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Fluxes of nitrous oxide and methane in different coastal *Suaeda salsa* marshes of the Yellow River estuary, China

Zhigao Sun [a,*], Lingling Wang [a], Hanqin Tian [b], Huanhuan Jiang [a], Xiaojie Mou [a], Wanlong Sun [a]

[a] *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, PR China*

[b] *International Center for Climate and Global Change Research, School of Forestry and Wildlife Sciences, Auburn University, Auburn, AL 36849, USA*

HIGHLIGHTS

- ▶ Fluxes of N_2O and CH_4 in coastal marsh had different spatial and temporal variations.
- ▶ Coastal marsh represented N_2O emission and CH_4 sink during sampling campaigns.
- ▶ N_2O comprised the principal part of total CO_2-e emissions during spring and winter.
- ▶ Contributions of CH_4 to total CO_2-e could not be ignored during summer and autumn.

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ABSTRACT

The spatial and temporal variations of the fluxes of nitrous oxide (N_2O) and methane (CH_4) and associated abiotic sediment parameters were quantified for the first time across the coastal marsh dominated by *Suaeda salsa* in the Yellow River estuary during 2009/2010. During all times of day and the seasons measured, N_2O and CH_4 fluxes from coastal marsh ranged from -0.0147 mgN_2O m^{-2} h^{-1} to 0.0982 mgN_2O m^{-2} h^{-1} and -0.7421 mgCH_4 m^{-2} h^{-1} to 0.4242 mgCH_4 m^{-2} h^{-1}, respectively. The mean N_2O fluxes in spring, summer, autumn and winter were 0.0325 mgN_2O m^{-2} h^{-1}, 0.0089 mgN_2O m^{-2} h^{-1}, 0.0119 mgN_2O m^{-2} h^{-1} and 0.0140 mgN_2O m^{-2} h^{-1}, and the average CH_4 fluxes were -0.0109 mgCH_4 m^{-2} h^{-1}, -0.0174 mgCH_4 m^{-2} h^{-1}, -0.0141 mgCH_4 m^{-2} h^{-1} and -0.0089 mgCH_4 m^{-2} h^{-1}, respectively, indicating that the coastal marsh acted as N_2O source and CH_4 sink. Both N_2O and CH_4 fluxes differed significantly between times of day of sampling. N_2O fluxes differed significantly between sampling seasons as well as between sampling positions, while CH_4 fluxes had no significant differences between seasons or positions. Temporal variations of N_2O emissions were probably related to the effects of vegetation (*S. salsa*) during summer and autumn and the frequent freeze/thaw cycle of sediment during spring and winter, while those of CH_4 fluxes were controlled by the interactions of thermal conditions and other abiotic factors (soil moisture and salinity). Spatial variations of N_2O and CH_4 fluxes were primarily affected by soil moisture fluctuation derived from astronomic tide, sediment substrate and vegetation composition. N_2O and CH_4 fluxes, expressed as CO_2-equivaltent (CO_2-e) emissions, showed that N_2O comprised the principal part of total calculated CO_2-e emissions during spring and winter, while the contributions of CH_4 could not be ignored during summer and autumn. This study highlights the importance of seasonal N_2O and CH_4 contributions, particularly during times of significant CH_4 consumption. For the accurate up-scaling of N_2O and CH_4 fluxes to annual rates, a careful sampling design at site-level is required to capture the potentially considerable temporal and spatial variations of N_2O and CH_4 emissions.

1. Introduction

Nitrous oxide (N_2O) and methane (CH_4) are key radiatively active greenhouse trace gases which have been recognized to contribute global warming by 5% and 25%, respectively (Mosier, 1998). Quantification of the trace gases is a subject of great interest because accurate information is required to determine the contribution of these gases to global greenhouse gas fluxes (Khalil et al., 2002). IPCC (2007) has reported increased concentrations in N_2O and CH_4 since industrial times, a concern since both gases, although present in lower concentrations to that of carbon dioxide (CO_2), have 298 (N_2O) and 25 (CH_4) times the global warming potential of CO_2 over a 100-year time period. The current global

* Corresponding author. Tel.: +86 535 2109121; fax: +86 535 2109000.
E-mail address: zgsun@yic.ac.cn (Z. Sun).

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atmospheric N_2O and CH_4 concentrations are 319 ± 0.12 ppb and 1774.62 ± 1.22 ppb, respectively, and are increasing approximately 0.26% and 1.1% per year (IPCC, 2007).

Emission of N_2O and CH_4 from various natural ecosystems has significant influences on the global climate change since they account for 44–54% of the total N_2O emissions and 30–40% of the total CH_4 emissions, respectively (IPCC, 2007). Tropical soil and wetlands play an important role in the global carbon (C) and nitrogen (N) biogeochemical cycles, and are considered significant natural sources of N_2O and CH_4, contributing approximately 22–27% (N_2O) and 24% (CH_4), respectively, towards this inventory (Whalen, 2005). Coastal marsh ecosystem is characterized by high temporal and spatial variations involved with topographic feature, 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 aquatic coastal ecosystems may represent a high dynamic interface of intense material processing and transport, with potentially high decomposition and associated greenhouse gases emission (Hirota et al., 2007). In the past two decades, considerable efforts have been made to quantify the N_2O and CH_4 fluxes in different coastal ecosystems, especially in estuarine salt marshes (Magalhães et al., 2007; Moseman-Valtierra et al., 2011), mangrove swamps (Allen et al., 2007; Ganguly et al., 2008), coastal lagoons (Gregorich et al., 2006; Hirota et al., 2007) and coastal marshes (Amouroux et al., 2002). In China, the studies on N_2O and CH_4 emissions from coastal marshes started quite late (in the 2000s), and the related research primarily focused on the coastal tundra marshes in Antarctica (Sun et al., 2002; Zhu et al., 2008) and the salt marshes in the Yangtze River estuary (Yang et al., 2006; Wang et al., 2007) and Min River estuary (Zeng et al., 2010; Mou et al., 2012). However, information on the coastal marshes in other estuaries is still very scarce.

