective and inexpensive source of denitrifying carbon after proper treatment. Previous studies have indicated that the corn straw has unique structures for strain adherence, structural stability and slow release of carbon from long-time operation [24]. The cellulose of raw corn straw is usually wrapped by refractory lignin which hinders the release of carbon and affects the utilization by microorganisms. Alkali treatment can effectively remove the lignin, and improve the enzymatic hydrolysis and saccharification rate of the biomass material [25,26]. Agricultural biomass is mainly used as an auxiliary substrate for CWs in previous studies [23]. However, it may cause problems such as rot, secondary pollution, and maintenance difficulty. In this study, the corn straw was pretreated with alkali and then it was respectively added to the hydrolysis acidification tank where is easy to substitute or save extra carbon sources. It not only provides new ideas to solve the problem of agriculture wastes management but also helps to improve the ecological treatment effect of tailwater at low-cost.

Thus, the main purpose of this study was to investigate the advanced treatment of sewage plant effluent by HSCWs with cellulosic carbon sources. The effects of synthetic cellulose carbon (composed of cellulose, hemicellulose, and lignin) and alkali-modified corn straw on the removal of typical pollutants such as nitrogen and phosphorus were evaluated. The operational effects and differences in CWs coupled with external carbon sources that consisted of polyethylene plastic filler, zeolite, fly ash ceramsite, sandstone, and fine sand were also explored. It helps to construct a low-cost ecological technology for advanced treatment of low carbon/nitrogen ratio and micro-polluted water such as sewage plant effluent.

## 2. Material and methods

### 2.1. Description of CWs systems and operation

The experiment was conducted in Nanjing, East China ( $32^{\circ}04'45''\text{N}$ ,  $118^{\circ}49'01''\text{E}$ ) with subtropical monsoon climate, from February to April 2018. Fig. 1 shows the structure of the experimental CWs systems, and each system was made of the PVC sheets with a length of 110 cm, a width of 25 cm and a height of 70 cm. Each device was divided into four units, spaced apart by a PVC sheet uniformly arranged with small holes of 10 mm in diameter. The inlet water tank was separately placed next to the units and was connected to the CWs systems through a rubber hose with a flow meter. There was a backwashing pipe (20 mm pipe diameter) at the bottom of the device, and a hydrolysis acidification tank was arranged at the front to enhance the biodegradability of the water body. The designed backwashing strength used in the study was  $5 \text{ L/m}^2\text{-s}$  and backwashing time was 10 min. Three sets of devices were set up respectively, equipped with 700 mm polyethylene plastic filler (called as HSCW-Plastic), 300 mm sandstone + 100 mm fly ash ceramsite + 100 mm zeolite + 200 mm fine sand (called as HSCW-Zeolite/Ceramsite),

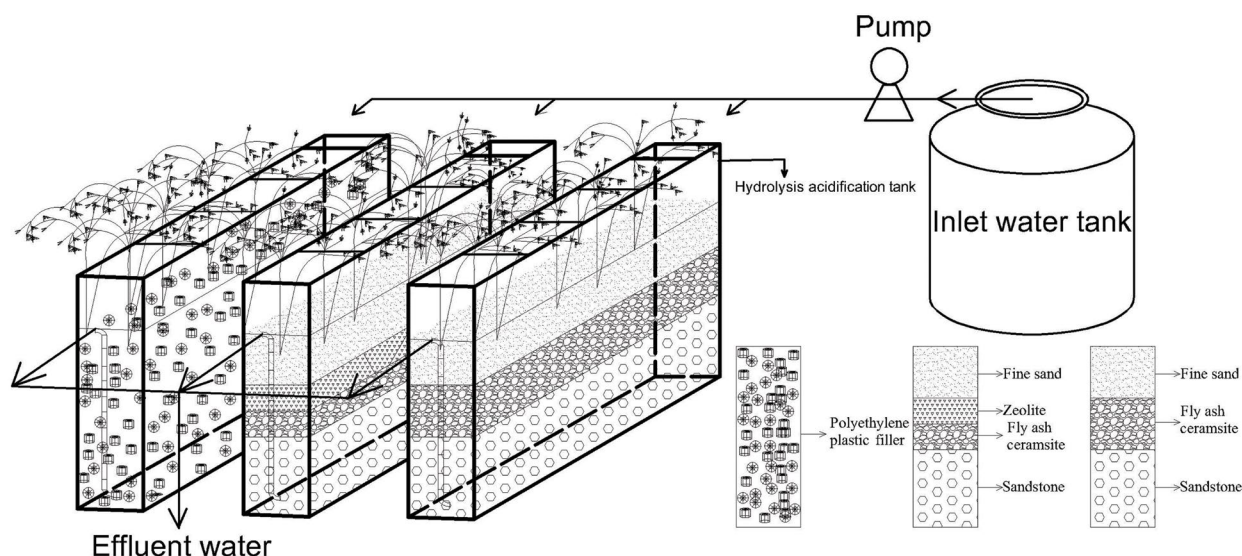


Fig. 1. Structure diagram of lab-scale horizontal subsurface-flow constructed wetland (HSCW).

300 mm sandstone + 200 mm fly ash ceramsite + 200 mm fine sand (called as HSCW-Ceramsite). *Oenanthe javanica* (*Oenanthe javanica* (BI.) DC.) was planted on the upper layer of the device, and the planting density was about 80 rhizomes per square meter.

Activated sludge was obtained from the oxidation ditch of Jiangning development district wastewater treatment plant (Nanjing, China) to inoculate the HSCWs. The activated sludge was acclimated under anaerobic domestication for 10 d, and the nutrient solution was regularly maintained to ensure the normal metabolic activities of the microorganisms. After acclimation, the sludge and glucose culture solution was used as the substrate for HSCWs in a ratio of 1:2. The HSCWs systems were fed with synthetic wastewater to promote microorganisms, plant growth, and stability. The synthetic wastewater based on sewage treatment plant effluent was composed of glucose,  $\text{NH}_4\text{Cl}$ ,  $\text{NaNO}_3$  and  $\text{KH}_2\text{PO}_4$  with tap water. The composition of the culture solution required for the experiment was shown in Table 1. After an acclimation period of one month, the experiment was officially launched from March 4th to April 30th, 2018.

In the first stage of this study, the C/N (COD/TN ratios) of wastewater was increased by adding synthetic cellulose carbon to synthetic tailwater, and the effect of synthetic cellulose carbon addition on CWs was studied. The synthetic cellulose carbon was prepared according to the proportion of corn straw fiber determined by the preliminary test results and consisted of 52.90% methyl cellulose, 19.82% lignin, 19.46% xylan. In the second stage, 40/20 g of alkali-modified corn straw was respectively added to study the removal efficiency of organic matter, nitrogen, and phosphorus. In phase I and III of this stage, the corn straw was not added and the devices were operated for 5 d. In phase II and IV, the devices were operated for 10 d with 40/20 g of alkali pretreated corn straw, respectively. The corn straw obtained from Hebei, China was cut into 2 cm and its epidermis was removed. It was pretreated in a solid–liquid ratio of 1:10 for 10 h with 2% NaOH, washed with water until the pH was neutral, and

then dried at  $105^\circ\text{C}$  for 2 h. In the experimental stage, the pre-treated corn straw was placed in a perforated polypropylene cylinder of 80 mm in diameter and 100 mm in height, and then added to the hydrolysis acidification tank at the front of CWs. Three submersible pumps (Model ML12VDC/3A, Shanghai Mingling Electromechanical Co. Ltd., China) were used to pump synthetic wastewater from an inlet water tank to CWs. The hydraulic retention time in the hydrolysis acidification tank and the whole system was 6.72 h and 26.88 h, respectively. The experimental apparatuses were maintained within a temperature range of  $15^\circ\text{C}$ – $25^\circ\text{C}$  during the laboratory experiments.

