

Seasonal and spatial variations of methane emissions from coastal marshes in the northern Yellow River estuary, China

Zhigao Sun · Huanhuan Jiang · Lingling Wang ·
Xiaojie Mou · Wanlong Sun

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Abstract

Aims and methods To evaluate the seasonal and spatial variations of methane (CH₄) emissions and understand the controlling factors, we measured CH₄ fluxes and their environmental variables for the first time by a static chamber technique in high *Suaeda salsa* marsh (HSM), middle *S. salsa* marsh (MSM), low *S. salsa* marsh (LSM) and bare flat (BF) in the northern Yellow River estuary throughout a year.

Results CH₄ emissions from coastal marsh varied throughout different times of the day and significant differences were observed in some sampling periods ($p < 0.05$). Over all sampling periods, CH₄ fluxes averaged between $-0.392 \text{ mgCH}_4 \text{ m}^{-2} \text{ h}^{-1}$

and $0.495 \text{ mgCH}_4 \text{ m}^{-2} \text{ h}^{-1}$, and emissions occurred during spring ($0.008 \text{ mgCH}_4 \text{ m}^{-2} \text{ h}^{-1}$) and autumn ($0.068 \text{ mgCH}_4 \text{ m}^{-2} \text{ h}^{-1}$) while sinks were observed during summer ($-0.110 \text{ mgCH}_4 \text{ m}^{-2} \text{ h}^{-1}$) and winter ($-0.009 \text{ mgCH}_4 \text{ m}^{-2} \text{ h}^{-1}$). CH₄ fluxes from the four marshes were not significantly different ($p > 0.05$), and emissions occurred in LSM ($0.026 \text{ mgCH}_4 \text{ m}^{-2} \text{ h}^{-1}$) and BF ($0.055 \text{ mgCH}_4 \text{ m}^{-2} \text{ h}^{-1}$) while sinks were observed in HSM ($-0.035 \text{ mgCH}_4 \text{ m}^{-2} \text{ h}^{-1}$) and MSM ($-0.022 \text{ mgCH}_4 \text{ m}^{-2} \text{ h}^{-1}$). The annual average CH₄ flux from the intertidal zone was $0.002 \text{ mgCH}_4 \text{ m}^{-2} \text{ h}^{-1}$, indicating that coastal marsh acted as a weak CH₄ source. Temporal variations of CH₄ emission were related to the interactions of abiotic factors (temperatures, soil moisture and salinity) and the variations of limited C and mineral N in sediments, while spatial variations were mainly affected by the vegetation composition at spatial scale.

Conclusions This study observed a large spatial variation of CH₄ fluxes across the coastal marsh of the Yellow River estuary (CV=7856.25 %), suggesting that the need to increase the spatial replicates at fine scales before the regional CH₄ budget was evaluated precisely. With increasing exogenous nitrogen loading to the Yellow River estuary, the magnitude of CH₄ emission might be enhanced, which should also be paid more attentions as the annual CH₄ inventory was assessed accurately.

Keywords Methane flux · Seasonal variation · Spatial variation · Coastal marshes · Yellow River estuary

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Z. Sun (✉) · L. Wang · X. Mou · W. Sun
Key laboratory of Coastal Zone Environmental Processes,
Yantai Institute of Coastal Zone Research (YIC), Chinese
Academy of Sciences (CAS); Shandong Provincial Key
Laboratory of Coastal Zone Environmental Processes,
YICCAS,
Yantai, Shandong 264003, People's Republic of China
e-mail: zgsun@yic.ac.cn

H. Jiang
Policy Research Center for Environment and Economy,
Ministry of Environmental Protection,
Beijing 100029, People's Republic of China

X. Mou · W. Sun
University of Chinese Academy of Sciences,
Beijing 100049, People's Republic of China

Introduction

Methane (CH₄) is an important greenhouse gas (GHG) that has more global warming potential than carbon dioxide (CO₂) and has been recognized to contribute global warming by 25 % (Mosier 1998). The globally averaged atmospheric CH₄ concentration increased from 715 ppb in 1750 to 1,774 ppb in 2005 (IPCC 2007), and increased by 8.3±0.6 ppb during 2007 and 4.4±0.6 ppb in 2008 (Dlugokencky et al. 2009). In 2010, the globally averaged CH₄ concentration reached 1,808 ppb, which exceeded the highest annual mean abundance so far (World Meteorological Organization 2011). Emission of CH₄ from various natural ecosystems has significant influences on the global climate change since it accounts for 30–40 % of the total CH₄ emissions (150~237 TgCH₄year⁻¹) (Allen et al. 2007). Tropical soil and wetlands play an important role in the global carbon biogeochemical cycles and are considered significantly natural sources of CH₄, contributing approximately 24 % towards this inventory (Whalen 2005).

CH₄ fluxes from wetlands are the result of CH₄ production, consumption and transportation from the anaerobic zone to the atmosphere (Bubier and Moore 1994). Considerable efforts have been widely made to investigate the CH₄ fluxes and key controlling factors (such as water table, temperature, vegetation, substrate availability for methanogens and redox condition) in different natural wetland ecosystems (Allen et al. 2007; Ganguly et al. 2008; Turetsky et al. 2008; Pennock et al. 2010; Danevčič et al. 2010; Sun et al. 2011). The complex processes of CH₄ fluxes and the variability of their controlling factors cause dramatic spatial and temporal variations of CH₄ fluxes. The accurate global CH₄ budget from different wetlands is very important for forecasting the future climate. However, one of the primary problems in attempting to develop accurate CH₄ budgets for global wetlands is the large spatial and temporal variability in CH₄ emission rates that are reported from all over the world. A clear understanding these spatial and temporal variations and their primary controlling factors in site measurements will be favorable for obtaining the optimal possible up-scaled emission estimation (Dinsmore et al. 2009).

Coastal marsh is characterized by high temporal and spatial variation related to topographic features, environmental factors and astronomic tidal fluctuation, and

is very sensitive to global climate changes and human activities. Above all, the intertidal zone between terrestrial and coastal ecosystems may represent a high dynamic interface of intense material processing and transport, with potentially high GHGs emission (Hirota et al. 2007). In the past two decades, considerable efforts have been made to quantify the CH₄ fluxes in different coastal ecosystems, especially in estuarine salt marshes (Shingo et al. 2000; Wang et al. 2009), mangrove swamps (Allen et al. 2007; Ganguly et al. 2008), coastal lagoons (Gregorich et al. 2006; Hirota et al. 2007) and coastal marshes (Sun et al. 2002; Amouroux et al. 2002). In China, the studies on CH₄ emission from coastal marshes started quite late (in the 2000s), and the related research mainly focused on the coastal tundra marshes in Antarctica (Sun et al. 2002; Zhu et al. 2008), the mangrove swamps in southeast coastal area (Lu et al. 2000; Ye and Lu 2001), and the salt marshes in the Yangtze River estuary (Yang et al. 2007; Cheng et al. 2007; Wang et al. 2009; Cheng et al. 2010) and the Min River estuary (Tong et al. 2009; Zeng et al. 2010). However, few studies have reported on coastal marshes in northern estuaries, such as Liao River estuary and Yellow River estuary.

The Yellow River is well known as a sediment-laden river. Every year, approximately 1.05×10^7 tons of sediment is carried to the delta (Cui et al. 2009) and deposited in the slow flowing landform, resulting in vast floodplain and natural marsh landscape (Xu et al. 2002). Sediment deposition is an important process for the formation and development of coastal marshes in the Yellow River Delta. The deposition rate of sediment in the Yellow River not only affects the formation rate of coastal marshes, but also, to some extent, influences the water or salinity gradient and the succession of plants from the land to the sea. With an area of 964.8 km², coastal marsh is the main type of marsh in the Yellow River Delta and accounts for 63.06 % of total area (Cui et al. 2009). *Suaeda salsa*, an annual C₃ plant, is one of the most prevalent halophytes in coastal marshes of the Yellow River estuary. As a pioneer plant, it has strong adaptations to environmental stresses, such as high salinity, tidal inundation and sediment burial (Han et al. 2005). In the intertidal zone, three phenotypes are generally formed due to the differences of water and salinity conditions in high, middle and low tidal flats (marshes). In recent years, the nitrogen (N) and organic matter (OM) loadings of the Yellow River estuary have significantly

increased due to the effects of human activities, and approximately 4,650 tons of nutrient and 4.33×10^5 tons of COD_{cr} are discharged into Bohai Sea every year (State Oceanic Administration of China 2010). Increases in N and OM loadings to estuarine and coastal marshes can stimulate microbial processes and associated GHGs emission (Seitzinger and Kroeze 1998; Purvaja and Ramesh 2001). However, CH_4 fluxes from different coastal *S. salsa* marshes in the Yellow River estuary remains poorly understood till now.

