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Original Article

Estimating greenhouse gas fluxes from constructed wetlands used for water quality improvement

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Abstract

Methane (CH_4), nitrous oxide (N_2O) and carbon dioxide (CO_2) fluxes were evaluated from constructed wetlands (CWs) used to improve domestic wastewater quality. Experiments employed subsurface flow (SF) and free water surface flow (FWS) CWs planted with *Cyperus* spp. Results showed seasonal fluctuations of greenhouse gas fluxes. Greenhouse gas fluxes from SF-CWs and FWS-CWS were significantly different ($p<0.05$) while pollutant removal efficiencies of both CWs were not significantly different. The average CH_4, N_2O and CO_2 fluxes from SF-CWs were 2.9±3.5, 1.0±1.7, and 15.2±12.3 mg/m^2/hr, respectively, corresponding to the average global warming potential (GWP) of 392 mg CO_2 equivalents/m^2/hr. For FWS-CWs, the average CH_4, N_2O and CO_2 fluxes were 5.9±4.8, 1.8±1.0, and 29.6±20.2 mg/m^2/hr, respectively, having an average GWP of 698 mg CO_2 equivalents/m^2/hr. Thus, FWS-CWs have a higher GWP than SF-CWs when they were used as a system for domestic water improvement.

Keywords: greenhouse gas fluxes, methane, nitrous oxide, carbon dioxide, constructed wetlands, domestic wastewater.

1. Introduction

Due to the main expectation that climate change will follow upon an increase in atmospheric concentration of greenhouse gases (e.g. CO_2, CH_4, N_2O), there is intense interest in the sources and sinks of these gases, and in the strength of their respective emissions and consumptions. Constructed wetland (CW) systems are the combination of natural wetlands and conventional wastewater treatment processes and are constructed in order to reduce input of nutrients and organic pollutants to water bodies. When wetlands are used for purification of wastewater, microbial processes and gas dynamics are likely to be altered. With increased inputs of nutrients and organic pollutants, the productivity of the ecosystem could increase as well as the production of greenhouse gases, which are by- or end-products of microbial decomposition processes. Constructed wetlands, therefore, can be sources of important greenhouse gases.

There are relatively few studies regarding CH_4, N_2O, and CO_2 fluxes emitted from constructed wetlands used for water quality controlling purposes since the total area of CWs worldwide is negligible as compare to all natural wetlands and agricultural areas. However, worldwide increase

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in the development of CWs necessitates an understanding of their potential atmospheric impact in light of the trend that natural wetlands in many countries are decreasing (e.g., Thailand) while environmental regulatory agencies are trying to stimulate an increase in CW acreage. Thus, insight knowledge to clarify the atmospheric impact of such wetlands is much needed.

CWs gas dynamics are greatly affected by climatic and weather conditions, especially by temperature and moisture (MacDonald *et al.,* 1998). Rate of photosynthesis (the source of energy and carbon in ecosystems) and microbial activities producing greenhouse gases increase with increasing temperature. Both denitrification and methane formation depend on the oxygen status of the soil or sediment and decomposition rates of organic matter. As a result, the temporal and seasonal variability of fluxes of CO_2, N_2O, and CH_4 are extremely high resulting from variations in the environmental factors regulating the microbial processes behind the gas fluxes (Liikanen *et al.,* 2006; Inamori *et al.,* 2007). In some seasons wetlands can act as a source or sink for carbon and there can be great differences in the CH_4 (Nykánen *et al.,* 1995) and N_2O fluxes (Huttunen *et al.,* 2002). Therefore, prospective studies are needed to obtain a holistic picture of the gas dynamics of constructed wetlands.

The objectives of this study were (1) to quantify CH_4, N_2O, and CO_2 fluxes from domestic wastewater treatment constructed wetlands, (2) to estimate seasonal fluctuations of CH_4, N_2O, and CO_2 fluxes from wastewater treatment constructed wetlands, and (3) to investigate the effect of constructed wetland types on CH_4, N_2O, and CO_2 fluxes.