## 2.2. Synthetic wastewater

Synthetic wastewater was used as the experimental influent in this study. In the first stage, the influent was prepared with glucose,  $\text{NH}_4\text{Cl}$ ,  $\text{NaNO}_3$ , and  $\text{KH}_2\text{PO}_4$ . The COD was 60 mg/L, and the TN was 18 mg/L. The amount of synthetic cellulose carbon was calculated according to the theoretical value of C/N at 6 (synthetic cellulose carbon consisted of 52.90% methylcellulose, 19.82% lignin and 19.46% xylan), and 44.12 mg/L synthetic cellulose carbon was added to group c. Two times and three times of synthetic cellulose carbon in group c were respectively added to group d and

Table 1  
Microbial culture solution formula

Water quality index	Concentration (mg/L)	Formula composition
COD	50–60	Glucose
Ammonia nitrogen	5–10	$\text{NH}_4\text{Cl}$
Nitrogen nitrate	8–10	$\text{NaNO}_3$
Total nitrogen	15–20	$\text{NH}_4\text{Cl}$ , $\text{NaNO}_3$
Total phosphorus	1	$\text{KH}_2\text{PO}_4$

group e. Table 2 shows the actual values of the water quality indices of the influent of each experimental group. In the second stage, the influent was prepared as the first stage. But the carbon source in the influent was only composed of glucose. The COD concentration of influent was 50–60 mg/L. The concentrations of nitrate–nitrogen and ammonia nitrogen were in the range of 8–10 mg/L, 5–10 mg/L, respectively, and the concentration of TP was about 1 mg/L.

### 2.3. Experimental sampling and statistical analysis

The samples from the inlet water tank and effluents were taken respectively after 48 h stable operation period in the first stage. But in the second stage, the samples were collected from CWs and the inlet water tank every 1 or 2 d with different alkali-modified corn straw dosage. The concentrations of COD,  $\text{NH}_4^+\text{-N}$ ,  $\text{NO}_3^-\text{-N}$ , TN, and TP in the water were analyzed according to the standard analysis methods issued by the American Public Health Association [27].

The removal efficiency of these pollutants was calculated using the formula shown in Eq. (1).

$$E = \frac{(C_{\text{influent}} - C_{\text{effluent}})}{C_{\text{influent}}} \times 100\% \quad (1)$$

where  $E$  - efficiency of these pollutants removal by HSCWs;  $C$  - concentration of COD,  $\text{NH}_4^+\text{-N}$ ,  $\text{NO}_3^-\text{-N}$ , TN or TP in the influent or effluent.

The statistical analysis was carried out using SPSS version 21.0 software (SPSS Inc., IBM, USA). One-way analysis of variance (ANOVA) tests and the “ $t$ ” test for independent-samples was performed to identify any significant differences between samples, with  $P < 0.05$  indicating significance. The operational stability of CWs was evaluated by the coefficient of variation  $C_v$  according to Eq. (2).

$$C_v = \frac{\sigma}{\mu} \quad (2)$$

where  $\sigma$  - standard deviation;  $\mu$  - average value.

## 3. Results and discussion

### 3.1. Extraction of pollutants by CWs coupling with synthetic cellulose carbon

Fig. 2 illustrated the extraction and efficiency of pollutants by CWs coupling with synthetic cellulose carbon. When the COD concentration of the influent was lower than 101.77 mg/L, the effluent varied in the range of 0–20 mg/L (Fig. 2a). With the dosage of 88.24 or 132.36 mg/L synthetic cellulose carbon, the removal efficiency of COD by CWs coupling with synthetic cellulose carbon decreased. Meanwhile, the microbial decomposition of organic matter and nitrifying bacteria competed for dissolved oxygen, which would lead to dissolved oxygen deficiency and reduce microbial removal of  $\text{NH}_4^+\text{-N}$  [17]. As shown in Fig. 2b, the average removal efficiency of  $\text{NH}_4^+\text{-N}$  decreased by about 25% with the increase of influent COD concentration. Contrary to the  $\text{NH}_4^+\text{-N}$  removal, the TN removal efficiency of CWs was gradually improved with the dosage of synthetic

Table 2

Influent water quality indicators and carbon source dosage in the first stage

Water quality index	Concentration (mg/L)				
	<i>a</i>	<i>b</i>	<i>c</i>	<i>d</i>	<i>e</i>
COD	21.05	67.59	101.77	148.9	176.82
Ammonia nitrogen	5.09	6.66	6.93	8.96	3.94
Total nitrogen	16.54	19.91	16.18	18.79	12.26
Total phosphorus	0.96	1.02	0.93	0.99	0.98
C/N	1.27	3.40	6.29	7.93	14.43
Synthetic cellulose carbon dosage(mg/L)	0	0	44.12	88.24	132.36

cellulose carbon increased, which indicated that the C/N was the limiting factor of microbial denitrification (Fig. 2c). The increase of C/N improved nitrogen removal. Overall, the TP removal performance of CWs was greatest when the dosage of synthetic cellulose carbon was 44.12 mg/L. But the removal efficiency reduced when the amount of dosage increased. It was mainly due to that proper amount of carbon source contributed to phosphorus uptake by phosphorus accumulating organisms (PAOs) in CWs, however, decomposition of organic matter competed for dissolved oxygen with excessive carbon dosage. It inhibited decomposition of the PHB (poly- $\beta$ -hydroxybutyric acid)-like substances in the aerobic zone and reduced the uptake of dissolved phosphate by PAOs [28,29].

All these results indicated that an appropriate dosage of synthetic cellulose carbon contributed to nitrogen and phosphorus removal in CWs when the influent maintained low C/N. However, organic matter removal would be affected by an excessive dosage of synthetic cellulose carbon [30]. Strong et al. [31] investigated that systemic microorganisms have different responses to specific forms of carbon sources, and used a liquor from the wet oxidation of waste activated sludge as a carbon source of the sequencing batch reactor, showing that the denitrification was excellent due to the variety of organic carbon substrates (particularly acetic acid) present within the liquor. Therefore, it was necessary to increase the content of bioavailable components in the carbon source to reduce the organic matter level in the effluent while effective denitrification was accomplished.

As shown in Fig. 2b, the  $\text{NH}_4^+\text{-N}$  concentration range of effluents from HSCW-Plastic, HSCW-Zeolite/Ceramsite, and HSCW-Ceramsite was 0.88–3.07 mg/L, 0.72–2.24 mg/L and 0.56–2.43 mg/L, respectively. The removal efficiency was separately in the range of 63.49%–84.52%, 63.49%–85.89%, and 55.56%–88.96%. As a whole, there was no significant difference ( $P = 0.732$ ) for effluent  $\text{NH}_4^+\text{-N}$  removal efficiency among CWs with different external carbon source. Under the same influent condition, the mean removal efficiency of  $\text{NH}_4^+\text{-N}$  by HSCW-Zeolite/Ceramsite and HSCW-Ceramsite (74.66% and 75.20%, respectively) was higher than that of HSCW-Plastic (72.64%) mainly due to adsorption of ammonia nitrogen partly by zeolite and fly ash ceramsite. Lu et al. [32] analyzed ammonia nitrogen adsorption and desorption characteristics in twenty-nine kinds of common CWs substrates, showing that natural zeolite is a high-quality matrix material with