In this paper, we investigated CH_4 fluxes from the coastal marshes in the Yellow River estuary during the spring, summer, autumn and winter of 2010/2011. The objectives were: *i*) to determine the spatial and temporal variations of CH_4 fluxes from different coastal *S. salsa* marshes and bare flat, *ii*) to identify the key factors controlling the variations and assess the potential effects of exogenous N loading on CH_4 emission.

Materials and methods

Site description

The study was carried out in the northern intertidal zone of the Yellow River estuary, which is located in the Nature Reserve of Yellow River Delta ($37^{\circ}35'N \sim 38^{\circ}12'N$, $118^{\circ}33'E \sim 119^{\circ}20'E$) in Dongying City, Shandong Province, China. The nature reserve is of typical continental monsoon climate with distinctive seasons. The annual average temperature is 12.1°C and the frost-free period is 196 day. The average temperature in spring, summer, autumn and winter are 10.7°C , 27.3°C , 13.1°C and -5.2°C , respectively. The annual evaporation is 1,962 mm, the annual precipitation is 551.6 mm, and about 70 % of precipitation occurring between June and August. The soils are dominated by intrazonal tide soil and salt soil (Tian et al. 2005). The main plant communities include *S. salsa-Phragmites australis*, *S. salsa-Tamarix chinensis* and *S. salsa*.

In the intertidal zone, natural geomorphology and depositing zones are distinct and high, middle and low tidal flats (marshes) develop from the land to the sea. The high tidal flat (marsh) is predominated by *S. salsa* (>90 %) and *P. australis* (<10 %), while middle tidal flat (marsh) is predominated by *S. salsa* (>95 %) and *T. chinensis* (<5 %). Low tidal flat (marsh) includes

two distinct ecosystem-types. One is pure *S. salsa* community (100 %), with sparse distribution in tidal flat, and the other is bare flat (Fig. 1). The coverage and maximum aboveground biomass of *S. salsa-P. australis*, *S. salsa-T. chinensis* and *S. salsa* communities are 95 %, 80 %, 60 % and 902.08 ± 195.81 , 564.89 ± 99.66 , $252.97 \pm 29.24 \text{ g m}^{-2}$, respectively (Mou 2010). In this study, four sampling sites were laid in high *S. salsa* marsh (HSM), middle *S. salsa* marsh (MSM), low *S. salsa* marsh (LSM) and bare flat (BF) on the northern coastal marshes of the Yellow River estuary (Fig. 1). The tide in the intertidal zone of the Yellow River estuary is irregular semidiurnal tide (twice a day) and the mean tidal range is 0.73–1.77 m (Li et al. 1991). The salinity of the floodwater in the intertidal zone is greatly affected by the runoff of Yellow River and the values throughout a year average between 2.2 % and 3.1 % (Tian et al. 2005).

Experimental design

Fluxes of CH_4 were measured by using opaque, static, manual stainless steel chambers and gas chromatography techniques. The chamber is an open-bottom a square box ($50 \text{ cm} \times 50 \text{ cm} \times 50 \text{ cm}$) and equipped with an electric fan installed on the top wall of each chamber to make turbulence when chamber was closed. Outside of the chamber was covered with 2 cm thickness white foam to reduce the impact of direct radiative heating during sampling. In addition, a thermometer sensor was installed to measure temperature, a three-way stopcocks was fixed to collect gas sample, and a balance pipe was used to equalize the air pressure between the inside and the outside of the chamber. In August 2010, the stainless steel base ($50 \text{ cm} \times 50 \text{ cm} \times 20 \text{ cm}$) with a water groove on top was installed at the four sampling sites. During observations, the chamber was placed over the base filled with water in the groove to ensure air-tightness and the plant was covered within the chamber.

Sampling campaigns were undertaken in autumn (September, October and November in 2010), winter (December in 2010), spring (April and May in 2011) and summer (June and July in 2011). Each measurement campaign consisted of 12 chambers set up at four sites (3 chambers per position): HSM and MSM were located near the shore and which were submerged only at high or middle tide, and LSM and BF were adjacent to the sea and which were frequently submerged at

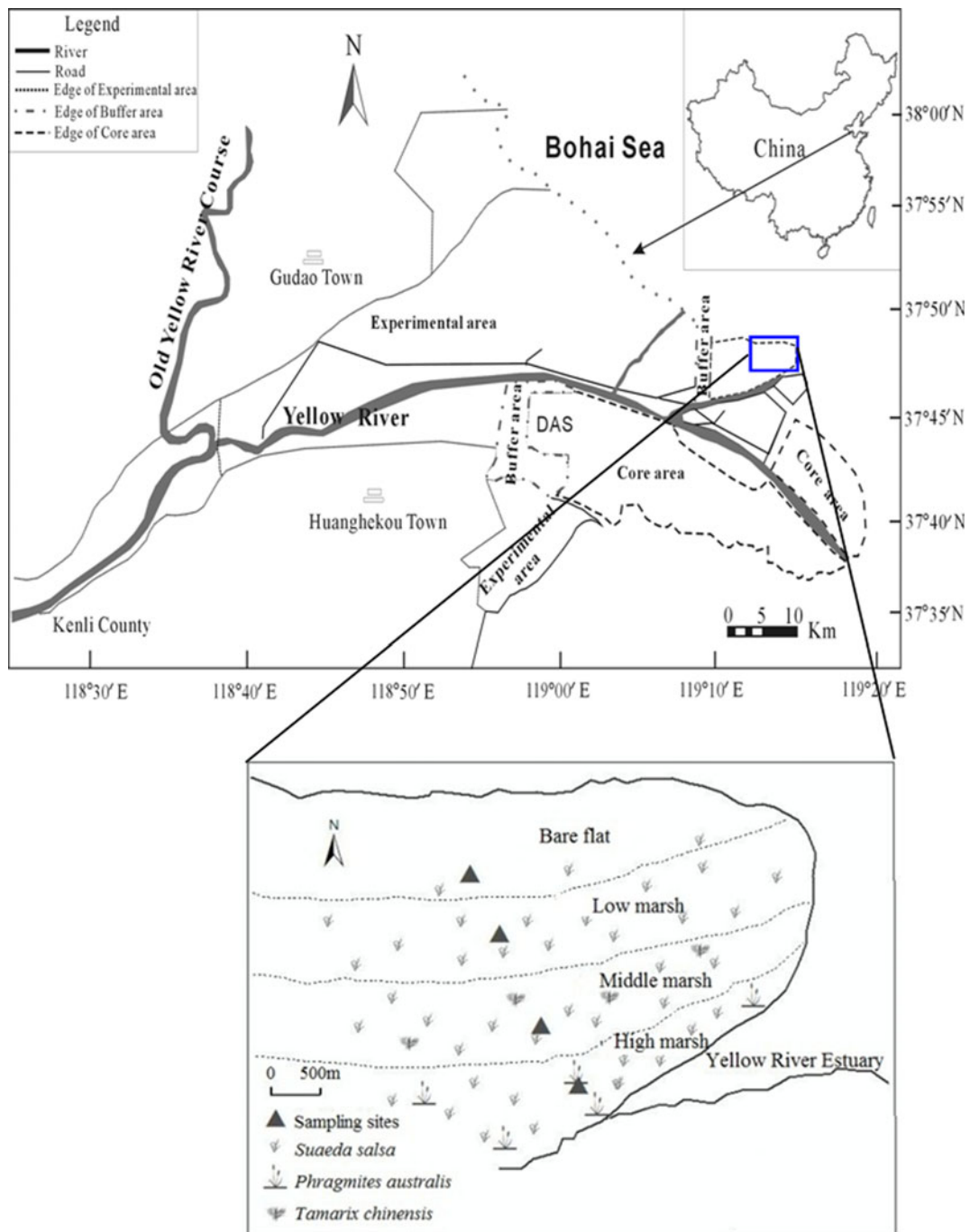


Fig. 1 Sketch of the Yellow River estuary and sampling sites

low tide. Since the tide in the Yellow River estuary is irregular semidiurnal tide, the sampling campaigns in the LSM and BF were sometimes affected by tidal inundation. In this study, the sampling campaigns in the LSM and BF in May and the BF in June were not

carried out due to the influence of tidal inundation. On each sampling date, measurements were conducted at 7:00, 9:30, 12:00, 14:30 and 17:00 h. Measurements representing different times of day were from sediments exposed during low tide, over a number of days.