2. Materials and Methods

2.1 Study site and constructed wetland systems

The experimental scale of constructed wetlands was conducted in four constructed wetlands (CWs) at 14°53'24.48"N and 102°00'23.11"E, Suranaree University of Technology, Nakhon Ratchasima Province in northeastern Thailand. This area of Thailand has three-season monsoonal climate, with a relatively cool dry season from November to late February, followed by a hot dry season from March to June and then a hot rainy season from July to October (TMD, 2011).

The CWs were built based primarily on criteria of aspect ratio (AR) or length to width of 4:1 to minimize short circuiting and force the flow to move closely to plug flow hydraulic regime (U.S. EPA, 2000). Each of the units was 2.0 m × 0.5 m × 0.8 m (length × width × depth) in dimension. Brick, cement, and mortar were the materials used for the construction of the CWs. Permanent transparent roof made from clear plastic was also constructed to prevent rain getting into the experiment setup while allowed direct sunlight exposure. Synthetic wastewater similar to domestic discharge from Thailand's Housing Estates was fed as an influent. The composition of the synthetic domestic wastewater was glucose, $FeCl_3$, $NaHCO_3$, KH_2PO_4, $MgSO_4"7H_2O$, and urea (Sirianuntapiboon and Tondee, 2000). Synthetic domestic wastewater was fed daily into the constructed wetlands containing 17-23 mg/l biochemical oxygen demand (BOD), 115-235 mg/l chemical oxygen demand (COD), 0.28-0.35 mg/l ammonia nitrogen (NH_3-N), and 0.03-0.33 mg/l total phosphorus (TP). Hydraulic loading rate, the ratio of flow and surface area (0.5 m x 2.0 m), was estimated to be about 0.04 m^3/d for each unit. Average organic loading rate was approximately 42 mg/d.

The experiment units were designed to have a duplicate of both sub-surface flow (SF) beds and free-water surface (FWS) beds. The monoculture emergent plant, *Cyperus* spp., was grown in each CW unit. The SF constructed wetlands had a media depth of 0.70 m since the water must filtrate down to the bottom and accumulate until it reached the outlet at 0.65 m. An additional 0.05 m was topsoil. The media depth in FWS constructed wetlands was approximately 0.45 m in height with the water depth of 0.20 m above the media, 0.65 m all together (Figure 1).

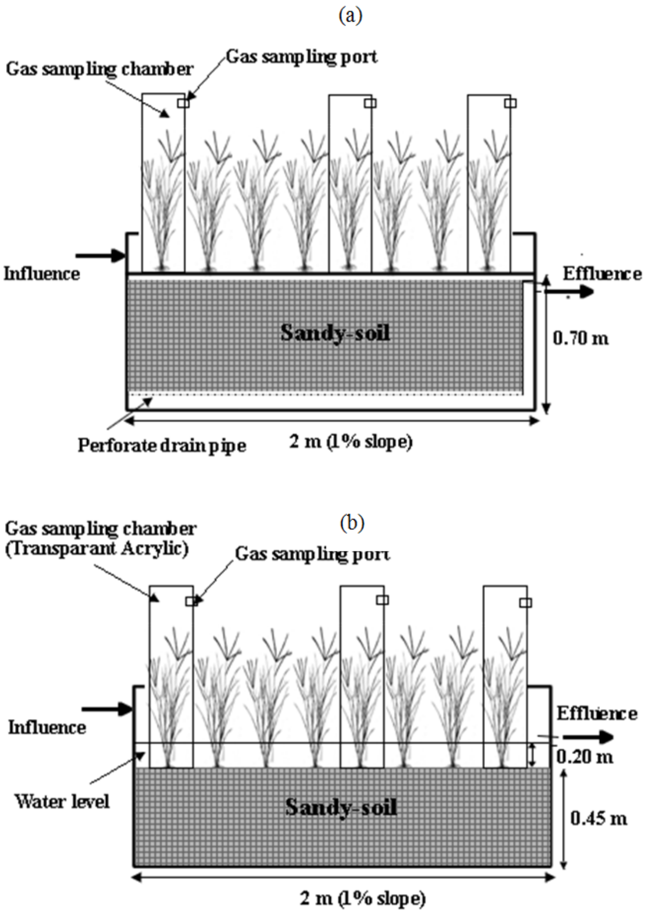

Figure 1. Schematic diagram of the experimental plots (a) sub-surface flow constructed wetland and (b) free-water surface constructed wetland. Gas sampling chambers were placed during gas sampling only.