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 estuary and deposited in the slow flowing landform, resulting in vast floodplain and special marsh landscape (Cui et al., 2009). 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 marsh, but also, to some extent, influences water or salinity status and plants succession. Coastal marsh is the main marsh type, with an area of 964.8 km^2, accounting for 63.06% of total area of the Yellow River Delta (Cui et al., 2009). *Suaeda salsa*, an annual C_3 plant, is one of the most prevalent halophytes in the coastal marshes of the Yellow River estuary. As a pioneer plant, it has strong adaptations to environmental stresses, such as high salinity, flooding and sediment burial (Han et al., 2005). *S. salsa* generally germinates in late April, blooms in July, matures in late September and completely dies in late November. In the intertidal zone, three phenotypes are formed due to the differences of water and salinity in high marsh, middle marsh and low marsh. In recent years, the N and organic matter loadings of the Yellow River estuary have significantly increased due to the effects of human activities, and approximately 4650 tons of nutrient and 4.33×10^5 tons of organic matter are discharged into Bohai Sea every year (Ocean Environmental Quality Communique of Shandong Province, 2009). Increases in N and organic matter loadings to estuarine and coastal environment can stimulate microbial processes and associated trace gases emission (Seitzinger and Kroeze, 1998). However, emissions of N_2O and CH_4 from different coastal *S. salsa* marshes in the Yellow River estuary remains poorly documented till now. In addition, because microbial processes affecting trace gas production are regulated by many parameters including oxygen availability, temperature, water content, sediment redox potential, salinity, pH and microbially available C and N sources (Bauza et al., 2002; Whalen, 2005), evaluating the influences of different environmental factors on the emissions of N_2O and CH_4 from coastal marsh will be of importance.

In this paper, we measured the N_2O and CH_4 fluxes from the intertidal zone of the Yellow River estuary using the closed chamber technique. The aims of this study are: (i) to quantify and compare the N_2O and CH_4 fluxes from different coastal *S. salsa* marshes and bare flat; (ii) to determine whether distinct spatial and temporal variation occurs in N_2O and CH_4 flux throughout the day and in different seasons; (iii) to examine how environmental factors influence N_2O and CH_4 emissions.

2. Materials and methods

2.1. Site description

The study was carried out in the intertidal zone of the Yellow River estuary, which is located in the Nature Reserve of Yellow River Delta (37°35′N–38°12′N, 118°33′E–119°20′E) in Dongying City, Shandong Province, China. The nature reserve is of typical continental monsoon climate with distinctive seasons. The temperature changes significantly during early spring and winter, and the freeze/thaw cycles frequently occur in topsoil in majority days, with the frozen depth ranged from 0 cm to 15 cm. The annual average temperature is 12.1 °C, and the frost-free period is 196 d. 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 1962 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 and the main vegetation types include *Phragmites australis*, *S. salsa*, *Triarrhena sacchariflora*, *Tamarix chinensis* and *Imperata cylindrica*.

Intertidal zone sediment is composed mainly of fine particles. Natural geomorphology and depositing zones are distinct. High marsh, middle marsh, and low marsh develop from the land to the sea. The high marsh is predominated by *S. salsa* (>90%) and *P. australis* (<10%), while middle marsh is predominated by *S. salsa* (>95%) and *T. chinensis* (<5%). Low marsh includes two distinct ecosystem-types. One is pure *S. salsa* community (100%), with sparse distribution in the intertidal zone, and the other is bare flat. 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 g m^{-2}, 564.89 ± 99.66 g m^{-2}, 252.97 ± 29.24 g m^{-2}, respectively. In this study, four typical sampling positions 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 marsh of the Yellow River estuary. The initial physical and chemical properties of topsoil (0–10 cm) in the four positions are shown in Table 1.

2.2. Experimental design

Fluxes of N_2O and CH_4 were measured by using static, manual stainless steel chambers and gas chromatography techniques. The chamber is an open-bottom a square box (50 cm × 50 cm × 50 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 August 2009, the stainless steel base (50 cm × 50 cm × 20 cm) with a water groove on top was installed at the four sampling positions. During observations, the chamber was placed over the base filled with water in the groove to ensure airtightness, and the plant was covered within the chamber.

Sampling campaigns were undertaken during four seasons in October 2009 (autumn), December 2009 (winter), April 2010

Table 1
Initial physical and chemical properties[a] of topsoil (0–10 cm) in the four coastal marshes in the Yellow River estuary.

Sites	Bulk density ($g\ cm^{-3}$)	Grain composition (%)			Electrical conductivity ($mS\ cm^{-1}$)	Water content ($cm^3\ cm^{-3}$)	TC (%)	TN (%)	NH_4^+—N ($mg\ kg^{-1}$)	NO_3^-—N ($mg\ kg^{-1}$)
		Clay	Silt	Sand						
HSM	1.28 ± 0.03	8.00	51.46	40.54	15.45 ± 1.44	0.353 ± 0.007	1.22 ± 0.02	0.039 ± 0.001	2.26 ± 0.71	1.31 ± 0.39
MSM	1.34 ± 0.06	8.67	49.67	41.66	13.84 ± 0.81	0.343 ± 0.005	1.29 ± 0.01	0.039 ± 0.002	3.62 ± 0.22	1.21 ± 0.27
LSM	1.29 ± 0.11	10.14	57.13	32.74	10.38 ± 2.08	0.347 ± 0.022	1.27 ± 0.02	0.036 ± 0.001	2.01 ± 0.94	1.10 ± 0.57
BF	1.43 ± 0.01	13.10	73.09	13.81	10.85 ± 0.44	0.358 ± 0.017	1.56 ± 0.02	0.045 ± 0.003	4.66 ± 2.43	1.19 ± 0.42

[a] Values are means (±S.E.); HSM, High *Suaeda salsa* marsh; MSM, Middle *S. salsa* marsh; LSM, Low *S. salsa* marsh; BF, Bare flat.

(spring) and July 2010 (summer). Each measurement campaign consisted of 12 chambers set up at four positions (three chambers per site): 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 low tide. 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. 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 to the laboratory and analyzed within 36 h using gas chromatography (Agilent 7890A) equipped with FID and ECD. The gas chromatography configurations for analyzing N_2O and CH_4 concentrations were according to the method of Wang and Wang (2003), and the fluxes calculation followed the description of Song et al. (2008).

2.3. Environmental measurements

Air temperature and soil temperatures (0, 5, 10, 15 and 20 cm) were measured in each position during gas sampling. Soil volumetric moisture and electrical conductivity (EC) in 0–5 and 5–10 cm depths were determined *in situ* by high-precision moisture measuring instrument (AZS-2) and soil and solution EC meter (Field Scout), respectively. Soil moisture and EC were not determined in December 2009 since the topsoil (0–10 cm) was frozen. On each sampling date, two soil samples (0–10 cm) per site were taken for analyzing TC and TN contents by element analyzer (Elementar Vario Micro, German) and NH_4^+—N and NO_3^-—N contents by sequence flow analyzer (San^{++} SKALAR, Netherlands).

2.4. Conversion to CO_2-equivalents

Global warming potentials of 298 for N_2O and 25 for CH_4 over 100 years (IPCC, 2007) were used to convert N_2O and CH_4 emissions to CO_2-e for comparing their greenhouse impacts.

