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Domestic wastewater treatment with a novel process of chemically enhanced primary treatment using fly ash-based coagulants and constructed wetland

Kai Hu[a,b,*], Qing-liang Zhao[c,d], Wei Chen[a,b], Feng Tang[d]

[a]*Key Laboratory of Integrated Regulation and Resource Development on Shallow Lakes, Ministry of Education, Hohai University, Nanjing 210098, China, Tel. +86 25 83786707; email: hukaihit@hhu.edu.cn (K. Hu)*
[b]*College of Environment, Hohai University, Nanjing 210098, China*
[c]*State Key Laboratory of Urban Water Resource and Environment, Harbin Institute of Technology, Harbin 150090, China, Tel. +86 451 86283017; email: zhql1962@163.com (Q.-l. Zhao)*
[d]*School of Municipal and Environmental Engineering, Harbin Institute of Technology, Harbin 210098, China, Tel. +86 451 86283017; email: 739178045@qq.com (F. Tang)*

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ABSTRACT

A novel integrated system of chemically enhanced primary treatment and constructed wetland was studied, aiming to provide an efficient domestic wastewater treatment method. In the coagulation pre-treatment, a composite coagulant was made from fly ash. This coagulant efficiently removes COD, suspended solids (SS) and total phosphorus (TP) from the wastewater. The removal efficacy depends on the dose of coagulant and pH of wastewater. At a dosage of 1 mL/L and pH of 6–7, the removal efficiencies of COD, SS and TP of domestic wastewater reached 64%, 93% and 91%, respectively. The operation of simulated subsurface-flow constructed wetland under different hydraulic loadings (0.03–0.10 $m^3/(m^2\ d)$, corresponded to 5.61–18.7 $g/(m^2\ d)$), which received effluents from the previous coagulation treatment, showed that COD might be further reduced 64%–77% with effluent COD values lower than 60 mg/L. However, the ammonium nitrogen was less effectively removed by limited oxygen transfer as a result of consumption through organic matter degradation. TP removal was compensated by the phosphorus release within the bed. Compared with single-stage, two-stage combinations of the subsurface horizontal flow beds are an alternative approach to improve the removal of ammonium nitrogen.

Keywords: Chemically enhanced primary treatment; Constructed wetland; Fly ash; Domestic wastewater; Coagulant

1. Introduction

Chemically enhanced primary treatment (CEPT) is a relatively simple and efficient wastewater treatment process [1]. As it offers satisfactory and stable performance, it is highly recommended for wastewater purification [2], especially suitable for the present situation in China. This method could be easily applied in most treatment plants. However, this method unavoidably presents certain drawbacks such as excessive sludge production and in certain cases, the expensive chemical coagulants [3,4]. On the other hand, constructed wetlands (CWs) are energy-saving and environmentally friendly polishing and treatment systems [5], especially for the removal of residual BOD and COD, sometimes for ammonium nitrogen (NH_4^+–N) and total phosphorus (TP), to likely discharge consent levels. The CWs possess such advantages as relatively low capital costs, minimal running costs and considerable flexibility to operation [6–8], yet the major encountered problems include declined removal efficiencies under low temperature and substrate clogging caused by high concentration of suspended solids (SS) in influent [9,10].

The objectives of this lab-scale study were to investigate into the combined processes of CEPT and CW as an efficient domestic wastewater treatment method. CEPT

* Corresponding author.

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was a physicochemical pretreatment in removing most SS, and TP and partial COD from raw wastewater, in order to reduce the influent organic loading of CW and possibility of clogging triggered by influent SS. One aim of the study was to examine the performance of the proposed method under various loadings; the other was to realize cost reduction of coagulants and integration of reutilization of fly ash (FA) with wastewater treatment, which is a widely generated by-product of coal-fired power plants. However, no investigation has been reported on the combined technology of CEPT and CW for domestic wastewater treatment. This study might present another effective way for FA reuse and propose an alternative process for domestic wastewater treatment.

2. Materials and methods

2.1. Characterization of FA

Three raw FA samples were collected from Harbin Third Power Plant in Northeast China. The size distribution indicated that FA particles were mainly in the range of 15–75 μm in diameter. The value of loss of ignition of FA was as low as 2.79–4.50 wt%.

The major elements in FA were determined by using a Axios pw4400 X-ray fluorescence spectrograph. The result is presented in Table 1. It was evident that the FA was rich in aluminum and silicon; the Al_2O_3 and SiO_2 contents were higher than 62 wt%; the Al_2O_3 and Fe_2O_3 contents were higher than 24.2 wt%. These compounds are identified as essential raw materials for the production of coagulants [11,12]. The contents of other trace metals (not listed in Table 1) of Bi, Cu, Cr, As and Cd were 0.01 wt%, 0.009 wt%, 0.006 wt%, 0.005 wt% and <0.001 wt%, respectively. According to the chemical compositions, the ash sample categorized into class F as defined in the standard C 618 by ASTM (American Society for Testing and Materials). The X-ray diffraction result of raw FA indicated that the main crystalline phases were quartz (SiO_2), mullite ($Al_6Si_2O_{13}$) and hematite (Fe_2O_3). The diffraction peak profiles in the range of 22°–35° ($2\theta_{max}CuK_{\alpha}$) suggested the presence of glass phases, which were unfavorable to acid leaching.