2.2 Gas fluxes measurement

The gas emissions were measured using a static chamber method described by Hutchingson and Moiser (1981). The chambers consisted of two parts. The upper part was constructed from 3.0 mm clear acrylic sheet and made gas-tight by heated glue doubling with silicon sealant. The acrylic chamber equipped with a thermometer, a fan, and two gas sampling points. The chamber had a total height of 1.5 m and 0.25 m in width and length in which the gases emitted from the constructed wetlands were trapped and sampled. A small fan was operated during the sampling period to create thorough gas mixing inside the chamber. The lower part was aluminum base to firmly attached acrylic chamber and soil surface. Four-side groove was made with 4.0 mm trench to accommodate the acrylic chamber during the gas sampling. The aluminum frame was firmly inserted into the top soil overnight prior to the measurement. At the beginning, the chamber was placed on top of the aluminum frame and water was filled in the groove to prevent gas leak. Each constructed wetlands accommodated three chambers, at the inlet, middle and outlet. Gas measurements were performed at these locations. The sampling periods began at 0, 15, 30, and 45 minute intervals.

Carbon dioxide was measured real-time with CO_2 gas analyzer (LI-820 model, LI-COR, Inc., U.S.A.). Data were recorded in a computer and retrieved for later analysis. The instrument used non-dispersive infrared (NDIR) as detector to determine the concentration of carbon dioxide.

While carbon dioxide was measured real-time in the field, two gas sampling vials were used to store gas samples. Glass vials were evacuated trapped air inside to generate negative pressure right before gas sampling. Gas samples were immediately store and kept under 4°C until the time of analysis. Conditions for gas analysis are listed in Table 1. The analysis of methane and nitrous oxide were performed with Agilent GC 6890 (Agilent, U.S.A.) equipped with FID and micro-ECD detectors. Agilent Chemstation A.08.03 software (Agilent, U.S.A.) was used for spectrum and gas concentration determination against standard gases (Scott Specialty Gases, The Netherlands). The sensitivity of N_2O detection was estimated to be about 0.2 ppmv.

2.3 Gas flux analysis

Emission rates were calculated based on the linear change of gas concentration over time. The gas concentration for each sample was plotted in a concentration versus time. The derivative represents the gas emission rate (ppm/hr) of the series, which were then converted to flux rates (mg/m^2/hr) and corrected for chamber volume and temperature (Healy *et al.*, 1996). Regressions were performed on each flux rate in Microsoft Excel® to determine linearity of flux. If the increase/decrease in the gas concentration was non-linear ($r^2 < 0.85$) the measurement was rejected (Altor and Mitsch, 2006).

Gas flux (mg/m^2/hr) was calculated by the following equation (at the reference air pressure of 1 atm)

$$E = \frac{XhM}{RT} \tag{1}$$

Where E = emission on the aerial basis (mg/m^2/hr), X = gas concentration increase in chamber (ppm/hr), h = height of chamber (m), M = molecular weight (g/mol), R = gas constant = 0.0821 (atm”L/K/mol), and T = absolute temperature (K).

2.4 Environmental factors monitoring

The study on seasonal variation of greenhouse gas fluxes from different constructed wetlands was performed monthly from June 2010 to May 2011. Measurements of greenhouse gas fluxes were conducted between 09:00 and 15:00 periods. Gas fluxes data were analyzed seasonally. During the gas sampling period, soil temperature at the 5 cm depth was also continuously monitored, whereas soil pH, soil ORP (at 5 cm depth) were measures by pH/ORP meter.