Four air samples inside the chamber were collected every 20 min over a 60 min period by using 100 ml syringe equipped with three-way stopcocks. Samples were injected into pre-evacuated packs, transported back to the laboratory and analyzed within 36 h using gas chromatography (Agilent 7890A) equipped with an FID. The CH₄ was separated from the other gases with a 2 m stainless-steel column, with an inner diameter of 2 mm 13XMS column (60/80 mesh). The oven, injector and detector temperatures were 80 °C, 200 °C and 200 °C, respectively. The flow rate of the carrier gas (N₂) was 30 mlmin⁻¹. Flame gases (H₂ and O₂) were set at 20 and 30 mlmin⁻¹. Gas concentrations were quantified by comparing peak areas of samples against standards run every 8 samples, ensuring each sample run maintained RSD below 6 %. CH₄ fluxes were calculated from the linear changes in the chamber concentration over time with an average chamber temperature (Song et al. 2006). Only samples with a regression determination coefficient R^2 greater than 0.95 were used for analysis (2.5 % measurements were rejected).

Environmental measurements

Air temperature and soil temperatures (0, 5, 10 and 15 cm) were measured in each position during gas sampling. Soil volumetric moisture and electrical conductivity (EC) in 0–5, 5–10 and 10–15 cm depths were determined *in situ* by high-precision moisture measuring instrument (AZS-2) and soil & solution EC meter (Field Scout), respectively. Soil moisture and EC were not determined in December 2010 since the topsoil (0–10 cm) was frozen. On each sampling date, two soil samples (0–10, 10–20 cm) per position were taken for analyzing total carbon (TC) and total nitrogen (TN) contents by element analyzer (Elementar Vario Micro, German) and ammonium (NH₄⁺ – N) and nitrate (NO₃⁻ – N) contents by sequence flow analyzer (San⁺⁺ SKALAR, Netherlands). The total sulfur (TS) and sulfate (SO₄²⁻) data in the coastal marshes of the Yellow River estuary were cited from Sun et al. (2009) and Fan et al. (2010).

Statistical analysis

The results were presented as means of the replications, with standard error (S.E). Statistical significance of differences at $p < 0.05$ between samples were

analyzed using analysis of variance (ANOVA). Multiple comparison of samples was undertaken by Tukey's test with a significance level of $p = 0.05$. Correlation analyses and stepwise linear regression analyses were used to examine the relationship between fluxes and the measured environmental variables. In all tests, differences were considered significantly only if $p < 0.05$.

Results

Spatial variation of CH₄ fluxes

Variation of CH₄ fluxes in spring

CH₄ fluxes in spring averaged between $-0.781 \text{ mgCH}_4 \text{ m}^{-2} \text{ h}^{-1}$ and $0.822 \text{ mgCH}_4 \text{ m}^{-2} \text{ h}^{-1}$, but the values among the four marshes showed no significant difference ($p > 0.05$) (Fig. 2). Similar variations of CH₄ flux in HSM and MSM were observed both in April and May. In April, with the exception of 12:00 sampling, the other sampling periods showed CH₄ consumptions. In May, both HSM and MSM showed consumptions before 9:30 sampling and emissions afterward. CH₄ fluxes from LSM ranged from $-0.067 \text{ mgCH}_4 \text{ m}^{-2} \text{ h}^{-1}$ to $0.190 \text{ mgCH}_4 \text{ m}^{-2} \text{ h}^{-1}$ and a significant peak occurred in 14:30. The BF was found to release CH₄ with the exception of 7:00 sampling, with the maximum occurred in 17:00. The mean CH₄ fluxes from HSM, MSM, LSM and BF in spring were -0.056 , 0.049 , 0.034 and $0.027 \text{ mgCH}_4 \text{ m}^{-2} \text{ h}^{-1}$, respectively, indicating that coastal marshes represented weak emission except HSM.

Variation of CH₄ fluxes in summer

CH₄ fluxes in summer ranged from $-1.128 \text{ mgCH}_4 \text{ m}^{-2} \text{ h}^{-1}$ to $0.380 \text{ mgCH}_4 \text{ m}^{-2} \text{ h}^{-1}$, but the values among the four marshes were not significantly different ($p > 0.05$) (Fig. 2). Similar variations of CH₄ flux in June and July were observed in HSM except for 17:00 sampling. CH₄ fluxes from MSM in June and July were -1.128 – 0.261 and -0.244 – $0.261 \text{ mgCH}_4 \text{ m}^{-2} \text{ h}^{-1}$, respectively, and they had no significant difference ($p > 0.05$). CH₄ fluxes from LSM in June and July were in opposite except for 14:30 sampling and the ranges were -0.762 – 0.328 and -0.239 – $0.218 \text{ mgCH}_4 \text{ m}^{-2} \text{ h}^{-1}$, respectively. With the exception of July 14:30 sampling, the BF was found to

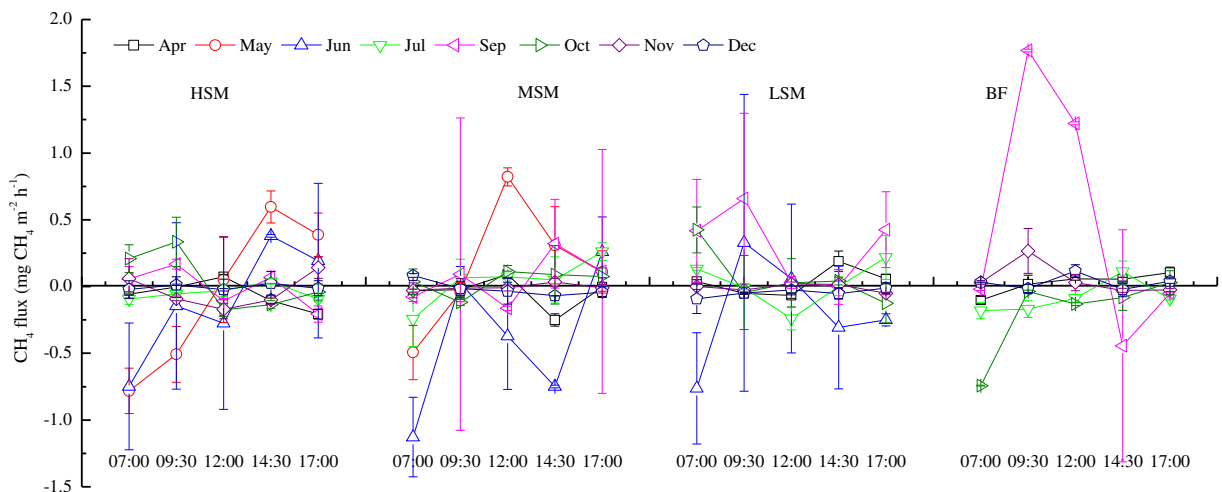


Fig. 2 Variations of CH_4 fluxes ($\text{mgCH}_4 \text{ m}^{-2} \text{ h}^{-1}$) from high *Suaeda salsa* marsh (HSM), middle *S. salsa* marsh (MSM), low *S. salsa* marsh (LSM) and bare flat (BF) in spring (April and

May), summer (June and July), autumn (September, October and November) and winter (December)

consume CH_4 in other periods. The mean CH_4 fluxes from HSM, MSM, LSM and BF in summer were -0.083 , -0.175 , -0.083 and $-0.083 \text{ mgCH}_4 \text{ m}^{-2} \text{ h}^{-1}$, respectively, indicating that coastal marshes acted as a CH_4 sink.