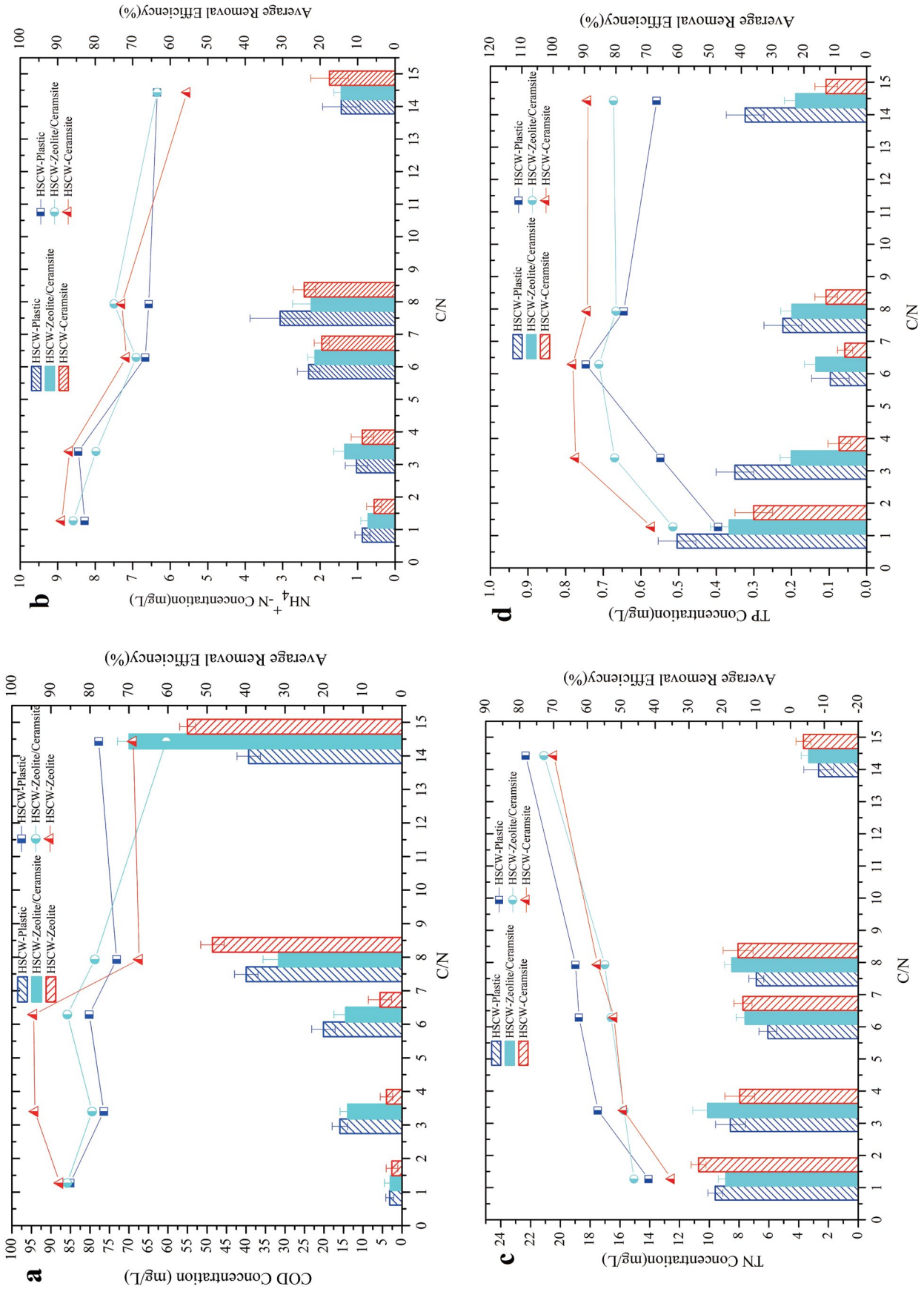


Fig. 2. Extraction of (a) COD, (b) NH<sub>4</sub><sup>+</sup>-N, (c) TN, and (d) TP at different C/N by CWs coupling with synthetic cellulose carbon.

good permeability, low desorption risk, long-term adsorption, and other advantages. Many previous studies have shown that the zeolite grains are very efficient at ammonium removal [16,23].

The TN concentration range of effluents from HSCW-Plastic, HSCW-Zeolite/Ceramsite, and HSCW-Ceramsite was 2.66–9.60 mg/L, 3.33–10.11 mg/L, and 3.68–10.72 mg/L, respectively. The removal efficiency was in the range of 41.96%–78.27%, 46.25%–72.84%, and 35.19%–69.94%, respectively (Fig. 2c). With the same influent, the mean TN removal efficiency of HSCW-Plastic (60.63%) was higher than that of HSCW-Zeolite/Ceramsite and HSCW-Ceramsite (55.24% and 52.73%, respectively,  $P = 0.345$ ). It indicated that the denitrification of HSCW-Plastic was relatively good mainly due to the superior biofilm formation on the plastic filler. Tatoullis et al. [16] evaluated a plastic substrate media (high-density polyethylene) with high porosity and hydraulic conductivity in CWs and found the hydraulic (0.08 m/d) and organic loads (620 g/m<sup>2</sup>/d) of units containing the plastic material and zeolite can be three and four times higher than that in CWs containing gravel and zeolite, respectively. This revealed that the units with plastic material received much higher organic loads.

According to the data in Fig. 2d, the TP concentration range of effluents from HSCW-Plastic, HSCW-Zeolite/Ceramsite, and HSCW-Ceramsite was 0.10–0.50 mg/L, 0.14–0.37 and 0.06–0.30 mg/L, respectively. The removal efficiency was separately in the range of 47.37%–89.58%, 61.83%–85.42%, and 68.65%–93.76%. On the whole, the average removal efficiency of TP recorded in this study (47.37%–93.76%) was higher than the results (48%–67%) of Wu et al. [33] who assessed the long-term performance in removing nitrogen and phosphorus from simulated polluted river water in surface-flow constructed microcosm wetlands in northern China. The calcium silicate in the fly ash ceramsite could form an ion exchange with the phosphate ions, thereby converting the phosphate ions into precipitates and depositing on the surface of the ceramsite. Therefore, fly ash ceramsite could enhance the removal of phosphorus. Cheng et al. [15] also shows that the active calcium in the sustainable ceramsite substrate material (by using coal fly ash and waterworks sewage as the main material) played the dominant role in the adsorption of phosphorus, and neutral-alkaline conditions promoted the adsorption by facilitating the formation of calcium phosphate precipitate. Moreover, the growth situation of *Oenanthe javanica* which had a certain contribution to the absorption of phosphorus in HSCW-Zeolite/Ceramsite and HSCW-Ceramsite was better than that in HSCW-Plastic. Wu et al. [33] assessed the uptake of phosphorus in CWs by plants constituted 10.76%–34.17% of the mass phosphorus removal and the main removal pathway in phosphorus was substrate accumulation. Consequently, HSCW-Zeolite/Ceramsite and HSCW-Ceramsite (77.67% and 86.67%, respectively) performed better than HSCW-Plastic (69.45%) in TP removal ( $P = 0.074$ ).