Table 1
Chemical composition of fly ash[a]

Component	wt (%)
SiO_2	41.1–55.7 (50.8[b])
Al_2O_3	20.9–25.9 (24.5)
CaO	3.7–5.7 (4.8)
Fe_2O_3	3.3–3.7 (3.6)
K_2O	1.5–2.4 (2.0)
TiO_2	0.73–0.76 (0.75)
MgO	0.2–0.6 (0.4)
P_2O_5	0.24–0.26 (0.25)
SO_3	0.20–0.50 (0.37)
Na_2O	0.20–0.99 (0.64)
MnO	0.076–0.16 (0.12)
BaO	0.062–0.12 (0.10)
SrO	0.058–0.12 (0.091)
ZrO_2	0.013–0.058 (0.038)
ZnO	0.012–0.035 (0.026)
PbO	0.004–0.015 (0.010)
Rb_2O	0.002–0.014 (0.008)
Nb_2O_5	0.0014–0.009 (0.006)
Y_2O_3	0.0013–0.006 (0.004)

[a]Element concentrations of C, N, B, He, Li and Be were not able to be detected using XRF method.
[b]Value in the brackets represented the average value of three raw FA samples.

2.2. Preparation of fly ash-based coagulant

The silicon and aluminium components of FA exist as the amorphous structures which resist to acid leaching under the prevailing reaction conditions. In order to facilitate amenable conditions for leaching by sulfuric acid, a preliminary treatment of FA sintered with Na_2CO_3 was conducted. The steps of the production of a FA-based coagulant were as follows [13]: FA sample (1 g) was blended with Na_2CO_3 (0.06 g) and then sintered in a furnace at 805°C for 1 h. The sintered product was cooled to the ambient temperature and afterwards milled. Sulfuric acid with 4 mol/L H^+ was chosen to leach the FA at L/S ratio (defined as the ratio of volume of leaching solution to mass of FA) of 3 mL/g under the condition of heating at boiling point and refluxing. The leaching process was conducted for 0.5 h. The slurry was then allowed to cool off for another 0.5 h to prepare the FA-based coagulant. The FA-based coagulant was composed of leached solution and ash residues. After filtration, the volume of ash residues could be determined while the concentrations of Al^{3+} and Fe^{3+} in the solution were measured. The FA-based coagulant demonstrated the following properties: concentration of Al^{3+} of 0.213 mol/L, concentration of Fe^{3+} of 0.042 mol/L; 1 mL such coagulant contained solution of 0.83 mL and FA residue of 0.17 mL.

2.3. Jar tests

Coagulation experiments were carried out by jar testing on a six paddle gang stirrer (C6F, VELP® Scientifica). The 250 mL test water was stirred at first for the sake of minimizing the heterogeneity of the water sample. Then the required dosage of the FA-based coagulant was injected into the test water, followed by a rapid stirring at 200 rpm for 1 min and a slow stirring at 60 rpm for 10 min. After the completion of coagulation, the water sample was allowed to settle for 30 min. Finally, a supernatant sample was withdrawn for measurement. The temperature at which the jar tests were performed was 18°C–21°C. For experiments on the effect of pH on the coagulation, the pH value of the test water was adjusted using 2 mol/L H_2SO_4 solution or 2 mol/L NaOH solution.

Domestic wastewater was obtained from the sewer pipelines in Harbin, China. The characteristics of raw wastewater can be seen in Table 2.

2.4. Operation of CW treatment systems

The experimental subsurface horizontal flow beds were functioned as secondary treatment, which were operated either in parallel as single stage or in cascade as two stages. The wetland systems were installed indoors (sheltered from rain and ambient temperature fluctuated between 18°C and 25°C) in Harbin Institute of Technology in the city of Harbin, China. Each bed was 1,200 mm long and 400 mm wide with an effective surface area of approximately 0.4 m^2. It was filled with three successive layers of granular materials: an upper filtration layer (50 mm with Ø 12–20 mm gravels); an intermediate layer (200 mm with Ø 20–30 mm gravels) and a lower drainage layer (150 mm with Ø 30–50 mm pebbles), which exhibited high hydraulic conductivity and was easy for maintenance [14]. On top of the small gravel, 200 mm soil was placed (Fig. 1(a)). The inlet zone was riprapped with gravels (Ø 30–50 mm rocks) so as to evenly distribute influent flow. Besides, the outlet zone was an additional riprap zone, same as the inlet, which prevented weed growth and resuspension of bed substrates [15]. The drainage pipes were set 100, 300, 500 mm, respectively, below the substrate surface.