Table 1. Gas chromatography conditions for gases analysis.

Conditions	CH_4 analysis	N_2O analysis
Column	Poraplot Q capillary column (10 m x 0.32 mm ID)	HP-Plot Q capillary column (15 m × 0.53 mm ID)
Column temperature	40°C	180°C
Injector temperature	250°C	200°C
Detector	flame ionization detector (FID)	micro electron capture detector (micro-ECD)
Detector temperature	300°C	300°C
Carrier gas	N_2	N_2
Gas flow rate	20 ml/min	15 ml/min
Spit ratio	0.7:1	None
Spit flow	15.0 ml/min	None

2.5 Wastewater analysis

Samples were analyzed according to the Standard Methods of Waste Water Protocols (APHA-AWWA-WEF, 2005). Daily influent and effluent samples were collected and analyzed for BOD until the steady-state conditions were achieved. Then, routine wastewater analysis was carried out to determine the removal efficiency of BOD, COD, NH_3-N, and TP for the period of once a month.

2.6 Data analysis

Statistical analysis was performed with SPSS® and Microsoft Excel® for Windows®. All data entering statistical comparisons were tested for homogeneity of variance and normal distribution using Levene and Kolmogorov–Smirnov Test. If assumptions of normal distribution were fulfilled, independent–samples t-test was carried out. Otherwise non-parametric Mann-Whitney U–Test was used. All results were considered statistically significant if p value was <0.05.

3. Results and Discussion

3.1 Performance of pollutant removals

During the operation of constructed wetland units the removal of BOD, COD, NH_3-N, and TP was investigated from June 2010 to May 2011. Descriptive statistics for pollutants removal rates of two constructed wetland types are shown in Table 2. Removal rates of BOD, COD, NH_3-N, and TP were in the ranges of 47-69%, 32-65%, 27-60%, and 20-68%, respectively. Since the data were normally distributed and showed homogeneity of the variances, independent–samples *t*-test was suitable for the analysis of variation of pollutants removal rates from different constructed wetlands. Overall performance of pollutant removals between SF and FWS constructed wetlands showed that pollutant removal efficiencies of both types constructed wetlands were not significantly different under experimental conditions. The average removal rates of COD in SF-CW were comparable to the results reported in Tanzania with the ranged of 33.6%-60.7% (Kaseva, 2004) while BOD removal rates was a bit lower than the results reported by Hadad *et al.*(2006) in Argentina at 76%. The highest removal rate of TP in this present study was similar to Hadad *et al.* (2006) as well.

3.2 Seasonal variations of greenhouse gas fluxes and environmental factors

3.2.1 Seasonal variations of methane fluxes from constructed wetlands

During one year of experiment, CH_4 fluxes from all constructed wetlands were noticeable higher in August and November than those in other months (Figure 2). The highest CH_4 fluxes from SF occurred in August with the average of 10.5 mg/m^2/hr whereas FWS occurred in October with the average of 15.3 mg/m^2/hr. The lowest CH_4 fluxes from SF occurred in May with the average of 0.7 mg/m^2/hr whereas FWS occurred in December with the average of 1.0 mg/m^2/hr. When seasonal variation was taken into account, the results showed that CH_4 fluxes from SF and FWS constructed wetlands were highest in hot rainy season (July-October) with the average of 5.2 and 10.8 mg/m^2/hr, respectively, followed by cold season (November-February) with the

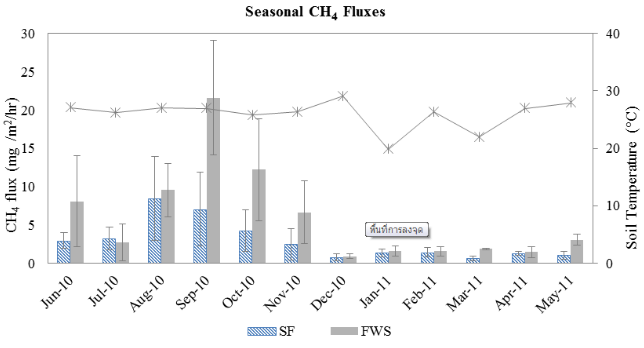

Figure 2. Seasonal variation of CH_4 fluxes from constructed wetland planted with *Cyperus* spp.