Variation of CH_4 fluxes in autumn

Although CH_4 fluxes in autumn covered a wide range, from $-0.742 \text{ mgCH}_4 \text{ m}^{-2} \text{ h}^{-1}$ to $1.767 \text{ mgCH}_4 \text{ m}^{-2} \text{ h}^{-1}$, the values among the four marshes were not significantly different ($p > 0.05$) (Fig. 2). Over all sampling periods, the variations of positive and negative CH_4 fluxes were observed irregularly in each coastal marsh and the maximum and minimum occurred in BF. CH_4 fluxes from each coastal marsh showed no significant difference among September, October and November ($p > 0.05$). The ranges of CH_4 flux in HSM, MSM, LSM and BF in autumn were $-0.176 \sim 0.335$, $-0.162 \sim 0.321$, $-0.125 \sim 0.660$ and $-0.742 \sim 1.767 \text{ mgCH}_4 \text{ m}^{-2} \text{ h}^{-1}$, and the means were 0.001 , 0.030 , 0.121 and $0.119 \text{ mgCH}_4 \text{ m}^{-2} \text{ h}^{-1}$, respectively, indicating that coastal marshes represented weak CH_4 emission.

Variation of CH_4 fluxes in winter

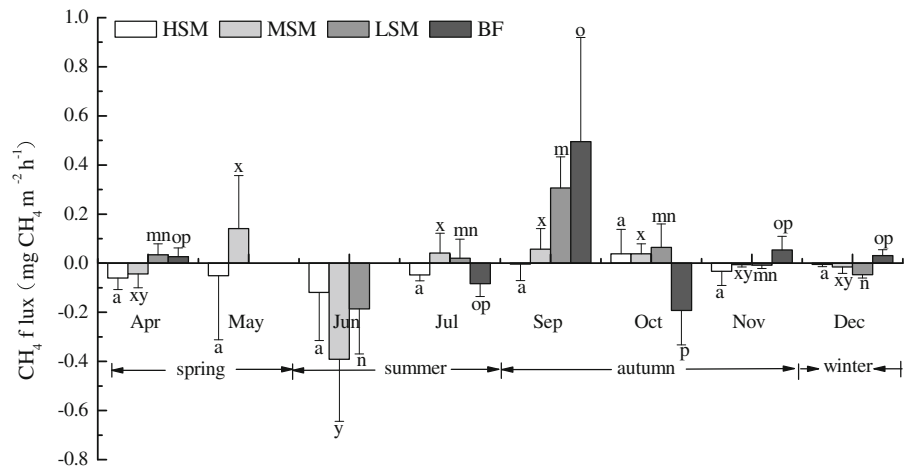
CH_4 fluxes in winter covered a range of $-0.092 \text{ mgCH}_4 \text{ m}^{-2} \text{ h}^{-1}$ to $0.117 \text{ mgCH}_4 \text{ m}^{-2} \text{ h}^{-1}$, but the values among the four marshes showed no significant difference ($p > 0.05$) (Fig. 2). With the exception of LSM that showed CH_4 consumption during all

times of day sampled, the other marshes were found to release CH_4 in some sampling periods. Although the variations of CH_4 flux in HSM and BF were opposite, both positive and negative values alternated regularly. With the exception of 7:00 sampling, the MSM was found to consume CH_4 in other sampling periods. The mean CH_4 fluxes from HSM, MSM, LSM and BF in winter were -0.004 , -0.016 , -0.046 and $0.031 \text{ mgCH}_4 \text{ m}^{-2} \text{ h}^{-1}$, respectively, indicating that coastal marshes acted as weak CH_4 sink except BF.

Temporal variation of CH_4 fluxes

CH_4 fluxes from different coastal marshes varied throughout different times of the day and significant differences were observed in some sampling periods ($p < 0.05$) (Fig. 2). Although the temporal variations of CH_4 fluxes were not significant in each coastal marsh ($p > 0.05$), significant differences still could be observed among different months ($p < 0.05$) (Fig. 3). Over all seasons, CH_4 fluxes averaged between $-0.392 \text{ mgCH}_4 \text{ m}^{-2} \text{ h}^{-1}$ and $0.495 \text{ mgCH}_4 \text{ m}^{-2} \text{ h}^{-1}$, with the maximum and minimum occurred in September (in BF) and June (in MSM), respectively. CH_4 emissions occurred during spring ($0.008 \text{ mgCH}_4 \text{ m}^{-2} \text{ h}^{-1}$) and autumn ($0.068 \text{ mgCH}_4 \text{ m}^{-2} \text{ h}^{-1}$) while CH_4 sink were observed during summer ($-0.110 \text{ mgCH}_4 \text{ m}^{-2} \text{ h}^{-1}$) and winter ($-0.009 \text{ mgCH}_4 \text{ m}^{-2} \text{ h}^{-1}$) (Fig. 3). Over all sampling periods, CH_4 fluxes from the four

Fig. 3 Temporal variations of CH_4 fluxes ($\text{mgCH}_4 \text{ m}^{-2} \text{ h}^{-1}$) from high *S. salsa* marsh (HSM), middle *S. salsa* marsh (MSM), low *S. salsa* marsh (LSM) and bare flat (BF). Bars with different letters (a, b for HSM; x, y for MSM; m, n for LSM; o, p for BF) are significantly different at the level of $p < 0.05$; bars with same letters are not significantly different at the level of $p < 0.05$



marshes were not significantly different ($p > 0.05$). CH_4 emissions occurred in LSM ($0.026 \text{ mgCH}_4 \text{ m}^{-2} \text{ h}^{-1}$) and BF ($0.055 \text{ mgCH}_4 \text{ m}^{-2} \text{ h}^{-1}$) while CH_4 sink were observed in HSM ($-0.035 \text{ mgCH}_4 \text{ m}^{-2} \text{ h}^{-1}$) and MSM ($-0.022 \text{ mgCH}_4 \text{ m}^{-2} \text{ h}^{-1}$). The annual average CH_4 flux from the intertidal zone was $0.002 \text{ mgCH}_4 \text{ m}^{-2} \text{ h}^{-1}$, indicating that coastal marsh acted as a weak source.

Environmental variables in coastal marsh

Similar variations of air temperature and ground temperature (0, 5, 10 and 15 cm) in the four marshes were observed over all sampling periods (Fig. 4a). Air temperature did not show significant difference among the four marshes ($p > 0.05$). Ground temperatures generally decreased with increasing soil depth, but no significant differences were found within the four marshes ($p > 0.05$). Significant correlations between CH_4 fluxes and temperatures were observed in different marshes during spring or summer ($p < 0.05$ or $p < 0.01$). By comparison, most correlations between CH_4 fluxes and temperatures were not significant during autumn or winter ($p > 0.05$) (Table 1). Dissimilar variations of soil moisture and EC (0–5 and 5–10 cm) in the four marshes were observed over all sampling periods (Fig. 4b,c). With increasing depth, soil moisture increased (Fig. 4b), while EC generally decreased (Fig. 4c). Soil moisture did not show significant differences among the four marshes ($p > 0.05$), while significant differences of EC were observed ($p < 0.05$). Although both positive and negative influences of soil moisture and EC on CH_4 emissions were observed within the four marshes, only the correlations between soil moisture (5–10, 10–15 cm) and CH_4 fluxes in

HSM ($p < 0.01$) and between EC (0–5 cm) and CH_4 fluxes in LSM ($p < 0.05$) were significant (Table 2).

Seasonal dynamics of sediment substrate in the four marshes were observed over all sampling period (Fig. 5). TC, TN and $\text{NH}_4^+\text{-N}$ in the surface (0–10 cm) and subsurface sediment (10–20 cm) of BF were generally higher than those in other marshes (Fig. 5a, b, c). Both TC in surface and subsurface sediment had significant differences among the four marshes ($p < 0.05$), while only TN in subsurface sediment showed significant difference ($p < 0.01$). Both $\text{NH}_4^+\text{-N}$ and $\text{NO}_3^-\text{-N}$ in sediment (0–10, 10–20 cm) were not significantly different within the four marshes ($p > 0.05$). Lacks of correlations between CH_4 fluxes and substrate variables were observed ($p > 0.05$) except the correlations occurred in subsurface sediment of HSM ($p < 0.01$) (Table 3). Moreover, the correlations between environmental variables determined during all times of day and CH_4 diurnal emissions were not significant ($p > 0.05$).