Table 2
Characterization of domestic wastewater

COD (mg/L)	349–641
SS (mg/L)	102–162
TP (mg/L)	4.16–6.14
NH_4^+–N (mg/L)	30–59
TN (mg/L)	42–72
pH	7–8
T (°C)	9–19

TN, total nitrogen.

The vegetation beds were automatically fed at a determined loading rate with pre-coagulated domestic wastewater per day and operated with batch feeding scheme (1 feeding/d). Effluent samples were taken from the bottom drainage pipe (sampling port) located 500 mm below the substrate surface on a daily basis and monitored by analyzing various parameters to assess CW performance. Two beds were planted with *Acorus calamus* (denoted as CW-A) and *Canna indica* (denoted as CW-B), respectively. The influence of the hydraulic loading on the pollutant elimination performance was investigated on each CW operated in parallel. Detailed operation parameters were given in Table 3.

Then, the two wetlands were operated as two successive stages (Fig. 1(b)), in the interest of intensifying the N reduction. Operation conditions for the cascade running stages were as follows: the *Acorus calamus* wetland was continuously fed with pre-coagulated domestic wastewater at a flow rate of 27.8 mL/min (40 L/d). The effluent was then introduced to the *Canna indica* wetland. The hydraulic loading of each CW was 0.05 $m^3/(m^2\ d)$. The influent quality of *Acorus calamus* wetland is listed in Table 4.

2.5. Analytical methods

The concentration of Fe^{3+} in the solution was measured using the 1,10-phenanthroline colorimetric method [16]. The concentration of Al^{3+} in the solution was analyzed using the EDTA titration method with NaF as the masking agent [17]. Determination of the metal ions in the solution was performed in duplicate. Therefore, each reported result represented an average of two determinations.

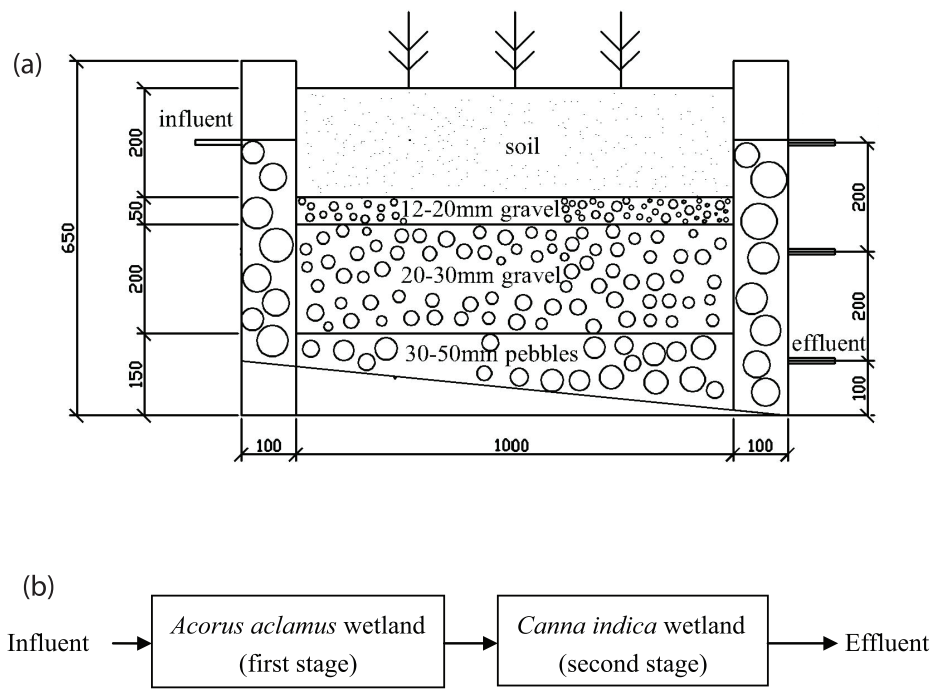

Fig. 1. (a) Configuration of CW (the numbers were counted by unit of mm) and (b) schematic diagram of two-stage wetlands.

Table 3
Operation parameters of CWs

Parameter	*Acorus calamus* wetland			*Canna indica* wetland	
Duration (d)	44	25	26	44	25
Treatment capacity (L/d)	12	20	40	12	20
Hydraulic loading ($m^3/(m^2·d)$)	0.03	0.05	0.1	0.03	0.05
Organic loading rate ($g/(m^2 d)$)	5.49	9.15	18.3	5.49	9.15
Feed/drainage time (h)	4.8	12	24	4.8	12
HRT (d)	6.52	3.91	1.95	6.73	4.04

HRT, hydraulic retention time.

Table 4
Influent quality of *Acorus calamus* wetland (raw wastewater after coagulation)

Parameter	Range	Average
T (°C)	15.9–18.9	17.2
pH	6.67–7.50	6.97
COD (mg/L)	112–227	187
TP (mg/L)	0.096–0.470	0.303
NH_4^+–N (mg/L)	31.60–49.32	43.12
TN (mg/L)	36.30–57.60	55.10

The COD, SS, NH_4^+–N, TN, TP and orthophosphate of the test water were determined using an APHA (the American Public Health Association) standard analysis method [16]. The pH was monitored by a pHS-3C digital pH meter (Wei-ye, Shanghai).

