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Effects of plant species on CH_4 emission from integrated vertical subsurface flow constructed wetlands

Xin Cao[a], Wei Huang[a,b], Deying Huang[c], Yunfei Tian[a], Xuyao Jiang[a], Jibiao Zhang[a,*]

[a]*Department of Environmental Science and Engineering, Fudan University, Shanghai 200433, China, Tel. +86-21-65642192; emails: jbzhang@fudan.edu.cn (J. Zhang), Tel. +86-21-65642948; emails: cxin615@163.com (X. Cao), 14210740047@fudan.edu.cn (Y. Tian), 15210740042@fudan.edu.cn (X. Jiang), Tel. +86-10-84913896; email: yixinghd6@163.com (W. Huang)*
[b]*State Key Laboratory of Environmental Criteria and Risk Assessment, Chinese Research Academy of Environmental Sciences, Beijing 100012, China*
[c]*Department of Chemistry, Fudan University, Shanghai 200433, China, email: huangdeying@fudan.edu.cn (D. Huang)*

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ABSTRACT

Methane (CH_4) emission from constructed wetlands (CWs) has increased the amount of greenhouse gases (GHG) and raised environmental concerns. The plant species could significantly affect the CH_4 emission in integrated vertical subsurface flow constructed wetlands (IVSSF CWs). This study evaluated the removal efficiency of water pollutants and determined the influence of plant species on CH_4 fluxes in IVSSF CWs planted with *Cyperus alternifolius*, *Canna indica*, *Acorus calamus*, and *Scirpus tabernaemontani*. Result indicated that the pollutant removal efficiency in IVSSF CWs planted with *C. alternifolius* was apparently higher than that in the wetlands planted with the three other plants. The mean removal efficiencies in CWs planted with *C. alternifolius* were 84.46% for chemical oxygen demand (COD), 85.80% for NH_4-N, 82.94% for total nitrogen, and 94.87% for total phosphorous. The average CH_4 fluxes were 5.45 mg m^{-2} h^{-1} (*A. calamus*), 2.49 mg m^{-2} h^{-1} (*S. tabernaemontani*), 9.26 mg m^{-2} h^{-1} (*C. indica*), and 3.25 mg m^{-2} h^{-1} (*C. alternifolius*). The CH_4 fluxes in IVSSF CWs planted with *C. alternifolius*, *C. indica*, and *S. tabernaemontani* were significantly correlated with temperature ($P < 0.05$). Moreover, the CH_4 fluxes in IVSSF CWs planted with *C. indica* and *A. calamus* significantly differed between up-flow and down-flow chambers ($P < 0.05$). The relationship between CH_4 flux and removal loading of COD was also analyzed. *S. tabernaemontani* and *A. calamus* were found to be the optimal plants for COD removal in IVSSF CWs in summer and winter, respectively. The results can be helpful for plants optimization in CWs and provide the data support for the control of GHG emission.

Keywords: Constructed wetland; Water purification; Greenhouse gas flux; Plants optimization

1. Introduction

Constructed wetlands (CWs) are used to remove pollutants from wastewater and reduce nutrient export to adjacent ecosystems [1,2]. Aerobic and anaerobic bacteria in CWs can degrade organic and inorganic compounds, as well as nutrients (nitrogen and phosphorus) [3,4].

CWs could be used to achieve cost-effective and efficient wastewater treatment. However, CWs system will produce greenhouse gases (GHG) during the wastewater treatment process and this could mitigate the environmental benefits of nutrient removal in these man-made ecosystems [5]. GHG, such as carbon dioxide (CO_2), nitrous oxide (N_2O), and methane (CH_4), could adversely affect the atmosphere [6].

Various environmental regulatory agencies do a large number of studies to improve CWs and current wetlands are still widely utilized in wastewater treatment [7,8].

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* Corresponding author.

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CWs are categorized into surface flow and subsurface flow (SSF) CWs [9]. SSF CWs include integrated vertical subsurface flow (IVSSF) CWs, which have been increasingly used because of the lower land requirement and higher pollutant removal efficiency than the other CWs types [4].