Table 2. Descriptive statistics and comparison of pollutants removal rates between SF and FWS constructed wetlands using t- test.

Pollutant/CW		Pollutants removal rates (%)			t-test	
		N	Mean	S.D.	Mean difference	Sig.(2-tailed)
BOD	SF	12	61.26	5.26	1.73	0.531
	FWS	12	59.53	7.83		
COD	SF	12	49.26	8.98	-1.45	0.688
	FWS	12	50.71	8.41		
NH_3	SF	12	39.92	9.05	-1.22	0.757
	FWS	12	41.13	9.94		
TP	SF	12	48.42	16.48	9.37	0.159
	FWS	12	39.05	14.97		

average of 1.5 and 2.6 mg/m^2/hr, respectively, and lowest in summer season (March-June) with the average of 1.0 and 2.1 mg/m^2/hr, respectively. Environmental parameters monitored during the experiment showed an unusual phenomenon in the area especially air temperature at the end of 2010 and early 2011. Monthly average of air temperature reached maximum in September (36.7°C) while the lowest mean was found in March (24.7°C). It is important to note that the lower air temperature in March was quite unusual but it occurred in March 2011. Soil temperatures, measured at 5 cm depth, were in the optimum range for microbial activities and changed in a narrow-range during three seasons. It peaked in December (29°C) with average of 29°C and reached its minimum value in January (19.9°C) before it rise again in summer season (March-June).

3.2.2 Seasonal variations of nitrous oxide fluxes from constructed wetlands

N_2O fluxes were high during April and October compared with the rest of the year (Figure 3). The highest N_2O fluxes from SF occurred in April with the average of 5.3 mg/m^2/hr whereas FWS occurred in August with the average of 6.9 mg/m^2/hr. Lowest N_2O fluxes from SF occurred in January with the average of 0.3 mg/m^2/hr whereas FWS occurred in December with the average of 0.3 mg/m^2/hr. When seasonal variation was taken into account, N_2O fluxes from SF was highest in summer season (March-June) with the average of 2.0 mg/m^2/hr, followed by hot rainy season (July-October) with the average of 1.0 mg/m^2/hr, and lowest in cold season (November-February) with the average of 0.4 mg/m^2/hr. While N_2O fluxes from FWS was highest in hot rainy season (July-October) with average of 2.6 mg/m^2/hr, followed by summer season (March–June) with average of 2.5 mg/m^2/hr and lowest in cold season (November-February) with average of 0.3 mg/m^2/hr.

3.2.3 Seasonal variations of carbon dioxide fluxes from constructed wetlands

Estimated CO_2 fluxes were low in December and February. The highest CO_2 fluxes from SF occurred in September with the average of 32.7 mg/m^2/hr whereas FWS occurred in May with the average of 60.2 mg/m^2/hr (Figure 4). The lowest CO_2 fluxes from SF occurred in February with the average of 3.2 mg/m^2/hr whereas FWS occurred in December with the average of 8.5 mg/m^2/hr. In the case of seasonal variations, CO_2 fluxes from SF and FWS were highest in summer season (March-June) with average of 21.2 and 49.7 mg/m^2/hr, respectively, followed by hot rainy season (July-October) with average of 19.6 and 28.5 mg/m^2/hr, respectively, and lowest in cool season (November-February) with average of 5.1 and 15.6 mg/m^2/hr, respectively.