3. Results and discussion

3.1. Coagulation performance of FA-based coagulant

3.1.1. Effect of pH

The impact of coagulation pH on the performance has been studied and the results are presented in Fig. 2. For this experiment, the ranges of pH, and coagulant dosage were from pH 4.48 to pH 10.53, and 0.169 mmol/L as Al + Fe, respectively.

At lower pH side (4.4–5.5), the addition of FA-based coagulant resulted in the pH of the solution dropping to an even lower range, that is, pH 3.0–3.8; therefore, no flocs were found at these conditions. A remarkable enhancement of pollutant removal was gained when pH of wastewater rose from 5.5 to 6.0. Meanwhile, the coagulation pH after adding FA-based coagulant rose from 3.8 to 4.7.

When FA-based coagulant was used at wastewater pH of 6.5, about 97% and 88% of SS and TP, respectively, could be removed. Effluent SS and TP were 4.0 and 0.688 mg/L, respectively. At higher pH side (>6.5), the SS and TP removal efficiencies decreased with increasing wastewater pH. However, the differences in COD removal efficiency at various pH values were slight. It can be seen that the optimum coagulation pH range is 4.7–7.5, corresponding to pH range of wastewater from 6.0 to 8.5.

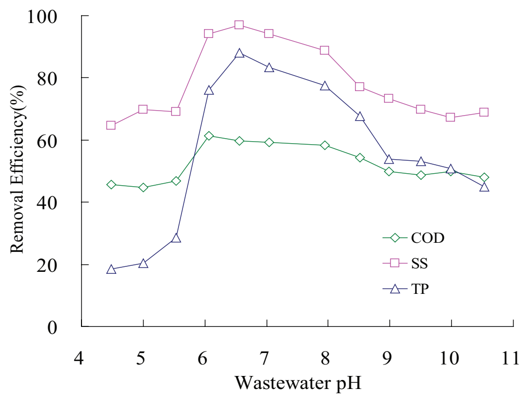

Fig. 2. Effect of coagulation pH on domestic wastewater treatment with fly ash-based coagulant.

3.1.2. Effect of dosage

Fig. 3 displays the coagulation performance by FA-based coagulant as a function of coagulant dosage at coagulation pH of 5.7–6.1.

The results revealed that increased dosage can also result in the enhancement of pollutant reduction; however, when the dosage was higher than 0.8 mL/L, the removal efficiency became stable. Effluent COD, SS and TP were 219, 12 and 0.556 mg/L, respectively, at an optimal dosage of 1.0 mL/L. The effluent concentrations of SS and TP met the class I B criteria of Chinese discharge standard for WWTPs [18] whereas it had been exceeded for COD. This requires further biological treatment (such as CW method) for COD reduction.

3.1.3. Comparison with other coagulants

One objective of this study was to assess the prepared FA-based coagulant in comparison with other commercial types (i.e., $Al_2(SO_4)_3·18H_2O$, $Fe_2(SO_4)_3·XH_2O$, PAC [polyaluminum chloride] and PFS [polyferric sulphate]). The coagulant dose was 0.211 mmol/L as Al and Fe for FA-based coagulant, or as Al alone, or Fe alone for other coagulants.

It was noteworthy that FA-based coagulant performed better than commercial coagulants; approximately 22%–43% greater removal of COD, TP and SS were achieved by FA-based coagulant (Fig. 4). This could be explained by the solubility of various chemical species associated with FA including Fe^{3+}, Al^{3+}, Ca^{2+}, Fe^{2+}, Mg^{2+} and H_2SiO_3; the adsorption capacity of chemical-treated FA and the quickly developed and large flocs with the accompany of FA residuals [13]. As a result of ash residuals contained in the coagulant, at a dosage of 1 mL/L, the volume of produced sludge was about 10% less than the volume of PAC-generated sludge. Meanwhile, the produced sludge contained about 4.8 g ash residuals per 1 L wastewater. According to the preparation procedures of FA-based coagulant described in section 2.2, a cost and energy analysis was performed regarding the production. Results showed that the cost of producing FA-based coagulant was estimated 18.5 USD/m^3 in China, while the price of industrial PAC coagulant equal to ca. 280 USD/kg in the Chinese market.

3.2. Post-treatment performance of CW systems operated in parallel

The wetland systems were designed for polishing of effluents from the coagulation pre-treatment. The combined treatment method was carried out by the pre-coagulation of wastewater at a FA-based coagulant dosage of 1 mL/L, followed by the application of CW treatment. Within the above coagulant dosage, the COD values were reduced from the range of 398–600 mg/L for raw wastewater to the range of 112–227 mg/L.