### 3.2. Extraction of pollutants by CWs coupling with alkali-modified corn straw

At present, the commonly used wetland denitrification external carbon sources are mainly divided into three

categories, that is, traditional low-molecular liquid carbon sources, waste carbon sources, and biodegradable polymers [34]. Traditional low-molecular carbon sources are utilized extensively and easy to decompose, but there are problems such as difficulty in controlling dosage, storage and transportation, self-toxicity, and accumulation of nitrite [30]. A degradable polymer is a stable and slow-release carbon source that is easy to biodegrade. At present, it is commonly used in the research of nitrate wastewater, nitrate-containing groundwater remediation and so forth [35,36]. Due to its high cost, it is unreasonable for low-cost ecological treatment technologies, such as CWs, in terms of economic benefits and construction concepts. Waste carbon sources mainly include agricultural waste, plant litter, landfill leachate, sludge and so on [34]. Agricultural waste has been extensively studied as a new type of solid carbon source in recent years. It has the advantages of non-toxicity, rich source, slow carbon release and low-cost. At present, carbon sources of agricultural wastes are mainly corn cob, rice husk, straw, nutshell, wood chips and so on [36]. However, agricultural waste, as a source of cellulose carbon, also has problems such as slow start-up, difficulty in addition and replacement, and secondary pollution. The original agricultural waste was directly filled as a wetland substrate in most of the current research [34–36]. Nevertheless, in this study, it was firstly treated with alkali and then fixed in a hydrolysis acidification tank at the front of CWs to evaluate the feasibility of the scheme and the effect of contaminant removal.

### 3.3. Effect of alkali-modified corn straw addition on COD concentration of effluent

The variations of COD concentrations in three systems during the experimental period are shown in Fig. 3. The effluent COD concentrations fluctuated greatly at different stages, but most of the effluent COD concentrations were below 30 mg/L.

COD is important for the assessment of secondary pollution problems in the construction of CWs coupled with cellulose carbon. Jia et al. [37] explored the feasibility and efficiency by using agricultural biomass such as wheat straw, apricot pit, and walnut shell as substrates in VFCWs to enhance nitrogen removal in treating low carbon sewage. It was noted that the conventional wetland was more efficient in organic matter degradation than the other three systems with the addition of biomass materials. Organic matters released from the biomass itself were partially used to enhance the removal of nitrogen and some components such as lignin which were difficult for microorganisms to degrade made the COD concentration increase.

The carbon source provides sufficient carbon to enhance denitrification and it is not suitable to produce a large amount of organic matter, especially organic matter that is difficult to degrade, resulting in an increase in COD of the effluent. Zheng et al. [38] studied the changes in structure and material composition of corn straw after wet state NaOH pretreatment. It was found that the main component of alkali-modified corn straw leachate was mainly biodegradable soluble small molecule organic matter and pretreatment effectively improved the biodegradability of corn straw, which is consistent with the research conclusions of

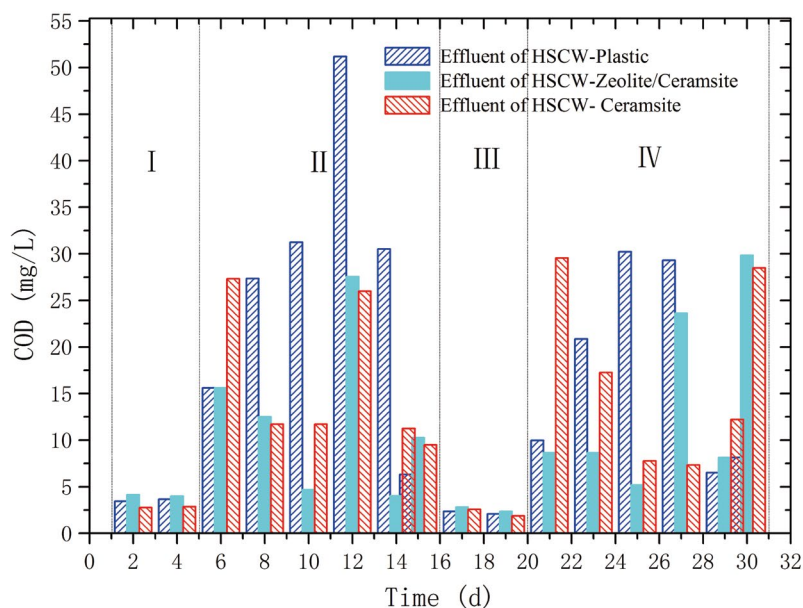


Fig. 3. Variation of COD concentration in effluents of CWs coupling with alkali-modified corn straw.

this research. The effluent organic matter content presented a downward trend after an initial increase with alkali-modified corn straw addition. Although the effluent COD concentrations reached 50 mg/L on the seventh day after adding 40 g alkali-modified corn straw, the organic matter in the effluents could be effectively decomposed and the overall water quality would not deteriorate through dosage control. Also, Yao et al. [39] developed a hybrid macrophyte residue denitrifying bioreactor for nitrate removal from surface water. Although the TOC concentrations in the effluent within the initial 50 d, especially the initial 20 d, were significantly higher than those in the influent, it can be eliminated through aeration.

It can be noted that the stability of HSCW-Plastic ( $C_v = 0.832$ ) was poor after adding a carbon source, and the effluent COD concentration reached 50 mg/L at most. HSCW-Zeolite/Ceramsite ( $C_v = 0.829$ ) and HSCW-Ceramsite ( $C_v = 0.742$ ) exhibited better stability in controlling the organic matter than HSCW-Plastic due to large porosity ratio and particle size of plastic filler, the high retention properties of sandstone, fly ash ceramsite and zeolite.

### 3.4. Nitrogen removal performance by different coupling systems

The difference in nitrogen removal performance of CWs coupling with alkali-modified corn straw was studied in this part. Directly adding plant biomass into CWs was different from the synthetic cellulose carbon dissolved in the influent water. The main problems included the unstable release of carbon source, the availability of organic matter by microorganisms, and the increase of COD concentration in the effluent water [40]. Therefore, it was necessary to study further the effects of different plant biomass dosage on the removal of pollutants from the system. As shown in Fig. 4, the concentrations of  $\text{NH}_4^+\text{-N}$ ,  $\text{NO}_3^-\text{-N}$  and TN varied with

the alkali-modified corn straw dosage. Fig. 5a exhibited the variation of  $\text{NH}_4^+\text{-N}$  and TN removal efficiency in effluents of CWs.

$\text{NH}_4^+\text{-N}$  in the influent varied slightly in the range of 7.59–8.86 mg/L, and final effluents in four stages varied in the range of 1.39–2.22, 0.89–4.95, 1.03–2.30, and 0.45–4.91 mg/L, respectively. Because of the microbial decomposition of organic matter and nitrifying bacteria competed for dissolved oxygen as mentioned in section 3.1. It can be seen that  $\text{NH}_4^+\text{-N}$  concentrations in the effluents mostly increased and the removal efficiency decreased when the alkali-modified corn straw was added. Transformation of  $\text{NH}_4^+\text{-N}$  into  $\text{NO}_3^-\text{-N}$  requires high DO concentration [17]. It's obvious from Fig. 5a that the addition of external biomass carbon sources affected the removal of  $\text{NH}_4^+\text{-N}$  to some extent. However, Jia et al. [37] have shown that the change of influent C/N ratios, as well as the addition of external biomass carbon sources, had little effect on the removal of  $\text{NH}_4^+\text{-N}$ . It could be mainly due to intermittent aeration providing sufficient oxygen to enhance nitrification. Therefore, further study should focus on oxygen concentration and aeration in HSCWs with agricultural biomass.

$\text{NO}_3^-\text{-N}$  and TN in the influent respectively varied slightly in the range of 13.69–14.99 and 20.55–21.72 mg/L in different stages (Figs. 4b and c). Effluent  $\text{NO}_3^-\text{-N}$  concentrations in four stages varied in the range of 6.29–9.11, 0.55–7.34, 6.39–10.22, and 0.34–11.64 mg/L, respectively. And in the meanwhile, the removal of TN has also been improved to a varying degree. The concentrations of effluent TN in four stages were in the range of 7.52–11.24, 1.60–10.89, 8.21–12.11, and 1.13–13.88 mg/L, respectively.