2.5. 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). Correlation analyses were used to examine the relationship between fluxes and the measured environmental variables. In all tests, differences were considered significantly only if $p < 0.05$.

3. Results

3.1. N_2O fluxes from different coastal marshes

Over all sampling periods, N_2O fluxes from the four positions ranged from $-0.0147\ mgN_2O\ m^{-2}\ h^{-1}$ to $0.0982\ mgN_2O\ m^{-2}\ h^{-1}$, thus both emission and consumption of N_2O occurred. Except for MSM, N_2O fluxes from the other positions (HSM, LSM and BF) differed significantly between seasons ($p < 0.05$) (Fig. 1). With the exception of HSM that were found to release N_2O during all times of day and the seasons measured, the other positions showed consumptions in some sampling times and seasons. Significantly higher N_2O emissions were observed in HSM compared to LSM during April 7:00 and 14:30 sampling, and for October 9:30, 17:00 and July 9:30 ($p < 0.05$). Moreover, N_2O fluxes from HSM were significantly higher than those from MSM during April 14:30 and July 9:30 sampling ($p < 0.05$), or those from BF during July 9:30 sampling ($p < 0.05$). N_2O fluxes also differed significantly among the four positions during spring (April), autumn (October) and winter (December) ($p < 0.01$), but not during summer (July) ($p > 0.05$) (Fig. 1). N_2O fluxes from MSM were quite lower than those from other positions over all sampling periods, and the values from HSM were significantly higher than those from MSM and BF in spring, summer and autumn ($p < 0.05$) or those from LSM in spring and autumn ($p < 0.01$). Significantly higher N_2O emission was observed in LSM in winter compared to MSM ($p < 0.01$) and BF ($p < 0.05$) (Fig. 1). The average N_2O fluxes from the four positions during spring, summer, autumn and winter were $0.0325\ mgN_2O\ m^{-2}\ h^{-1}$, $0.0089\ mgN_2O\ m^{-2}\ h^{-1}$, $0.0119\ mgN_2O\ m^{-2}\ h^{-1}$ and $0.0140\ mgN_2O\ m^{-2}\ h^{-1}$, respectively, indicating that coastal marsh represented N_2O emission across all the seasons sampled.

3.2. CH_4 fluxes from different coastal marshes

CH_4 fluxes from the four positions averaged between $-0.7421\ mgCH_4\ m^{-2}\ h^{-1}$ and $0.4242\ mgCH_4\ m^{-2}\ h^{-1}$ and had no significant differences between seasons ($p > 0.05$) (Fig. 2). During all times of day and the seasons measured, both emission and consumption of CH_4 occurred in the four positions, but the consumption was predominated. CH_4 fluxes from HSM were significantly different with those from LSM during April 7:00, 12:30, 14:30 and 17:00 sampling, and for July 7:00, October 14:30 and December 10:30 ($p < 0.05$). Significant differences were also observed between MSM and LSM during April 7:00, 12:30, 14:30 and July 12:30 sampling ($p < 0.05$), or between MSM and BF during April 14:30, July 14:30, October 7:00, 12:00 and December 7:00 sampling ($p < 0.05$). Over all sampling seasons, CH_4 fluxes from the four positions had no significant differences ($p > 0.05$) (Fig. 2). Moreover, CH_4 fluxes from MSM and LSM were generally higher than those from HSM and BF in some sampling times and seasons. The mean CH_4 fluxes from the four positions during spring, summer, autumn and winter were $-0.0109\ mgCH_4\ m^{-2}\ h^{-1}$, $-0.0174\ mgCH_4\ m^{-2}\ h^{-1}$, $-0.0141\ mgCH_4\ m^{-2}\ h^{-1}$ and $-0.0089\ mgCH_4\ m^{-2}\ h^{-1}$, respectively, indicating that coastal marsh acted as CH_4 sink across all the seasons sampled.

3.3. Global warming potential (GWP)

Across all the seasons sampled, the coastal marsh was a net source of N_2O and a net sink of CH_4. As N_2O and CH_4 emissions were expressed as CO_2-e emissions, N_2O emissions contributed