CH_4, as one of the main GHGs, has a lifetime of 8.4 years, which increase at the rate of almost 1% per year [10]. On a 100-year time horizon, CH_4 exhibits a global warming potential of 23 relative to CO_2 and is responsible for about 20% of the predicted warming [11]. The increase in CH_4 emission can reduce the oxidative power of the troposphere and induce the production of other GHGs, such as ozone, CO and CO_2. Natural and anthropogenic processes, particularly wastewater treatment, contribute to emission of CH_4 in the atmosphere [12]. Although CWs have high CH_4 emission potential, CH_4 formation in CWs depends on the oxygen status of the soil or sediments [13]. With increasing utilization of CWs, studies have focused on CH_4 flux in CWs. Results indicated that SSF CWs were under anoxic condition and provided a good environment for CH_4 generation in the methanogenesis process [14–16]. The root growth of plants in CWs can change the oxidation–reduction potential of the soil, thereby increasing CH_4 consumption to compensate for enhanced turnover of root materials and oxygen release. Hence, aquatic plants play an important role in soil microbial processes in CWs [17–19]. Scholars have studied the different effects of CH_4 generation and oxidation in CWs on C/N ratio [20], vegetation zone [21], and spatial and temporal variations [22]. Although the pollutant removals in CWs using plants were reported [23,24], the effects comparison of plant species (*Cyperus alternifolius*, *Canna indica*, *Acorus calamus*, and *Scirpus tabernaemontani*) on CH_4 flux and chemical oxygen demand (COD) removal in SSF CWs have been rarely investigated.

In this study, IVSSF CWs were used to purify synthetic wastewater. The effects of different plant species (*C. alternifolius*, *C. indica*, *A. calamus* and *S. tabernaemontani*) on wastewater treatment and CH_4 emission in up-flow and down-flow chambers were studied. The relationship between CH_4 flux and removal loading of COD was also analyzed. The comprehensive environmental effect of IVSSF CWs was discussed.

2. Materials and methods

2.1. Description of the CW

Four IVSSF CWs were planted with *C. alternifolius* (height: 18.86 ± 3.67 cm), *C. indica* (height: 9.14 ± 3.26 cm), *A. calamus* (height: 32.15 ± 2.97 cm), and *S. tabernaemontani* (height: 26.26 ± 2.65 cm), respectively. The IVSSF CWs were built in May 2015, and the planting time was from May 2015 to January 2016. Each IVSSF CW, measuring 150 cm × 90 cm × 90 cm (length × width × height), was made up of up-flow and down-flow chamber. The synthetic wastewater was introduced into down-flow chamber and discharged from up-flow chamber (Fig. 1). *C. alternifolius*, *C. indica*, *A. calamus* and *S. tabernaemontani* were mainly used for pollutant removal. The down-flow chamber was filled with gravel (diameter of 20–40 mm, depth of 0.3 m), fine sand (diameter of 2–5 mm, depth of 0.2 m), and soil (depth of 0.2 m from down to top layer). The up-flow chamber was filled with gravel (diameter of 20–40 mm, depth of 0.3 m), modified gravel (diameter of 1–2 mm, depth of 0.1 m), as reported in our previous study [25], and soil (depth of 0.3 m).

2.2. Experimental procedure

The IVSSF CWs were irrigated with synthetic wastewater consisting of COD (138–195 mg L^{-1}), NH_4-N (22–30 mg L^{-1}), total nitrogen (TN; 46–53 mg L^{-1}), and total phosphorous (TP; 4.5–6.0 mg L^{-1}). The buildings of IVSSF CWs were completed in May 2015 and filled with fillers. The four plants were planted with a density of 20 plants m^{-2}. In August 2015, all the plants resumed growth and the CWs reached maturity state. Synthetic wastewater was introduced by the PVC pipe with hydraulic loading of 0.3 m^3 m^{-2} d^{-1} using peristaltic pump (Fig. 1).