In phases I and III of this part, the mean TN removal efficiency was 60.67%, 45.80% and 50.42% in the HSCW-Plastic, HSCW-Ceramsite, and HSCW-Zeolite/Ceramsite without alkali-modified corn straw. Moreover, the effluent TN concentrations were mostly above 8 mg/L. According to

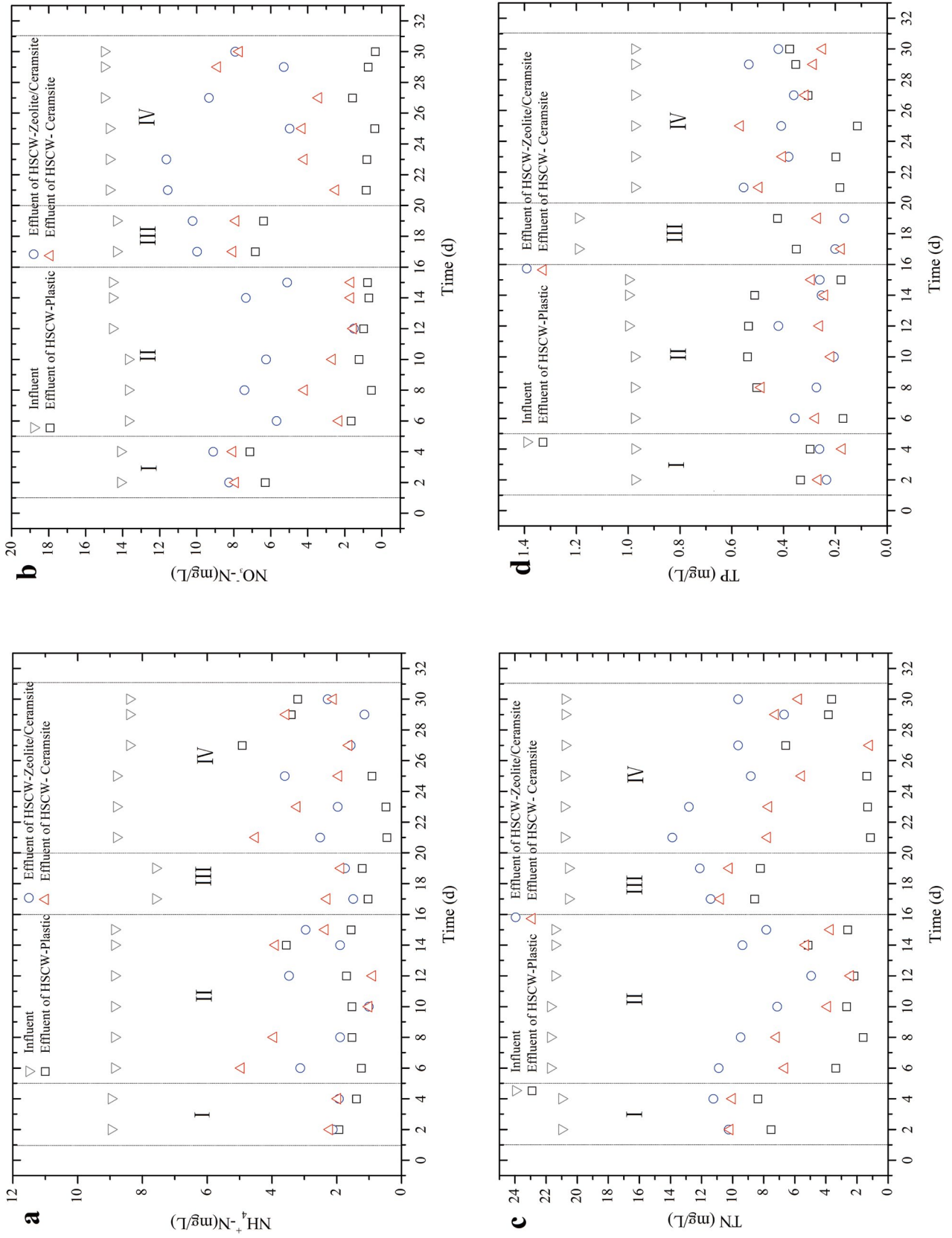


Fig. 4. Variations of (a)  $NH_4^+-N$ , (b)  $NO_3^-N$ , (c) TN, and (d) TP concentrations in effluents of CWs coupling with alkali-modified corn straw.



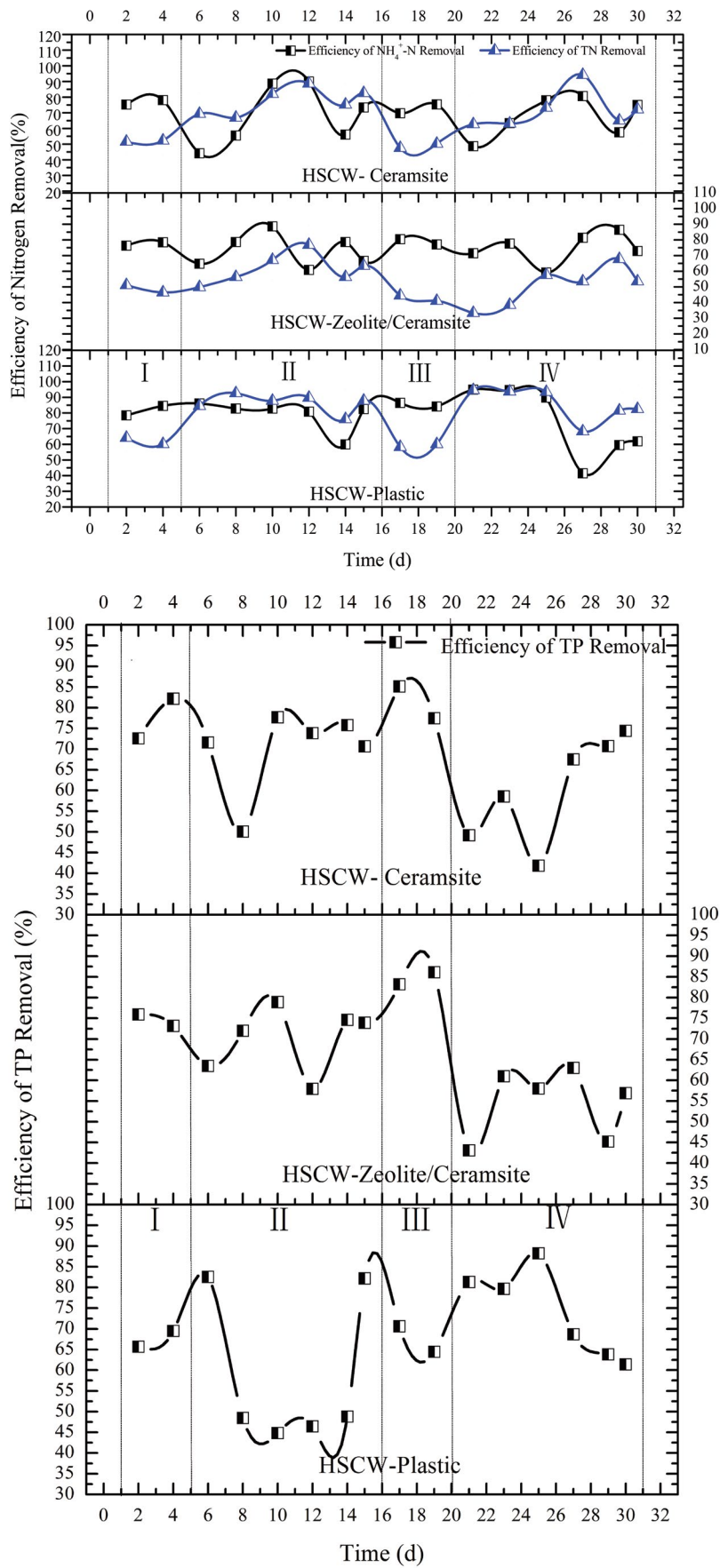


Fig. 5. Variation of (a) nitrogen and (b) TP removal efficiency in effluents of CWs coupling with alkali-modified corn straw.