GHG fluxes from CWs were observed but the pattern of seasonal CO_2 variations and environmental factors was not distinct. It was possible that important environmental factors such as soil temperature, soil pH, and soil ORP were in the optimum range for GHG production and changed occurred in a narrow-range during the experiment. Another reason was that GHG fluxes were not the result of a one-factor action but of the interaction of more biotic and abiotic factors. Soil pH was in the range of 6.70 to 8.02, and soil ORP was in negative range (-168 to -232 mV). In cold climate, SØvic and KlØve (2007) reported seasonal variations differences of N_2O from FWS-CWs in which N_2O emissions were highest in autumn while CH_4 did not show significant seasonal difference.

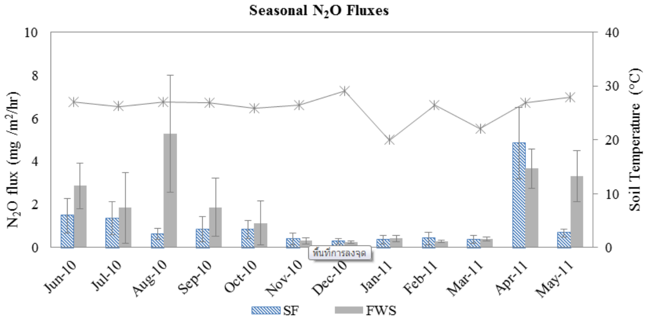

Figure 3 Seasonal variation of N_2O fluxes from constructed wetland planted with *Cyperus* spp.

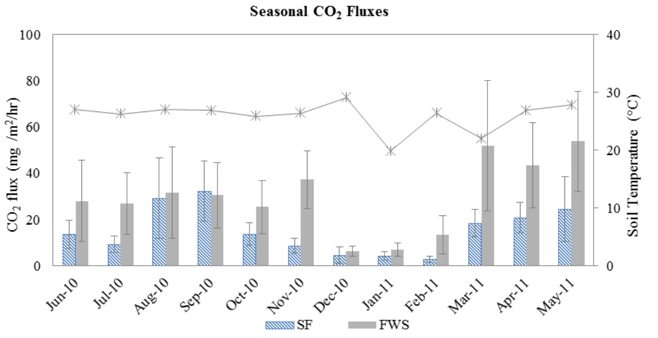

Figure 4. Seasonal variation of CO_2 fluxes from constructed wetland planted with *Cyperus* spp.

3.3 Greenhouse gas fluxes and constructed wetland type

The investigation of GHG fluxes from different type of constructed wetlands found that CH_4, N_2O and CO_2 fluxes released from SF-CWs had the average of 2.9±2.5, 1.0±0.7, and 15.2±12.3 mg/m^2/hr, respectively. In FWS-CWs, CH_4, N_2O, and CO_2 fluxes had the average of 5.9±4.8, 1.8±1.0, and 29.6±20.2 mg/m^2/hr, respectively. Kaewkamthong (2002) reported the flux of CH_4 from FWS-CWs planted with *Digitaria bicornis* and *Typha angustifolia* in the range of 2.7–75.7 mg/m^2/hr. In Japan, Zhu *et al.* (2007) used *Phargmites communit* in FWS-CWs and CH_4 flux varied from 0-4 mg/m^2/hr. Estimated N_2O flux from SF-CWs in another experiment in Japan was 0-1.4 mg/m^2/hr (Wang *et al.*, 2008).

Table 3. Seasonal variation of GHG fluxes from different constructed wetlands.

CW type/Season		CH_4 emission (mg/m^2/hr)		N_2O emission (mg/m^2/hr)		CO_2 emission (mg/m^2/hr)	
		Mean	S.D.	Mean	S.D.	Mean	S.D.
SF	Hot rainy	5.17	4.48	1.03	0.70	19.63	13.53
	Cool	1.48	1.22	0.37	0.15	5.06	3.31
	Summer	0.96	0.55	1.96	3.11	21.25	9.39
FWS	Hot rainy	10.83	13.58	2.60	2.44	28.47	14.05
	Cool	2.65	3.07	0.32	0.12	15.99	17.88
	Summer	2.12	1.47	2.46	1.71	49.68	15.89

Table 4. Comparison of GHG fluxes from different constructed wetlands using Mann-Whitney U-Test including descriptive statistics.