Discussion

Temporal variations of CH_4 fluxes

The magnitudes of CH_4 fluxes from coastal marshes in the Yellow River estuary were in the range of $-0.392 \text{ mgCH}_4 \text{ m}^{-2} \text{ h}^{-1}$ to $0.495 \text{ mgCH}_4 \text{ m}^{-2} \text{ h}^{-1}$, which were generally lower than those from salt marshes in the Yangtze River estuary and the Min River estuary, and mangrove swamps in the Brisbane River and Puerto Rico, but approximated emissions

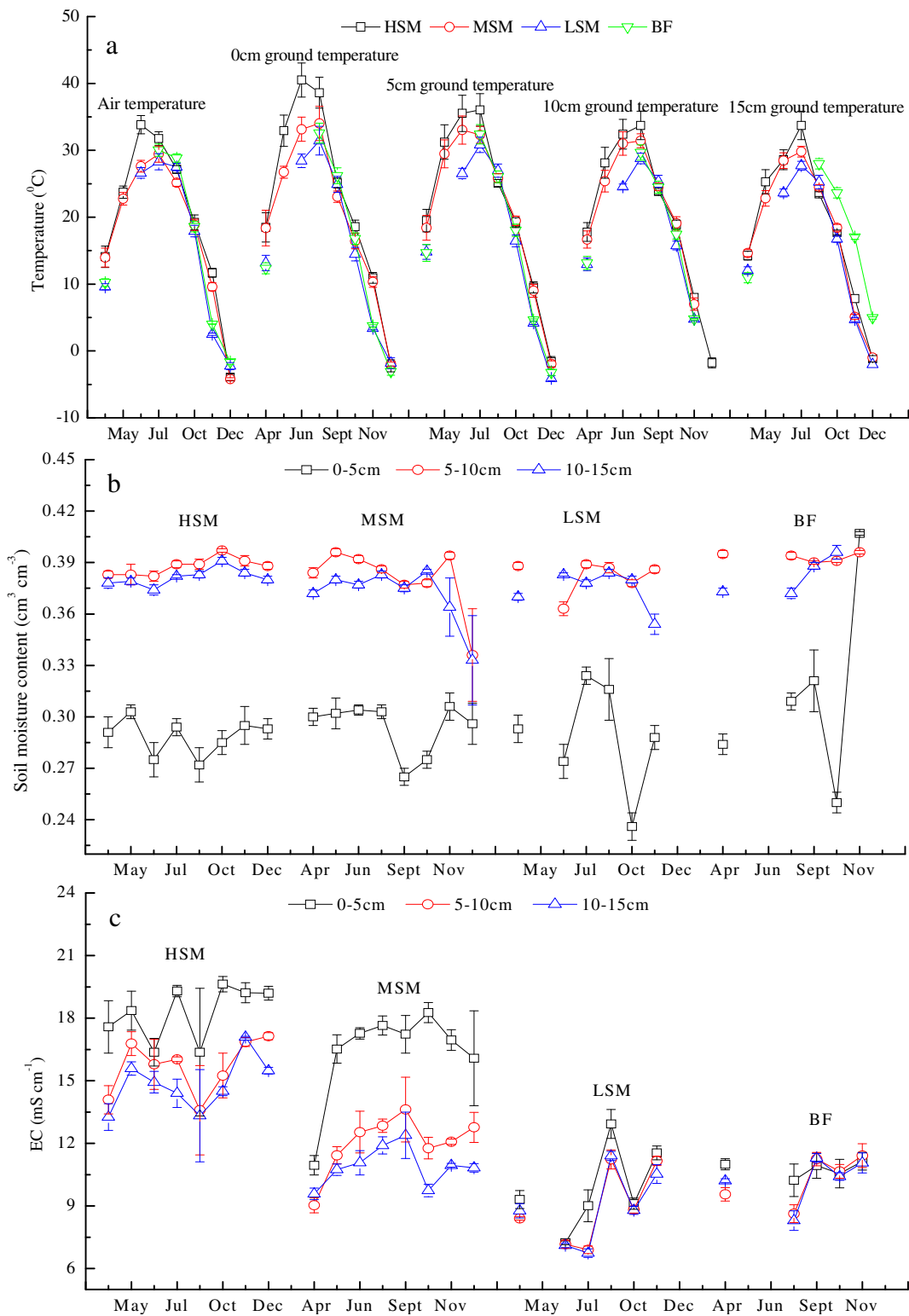


Fig. 4 Variations of environmental temperatures (a), soil moisture content (b) and electrical conductivity (EC) (c) in high *S. salsa* marsh (HSM), middle *S. salsa* marsh (MSM), low *S. salsa* marsh (LSM) and bare flat (BF)

Table 1 Pearson correlation analysis between CH₄ fluxes and temperatures

Month	Air temperature						0 cm ground temperature						5 cm ground temperature						10 cm ground temperature						15 cm ground temperature					
	HSM	MSM	LSM	BF	HSM	MSM	LSM	BF	HSM	MSM	LSM	BF	HSM	MSM	LSM	BF	HSM	MSM	LSM	BF	HSM	MSM	LSM	BF	HSM	MSM	LSM	BF		
April	0.734	-0.012	0.283	0.455	0.421	-0.177	0.382	0.585	-0.049	-0.374	0.538	0.874	-0.454	-0.544	0.371	0.917*	-0.570	-0.456	0.365	0.922*										
May	0.485	0.938*	-	-	0.647	0.849	-	-	0.945*	0.857	-	-	0.987*	0.781	-	-	0.981*	0.544	-	-										
June	0.885*	0.497	0.339	-	0.697	0.249	0.691	-	0.886*	0.396	0.445	-	0.891*	0.495	0.366	-	0.861	0.617	0.168	-										
July	0.769	-0.320	-0.811	0.811	0.831	-0.167	-0.813	0.822	0.597	0.311	-0.954*	0.766	0.448	0.617	-0.632	0.738	0.448	0.789	-0.331	0.668										
September	0.258	0.517	0.274	0.787	0.288	0.463	-0.087	0.873	-0.264	0.563	-0.153	0.527	-0.502	0.846*	-0.058	0.329	-0.606	0.788*	0.068	0.050										
October	0.306	-0.230	-0.081	0.577	-0.116	-0.061	-0.329	0.604	-0.591	0.353	-0.108	0.570	-0.658	0.571	-0.256	0.512	-0.665	0.567	-0.355	-0.035										
November	-0.711	0.806	-0.047	0.485	-0.822	0.546	0.676	0.720	-0.311	0.720	0.348	-0.385	-0.345	0.690	-0.074	-0.254	-0.240	0.680	-0.074	0.650										
December	0.898*	-0.510	0.555	-0.147	0.639	-0.162	0.717	-0.065	0.325	-0.883*	0.628	-0.147	0.249	-	-	-	0.179	-	-	-										

Values with bold and asterisk symbol indicate that correlations are significant at the 0.05 level (one asterisk, *). HSM, High *Suaeda salsa* marsh; MSM, Middle *S. salsa* marsh; LSM, Low *S. salsa* marsh; BF, Bare flat. Pair sample size, n=8 for air temperature and ground temperatures (0, 5, 10 and 15 cm) in HSM; n=8 for air temperature and 0, 5 and 15 cm ground temperatures, and n=7 for 10 cm ground temperature in MSM; n=7 for air temperature and ground temperatures (0, 5, 10 and 15 cm) in LSM; n=6 for air temperature and 0 and 5 cm ground temperatures, and n=5 for 10 and 15 cm ground temperatures in BF

recorded at the Macrotidal Salt Marsh Bay, the sand shore of Lake Nakaumi, the intertidal zone of Maritime Antarctica, and the mangrove swamps in the Moreton Bay and the Dong zhai Harbor (Table 4). Differently, CH₄ emissions from the salt marshes (*Carex rugulosa*, *P. australis*, *Solidago altissima*) of Lake Nakaumi under light condition recorded by Hirota et al. (2007) were 184~495 times greater than the maximum CH₄ emission reported by our study, which was probably owing to large aerobic respiration by plant and microbes with continuous anaerobic condition. Existing abundant both live and dead aboveground biomass in salt marsh were important supporting evidences (Hirota et al. 2007).