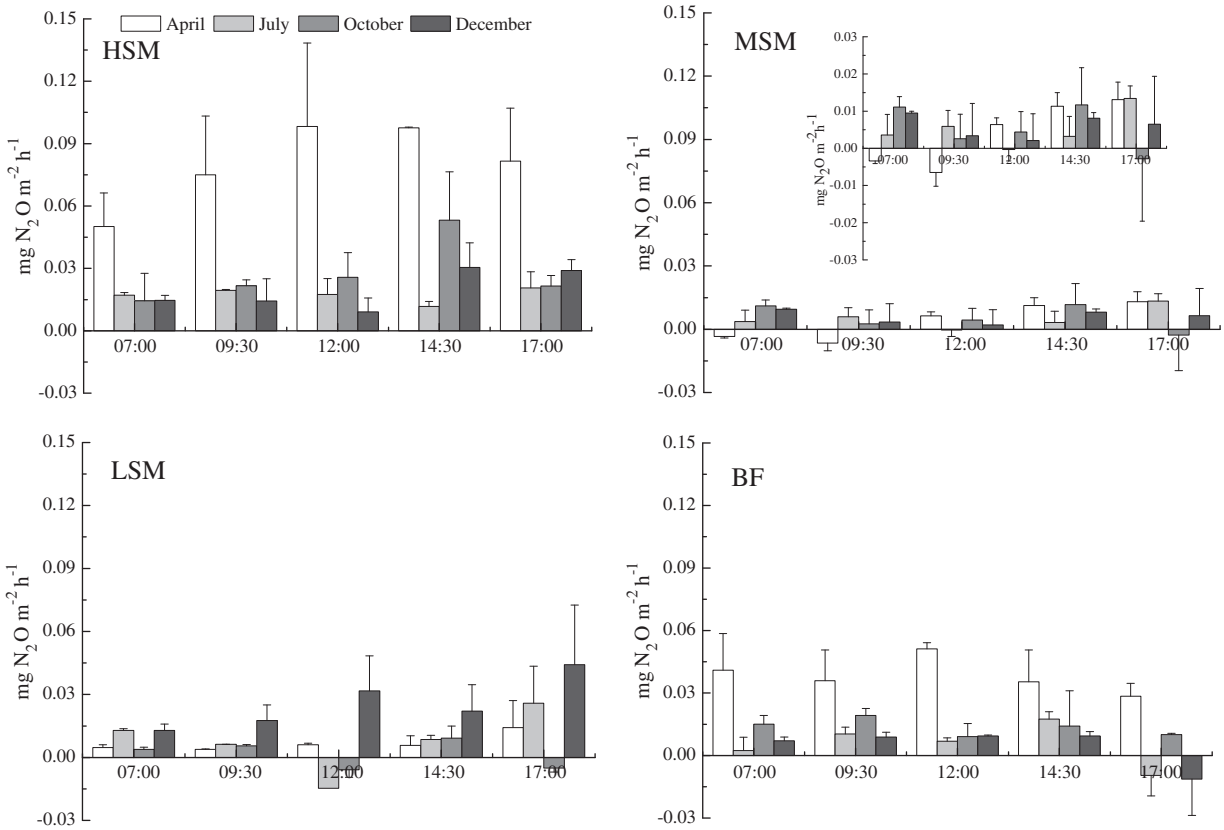

Fig. 1. Nitrous oxide flux (mgN_2O m^{-2} h^{-1}) from high *Suaeda salsa* marsh (HSM), middle *S. salsa* marsh (MSM), low *S. salsa* marsh (LSM) and bare flat (BF) in spring (April 2010), summer (July 2010), autumn (October 2009) and winter (December 2009) in the Yellow River estuary. Values are means (±S.E., $n = 3$).

47.29% and 20.45% of net CO_2-e emissions measured during spring and winter (Table 2), which were much higher than the contributions of net CO_2-e emissions measured during summer (14.42%) and autumn (17.84%). In contrast, CH_4 emissions contributed 27.09% and 35.18% of net CO_2-e emissions measured during summer and autumn (Table 2), which were much higher than the contributions of net CO_2-e emissions measured during spring (23.14%) and winter (14.58%). The contributions of CH_4 to the total calculated CO_2-e emissions were significantly lower than those of N_2O during spring and winter ($p < 0.05$), but not during summer and autumn ($p > 0.05$) (Table 2).

3.4. Environmental variables of coastal marsh

Total C, total N and NH_4–N in the sediment of BF were generally higher than those in the other positions over all sampling periods (Table 3). Significant differences in total C between BF and other position (HSM, MSM or LSM), and total N between BF and HSM (or LSM) were observed ($p < 0.05$). No significant differences in NH_4–N and NO_3–N content were observed in the four positions ($p > 0.05$). During all times of day and the seasons measured, air temperatures and sediment temperatures did not differ between positions ($p > 0.05$). Although water contents in subsurface sediment (5–10 cm) were higher than those in surface sediment (0–5 cm), no significant differences were found among the four positions ($p > 0.05$). Both EC in surface and subsurface sediments (0–5, 5–10 cm) had significant differences among the four positions ($p < 0.01$), in the order of HSM > MSM > LSM > BF.

Pearson correlation analysis between N_2O (or CH_4) fluxes and independent variables indicated that most of the correlations between N_2O (or CH_4) fluxes and environmental parameters in the four positions were not significant ($p > 0.05$) (Table 4). In HSM, N_2O fluxes showed significant positive correlations with 5, 10 and 20 cm sediment temperatures ($p < 0.05$), while CH_4 fluxes showed significant positive correlation with NH_4–N ($p < 0.05$). CH_4 fluxes from MSM showed significant negative correlations with NO_3–N and air temperature ($p < 0.05$), while those from LSM had significant negative correlation with 0 cm sediment temperature ($p < 0.05$). In BF, N_2O fluxes had no significant correlations with environment parameters, while CH_4 fluxes showed significant positive correlations with 5, 10 and 20 cm sediment temperatures ($p < 0.05$). In addition, the correlations between environmental parameters determined during all times of day and N_2O (or CH_4) diurnal emissions were not significant ($p > 0.05$).

4. Discussion

4.1. Temporal variations of N_2O and CH_4 fluxes

This study demonstrated that N_2O and CH_4 emissions from coastal marshes varied significantly throughout different times of the day (Fig. 1, Fig. 2). Diurnal variations in trace gas flux have been reported in other coastal marsh studies. Lu et al. (1999) showed that CH_4 fluxes from *Bruguiera sexangula* mangrove swamp sediment had large diurnal fluctuations, which were caused by the changes of tidal inundation rather than the changes of air or sediment temperatures. Zhu et al. (2008) indicated that N_2O emission from the coastal tundra marsh in eastern Antarctica greatly differed at different times of the day, with emission peaks coincided with the maximum of ground temperature. Significant diel cycle of N_2O emission was also observed in the mangrove (*Rhizophora*