Fig. 5 shows the effluent concentrations of COD, NH_4^+–N and TP for two parallel beds at various hydraulic loadings. As shown in Fig. 5(a), the two CWs planted with different sorts of vegetation demonstrated similar excellent treatment efficiency of organic matters (i.e., COD removal rates of 64%–77%). The effluent COD concentrations from wetlands varied slightly under various hydraulic loadings (0.03–0.10 m^3/(m^2 d)). During the operating period of

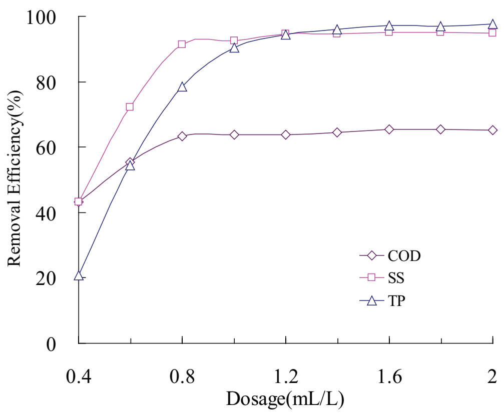

Fig. 3. Effect of dosage on domestic wastewater treatment with fly ash-based coagulant.

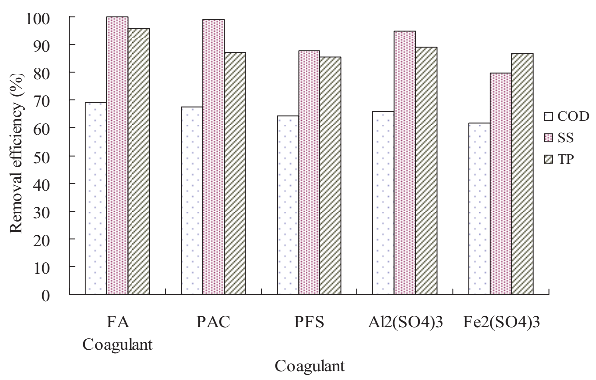

Fig. 4. Domestic wastewater treatment performance of fly ash-based coagulant compared with other coagulants.

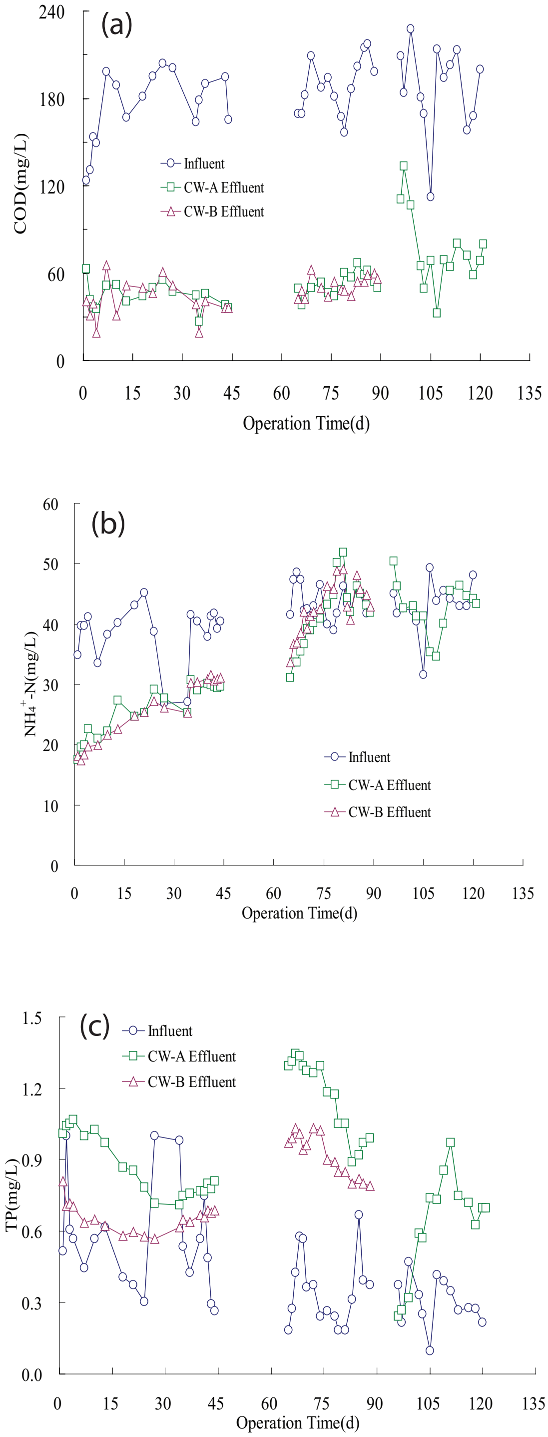

Fig. 5. Influent and effluent (a) COD, (b) NH_4^+–N and (c) TP variations of CWs vs. operation time.

hydraulic loadings of 0.03 and 0.05 $m^3/(m^2 d)$, an effluent with residual COD concentration below 60 mg/L was readily achieved, which met the class I B criteria of the Chinese discharge standard for WWTPs [18]. As hydraulic loading increased to 0.1 $m^3/(m^2 d)$, at the initial stage, the effluent COD increased sharply as a result of increasing hydraulic loading. Then it gradually decreased with time. After 1 week or so, the effluent COD became constant and was around 64 mg/L. It could be deduced that when influent COD loading was increased by 220%, the effluent COD only increased by 45.45%, which suggested that the wetland system resisted to shock loading.