GHG/CW		GHG fluxes (mg/m^2/hr)			Mean difference	Sig. (2-tailed)
		N	Mean	S.D.		
CH_4	SF	72	2.89	2.55	-3.03	0.022*
	FWS	72	5.92	4.82		
	Total	144	4.41	7.51		
N_2O	SF	72	1.05	0.70	-0.76	0.108
	FWS	72	1.80	1.06		
	Total	144	1.42	1.92		
CO_2	SF	72	15.18	12.32	-14.43	0.000**
	FWS	72	29.61	20.25		
	Total	144	22.39	18.20		

*significant at the 0.05 level, ** significant at the 0.01 level.

Mander *et al.* (2008) reported the range of CH_4 and N_2O fluxes from FWS-CWs used in Estonia to be about 0.06-0.17 and 0.05-0.06 mg/m^2/hr, respectively. Our results were in the range reported in hot climate but somewhat higher than the values found in European studies. However, the effect of different plants on GHGs fluxes remains debated (Maltais-Landry *et al.*, 2009).

When compare the results from SF-CWs and FWS-CWs, CH_4 and CO_2 fluxes were significantly different ($p< 0.05$) (Table 4). Average CH_4 and CO_2 fluxes from FWS-CWs were significantly higher than the fluxes from SF-CWs. However, there was no significant difference ($p>0.05$) in the average N_2O fluxes from SF and FWS constructed wetlands.

3.4 Global warming potential of greenhouse gases from SF and FWS constructed wetlands

Due to differences in potential to cause global warming, it was important to estimate the impact of greenhouse gases in terms of global warming potential (GWP) described by IPCC (2001). The average GWP of CH_4 and N_2O were 23 and 296 times of CO_2, respectively. In the present study, estimated GWP of SF-CWs was about 66 and 311 mg CO_2 equivalents/m^2/hr for CH_4 and N_2O, respectively. Overall, the greenhouse gas flux from SF-CWs was approximately 392 mg CO_2 equivalents/m^2/hr. Estimated GWP of FWS-CWs was about 136 and 533 mg CO_2 equivalents/m^2/hr for CH_4 and N_2O, respectively. Combined GWP of all greenhouse gases was approximately 698 mg CO_2 equivalents/m^2/hr. Thus, the results indicated that the estimated GWP of GHG fluxes from FWS-CWs was higher than SF-CWs about1.8 times. It indicated that the FWS-CWs planted with *Cyperus* spp. has higher potential in response to global warming compared to SF-CWs when they were used to improve water quality.

4. Conclusions

The average CH_4, N_2O and CO_2 fluxes from SF-CWs planted with *Cyperus* spp., were 2.9±2.5, 1.05±0.7, and 15.2±12.3 mg/m^2/hr, respectively. Higher flux was found from FWS-CWs with the average of CH_4, N_2O, and CO_2 fluxes at 5.9±4.8, 1.8±1.0, and 29.6±20.2 mg/m^2/hr, respectively. Seasonal fluctuations of GHG fluxes from wastewater treatment constructed wetlands could be observed. However, the pattern of seasonal CO_2 variations and environmental factors was not distinct.

GHG fluxes from SF and FWS constructed wetland showed significantly different ($p<0.05$) while pollutant removal efficiencies of both constructed wetlands were not significantly different. The average GWP of SF-CWs and FWS-CWs were approximately 392 and 698 mg CO_2 equivalents/m^2/hr, respectively. Thus, GWP released for FWS was higher than for SF constructed wetlands.

For a better understanding of GHG fluxes from constructed wetlands, further studies should be conducted on influencing factors related to the rate of GHG production, entrapment, oxidation and emission. Additionally, GHG fluxes from constructed wetlands maybe influenced by other factors, such as type and amount of substrates, water chemistry (e.g. dissolved SO_4^{-2} or NO_3^-) and the activity of microorganism.

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