2.3. Sampling and measurement of water quality and CH_4 level

During the experimental period, influent and effluent water in the IVSSF CWs was sampled in opaque plastic bottles (100 mL) every 15 d for water quality determination.

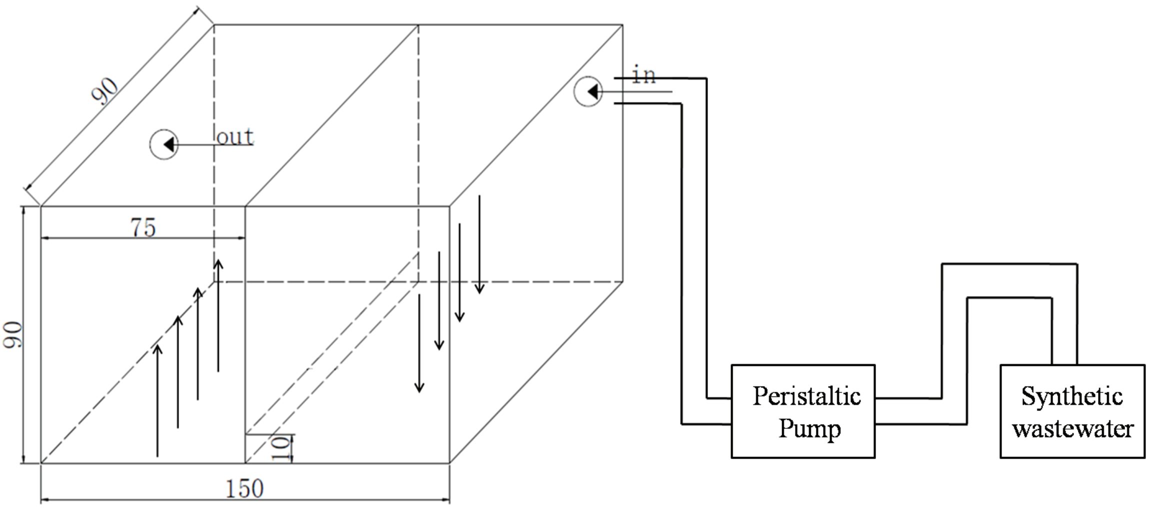

Fig. 1. Schematic diagram of integrated vertical subsurface flow constructed wetland.

COD, TN, TP, and NH_4-N were measured using the method described in Standard Methods for the Examination of Water and Wastewater [26].

CH_4 was sampled using a device with stainless steel base (50 cm × 50 cm × 30 cm) and organic glass cabinet (50 cm × 50 cm × 50 cm) and equipped with a measurement port in the upper part of the collector for sampling. The base was inserted into the soil surface up to 10 cm depth and sealed by a water-filled ring. Gas collectors were installed only during measurement by creating a hole in the soil and removing all rhizomes and roots of the plants. The fan at the top of the organic glass cabinet was used to uniformly mix the gas samples. The sampling connector was located in the middle of the device, and gas samples were collected by a gas-tight syringe and three-way valve. CH_4 emission was measured every 15 d between 9:30 a.m. and 11:30 a.m. and collected at 10 min intervals for 1 h (the average value was used as the final outcome of the measurement). All the gas samples were analyzed by gas chromatography (7890 B, Agilent) equipped with a flame ionization detector (working temperature of 250°C).

2.4. Data analysis

The removal efficiency, η (%), was calculated by the following equation:

$$\eta = \frac{C_0 - C_t}{C_0} \times 100\% \tag{1}$$

where C_0 (mg L^{-1}) is the initial concentration of the pollutants and C_t (mg L^{-1}) is the blank-corrected concentration of the pollutants at time t.

CH_4 flux was calculated by the following equation [27]:

$$J = \frac{dc}{dt}\frac{M}{V_0}\frac{P}{P_0}\frac{T}{T_0}H \tag{2}$$

where J (mg m^{-2} h^{-1}) is the CH_4 flux, dc/dt is the gas concentration gradient (mm^3 m^{-3} h^{-1}), M is the relative molecular mass, P (Kpa) is the pressure in the device, T (K) is the average temperature in the device, V_0 is the standard molar volume, P_0 is the standard atmospheric pressure, T_0 is the gas temperature in standard state, and H (m) is the height from the water surface to the top of the device.