Effluent NH_4^+–N of both CWs increased continuously with time (Fig. 5(b)). Particularly during the periods at hydraulic loadings of 0.05 and 0.1 $m^3/(m^2 d)$, effluent NH_4^+–N might be as high as that of influent (average value of 44.3 mg/L). The unfavorable performance concerning the NH_4^+–N removal was possibly ascribed to the ammonification of influent organic N and limited oxygen transfer within the bed. Removal of N compounds in CW is governed mainly by microbial nitrification and denitrification. Nitrification occurs when dissolved oxygen (DO) concentration satisfies certain level, that is, in order to obtain the nitrification of NH_4^+–N of 1.0 mg/L, DO concentration in the wetland should be no less than 4.6 mg/L. However, influent DO concentration of the wetland was less than 2 mg/L, which was consumed by organic matter degradation in majority. Consequently, the CW beds were in the anaerobic state and unable to provide sufficient oxygen for nitrification. In this situation, nitrification was considered as a rate controlling step for degradation of N compounds. In comparison with the *Acorus calamus* wetland, the *Canna indica* wetland showed a little better removal efficiency of NH_4^+–N. But the subtle difference between the two wetlands regarding NH_4^+–N removal was found with increment of hydraulic loading.

Fig. 5(c) depicts the TP removal as a function of hydraulic loading and vegetation. As the hydraulic loading increased, at the initial period (first 8–10 d), an elevated TP concentration in the wetland effluent was observed. Then the TP concentration gradually decreased with time. When operating at hydraulic loading of 0.1 $m^3/(m^2 d)$ for 21 d, the effluent TP concentration of CW-A was maintained at around 0.7 mg/L. Plants growth condition within the CW also had some effect on phosphorus elimination. With a better growing condition, the CW-B exhibited a stronger capacity than CW-A in removing TP under various hydraulic loadings.

Although influent TP concentration was low (0.303–0.554 mg/L) due to the excellent pre-treatment performance of CEPT, the effluent TP concentration from CW gradually increased, sometimes even higher (1.0 mg/L) than that of influent. It is likely the phenomenon of phosphorus release [19], which had been reported by previous researchers [20–22] that matrix/substrate adsorption of phosphorus in the wetland was reversible. In CWs, substrates have a crucial function in the phosphorus removal through adsorption and precipitation. When influent phosphorus was insufficient, the microorganisms in CW utilized the accumulated phosphorus absorbed on the matrix. To some extent, the wetland matrix was termed as a "phosphorus buffer", which adjusted phosphorus concentration in the wetland. Further investigation into mechanisms of nitrogen and phosphorus release within the CWs is still in progress.

3.3. Post-treatment performance of CW systems operated in cascade

Results showed that the effluent NH_4^+–N increased with an increase in hydraulic loading for subsurface-flow CW. At the end of the periods of hydraulic loading of 0.05 $m^3/(m^2 d)$, the effluent NH_4^+–N concentrations from CW-A and CW-B were 43.17 and 44.78 mg/L, respectively, which were almost the same as the influent NH_4^+–N concentration.

Generally, at the front of CW, the DO concentration was relatively low as a result of the degradation of organic matter. However, the concentration of organic matter decreased with the flow path. In addition, due to the oxygenation of the whole bed through oxygen release from roots and reaeration, at the back of CW, a relatively high DO concentration was detected [23,24]. As the NH_4^+–N removal is highly dependent on the DO concentration, for the purpose of improving the NH_4^+–N reduction, two CWs operated in cascade to increase the flow path were investigated through DO optimization. In the previous study, the growth status of *Canna indica* wetland was superior to that of *Acorus calamus* wetland, indicating a better oxygen loss generated by wetland plants. So the *Canna indica* wetland was inserted after the *Acorus calamus* wetland (Fig. 1(b)). The performance of each CW is shown in Figs. 6–8.

3.3.1. COD removal

The influent and effluent COD of each CW is presented in Fig. 6.

The difference between effluent COD from the first stage and from the second stage of CWs was about 20 mg/L. The COD removal rates were 65.6% and 32.1%, respectively, for the first and second stage of CWs. Effluent COD from the second CW stage satisfied the Class I A level of the Chinese discharge standard for WWTPs [18], which is the most stringent level of discharge standard concerning COD.