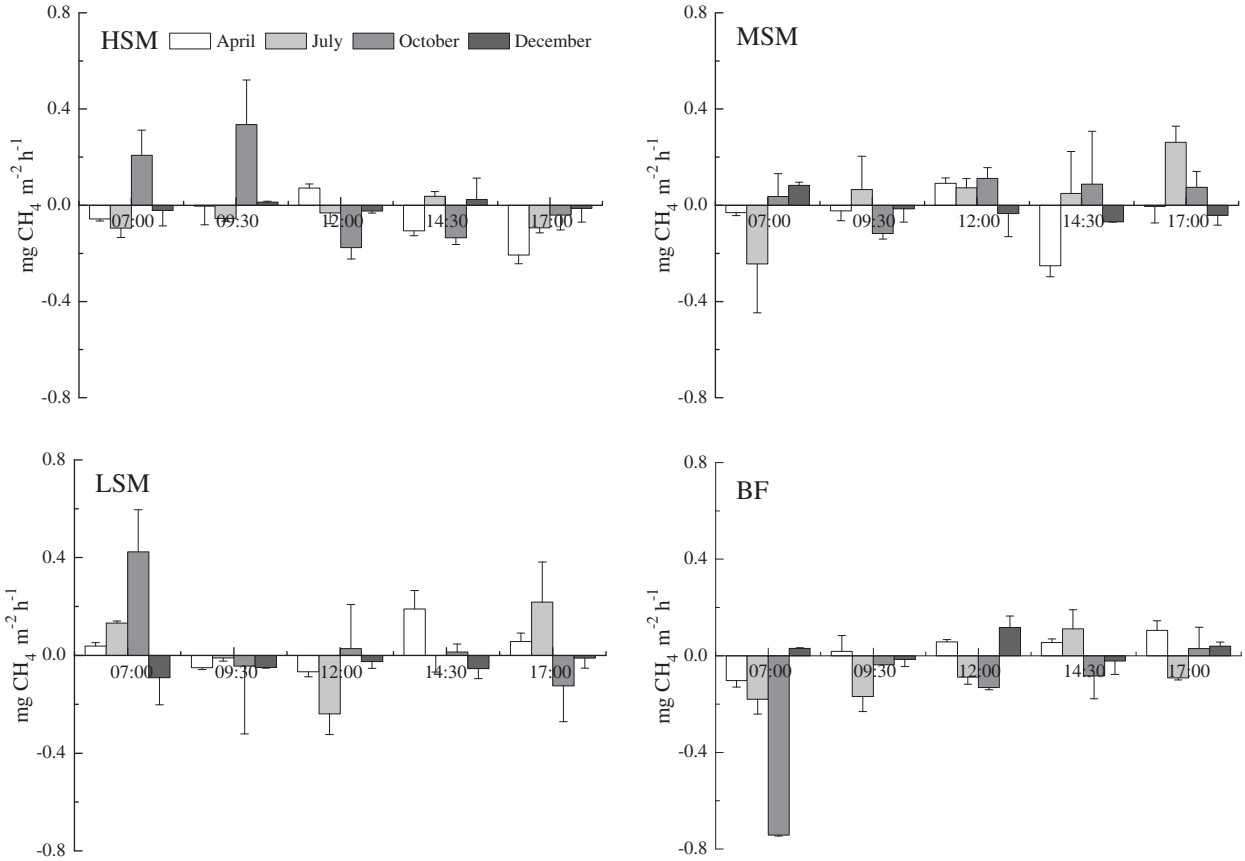

Fig. 2. Methane flux (mgCH$_4$ m^{-2} h^{-1}) from high *Suaeda salsa* marsh (HSM), middle *S. salsa* marsh (MSM), low *S. salsa* marsh (LSM) and bare flat (BF) in spring (April 2010), summer (July 2010), autumn (October 2009) and winter (December 2009) in the Yellow River estuary. Values are means (±S.E., $n = 3$).

Table 2
Estimated total N_2O and CH_4 emission (mgCO$_2$ equivalents m^{-2})[a] during the sampling periods in the coastal marshes of the Yellow River estuary.

Items	Spring		Summer		Autumn		Winter	
	N_2O	CH_4	N_2O	CH_4	N_2O	CH_4	N_2O	CH_4
HSM	50297 ± 13189	−3568 ± 184	11390 ± 1978	−2700 ± 329	17770 ± 6340	2654 ± 2628	12425 ± 2651	102 ± 1105
MSM	3117 ± 1009	−2410 ± 1116	3297 ± 1673	2716 ± 4445	4079 ± 2927	2904 ± 3211	3865 ± 2995	−1246 ± 1711
LSM	4212 ± 2668	1316 ± 833	6959 ± 1178	648 ± 263	1744 ± 1920	1436 ± 2943	16529 ± 8552	−2237 ± 351
BF	25273 ± 6868	1483 ± 1102	3630 ± 2409	−4384 ± 1282	7684 ± 4078	−11826 ± 347	3019 ± 2518	1378 ± 550
Coastal marsh	20725 ± 6634	−795 ± 774	6319 ± 1264	−930 ± 1297	7819 ± 2559	−1208 ± 2151	8960 ± 2703	−501 ± 614
	(47.29%)[b]	(23.14%)[b]	(14.42%)[b]	(27.09%)[b]	(17.84%)[b]	(35.18%)[b]	(20.45%)[b]	(14.58%)[b]

HSM, High *Suaeda salsa* marsh; MSM, Middle *S. salsa* marsh; LSM, Low *S. salsa* marsh; BF, Bare flat.
[a] Estimates were derived by weighted averages of 60 date points from morning to late afternoon (7:00, 9:30, 12:00, 14:30, 17:00 h) for each sampling month. The IPCC (2007) conversion of N_2O and CH_4 emissions to CO_2 equivalents is 298 and 25, respectively, over a 100-year timeframe.
[b] Percentage contribution of N_2O or CH_4 during each season is given in parentheses.

mangle) swamp at Magueys Island, with emissions peaks coinciding with increased ammonium production (Bauza et al., 2002). Since environmental factors are involved directly with different microbe and plant activities, such as photosynthesis, aerobic and anaerobic respiration and nitrification–denitrification and are changeable in relative short term (diurnal) scale, the diurnal variations of N_2O and CH_4 emissions from different coastal marshes are generally affected by different principal factors. However, our study showed that any environmental factors determined during all times of day had no significant correlation with N_2O or CH_4 diurnal emissions. Similar result was drawn by Hirota et al. (2007) who found that any environmental factors seemed to have no effects on the diurnal variations of N_2O and CH_4 flux under light condition. We considered that there were two probable reasons: (i) the variations of environmental factors in the coastal marshes of the Yellow River estuary might be more complicated than those in other coastal marshes, and N_2O or CH_4 diurnal emissions were controlled by multiple factors; (ii) since our study did not measure N_2O or CH_4 fluxes and environmental factors over the complete tidal inundation cycles, the variations of some major factors during all times of night might be missed, which, to some extent, covered the main factors. Whether N_2O and CH_4 fluxes undergo strong diel cycles and which factor controlled their diurnal cycle requires further study.

Seasonal variations in N_2O and CH_4 emissions from coastal marshes were observed in our study (Fig. 1, Fig. 2) and also reported by others (Wang et al., 2007; Hirota et al., 2007; Song et al., 2008). Whalen (2005) observed that, in arctic, boreal and temperate regions, seasonal patterns of trace gas emission were governed by seasonal variability in temperatures affecting water

Table 3
Environmental parameters[a] of coastal marshes over the 2009/2010 sampling campaign.

Environmental parameters	HSM	MSM	LSM	BF
TC (%)	1.29 ± 0.06[b]	1.49 ± 0.18[b]	1.35 ± 0.08[b]	1.57 ± 0.11[c]
TN (%)	0.04 ± 0.01[b]	0.04 ± 0.01[bc]	0.04 ± 0.01[b]	0.05 ± 0.01[c]
NH_4–N (mg kg^{-1})	2.39 ± 1.06[b]	2.90 ± 0.68[b]	2.57 ± 0.67[b]	3.09 ± 0.88[b]
NO_3–N (mg kg^{-1})	1.49 ± 0.50[b]	1.91 ± 0.93[b]	1.31 ± 0.97[b]	1.18 ± 0.40[b]
Air temperature (°C)	15.32 ± 14.82[b]	14.57 ± 14.07[b]	13.39 ± 12.94[b]	14.28 ± 13.37[b]
Sediment temperature (°C)				
0 cm	18.44 ± 16.68[b]	16.80 ± 14.73[b]	12.56 ± 11.73[b]	15.00 ± 14.76[b]
5 cm	18.33 ± 15.42[b]	17.09 ± 14.17[b]	13.47 ± 13.44[b]	15.25 ± 14.59[b]
10 cm	17.17 ± 14.57[b]	16.44 ± 13.74[b]	14.33 ± 12.68 [b]	14.23 ± 12.89[b]
15 cm	16.15 ± 14.31[b]	15.45 ± 12.72[b]	14.14 ± 12.73[b]	13.15 ± 12.11[b]
20 cm	15.23 ± 12.11[b]	14.68 ± 12.20[b]	13.38 ± 12.38[b]	13.07 ± 11.03[b]
Water content (cm^3 cm^{-3})				
0–5 cm	0.29 ± 0.02[b]	0.29 ± 0.02[b]	0.28 ± 0.04[b]	0.28 ± 0.03[b]
5–10 cm	0.39 ± 0.01[b]	0.38 ± 0.03[b]	0.39 ± 0.01[b]	0.39 ± 0.01[b]
EC (mS cm^{-1})				
0–5 cm	18.91 ± 1.7[b]	15.67 ± 3.59[c]	9.14 ± 0.98[d]	10.59 ± 0.97[e]
5–10 cm	15.76 ± 2.0[b]	11.37 ± 2.34[c]	8.04 ± 1.09[d]	9.61 ± 1.20 [e]