Table 3  
Estimated cost of common denitrification carbon sources

Carbon source	Price (CNY/kg)	Consumption of Substance (kg/kg N)	Nitrogen removal costs (CNY/kg N)	Reference
Methanol	7.70	2.08–3.98	16.02–30.65	[47]
Ethanol	9.25	2.0	18.50	[47]
Acetic acid	18.49	3.5	64.72	[47]
Glucose	2.40	4.6	11.04	[30,47]
Straw	0.19	7.8	1.43	[34]
Cotton	5.70	2.8	15.97	[48]
Alkali-modified corn straw	0.35	12.93	4.53	This study
PCL	68.00	2.34	159.1	[49]
PHB	77.06	2.1–2.7	161.83–208.06	[48]
PBS	2.11	24.8	52.33	[50]

the first stage in the experiment, it could be found that the removal was relatively poor. With 40/20 g of alkali-modified corn straw, there was a slight difference in the TN removal in HSCW-Zeolite/Ceramsite ( $P = 0.122$ ). However, the TN removal efficiency was significantly greater than that without alkali-modified corn straw in HSCW-Plastic and HSCW-Ceramsite ( $P = 0.000$ ). The mean TN removal efficiency was separately 86.44%, 61.67%, and 77.49% in HSCW-Plastic, HSCW-Zeolite/Ceramsite, and HSCW-Ceramsite with 40 g alkali-modified corn straw, while that was 85.69%, 50.72%, and 71.77% with 20 g alkali-modified corn straw, respectively. The TN removal in HSCW-zeolite/ceramsite and HSCW-ceramsite was generally similar ( $P = 0.108$ ), and the better overall operation was achieved after adding 40 g alkali-modified corn straw. HSCW-Plastic performed better than the others ( $P = 0.000$ ) in the nitrogen removal on the whole due to the superior biofilm and sufficient dissolved oxygen content [16]. The maximum TN removal efficiency of HSCW-Plastic coupling with 40/20 g alkali-modified corn straw was up to 92.63% and 94.59%, respectively, while the TN removal efficiency of HSCW-Plastic without alkali-modified corn straw was only about 60.67%. Natural organic substances, such as apricot pit (removal efficiency improved from 3.98% to 19.42%), wheat straws (removal efficiency improved from 3.98% to 96.89%) and walnut shell (removal efficiency improved from 3.98% to 62.88%), can all be used as organic carbon sources for denitrification in Jia's study [37]. It proved that agricultural biomass such as wheat straw could improve the nitrate removal rate greatly. Gao et al. [41] established an electrolysis-integrated horizontal sub-surface-flow constructed wetland and the removal rate of nitrate could be up to 75.5% during the entire experiment. The results in this study demonstrated a great improvement for the TN removal in HSCWs with agricultural biomass. The effluent TN concentrations of the HSCW-Plastic, HSCW-Ceramsite and HSCW-Zeolite/Ceramsite with alkali-modified corn straw could be stabilized below 7, 8 and 14 mg/L. However, compared with the experiment in section 3.1, the advantages of adsorption properties of Zeolite and Ceramsite diminished due to adsorption saturation.

The TN removal of the HSCW-Zeolite/Ceramsite and HSCW-Ceramsite was gradually improved after adding alkali-modified corn straw and had some hysteresis compared with that of HSCW-Plastic.

Compared with synthetic cellulose carbon, the TN removal efficiency of HSCW-Plastic, HSCW-Zeolite/Ceramsite and HSCW-Ceramsite reached 63.55%, 54.86%, and 57.03%, respectively, and the effluent COD was in the range of 30–50 mg/L. However, the effluent COD with alkali-modified corn straw was generally stable below 30 mg/L, and the TN removal efficiency was up to 94.59%, 76.89%, and 94.18%, respectively. The removal of nitrogen in CWs after adding alkali-modified corn straw was better than that using synthetic cellulose carbon. Synthetic cellulose carbon increased the viscosity of the influent, causing partial blockage of CWs and exacerbating the system's dissolved oxygen deficiency. The bioavailability of the unmodified synthetic cellulose carbon was worse than that of the modified corn straw so that the actual influent biodegradable carbon of the modified corn straw was more and system performance was better.

### 3.5. Phosphorus removal performance by different coupling systems

In CWs, phosphorus removal mainly depended on adsorption and precipitation within the filter media [42,43]. Also, it was related to biodegradation, plants' uptake and root exudates. Consequently, HSCW-Zeolite/Ceramsite and HSCW-Ceramsite ( $C_v = 0.113$  and  $C_v = 0.129$ , respectively) performed more stably and better than HSCW-Plastic ( $C_v = 0.231$ ) in TP removal at early stages ( $P = 0.000$ ). Fig. 4d and Fig. 5b illustrates the variation of TP concentration and removal efficiency, respectively. The removal efficiency of TP was in the range of 57.94%–86.10%, and the concentration of effluent TP was below 0.5 mg/L in the first three stages. Li et al. [3] analyzed sixty-one HSCWs in China and the data were within 3 y after CWs operation. The average TP removal efficiency was about 70.9% in HSCWs. The total phosphorus removal was deteriorated in the final stage due

to the blockage or adsorption saturation. Considering that the influent was mainly formulated with cellulose, glucose, nitrogen and phosphorus nutrients, the blockage of the system was mainly caused by cellulose and other organic substances, which could be solved by backwashing the substrate or resting operation on bio-clogging [45].

The removal efficiency of TP in HSCW-Plastic was in the range of 43.09%–74.39%, and the concentration of effluent TP varied between 0.25 and 0.55 mg/L. After adding 40/20 g of alkali-modified corn straw, the removal efficiency of TP decreased by 24.7% and 9.32%, respectively. It could be observed in Fig. 4b that appropriate addition of carbon source contributed to the improvement of phosphorus removal in HSCW-Plastic. Excessive carbon source caused the competition for dissolved oxygen due to the decomposition of organic matter, which would affect the absorption of dissolved phosphate by PAOs [29]. Xiong et al. [44] prepared a novel solid slow-release carbon source using polycaprolactone and peanut shells as carbon sources with polyvinyl alcohol-sodium alginate as hybrid scaffolds. The utilization efficiency of the carbon source was reduced when the carbon release rate was too fast. Therefore, according to the amount of wastewater and nitrate concentration, adjusting the release rate close to consumption rate was the key to improve the utilization of external carbon sources. In general, the amount of carbon added to CWs must be controlled especially for HSCW-Plastic.

### 3.6. Analysis of economic benefits of cellulosic carbon source

The cost of nitrogen removal and the denitrification rate of alkali-modified corn straw are presented in Table 3, which have been compared with other common denitrification carbon sources. The unit material prices of the carbon sources were based on the Chinese market in November 2019. Crop straw is a valuable biomass resource produced in crop production. The total amount of food crop straw in China was up to 598 million tons in 2014, and corn straw surpassed rice straw as the most abundant straw, accounting for 36.1% of the national straw yield in 2012 [46]. According to the statistics from the ministry of agriculture and rural affairs of the People's Republic of China, the comprehensive utilization rate of straw in China has reached 81.68%, but there are still about  $1.5 \times 10^8$  t straw burned or abandoned. Straw burning produces huge carbon emissions, which will have a serious impact on the ecological environment. In this study, alkali-modified corn straw was used as a denitrification carbon source in CWs, which provided a new idea for the recycling of corn straw waste. Corn straw as a kind of agricultural waste, the price is low, according to the agricultural products information network ([www.nongnet.com](http://www.nongnet.com)), the price of corn straw is RMB 150 yuan/t. NaOH used in modification with an average price of about RMB 4,000 yuan/t according to the China chemical industry website ([www.china.chemnet.com](http://www.china.chemnet.com)), and it can be recycled and calculated according to the 75% recovery rate. The results showed that the modification cost of corn straw was about RMB 200 yuan/t when it was soaked in NaOH with a 2% mass concentration for 10 h according to the solid-liquid ratio of 1:10, and the total cost of alkali-modified corn straw was about RMB 350 yuan/t. Compared

with methanol, acetic acid, and other commonly used carbon sources, the additional cost is lower and is conducive to solving the waste of agricultural waste resources of corn straw, which has the advantages of low operating cost, environmental friendliness and good application prospects [36,37].