Seasonal variations in CH₄ emissions from coastal marshes were observed in this study (Fig. 3) and also reported by others (Inubushi et al. 2003; Allen et al. 2007; Zhu et al. 2008). Whalen (2005) observed that seasonal patterns of trace gas emission were influenced by latitude, with arctic, austral and some temperate regions characterized by pronounced CH₄ emission (Gregorich et al. 2006; Allen et al. 2007), which are governed by seasonal variability in temperatures affecting water availability, production of substrate precursors and microbial activity. However, CH₄ emissions from the coastal marshes in the Yellow River estuary seemed not to be affected by seasonal variability in temperatures though the estuary located in temperate region (37°35'N~38°12'N). CH₄ emissions generally occurred during spring and autumn while CH₄ sink were observed during summer and winter. We considered that the seasonal variations in CH₄ emissions observed in this paper were probably related to the complex interactions of temperatures and other biotic/abiotic factors, such as water and salinity status (Whalen 2005; Tong et al. 2009), plants (Tong et al. 2009) and sediment substrate (Allen et al. 2007). Although significant positive/negative correlations between CH₄ fluxes and temperatures were observed in some periods (Table 1), the effects of seasonal variability in temperatures on CH₄ emissions, to a great extent, might be covered by above biotic/abiotic parameters.

We found that CH₄ emissions occurred during spring and autumn while CH₄ sink were observed during summer and winter. Because the environmental variables determined in coastal marshes were all excluded in the stepwise liner regression, we considered that CH₄ fluxes in different seasons were controlled by

Table 2 Pearson correlation analysis between CH₄ fluxes and soil moisture or electrical conductivity (EC)

Sites	Soil moisture			Electrical conductivity (EC)		
	0–5 cm	5–10 cm	10–15 cm	0–5 cm	5–10 cm	10–15 cm
HSM	-0.029	0.855**	0.910**	0.542	-0.109	-0.073
MSM	-0.304	-0.075	0.002	0.007	-0.003	0.128
LSM	0.356	0.601	0.226	0.830*	0.642	0.722
BF	0.537	-0.457	0.155	0.492	0.503	0.594

Values with bold and asterisk symbol indicate that correlations are significant at the 0.05 level (one asterisk, *) or at the 0.01 level (two asterisks, **). HSM, High *S. salsa* marsh; MSM, Middle *S. salsa* marsh; LSM, Low *S. salsa* marsh; BF, Bare flat

Pair sample size, $n=8$ for soil moisture and EC in 0–5, 5–10 and 10–15 cm depths in HSM and MSM; $n=6$ for soil moisture and EC in 0–5, 5–10 and 10–15 cm depths in LSM; $n=5$ for soil moisture and EC in different depths in BF except the soil moisture in 10–15 cm depth ($n=4$)

the interactions of multiple factors. Among them, the variations of limited C and mineral N in the sediments might have significant influences on CH₄ emissions. As was shown in Fig. 5, TC, TN and NH₄⁺-N in different coastal marsh sediments (0–10, 10–20 cm) were higher in spring and autumn (especially in autumn), while NO₃⁻ - N were generally higher in summer and winter, indicating that the increase of C and N (especially NH₄⁺ - N) during spring and autumn might promote CH₄ production and inhibit CH₄ uptake, while the increase of NO₃⁻ - N during summer and winter was unfavorable for CH₄ production. Similar results were drawn by Mancinelli (1995) and Bodelier and Laanbroek (2004). For one thing, the increase of C and N (NH₄⁺-N) improved the limited status of C and N in sediment and provided enough C and N sources for the growth of methanogens, which was favorable for the production of CH₄. For another, the NH₄⁺ - N addition could inhibit the uptake of CH₄, which was related to two underlying mechanisms: *i*) NH₄⁺ is a competitive inhibitor of CH₄ oxidation due to lack of specificity of methane monooxygenase (MMO) in methanotroph (Saari et al. 2004); *ii*) hydroxylamine and nitrite produced by methanotrophic ammonia oxidation are toxic to methanotrophic bacteria (Jiang et al. 2010). The inhibition of NO₃⁻ - N on CH₄ production has been reported by many studies (Banik et al. 1996; Kluber and Conrad 1998; Chidthaisong and Conrad 2000), which could be partly applied to explain the formation of CH₄ sink during summer and winter.

This study also showed that the coastal marsh in the Yellow River estuary represented weak CH₄ emission (0.002 mgCH₄ m⁻²h⁻¹) throughout the year and there

were three probable causes. Firstly, although the high soil moisture in sediment (Fig. 4b) was favorable for CH₄ production, the interaction of moisture and salinity might reduce CH₄ emissions. Magenheimer et al. (1996) showed that the CH₄ fluxes from a macrotidal salt marsh (Bay of Fundy) were inversely correlated ($r^2=0.23$, $p=0.001$) with salinity of the upper porewater at the sampling site. Chidthaisong and Conrad (2000) indicated that high salinity could inhibit the activities of methanogens or did harm to methanogens which reduced CH₄ emission. In this paper, the salinity (represented by EC) of the coastal marshes in the Yellow River estuary were high (0–5 cm, 14.44±3.96 mS cm⁻¹; 5–10 cm, 12.10±2.96 mS cm⁻¹; 10–15 cm, 11.50±2.63 mS cm⁻¹) (Fig. 4c), which might reduce CH₄ production. Secondly, the SO₄²⁻ concentrations in sediment substrate resulting from high SO₄²⁻ content in seawater (Ivanou 1992) were also considered dominant factor controlling CH₄ emissions from coastal marsh (Kreuzwieser et al. 2003). Because sulfate reducing bacteria (SRB) compete for H₂/CO₂ and ethylic acid (CH₃COOH) with methanogen and the former had more strong affinity to the reaction substrates, the coexistence of SRB and SO₄²⁻ would inhibit the production and emission of CH₄ (van der Gon et al. 2001). In this study, the TS and SO₄²⁻ contents in the coastal marshes of the Yellow River estuary were 3.0~6.2 % (Sun et al. 2009) and 0.62~1.50 % (Fan et al. 2010), respectively, indicating that the high S content and the anoxic condition in sediment could enhance the dissimilatory reduction of SO₄²⁻, which inhibited the CH₄ emission from *S. salsa* marsh. Thirdly, the microbially mediated AOM (anaerobic oxidation of methane) process occurring