HSM, High *Suaeda salsa* marsh; MSM, Middle *S. salsa* marsh; LSM, Low *S. salsa* marsh; BF, Bare flat.
[a] Values are means (±S.E.) of samples ($n = 24$ for TC, TN, NH_4–N and NO_3^-—N; $n = 18$ for water content and EC; $n = 60$ for air temperature and soil temperature) collected from high *S. salsa* marsh (HSM), middle *S. salsa* marsh (MSM), low *S. salsa* marsh (LSM) and bare flat (BF) over all sampling periods. Statistically significant differences among sampling positions ($p < 0.05$) were calculated for multiple comparisons using Kruskal–Wallis ANOVA and are indicated by different letters within each row.

Table 4
Pearson correlation analysis between N_2O or CH_4 fluxes and environmental parameters[a].

Environmental parameters	HSM		MSM		LSM		BF	
	N_2O	CH_4	N_2O	CH_4	N_2O	CH_4	N_2O	CH_4
TC	−0.237	0.088	−0.038	−0.881	0.573	−0.614	0.441	0.412
TN	0.484	−0.051	−0.367	−0.814	−0.021	0.057	0.635	0.139
NH_4–N	−0.616	0.983[b]	−0.092	0.293	0.445	−0.460	−0.741	−0.504
NO_3–N	−0.294	0.282	−0.524	−0.984[b]	−0.062	−0.104	−0.525	0.497
Air temperature	0.410	−0.059	0.042	−0.488[b]	−0.124	−0.334	−0.270	0.263
Sediment temperature								
0 cm	0.433	−0.004	−0.058	−0.345	−0.004	−0.495[b]	0.163	0.319
5 cm	0.536[b]	−0.032	0.221	−0.235	−0.032	−0.233	0.269	0.479[b]
10 cm	0.565[b]	−0.228	0.291	−0.093	−0.228	−0.294	−0.387	0.894[b]
15 cm	0.443	−0.374	0.313	−0.113	−0.374	−0.026	−0.022	0.348
20 cm	0.657[b]	−0.417	0.310	0.086	−0.016	0.017	−0.040	0.503[b]
Water content								
0–5 cm	−0.930	0.514	0.796	0.664	0.971	−0.996	−0.140	0.579
5–10 cm	−0.644	0.876	−0.731	0.310	0.996	−0.972	0.420	0.929
EC								
0–5 cm	−0.943	0.725	0.751	0.914	0.071	0.079	0.981	0.581
5–10 cm	−0.848	0.306	0.862	0.679	−0.755	0.844	0.186	−0.539

HSM, High *Suaeda salsa* marsh; MSM, Middle *S. salsa* marsh; LSM, Low *S. salsa* marsh; BF, Bare flat.
[a] Pair sample size, $n = 4$ for TC, TN, NH_4–N and NO_3^-—N; $n = 3$ for water content and EC; $n = 20$ for air temperature and soil temperature.
[b] $p < 0.05$. 95% Confidence level was given during Pearson correlation analysis.

availability, production of substrate precursors and microbial activity. However, N_2O and CH_4 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). For instance, N_2O emissions from HSM and BF were the lowest in summer and the highest in spring. N_2O fluxes from HSM and BF during spring were also higher than those during winter, while emissions from MSM and LSM during winter were higher than those during spring (Fig. 1). Moreover, CH_4 fluxes from MSM and LSM were positive during summer while those from HSM and BF were negative in the meantime (Fig. 2). We considered that the seasonal variations in N_2O and CH_4 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), plants (Hirota et al., 2007) and sediment substrate (Allen et al., 2007). Similar result was drawn by Hirota et al. (2007) who found that the temporal variations of N_2O and CH_4 fluxes from coastal lagoon were mainly controlled by temperatures and aboveground biomass.

We found that high N_2O emissions occurred during spring and winter and low fluxes occurred during summer and autumn. The results were different with others which showed that N_2O emissions were high in summer and autumn and low in spring and winter (Wang et al., 2007; Søvik and Kløve, 2007). Many studies have indicated that, in the mid-high latitude and high altitude regions, freeze/thaw cycle occurred in late autumn, winter or early spring was a very important process to increase N_2O production and emission since it could affect soil physical and biological properties greatly (Teepe et al., 2001; Song et al., 2008). As mentioned previously, the coastal marshes of the Yellow River estuary locate in the mid-latitude region and the freeze/thaw cycles frequently occur in topsoil in majority days during spring and winter (frozen depth ranged from 0 cm to 15 cm). Thus, the high N_2O emissions occurred during spring and winter might be related to the frequent freeze/thaw cycles. Similar results were drawn by Song et al. (2008) who found significant N_2O emissions from freshwater marsh in northeast China during the freeze/thaw cycle as the temperature increased in late spring. Although higher temperatures were observed in summer and autumn compared to spring and