All statistical analyses were performed using SPSS 22.0 software. The effects of plant species, CH_4 flux in up-flow and down-flow chambers, and environmental temperature were tested by two-way analysis of variance.

3. Results and discussion

3.1. Effects of plant species on water purification

Fig. 2(a) shows that the removal efficiencies for TN in CWs planted with *C. alternifolius*, *C. indica*, *A. calamus*, and *S. tabernaemontani* were 82.9%, 75.4%, 67.4%, and 51.2%,

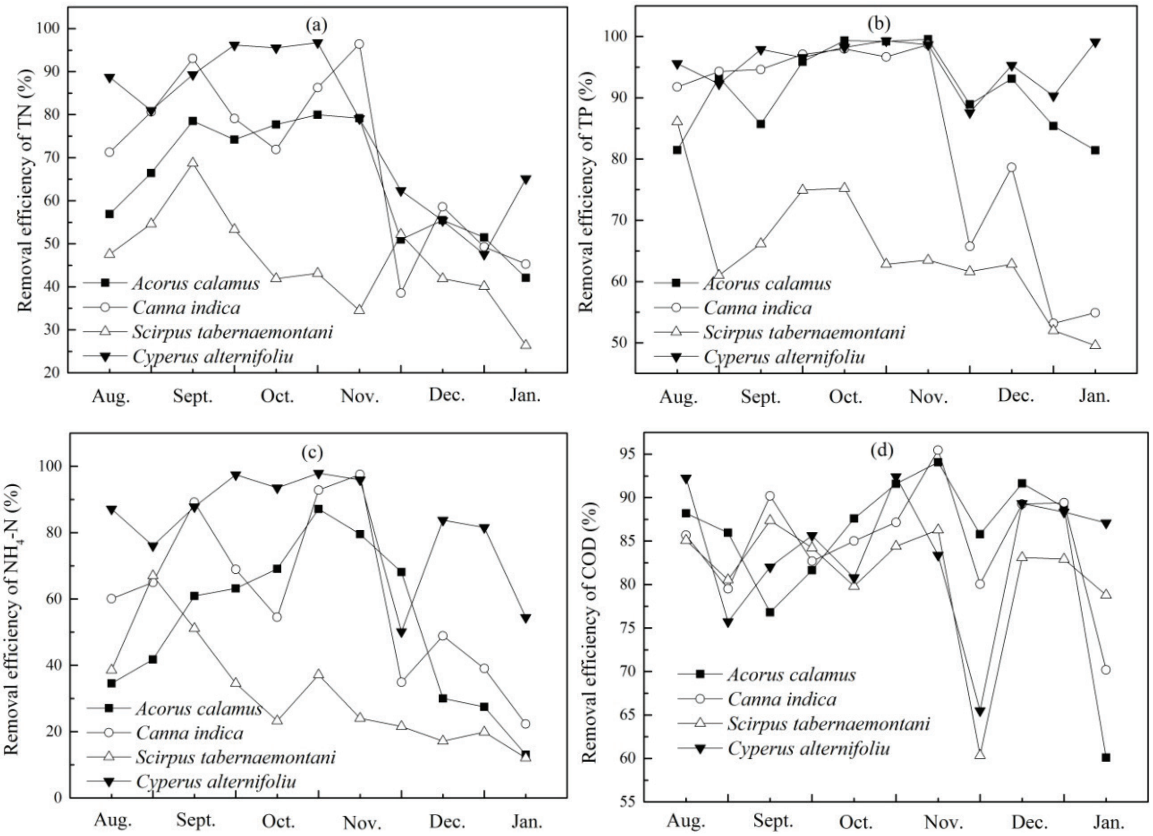

Fig. 2. Removal efficiency of TN (a), TP (b), NH_4-N (c), and COD (d) planted with four plants.

respectively. The trends of TN removal efficiency with increasing time were similar among CWs with the four plants. The highest removal efficiencies for TN in CWs planted with *C. alternifolius*, *C. indica*, and *A. calamus* were detected in November. However, the highest value in CWs planted with *S. tabernaemontani* was observed in September. TN removal efficiency was not significantly different ($P > 0.05$) among CWs planted with the four plants. In addition, TN removal efficiency was higher in summer than that in winter. In summer, the plants grew rapidly and had the denser roots. *C. alternifolius* had the highest TN removal efficiency in summer because of its developed root system. Hence, *C. alternifolius* might be the most suitable plant among the four plants for TN removal [28].