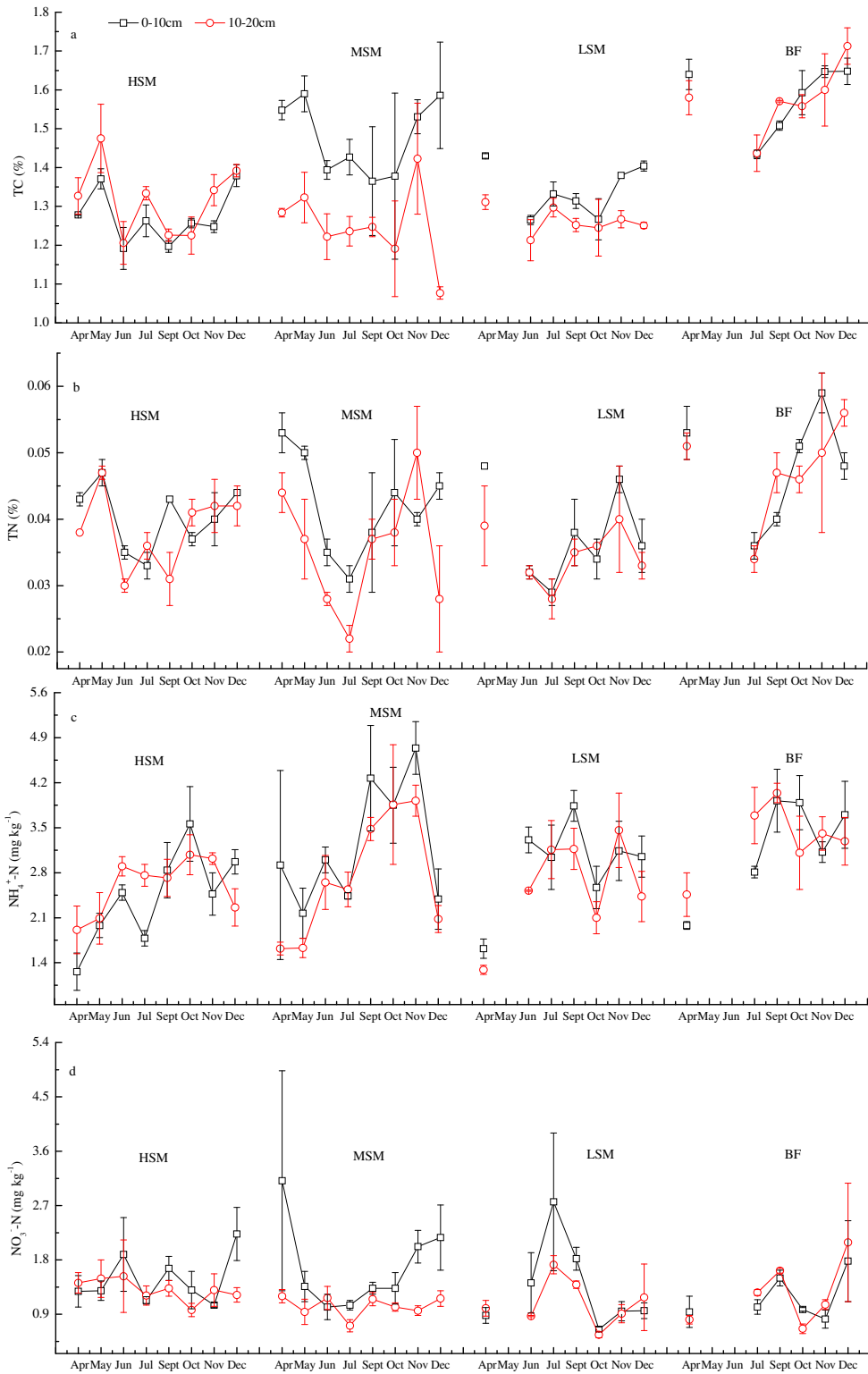


Fig. 5 Variations of TC (a), TN (b), $\text{NH}_4^+ - \text{N}$ (c) and $\text{NO}_3^- - \text{N}$ (d) contents in high *S. salsa* marsh (HSM), middle *S. salsa* marsh (MSM), low *S. salsa* marsh (LSM) and bare flat (BF) sediments

Table 3 Pearson correlation analysis between CH₄ fluxes and soil substrate

Sites	TC		TN		NH ₄ ⁺ -N		NO ₃ ⁻ -N	
	0–10 cm	10–20 cm	0–10 cm	10–20 cm	0–10 cm	10–20 cm	0–10 cm	10–20 cm
HSM	0.070	-0.235	-0.063	0.310	0.693	0.355	-0.134	-0.892**
MSM	0.305	0.187	0.308	0.231	0.006	-0.026	0.109	-0.422
LSM	-0.031	0.194	0.153	0.171	0.286	0.219	0.200	0.322
BF	-0.168	0.177	-0.265	0.203	0.295	0.543	0.451	0.450

Value with bold and asterisk symbol indicates that correlation is significant at the 0.01 level (two asterisks, **); HSM, High *S. salsa* marsh; MSM, Middle *S. salsa* marsh; LSM, Low *S. salsa* marsh; BF, Bare flat. Pair sample size, $n=8$ for TC, TN, NH₄-N and NO₃⁻-N in 0–10 and 10–20 depths in HSM and MSM; $n=7$ for TC, TN, NH₄-N and NO₃⁻-N in 0–10 and 10–20 depths in LSM; $n=6$ for TC, TN, NH₄-N and NO₃⁻-N in 0–10 and 10–20 depths in BF

mainly in anoxic sediments also has crucial effects on CH₄ emission (Raghoebarsing et al. 2006). AOM coupled to SO₄²⁻ reduction is performed by a consortium of anaerobic methanotrophic archaea (ANME) and SRB (Strous and Jetten 2004; Mileto et al. 2008; Knittel and Boetius 2009). During AOM, CH₄ is oxidized with SO₄²⁻ as the terminal electron acceptor (CH₄ + SO₄²⁻ → HCO₃⁻ + HS⁻ + H₂O) (Knittel and Boetius 2009). As mentioned above, the high S content in sediment might enhance the AOM process under anoxic condition, which further inhibited the CH₄ emission from *S. salsa* marsh.

We also demonstrated that CH₄ emissions from coastal marshes varied throughout different times of the day and significant differences were observed in some sampling periods ($p<0.05$) (Fig. 2). Diurnal variations in trace gas flux have been reported in other coastal marsh studies. Hirota et al. (2007) indicated that the diurnal variation of CH₄ fluxes from salt marsh in dark condition was influenced by soil temperature ($r=0.47$, $p<0.05$). Lu et al. (1999) showed that CH₄ fluxes from *Bruguiera sexangula* mangrove swamp sediment had large diurnal fluctuation, which was caused by the changes of tidal inundation rather than the changes of air or sediment temperatures. Since environmental factors are involved directly with different microbe and plant activities and are changeable in relative short term (diurnal) scale, the diurnal variations of CH₄ emission from different coastal marshes are affected by different principal factors. But this study showed that any environmental variables determined during all times of day had no significant correlations with CH₄ diurnal emissions ($p>0.05$). Similar result was drawn by Hirota et al. (2007) who found that any environmental factors

seemed to not affect diurnal variation of CH₄ flux under light condition. Because our study did not measure CH₄ fluxes and environmental variables over the complete tidal inundation cycles, the variations of some major factors during all times of night might be missed, which probably covered the main factors. Besides, the variations of environmental variables in the coastal marshes of the Yellow River estuary might be more complicated than those in other coastal marshes, which caused the CH₄ diurnal emissions to be affected by two or more factors. However, these explanations require to be verified in the following study.

Spatial variations of CH₄ fluxes

Over all sampling periods, we found that the physical (temperature, soil moisture and EC) and chemical (TC, TN, NH₄-N and NO₃-N) parameters of sediment differed in their magnitude among the four marshes. Significant differences in TC, TN and EC in sediment were observed ($p<0.05$). Such differences among the four marshes would be due to the site-specific conditions such as topography, slope, hydrology and species composition which determine the magnitudes and variations of CH₄ at spatial scale (Allen et al. 2007; Hirota et al. 2007). From the stepwise linear regression analysis, soil moisture (X_1) and EC (X_2) were the dominant factors that controlled the CH₄ emissions (Y) in HSM ($Y=-3.091+0.080X_1$, $R^2=0.773$, $p=0.004$) and LSM ($Y=-0.566+0.060X_2$, $R^2=0.682$, $p=0.043$), respectively, while in MSM and BF, the environmental variables determined during sampling periods were all excluded, indicating that CH₄ fluxes were controlled by multiple factors.

Table 4 Literature data of CH₄ emissions from different marshes

Marsh types	Location	Vegetations	CH ₄ fluxes ^a (mgm ⁻² h ⁻¹)	Observation period	References
Coastal marsh	Yellow River estuary, China	<i>Suaeda salsa</i> , <i>Phragmites australis</i>	-0.035 (-0.392-0.495) ^b	September, October, November and December 2010; April, May, June and July 2011	This study
		<i>S. salsa</i> , <i>Tamarix chinensis</i>	-0.022		
		<i>S. salsa</i> ,	0.026		
		Bare flat	0.055		
	Yangtze River estuary, China	<i>Scirpus mariqueter</i>	2.06	May 2004~April 2005	Yang et al. (2007)
		Bare flat	0.04		
	Minjiang River estuary, China	<i>Cyperus malaccensis</i>	0.60	May 2007~April 2008	Zeng et al. (2010)
	Macrotidal Salt Marsh Bay, Canada	<i>Spartina alterniflora</i> , <i>Plantago maritima</i> ,	0.067 (0.008-0.458) ^b	July~September 1993	Magenheimer et al. (1996)
		<i>Spartina patens</i>			
	Lake Nakaumi (coastal lagoon), Japan	<i>Carex rugulosa</i> , <i>P. australis</i> , <i>Solidago altissima</i>	(91-245) ^b	August 2003	Hirota et al. (2007)
		Sand shore	(0.01-0.34) ^b		
	Fildes Peninsula, Maritime Antarctica	<i>Adenocystis utricularis</i>	0.0445 (0.0016-0.1234) ^b	January~March 2000	Sun et al. (2002)
Mangrove swamp	Changning River estuary, China	<i>Bruguiera gymnorhiza</i> , <i>Bruguiera sexangula</i>	(0.0249-0.0482) ^b	May 1996	Ye et al. (2000)
	Moreton Bay, Queensland, Australia	<i>Avicennia marina</i>	(0.020-0.350) ^b	July and August 1998; September and October 1999	Kreuzwieser et al. (2003)
	Jiulongjiang estuary, China	<i>Kandelia candel</i>	(0.0038-0.0495) ^b	July and September 2002	Alongi et al. (2005)
	Brisbane River, Queensland, Australia	<i>Avocennia marina</i> , <i>Aegiceras corniculatum</i>	(0.003-17.4) ^b	April 2004;	Allen et al. (2007)
				February and July 2005	
	Dongzhai Harbour, China	<i>B. sexangula</i> , <i>B. gymnorhiza</i> , <i>K. candel</i> ,	(-0.0056-0.1870) ^b	May 1996; February, May, August and November 1997	Lu et al. (1999, 2000)
		<i>A. corniculatum</i>			Ye and Lu (2001)
	Puerto Rico	<i>Avicennia germinans</i> , <i>Laguncularia racemosa</i>	0.17	April~July 1991;	Sotomayor et al. (1994)
				September 1991~January 1992	
		<i>Rhizophora mangle</i>	1.75		
		<i>R. mangle</i> , <i>A. germinans</i>	3.42		
		<i>Schoenus nigricans</i> , <i>Molinia caerulea</i>	0.80		
		<i>Rhynchospora alba</i>	0.87		
		<i>Sphagnum cuspidatum</i> , <i>Sphagnum auriculatum</i>	(0.48-2.10) ^b		