## 4. Conclusions

The controlled dosage of alkali-modified corn straw which had better biodegradability than synthetic cellulose carbon could effectively improve the removal of nitrogen and phosphorus without causing deterioration of water quality. HSCW-Zeolite/Ceramsite and HSCW-Ceramsite had better overall operation after adding 40 g alkali-modified corn straw. The maximum TN removal efficiency was 76.89% and 88.69%, respectively, while the maximum TP removal efficiency was 78.90% and 77.68%, respectively. HSCW-Plastic performed better after adding 20 g alkali-modified corn straw. The TN and TP removal efficiency was up to 92.63% and 88.21%. The combination of alkali-modified corn straw with CWs could be used to promote the removal of typical pollutants such as nitrogen and phosphorus.

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## References

- [1] J. Tournebize, C. Chaumont, Ü. Mander, Implications for constructed wetlands to mitigate nitrate and pesticide pollution in agricultural drained watersheds, *Ecol. Eng.*, 103 (2017) 415–425.
- [2] N. Dal Ferro, H.M.S. Ibrahim, M. Borin, Newly-established free water-surface constructed wetland to treat agricultural waters in the low-lying Venetian plain: performance on nitrogen and phosphorus removal, *Sci. Total Environ.*, 639 (2018) 852–859.
- [3] X. Li, A. Ding, L. Zheng, B.C. Anderson, L. Kong, A. Wu, L. Xing, Relationship between design parameters and removal efficiency for constructed wetlands in China, *Ecol. Eng.*, 123 (2018) 135–140.
- [4] H. Wu, J. Fan, J. Zhang, H.H. Ngo, W. Guo, Z. Hu, J. Lv, Optimization of organics and nitrogen removal in intermittently aerated vertical flow constructed wetlands: effects of aeration time and aeration rate, *Int. Biodeterior. Biodegrad.*, 113 (2016) 139–145.
- [5] R. Wang, X. Zhao, H. Liu, H. Wu, Elucidating the impact of influent pollutant loadings on pollutants removal in agricultural waste-based constructed wetlands treating low C/N wastewater, *Bioresour. Technol.*, 273 (2019) 529–537.
- [6] J. Fan, W. Wang, B. Zhang, Y. Guo, H.H. Ngo, W. Guo, J. Zhang, H. Wu, Nitrogen removal in intermittently aerated vertical flow constructed wetlands: impact of influent COD/N ratios, *Bioresour. Technol.*, 143 (2013) 461–466.
- [7] N. Russo, A. Marzo, C. Randazzo, C. Caggia, A. Toscano, G.L. Cirelli, Constructed wetlands combined with disinfection systems for removal of urban wastewater contaminants, *Sci. Total Environ.*, 656 (2019) 558–566.
- [8] C. Vohla, M. Kõiv, H.J. Bavor, F. Chazarenc, Ü. Mander, Filter materials for phosphorus removal from wastewater in treatment wetlands—a review, *Ecol. Eng.*, 37 (2011) 70–89.
- [9] Z. Wang, Z. Wang, L. Chen, Z. Lin, Y. Liu, Y. Liu, Using an attapulgite-activated carbon composite ceramsite biofilter to