The average removal efficiencies for TP in CWs planted with the four plants were 94.9% (*C. alternifolius*), 89.9% (*A. calamus*), 85.0% (*C. indica*), and 60.6% (*S. tabernaemontani*; Fig. 2(b)). CWs with *C. alternifolius* exhibited the highest TP removal efficiency during the experimental period, except in early June and late November. The removal efficiency was not significantly different among CWs planted with the other three species in different time periods ($P > 0.05$). CWs with *S. tabernaemontani* exhibited the lowest TP removal efficiency and the removal efficiency decreased from late June (95.1%) to early January (48.9%).

In the experiments, the IVSSF CWs were irrigated with synthetic wastewater and the concentration of NH_4-N was from 22 to 30 mg L^{-1} accounting for approximately 50% of TN. The results showed that the average NH_4-N removal efficiency in CWs planted with the four plants decreased gradually from June to January (Fig. 2(c)). CW planted with *C. alternifolius* exhibited the highest removal efficiency for NH_4-N (average of 85.8%) and the highest value was detected in late October (96.1%). CW planted with *S. tabernaemontani* exhibited the lowest NH_4-N removal efficiency (average 37.3%) and the highest removal efficiency (66.4%) was detected in early August. The removal efficiencies of the other two CWs showed the irregular alterations as time changed. The average removal efficiencies for NH_4-N in CWs planted with the four plants had the similar trend with TN. Therefore, CWs planted with the four plants were suitable for the removal of synthetic wastewater with high NH_4-N concentration.

The four CWs exhibited high removal efficiency for COD (Fig. 2(d)). The CWs showed the highest removal efficiency in late October and the lowest removal efficiency in late November. In addition, the removal efficiency for COD in CWs planted with *C. indica* and *A. calamus* decreased in January. The growth of the plants reached their peaks in October and the developed roots provided active sites to promote microorganism growth, thereby increasing the removal efficiency for COD. The environmental temperature decreased rapidly in November, resulting in reduced activity of microorganisms in the CWs and decreased removal efficiency for COD. Although the temperature in January was low, the removal efficiency in CWs planted with *C. alternifolius* and *S. tabernaemontani* remained higher than that with *C. indica* and *A. calamus*. In addition, *C. alternifolius* and *A. calamus* CWs had the higher COD removal efficiency than CWs planted with *S. tabernaemontani* and *C. indica* in August. Hence, *C. alternifolius* was suitable for planting in both summer and winter. *S. tabernaemontani* should be planted in the CWs in winter, while *A. calamus* was more suitable for planting in summer.

The previous studies also indicated that the other kinds of CWs also had the high removal efficiencies of pollutants. The tidal flow operated CWs had the NH_4-N removal efficiency of 95%–99% [29], and the value was close to the NH_4-N removal result using *C. alternifolius* in late October. The oxidation pond–constructed wetland, a wastewater treatment system which was a combination of CW and oxidation pond, had the high COD removal efficiency (74.41%–80.61%) [30]. The COD removal of IVSSF CW using different plants was all higher than 80% using four plants. Therefore, the IVSSF CWs planted with *C. alternifolius*, *C. indica*, *A. calamus*, and *S. tabernaemontani* had higher COD removal efficiencies and TN and TP removal efficiencies in this study were also higher than that of other CWs [31,32].