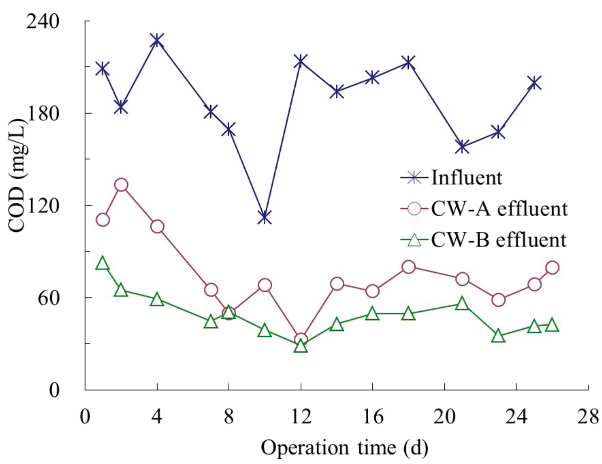

Fig. 6. Influent and effluent COD variation with time.

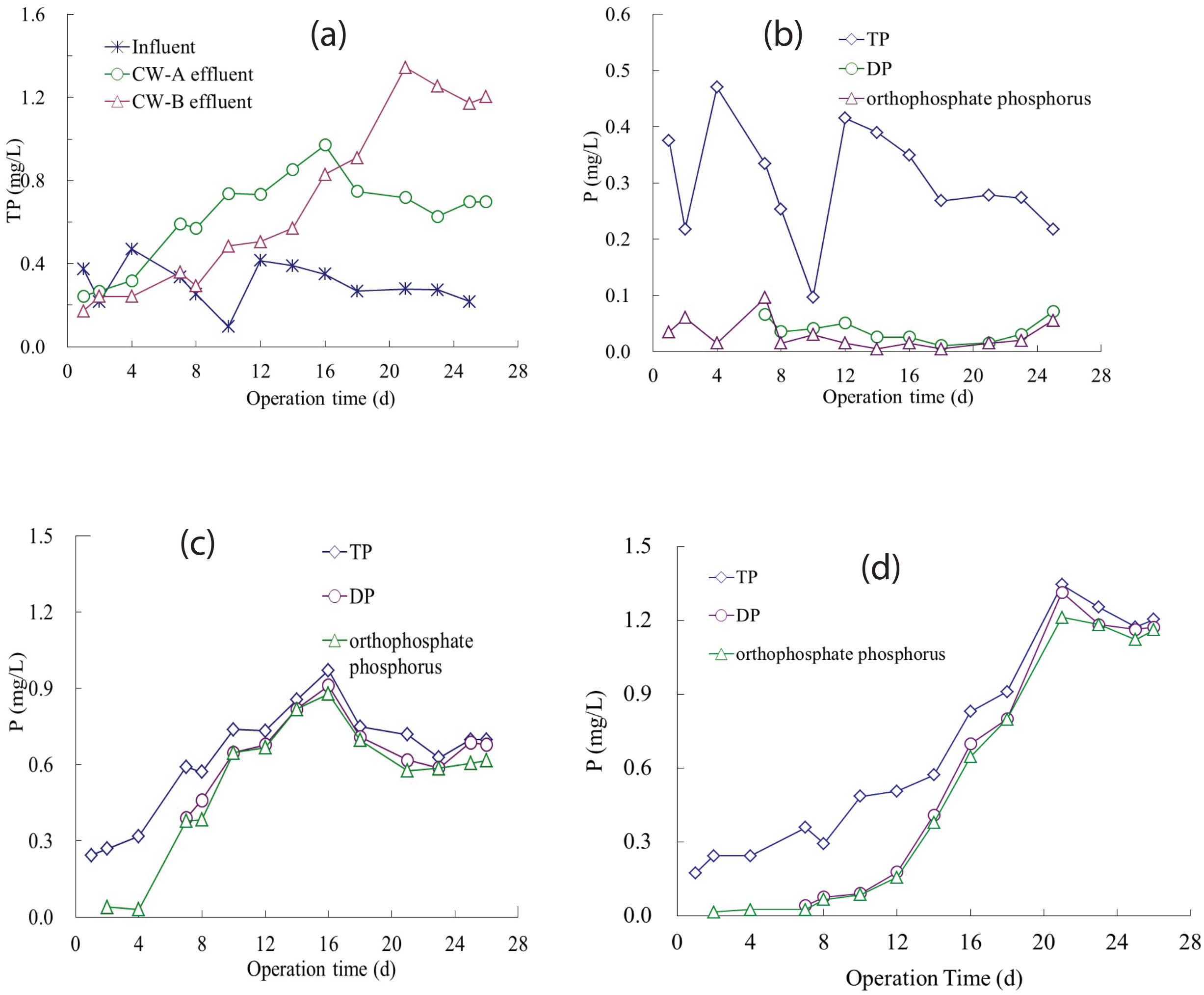

Fig. 7. Influent and effluent TP, DP and orthophosphate variations with time. (a) Influent and effluent TP, (b) TP, DP, and orthophosphate of influent, (c) TP, DP, and orthophosphate of CW-A (first stage) effluent, (d) TP, DP, and orthophosphate of CW-B (second stage) effluent.