- remove dibutyl phthalate from source water, *Pol. J. Environ. Stud.*, 27 (2018) 897–903.
- [10] Y. Yang, Y. Zhao, R. Liu, D. Morgan, Global development of various emerged substrates utilized in constructed wetlands, *Bioresour. Technol.*, 261 (2018) 441–452.
- [11] S. Lu, X. Zhang, J. Wang, L. Pei, Impacts of different media on constructed wetlands for rural household sewage treatment, *J. Cleaner Prod.*, 127 (2016) 325–330.
- [12] Z. Wang, J. Dong, L. Liu, G. Zhu, C. Liu, Screening of phosphate-removing substrates for use in constructed wetlands treating swine wastewater, *Ecol. Eng.*, 54 (2013) 57–65.
- [13] Z. Wang, Z. Wang, K. Xu, L. Chen, Z. Lin, Y. Liu, Performance evaluation of adsorptive removal of Ni(II) by treated waste granular-activated carbon and new granular-activated carbon, *Desal. Water Treat.*, 161 (2019) 315–326.
- [14] M.A. Shavandi, Z. Haddadian, M.H.S. Ismail, N. Abdullah, Z.Z. Abidin, Removal of Fe(III), Mn(II) and Zn(II) from palm oil mill effluent (POME) by natural zeolite, *J. Taiwan Inst. Chem. Eng.*, 43 (2012) 750–759.
- [15] G. Cheng, Q. Li, Z. Su, S. Sheng, J. Fu, Preparation, optimization, and application of sustainable ceramsite substrate from coal fly ash/waterworks sludge/oyster shell for phosphorus immobilization in constructed wetlands, *J. Cleaner Prod.*, 175 (2018) 572–581.
- [16] T. Tatoulis, C.S. Akratos, A.G. Tekerlekopoulou, D.V. Vayenas, A.I. Stefanakis, A novel horizontal subsurface flow constructed wetland: reducing area requirements and clogging risk, *Chemosphere*, 186 (2017) 257–268.
- [17] D. Chen, X. Gu, W. Zhu, S. He, F. Wu, J. Huang, W. Zhou, Denitrification- and anammox-dominant simultaneous nitrification, anammox and denitrification (SNAD) process in subsurface flow constructed wetlands, *Bioresour. Technol.*, 271 (2019) 298–305.
- [18] Z. Wang, M. Zhong, J. Wan, G. Xu, Y. Liu, Development of attapulgite composite ceramsite/quartz sand double-layer biofilter for micropolluted drinking source water purification, *Int. J. Environ. Sci. Technol.*, 13 (2016) 825–834.
- [19] X. Song, S. Wang, Y. Wang, Z. Zhao, D. Yan, Addition of Fe<sup>2+</sup> increase nitrate removal in vertical subsurface flow constructed wetlands, *Ecol. Eng.*, 91 (2016) 487–494.
- [20] Y. Shen, L. Zhuang, J. Zhang, J. Fan, T. Yang, S. Sun, A study of ferric-carbon micro-electrolysis process to enhance nitrogen and phosphorus removal efficiency in subsurface flow constructed wetlands, *Chem. Eng. J.*, 359 (2019) 706–712.
- [21] E.D. Roy, Phosphorus recovery and recycling with ecological engineering: a review, *Ecol. Eng.*, 98 (2017) 213–227.
- [22] W. Li, X. Shan, Z. Wang, X. Lin, C. Li, C. Cai, G. Abbas, M. Zhang, L. Shen, Z. Hu, H. Zhao, P. Zheng, Effect of self-alkalization on nitrite accumulation in a high-rate denitrification system: performance, microflora, and enzymatic activities, *Water Res.*, 88 (2016) 758–765.
- [23] G. Yu, H. Peng, Y. Fu, X. Yan, C. Du, H. Chen, Enhanced nitrogen removal of low C/N wastewater in constructed wetlands with co-immobilizing solid carbon source and denitrifying bacteria, *Bioresour. Technol.*, 280 (2019) 337–344.
- [24] D. Cai, P. Li, C. Chen, Y. Wang, S. Hu, C. Cui, P. Qin, T. Tan, Effect of chemical pretreatments on corn stalk bagasse as immobilizing carrier of *Clostridium acetobutylicum* in the performance of a fermentation-pervaporation coupled system, *Bioresour. Technol.*, 220 (2016) 68–75.
- [25] D.M. Donnelly, L.C. de Resende, D.E. Cook, R.H. Atalla, D.K. Combs, Technical note: a comparison of alkali treatment methods to improve neutral detergent fiber digestibility of corn stover, *J. Dairy Sci.*, 101 (2018) 9058–9064.
- [26] H. Liu, B. Pang, Y. Zhao, J. Lu, Y. Han, H. Wang, Comparative study of two different alkali-mechanical pretreatments of corn stover for bioethanol production, *Fuel*, 221 (2018) 21–27.
- [27] APHA, Standard Methods for the Examination of Water and Wastewater, 22nd ed., Washington, DC, 2012.
- [28] P.D. Hiley, The reality of sewage treatment using wetlands, *Water Sci. Technol.*, 32 (1995) 329–338.
- [29] L. Du, Q. Chen, P. Liu, X. Zhang, H. Wang, Q. Zhou, D. Xu, Z. Wu, Phosphorus removal performance and biological dephosphorization process in treating reclaimed water by integrated vertical-flow constructed wetlands (IVCWs), *Bioresour. Technol.*, 243 (2017) 204–211.
- [30] D. Chen, X. Gu, W. Zhu, S. He, J. Huang, W. Zhou, Electrons transfer determined greenhouse gas emissions in enhanced nitrogen-removal constructed wetlands with different carbon sources and carbon-to-nitrogen ratios, *Bioresour. Technol.*, 285 (2019) 121313.
- [31] P.J. Strong, B. McDonald, D.J. Gapes, Enhancing denitrification using a carbon supplement generated from the wet oxidation of waste activated sludge, *Bioresour. Technol.*, 102 (2011) 5533–5540.
- [32] S. Lu, Z. Wan, F. Li, X. Zhang, Ammonia nitrogen adsorption and desorption characteristics of twenty-nine kinds of constructed wetland substrates, *Res. Environ. Sci.*, 29 (2016) 1187–1194 (in Chinese).
- [33] H. Wu, J. Zhang, P. Li, J. Zhang, H. Xie, B. Zhang, Nutrient removal in constructed microcosm wetlands for treating polluted river water in northern China, *Ecol. Eng.*, 37 (2011) 560–568.
- [34] Z. Si, X. Song, Y. Wang, X. Cao, Y. Zhao, B. Wang, Y. Chen, A. Arefe, Intensified heterotrophic denitrification in constructed wetlands using four solid carbon sources: denitrification efficiency and bacterial community structure, *Bioresour. Technol.*, 267 (2018) 416–425.
- [35] X. Wang, W. Wang, Y. Zhang, Z. Sun, J. Zhang, G. Chen, J. Li, Simultaneous nitrification and denitrification by a novel isolated *Pseudomonas* sp. JQ-H3 using polycaprolactone as carbon source, *Bioresour. Technol.*, 288 (2019) 121506.
- [36] D. Liu, J. Li, C. Li, Y. Deng, Z. Zhang, Z. Ye, S. Zhu, Poly (butylene succinate)/bamboo powder blends as solid-phase carbon source and biofilm carrier for denitrifying biofilters treating wastewater from recirculating aquaculture system, *Sci. Rep.*, 8 (2018) 3289.
- [37] L. Jia, R. Wang, L. Feng, X. Zhou, J. Lv, H. Wu, Intensified nitrogen removal in intermittently-aerated vertical flow constructed wetlands with agricultural biomass: effect of influent C/N ratios, *Chem. Eng. J.*, 345 (2018) 22–30.
- [38] M. Zheng, X. Li, L. Li, X. Yang, Y. He, Enhancing anaerobic biogasification of corn stover through wet state NaOH pretreatment, *Bioresour. Technol.*, 100 (2009) 5140–5145.
- [39] Z. Yao, C. Wang, N. Song, H. Jiang, Development of a hybrid biofilm reactor for nitrate removal from surface water with macrophyte residues as carbon substrate, *Ecol. Eng.*, 128 (2019) 1–8.
- [40] C. Zhang, Q. Yin, Y. Wen, W. Guo, C. Liu, Q. Zhou, Enhanced nitrate removal in self-supplying carbon source constructed wetlands treating secondary effluent: the roles of plants and plant fermentation broth, *Ecol. Eng.*, 91 (2016) 310–316.
- [41] Y. Gao, Y.W. Xie, Q. Zhang, A.L. Wang, Y.X. Yu, L.Y. Yang, Intensified nitrate and phosphorus removal in an electrolysis-integrated horizontal subsurface-flow constructed wetland, *Water Res.*, 108 (2017) 39–45.
- [42] Y. Yang, J. Liu, N. Zhang, H. Xie, J. Zhang, Z. Hu, Q. Wang, Influence of application of manganese ore in constructed wetlands on the mechanisms and improvement of nitrogen and phosphorus removal, *Ecotoxicol. Environ. Saf.*, 170 (2019) 446–452.
- [43] A.O. Babatunde, Y. Zhao, A.M. Burke, M.A. Morris, J.P. Hanrahan, Characterization of aluminum-based water treatment residual for potential phosphorus removal in engineered wetlands, *Environ. Pollut.*, 157 (2009) 2830–2836.
- [44] R. Xiong, X. Yu, L. Yu, Z. Peng, L. Cheng, T. Li, P. Fan, Biological denitrification using polycaprolactone-peanut shell as slow-release carbon source treating drainage of municipal WWTP, *Chemosphere*, 235 (2019) 434–439.
- [45] G. Hua, Y. Zeng, Z. Zhao, K. Cheng, G. Chen, Applying a resting operation to alleviate bioclogging in vertical flow constructed wetlands: an experimental lab evaluation, *J. Environ. Manage.*, 136 (2014) 47–53.
- [46] H. Li, Y. Cao, X. Wang, X. Ge, B. Li, C. Jin, Evaluation on the production of food crop straw in China from 2006 to 2014, *Bioenergy Res.*, 10 (2017) 949–957.

- [47] A. Boley, W.-R. Müller, G. Haider, Biodegradable polymers as solid substrate and biofilm carrier for denitrification in recirculated aquaculture systems, *Aquacult. Eng.*, 22 (2000) 75–85.
- [48] R. Messalem, A. Brenner, Y. Leroux, I. Soares, A. Abeliovich, M. Soares, Denitrification of groundwater: pilot-plant testing of cotton-packed bioreactor and post-microfiltration, *Water Sci. Technol.*, 42 (2000) 353–359.
- [49] P. Li, J. Zuo, Y. Wang, J. Zhao, L. Tang, Z. Li, Tertiary nitrogen removal for municipal wastewater using a solid-phase denitrifying biofilter with polycaprolactone as the carbon source and filtration medium, *Water Res.*, 93 (2016) 74–83.
- [50] S. Zhu, Y. Deng, Y. Ruan, X. Guo, M. Shi, J. Shen, Biological denitrification using poly (butylene succinate) as carbon source and biofilm carrier for recirculating aquaculture system effluent treatment, *Bioresour. Technol.*, 192 (2015) 603–610.