3.2. Effect of plant species on CH_4 emission in up- and down-flow chambers

Fig. 3(a) shows CH_4 emission in CWs planted with *A. calamus*. The CH_4 flux decreased gradually from August to January and then decreased to 2 mg m^{-2} h^{-1} after October. The average CH_4 flux significantly differed between down-flow and up-flow chambers ($P < 0.01$). In addition, the CH_4 flux in the up-flow chamber was higher than that in the down-flow chamber in late August and early September. This finding might be explained by the fact that the anaerobic metabolism of microorganisms was reduced in the down-flow chamber, and the number of inorganic compounds transformed into organic compounds decreased [33,34]. Hence, processes in which methanogen produced CH_4 were delayed, resulting in increased CH_4 flux in the up-flow chamber.

The CH_4 flux in CWs planted with *S. tabernaemontani* is shown in Fig. 3(b). The flux was not significantly different ($P > 0.05$) between the samples from the up-flow and down-flow chambers, and the flux was affected by temperature ($P < 0.05$). The CH_4 flux in late September reached the highest value (26.07 mg m^{-2} h^{-1}) and the CH_4 flux was 1.10 mg m^{-2} h^{-1} for late December. Furthermore, CH_4 flux decreased to 1.00 mg m^{-2} h^{-1} from September to January.

The down-flow bed was the entrance of the water. The content of organic matters was high and the available carbon source of microbes was higher than the up-flow bed. Therefore, the CH_4 flux in CW planted with *C. indica* was higher in the down-flow chamber than that in the up-flow chamber from August to January. The average CH_4 flux decreased as the temperature decreased (Fig. 3(c)). The CH_4 flux in CW planted with *C. indica* was the highest in early August (30.41 mg m^{-2} h^{-1}) and decreased to less than 2.00 mg m^{-2} h^{-1} in late November. The CH_4 flux in the up-flow chamber significantly differed from that in the down-flow chamber and was affected by temperature ($P < 0.01$).

Fig. 3(d) shows the CH_4 flux in CW planted with *C. alternifolius*. The CH_4 flux in the up-flow chamber increased to the maximum (14.76 mg m^{-2} h^{-1}) and then decreased as the time changed. The CH_4 flux in the down-flow chamber exhibited similar tendency. In addition, the CH_4 flux in the up-flow chamber is −0.24 mg m^{-2} h^{-1} and the CW planted with *C. alternifolius* acted as a weak "sink" of CH_4. The CH_4