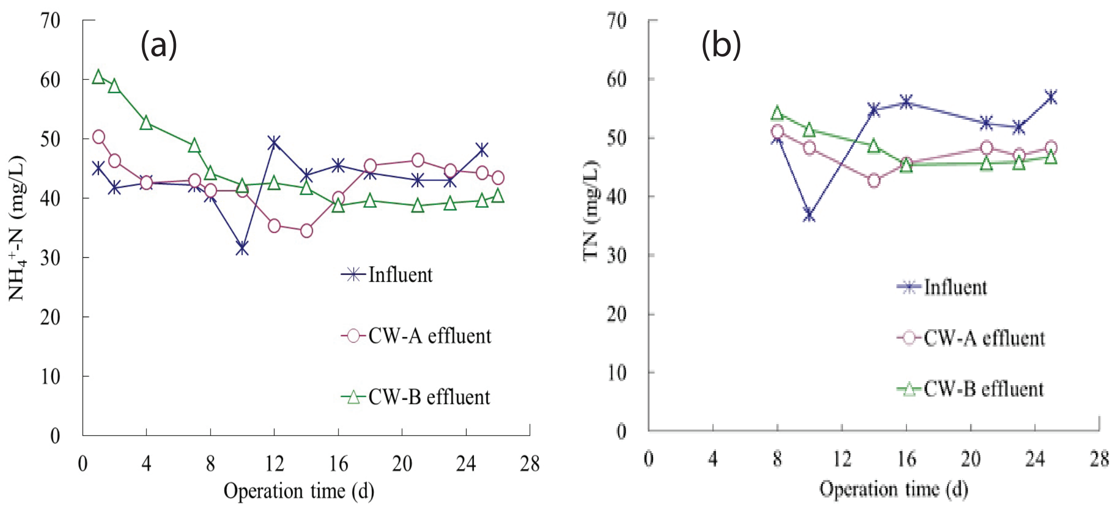

Fig. 8. Influent and effluent (a) NH_4^+–N and (b) TN variations with time (CW-A: first stage; CW-B: second stage).

3.3.2. Phosphorus removal

During the cascade operation, the variations of TP, dissolved P (DP) and orthophosphate of each CW stage were studied (Fig. 7).

A release of phosphorus was observed in both CWs (Fig. 7(a)). The effluent TP concentration of the first CW stage increased first and then decreased with time (Fig. 7(c)), whereas this concentration of the second CW stage increased (Fig. 7(d)). At day 17, the effluent TP concentration from the second CW stage was higher than that from the first CW stage (Fig. 7(a)).

The TP, DP and orthophosphate concentrations in the influent of *Acorus calamus* wetland (CW-A) were around 0.303, 0.037 and 0.030 mg/L, respectively (Fig. 7(b)). It can be seen that TP contained 12.2% DP and orthophosphate consisted of 81.1% DP (Fig. 7(b)). In contrast, in the effluent of *Canna indica* wetland (CW-B), the proportion of DP to TP increased with time (Fig. 7(d)). In the end of the operation, the proportion rose to above 97%. Still, DP consisted predominantly of orthophosphate (>91%). Moreover, orthophosphate removal in CWs is generally limited as a result of the low sorption capacity of substrate materials. Over time, the release of reactive phosphorus derived from the part of phosphorus adsorbed on the wetland matrix occurs [25].

3.3.3. Nitrogen removal

The variations of NH_4^+–N and TN of each CW stage are presented in Fig. 8.

As shown in Fig. 8, the effluent NH_4^+–N and TN from each CW demonstrated a declining trend with time. The variations regarding NH_4^+–N and TN concentrations were more obvious in the *Canna indica* wetland (CW-B). When the beds in cascade were operated at hydraulic loading of 0.05 $m^3/(m^2$ d), which corresponded to NH_4^+–N loading of 2.156 $g/(m^2$ d) and HRT of 3.97 d, effluent NH_4^+–N and TN were 40.46 and 46.80 mg/L, corresponding to removal rates of 11.97% and 15.44%, respectively. The remove rates of NH_4^+–N and TN contributed by the first CW stage were 1.60% and 12.87%, respectively. This result implied that CWs operated in cascade is an effective method to improve the N reduction, which is consistent with the finding [26].

4. Conclusions

A raw FA sample collected for this research was successfully used for the production of a composite coagulant by leaching with sulfuric acid. Experiments on CEPT of domestic wastewater using FA-based coagulant showed high removal efficiencies of pollutants. The polishing of effluent from coagulation pre-treatment adopting CW was then investigated to testify the reliability of this combined process. The operation of CW system under various hydraulic loadings (0.03–0.10 $m^3/(m^2$ d)) showed that COD might be reduced 64%–77% with the final effluent COD less than 60 mg/L. When influent COD loading was increased by 220%, the effluent COD only increased by 45.45%, which suggested that the wetland system resisted to shock loading. It was also found that the NH_4^+–N and TP were not effectively removed in the wetland system. Nevertheless, the CWs could be cascade joined to enhance nitrogen removal.

Acknowledgments

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