^a Means in different observation periods^b Values in bracket are the range of CH₄ fluxes

This study observed a large spatial variation of CH₄ fluxes in the coastal marshes of the Yellow River estuary. The coefficient of variations (CVs) of CH₄ fluxes in the four marshes were 133.70 %, 712.06 %, 562.02 % and 425.49 %, respectively, while the value across the coastal marsh was 7856.25 %, indicating that, to evaluate the regional budget of CH₄ emissions precisely, measurements should be designed at fine scales and the number of spatial replicates should be increased. Although CH₄ fluxes from the four marshes had no significant differences ($p > 0.05$), CH₄ emission patterns were different during all times of day and the seasons measured (Fig. 2). Previous studies have indicated that temperatures had great influence on CH₄ emissions at spatial scale (Sun et al. 2002; Gregorich et al. 2006). Although air temperature and ground temperatures did not show significant difference among the four marshes during all the seasons measured ($p > 0.05$), strong positive/negative correlations still could be found in some sampling periods ($p < 0.05$) (Table 1). This indicated that thermal condition was an important factor affecting CH₄ emission across the coastal marsh, but its function might be covered by the interactions of other biotic or abiotic factors, such as moisture, salinity, sediment substrate and plant. For spatial variations of CH₄ fluxes in coastal marsh, we considered that the differences of vegetation composition were the main driving forces. van den Pol-van Dasselaar et al. (1999) suggested that vegetation was the most important factor in predicting the spatial variability of CH₄ fluxes because it was a comprehensive reflection of environmental conditions such as climate, soil moisture and nutrient status. The importance of vegetation for carbon fluxes in wetlands also has been described in many studies (Joabsson et al. 1999; Hirota et al. 2006). Especially, it has been well known that wetland plants have complex gas transport system via their body and emit CH₄ from soil to the atmosphere (Schimel 1995; Hirota et al. 2004). As mentioned before, because the vegetation composition and biomass in the four marshes were different, the plant distributed continuously across the coastal marsh would be one of the key factors for the CH₄ emissions at spatial scale.

Site-level control of CH₄ emission was also attributed to the effects of soil moisture, salinity and nutrient status. Although soil moisture did not show significant differences among the four marshes ($p > 0.05$), strong positive correlations were observed

between soil moisture (5–10, 10–15 cm) and CH₄ fluxes in HSM ($p < 0.01$) (Table 2). This is common because higher moisture creates a reduction condition that is beneficial for the production and emission of CH₄. In contrast, EC showed significant differences within the four marshes ($p < 0.05$) and significant positive correlation was observed between EC and CH₄ emission in LSM ($p < 0.05$) (Table 2). Generally, both positive and negative impacts of soil moisture and EC on CH₄ emissions were found in coastal marshes (Table 2), which induced the formation of different CH₄ emission patterns at spatial scale. As discussed before, the negative impact of soil moisture or the positive effect of EC on CH₄ emission might be related to the interaction of soil moisture and salinity in sediment. Similar result was drawn by Hirota et al. (2007) who found that, in coastal ecosystems subjected to the fluctuation of water level (or soil moisture) by astronomic tide, there will be both positive and negative impact on CH₄ emissions. Site-level nutrient status also influenced the spatial variations of CH₄ fluxes (Ding et al. 2004). In this study, both positive and negative correlations between CH₄ emission and nutrient status were observed, and significant correlation between CH₄ emission and NO₃-N occurred in HSM ($p < 0.01$) (Table 3). Although lacks of correlations between CH₄ fluxes and substrate variables were observed ($p > 0.05$), local nutrient differences due to topography, aspect, slope, hydrology and vegetation, to some extent, influenced the spatial differences of CH₄ emissions at spatial level.

Our study showed that the coastal marsh acted as a weak CH₄ source in the present N loading of the Yellow River estuary. Numerous studies have demonstrated that N was an important regulatory factor for the production and consumption of CH₄ (Bodelier and Laanbroek 2004; Lebauer and Treseder 2008), and the response of CH₄ flux to N enrichment, both the magnitude and the direction, might vary due to the N addition level and N forms (Liu and Greaver 2009). Jiang et al. (2012) studied the responses of N enrichment (NH₄⁺ - N and NO₃⁻ - N) on CH₄ production and consumption of the different coastal marsh sediments in the Yellow River estuary, and found that NH₄⁺ - N addition caused a general stimulation on CH₄ emission while NO₃⁻ - N addition generally inhibited CH₄ production. Moreover, with increasing NH₄⁺ - N addition, the suppression of CH₄ uptake enhanced (Jiang 2012). Similar results were drawn

by Le Mer and Roger (2001), and Bodelier and Laanbroek (2004). The increase in CH₄ emission under N addition probably was caused by the activities of both methanogenic archaea and methanotropic bacteria (Liu and Greaver 2009). At present, the exogenous N loading (NH₄⁺ – N is dominated) of the Yellow River estuary is increasing due to human activities (State Oceanic Administration of China 2010). Since N is a very limited nutrient in the coastal marshes of the Yellow River estuary (Mou 2010), increases in exogenous N loading to estuarine and coastal marshes will stimulate microbial processes and CH₄ emission. In addition, increases in exogenous NH₄⁺ – N loading will also enhance the suppression of CH₄ uptake (Jiang 2012) and the multiple mechanisms have been declared as mentioned before. Based on the above analysis, we can conclude that the CH₄ emission in the future will be enhanced with increasing N loading to the Yellow River estuary (especially NH₄⁺ – N is the major pollutant), and the magnitude of CH₄ emission should be paid more attentions as the annual CH₄ inventory was assessed accurately.

Conclusions

This paper studied the seasonal and spatial variations of CH₄ emissions and associated environmental factors in the coastal marshes (HSM, MSM, LSM and BF) of the Yellow River estuary. Results have demonstrated that: *i*) CH₄ fluxes averaged between $-0.392 \text{ mgCH}_4 \cdot \text{m}^{-2} \cdot \text{h}^{-1}$ and $0.495 \text{ mgCH}_4 \cdot \text{m}^{-2} \cdot \text{h}^{-1}$, and emissions occurred during spring and autumn while sinks were observed during summer and winter. CH₄ emissions occurred in LSM and BF while sinks were observed in HSM and MSM. The annual average CH₄ flux from the intertidal zone was $0.002 \text{ mgCH}_4 \cdot \text{m}^{-2} \cdot \text{h}^{-1}$ and coastal marsh acted as a weak CH₄ source; *ii*) Temporal variations of CH₄ emission were related to the interactions of abiotic factors (temperatures, soil moisture and salinity) and the variations of limited C and mineral N in sediments, while spatial variations were mainly affected by the vegetation composition and biomass at spatial scale; *iii*) Both the large spatial variation of CH₄ fluxes across the coastal marsh (CV=7856.25 %) and the potential effect of exogenous N loading to the Yellow River estuary on CH₄ emission should be considered before the annual CH₄ inventory was evaluated accurately.

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