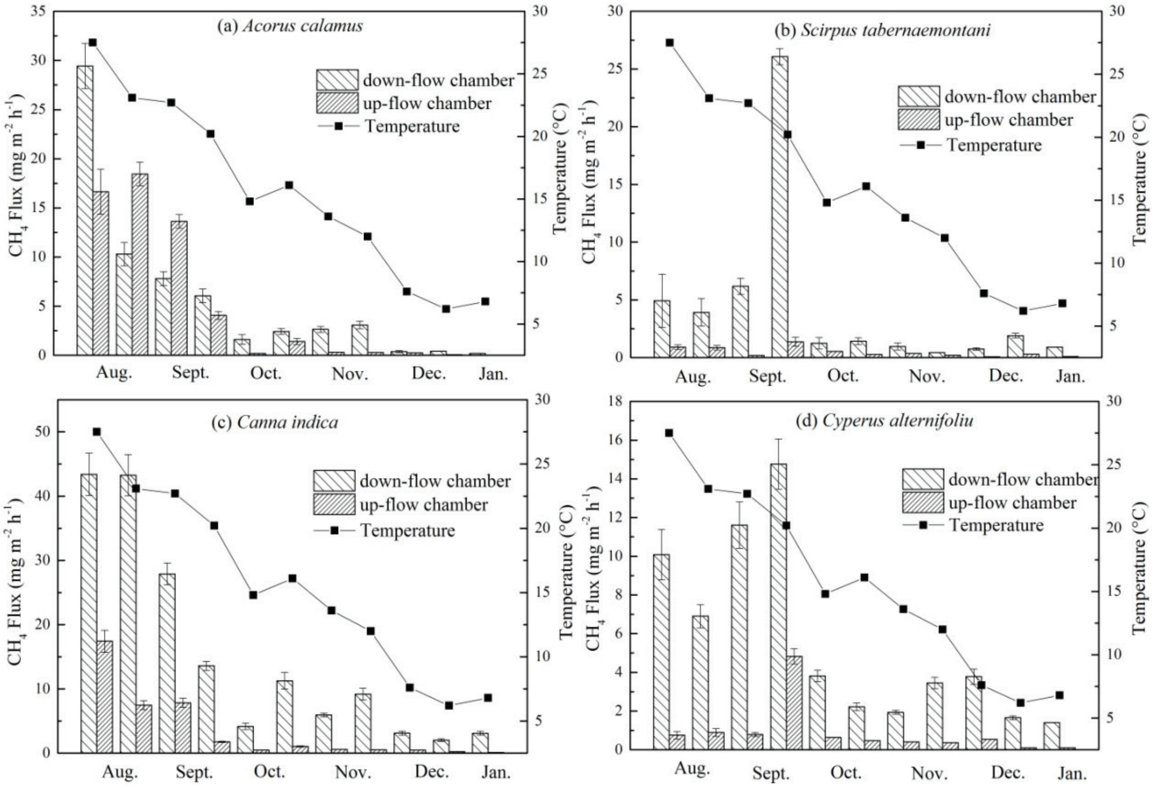

Fig. 3. CH_4 flux for constructed wetlands planted with *Acorus calamus* (a), *Scirpus tabernaemontani* (b), *Canna indica* (c), and *Cyperus alternifolius* (d) in down-flow and up-flow chamber.

flux in the down-flow chamber significantly varied with temperature ($P < 0.01$) but was not significantly different from that in the up-flow chamber.

The IVSSF CW system is one of the CH_4 sources in atmosphere. CH_4 emission is a complex process that is closely related to the activity and composition of microbes and is influenced by water loading, the temperatures, and other environmental conditions. Plant species is one of the most important influence factors on CH_4 emission in IVSSF CW, and different plants could cause the different CH_4 fluxes. The results indicated that IVSSF CWs planted with *C. indica* and *A. calamus* had the similar CH_4 emission fluxes, and the CH_4 fluxes were higher than other two IVSSF CWs. Therefore, CH_4 emission should be paid attention when using these two plants in IVSSF CW. *C. alternifolius* and *S. tabernaemontani* are the tracheophyte plants that can promote the CH_4 transmission, and CH_4 fluxes of IVSSF CWs planted with *C. alternifolius* and *S. tabernaemontani* reached the highest values in September. These two IVSSF CWs had the similar CH_4 emission fluxes because of the tracheophyte characteristics.

3.3. Relationship between CH_4 flux and removal loading of COD

$K_{COD/CH4}$ was the ratio of COD removal loading to the CH_4 flux. IVSSF CWs planted with different plants and with high $K_{COD/CH4}$ exhibited low CH_4 flux and the same COD removal loading. Fig. 4 shows the variations in $K_{COD/CH4}$ in CWs planted with the four plants. The values of $K_{COD/CH4}$ in three seasons were arranged as winter > autumn > summer. The values of $K_{COD/CH4}$ in August and September are lower than that in the other time periods and the CH_4 flux was higher than that in the other months. Furthermore, CWs planted with *S. tabernaemontani* exhibited higher $K_{COD/CH4}$ than that of CWs planted with the other plants in August and September. CWs planted with *S. tabernaemontani* showed higher capacity for COD removal than the other CWs. The removal loading of COD in late November decreased rapidly (Table 1) and the value of $K_{COD/CH4}$ decreased, except for CWs planted with *S. tabernaemontani*. In early November, the temperature began to decrease and the growths of *C. alternifolius*, *C. indica*,