winter, the N_2O emissions were very low. The low N_2O emissions during summer and autumn might be related to the variations and interactions of environmental factors in coastal marsh which concealed the significant effects of temperature. In this study, we considered that N_2O emissions during summer and autumn were controlled by multiple factors, and among them, *S. salsa*, had significant influences. As mentioned previously, *S. salsa* is the most prevalent halophyte in the coastal marshes of the Yellow River estuary and it has strong adaptation to the tidal inundation due to the transportation mechanism of oxygen (O_2) from aboveground parts to roots by aerenchyma (Han et al., 2005). First, part of O_2 transported to the roots can be released to the rhizosphere soil, which produces oxidizing microenvironment around *S. salsa* rhizosphere (Han et al., 2005; Kong et al., 2008). Second, soil microbes in the rhizosphere have great effects on the turnovers of available C and N in rhizosphere soil (Toal et al., 2000). Wang et al. (2010) studied the rhizosphere effects of *S. salsa*, *T. chinensis* and *I. cylindrica* in the Yellow River estuary, and found that the microbe amount and microbial activity in rhizosphere soil were much higher than those in non-rhizosphere soil and significantly higher values were observed in *S. salsa* rhizosphere soil compared to *T. chinensis* and *I. cylindrica* ($p < 0.01$), which was more favorable for the turnovers of available C and N (Herman et al., 2006; Cheng, 2009). Third, the *S. salsa* roots can also excrete some small molecular compounds (glucide, organic acid, amino acid and phenolic compounds), which provides plentiful available C for the rhizosphere microbes (Cheng, 2009; Wang et al., 2010). Thus, the presence of *S. salsa* probably accelerated the nitrification- denitrification rate as soil microbes in the rhizosphere were supplied with plentiful organic C and proper amount of O_2, and N_2O might be reduced to N_2 by denitrification regardless of whether N_2O was produced by nitrification or denitrification. In addition, the proper amount of O_2 around the rhizosphere could also be partly diffused to the non-rhizosphere soil though the topsoil was under anoxic conditions, which, to some extent, reduced N_2O emissions. Since studies on the N_2O absorption and emission of different plants have been widely reported (Yang and Chen, 1995; Chen et al., 2003), the low N_2O emission might also be linked to the N_2O absorption of *S. salsa*, which required to be verified.

We also found that the coastal marsh in the Yellow River estuary acted as CH_4 sink across all the seasons sampled, and higher CH_4 sinks occurred during summer and autumn than during spring and winter. As mentioned previously, the seasonal variations in CH_4 emissions from coastal marshes were probably affected by the complex interactions of temperatures and other biotic/abiotic factors. We considered that high CH_4 sinks occurred during summer and autumn, to some extent, were dependent on the interactions of thermal conditions and other abiotic factors (such as soil moisture, salinity and soil substrate), which might be more favorable for the activities of methanotrophs than methanogens. Moreover, the CH_4 sink formed in the coastal marsh depended on the equilibrium of CH_4 production and oxidation which were related to the variations and interactions of abiotic and biotic factors (Ding et al., 2004; Hirota et al., 2007). Among them, soil moisture and salinity had great effects on the production/consumption of CH_4 (Megonigal et al., 2003). In this study, we observed CH_4 consumption during all the seasons sampled although the soil moistures in sediment were high (0–5 cm, 0.28–0.29 $cm^3\ cm^{-3}$; 5–10 cm, 0.38–0.39 $cm^3\ cm^{-3}$) (Table 3). The result was contrary to mostly previous studies which described that CH_4 flux increased with higher moisture (Moore and Dalva, 1993; Ye et al., 2000), but was similar with the conclusion drawn by Hirota et al. (2007) who reported a negative partial correlation between CH_4 flux and soil moisture in the sandy shore of Lake Nakaumi. We considered that the negative impact of soil moisture on CH_4 emission was related to the interaction of moisture and salinity in sediment. Magenheimer et al. (1996) showed that the CH_4 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. Similar results were drawn by Chidthaisong and Conrad (2000) and Zeng et al. (2008) who found that high salinity inhibited the activities of methanogens or did harm to methanogens, which reduced CH_4 emission. In this study, the salinity (represented by EC) of the coastal marsh in the Yellow River estuary were high (0–5 cm, 9.14–18.91 mS cm^{-1}; 5–10 cm, 8.04–15.76 mS cm^{-1}) (Table 3), which might induce CH_4 consumption. The sulfate (SO_4^{2-}) concentrations in sediment substrate resulting from high SO_4^{2-} content in seawater (Ivanou, 1992) were also considered dominant factor controlling CH_4 emissions from coastal marsh (Kreuzwieser et al., 2003). Because sulfate (SO_4^{2-}) reducing bacteria (SRB) could compete H_2/CO_2 and ethylic acid (CH_3COOH) with methanogen and the former had more strong affinity to the reaction substrates, the coexistence of SRB and SO_4^{2-} would inhibit the production and emission of CH_4 (van der Gon et al., 2001). In this study, the SO_4^{2-} concentration (0.62–1.50%) (Fan et al., 2010) in the coastal marshes of the Yellow River estuary were very high and the anoxic condition in sediment resulting from high moisture could enhance the dissimilatory reduction of SO_4^{2-}, which would inhibit the CH_4 emission of *S. salsa* marsh. Similar results were drawn by Magenheimer et al. (1996) who also found the dominant role of SO_4^{2-} in inhibiting methanogenesis in salt-marsh sediments.

This study also showed that, across all the seasons sampled, the coastal marsh was a net source of N_2O and a net sink of CH_4. When N_2O and CH_4 emissions were converted to CO_2-e emissions, N_2O comprised the principal part of total calculated CO_2-e emissions, with largest emissions occurring during spring and winter (Table 2). Although this contrasted the standpoint proposed for *Carex Lasiocarpa* freshwater marsh ecosystem, where both N_2O and CH_4 contributed the main part of total calculated CO_2-e emissions measured during summer and autumn (Song et al., 2008), approximated the notion reported by Allen et al. (2007) who found that N_2O dominated winter CO_2-e emissions at both fringe and grey mangrove *(Avicennia marina)* positions when CH_4 emissions were lowest. This finding highlights the importance of adequately capturing seasonal variation of both N_2O and CH_4 in the future in the coastal marshes of the Yellow River estuary, particularly during times of CH_4 consumption, where the high global warming potential of N_2O can dominate trace gas emissions.

4.2. Spatial variations of N_2O and CH_4 fluxes

In this study, we observed that the physical and chemical parameters of sediment differed in their magnitude among the four positions during all times of day and the seasons measured. Especially, significant differences in total C between BF and other position (HSM, MSM or LSM), total N between BF and HSM (or LSM), and EC among the four positions were observed ($p < 0.05$) (Table 4). Such differences would be due to the site-specific conditions such as topography, aspect, slope, hydrology and species composition which determine the magnitudes and variations of N_2O or CH_4 at spatial scale (Allen et al., 2007; Hirota et al., 2007).