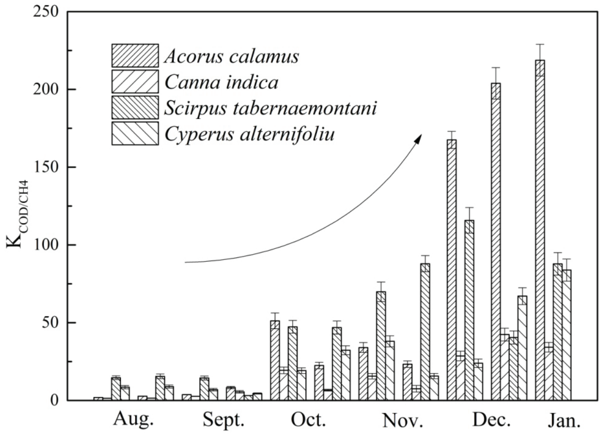

Fig. 4. $K_{COD/CH4}$ values of constructed wetlands planted with four plants from August to January.

Table 1
Removal loading of IVSSF CWs planted with four plants from August to January

Time	Removal loading (g COD m^{-2} h^{-1})			
	Acorus calamus	*Scirpus tabernaemontani*	*Canna indica*	*Cyperus alternifolius*
E-August[a]	44.14 ± 4.21	42.58 ± 4.23	42.88 ± 3.28	46.18 ± 3.76
L-August[b]	39.51 ± 3.37	37.03 ± 3.12	36.54 ± 3.33	34.82 ± 2.78
E-September	40.41 ± 4.39	45.96 ± 3.78	47.45 ± 4.78	43.15 ± 3.67
L-September	41.98 ± 4.56	43.31 ± 3.67	42.51 ± 4.01	44.04 ± 4.56
E-October	46.40 ± 3.17	42.28 ± 4.02	45.04 ± 4.27	42.80 ± 4.14
L-October	43.08 ± 5.38	39.70 ± 3.76	40.99 ± 3.28	43.46 ± 3.67
E-November	50.22 ± 5.65	46.07 ± 3.96	50.96 ± 6.23	44.52 ± 3.56
L-November	39.28 ± 4.17	27.63 ± 2.18	36.66 ± 2.89	29.98 ± 2.35
E-December	53.12 ± 4.11	48.18 ± 5.32	51.76 ± 4.23	51.79 ± 4.24
L-December	47.7 ± 3.40	44.49 ± 3.89	47.97 ± 5.34	47.40 ± 4.23
E-January	30.46 ± 3.27	44.96 ± 4.13	36.06 ± 2.98	44.73 ± 5.28

[a] E-August stands for early August.
[b] L-August stands for late August.

and *A. calamus* were inhibited. Although the temperature decreased in winter, *S. tabernaemontani* became more resistant to cold and kept its growth. CWs planted with *A. calamus* exhibited higher COD removal loading in early December (51.79 g m^{-2} h^{-1}) than that in the other months (Table 1). The $K_{COD/CH4}$ value was high in December and January. In addition, the $K_{COD/CH4}$ value in CWs planted with *A. calamus* was higher than that of other CWs. Hence, *A. calamus* was the optimal plant for COD removal in IVSSF CWs in winter [35,36].

4. Conclusions

Plant species in IVSSF CWs considerably influenced CH_4 emission and pollutant removal efficiency. The effects of plant species on water quality improvement and CH_4 emission in up-flow and down-flow chambers were studied, and the relationship between CH_4 flux and removal loading of COD was analyzed. CW with *C. alternifolius* had the highest removal efficiency of TP, TN and NH_4-N. In addition, four plants exhibited high removal efficiency for COD. CWs with *S. tabernaemontani* had the lowest CH_4 flux during the pollutants removal process. The plants significantly influenced CH_4 flux. The CH_4 fluxes of IVSSF CWs planted with *C. alternifolius*, *C. indica,* and *S. tabernaemontani* were significantly correlated with temperature. Moreover, the CH_4 fluxes of IVSSF CWs planted with *C. indica* and *A. calamus* significantly differed between the up-flow and down-flow chambers. Overall, *S. tabernaemontani* and *A. calamus* were found to be the optimal plants for COD removal in IVSSF CWs in summer and winter, respectively. This study could provide the theoretical foundation for plants selection according to effects of plant species on CH_4 emission.

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