This study showed that N_2O fluxes differed significantly among the four positions ($p < 0.01$) (Fig. 1), and the values from HSM were generally high and those from MSM were very low. Previous studies have indicated that temperatures had great effects on N_2O emissions at spatial scale (Alongi et al., 2005; Gregorich et al., 2006). During all times of day and the seasons measured, although air temperatures and sediment temperatures did not differ between positions ($p > 0.05$), significant relationships still could be found in HSM. This indicated that thermal condition was an important factor affecting N_2O 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 vegetation. In this paper, we found that the moisture in topsoil (0–10 cm) was very close and had no significant difference among the four positions (Table 3), which were mainly correlated with the low spatial heterogeneity of sediment moisture and the strong evaporation of the coastal marsh. Over all sampling seasons, the coefficient of variation (CV) of sediment moisture at 0–5 and 5–10 cm depths in HSM, MSM, LSM and BF were 1.38%, 4.31%, 15.7%, 10.54% and 1.49%, 6.36%, 1.60%, 0.53%, respectively, indicating that the spatial variability of sediment moisture (0–5 or 5–10 cm) among the four positions was not very high. As mentioned previously, the annual evaporation in the Yellow River estuary is 1962 mm and the annual precipitation is 551.6 mm (evaporation/precipitation ratio, 3.52), implying that the strong evaporation might cause insignificant difference of the topsoil moistures as the spatial variability of sediment moisture across the coastal marsh was not high and the measurements were carried out during low tide over a number of days. Because soil evaporation was mainly affected by surface sediment moisture and the evaporation of surface sediment (0–5 cm) was more vigorous than that of subsurface sediment (Yang et al., 1997), high sediment moisture was observed in subsurface sediment (5–10 cm) in our study (Table 3). We also found that both positive and negative impacts of moisture on N_2O emissions occurred in salt marshes (Table 4), which were different with mostly previous studies that N_2O emissions had negative correlation with moisture (Regina et al., 1996; Wang et al., 2005). We considered that N_2O emissions in different positions depended on the fluctuation of soil moisture resulting from astronomic tide. For one thing, proper soil moisture contributes to a favorable aerobic–anerobic status for N_2O production by denitrification or nitrification or both. For another, N_2O may be transported from surface seawater to the salt marsh by tidal fluctuation (Senga et al., 2001). Comparatively, the EC in topsoil (0–10 cm) had significant difference among the four positions (Table 3), which was mainly related to the spatial variability of sediment texture. Nadler (1991) indicated that soil texture significantly affected the soluble capacity of salinity even if soil moisture was approximate, and high EC was generally observed in coarse texture soil (Lin et al., 2005). As mentioned above, the sediment moisture across the coastal marsh had no significant difference while the grain composition had high spatial variability (the CVs of clay, silt and sand across the coastal marsh were 22.71%, 18.42% and 40.01%, respectively) (Table 1), which probably resulted in significant difference of EC among the four positions. Because the strong evaporation in coastal marsh could cause considerable salinity to be accumulated in surface sediment (Fan et al., 2010) and high moisture in subsurface sediment was favorable for the transference of salinity from subsurface sediment to deep layers (Lin et al., 2005), high EC was observed in surface sediment (Table 3). This paper also indicated that the correlations (positive and negative) between EC and N_2O emission were not significant (Table 4), which were different with mostly previous studies that N_2O emissions had negative correlation with EC (Dalal et al., 2003; Wang et al., 2009). One possible explanation for positive effect was that the salinity in MSM, LSM and BF might not completely inhibit the N turnover in salt marsh and the activities of nitrifiers and denitrifiers in sediment (Lv et al., 2008). Site-level control of N_2O emission was also attributed to the effects of nutrient status. Although significant differences in total C and total N were observed among the four positions, the positive/negative correlations between total C or total N and N_2O emission were not significant (Table 4). Positive/negative relationships between N_2O emissions and total C or total N have been reported in various studies (Allen et al., 2007; Søvik and Kløve, 2007) due to C/N regulations and interactions of other abiotic variables at spatial scale (Huang et al., 2002). NH_4–N and NO_3–N content mostly had negative correlations with N_2O emission in the four positions (Table 4), which was similar with the conclusions drawn by Huang et al. (2002) and Tauchnitz et al. (2008), but was contrary to mostly previous studies (Aelion et al., 1997; Muñoz-Hincapié et al., 2002). In HSM, MSM and LSM, one possible reason was related to the interactions of vegetation (*S. salsa*) and microorganism (nitrifiers and denitrifiers) that produced negative influences on the transformation of NH_4–N or NO_3–N during N_2O production. Li et al. (2002) indicated that vegetation had great impacts on N_2O emission through influencing the activities of soil microorganism. Because available C, NH_4–N and NO_3–N were important substrates participating in the processes of nitrification and denitrification (Tauchnitz et al., 2008), the production of N_2O might be inhibited by the available substrates as they were significantly competed by both vegetation absorption and microorganism utilization (Li et al., 2002). In BF, one possible reason was correlated with the strong horizontal and vertical movement of NH_4–N or NO_3–N in surface sediment due to the effects of tide (Mou, 2010). Strong movement of NH_4–N or NO_3–N would decrease the available N participating in the processes of nitrification and denitrification, which was unfavorable for the production and emission of N_2O. In addition, since the species composition, biomass and coverage of vegetation in the four positions were different as mentioned previously, the vegetation distributed continuously across the coastal marsh would be one of the key factors for the N_2O emissions at spatial scale.

Although CH_4 fluxes from the four positions had no significant differences ($p > 0.05$), CH_4 emission patterns were different during all times of day and the seasons measured (Fig. 2). Significant negative relationships between CH_4 emission and air temperature in MSM ($p < 0.05$) and between CH_4 emission and 0 cm sediment temperature in LSM ($p < 0.05$) were observed, while significant positive correlations occurred between CH_4 emission and sediment temperatures (5, 10 and 20 cm) in BF ($p < 0.05$) (Table 4). These indicated that thermal condition was also an important factor affecting CH_4 emission across the coastal marsh, but its function might be covered in some coastal marshes by other biotic or abiotic factors. For spatial variations of CH_4 fluxes in coastal marsh, we considered that the differences of coverage, aboveground biomass and species composition were the main driving forces. Significantly lower CH_4 emissions occurred in BF compared to HSM, MSM and LSM due to the inexistence of vegetation (Fig. 2). The importance of vegetation for C fluxes in wetlands 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 CH_4 from soil to the atmosphere (Schimel, 1995). Since the productions of CH_4 in sediment were very low as mentioned previously, CH_4 emissions from soil to atmosphere transported by vegetation were also very low. Moreover, the limited CH_4 production could also be partly or completely oxidized before it was transported to atmosphere. These indicated that the species composition, biomass and coverage of vegetation across the coastal marsh, to some extent, affected CH_4 emission patterns at spatial scale. Not only temporal but also spatial variations of CH_4 fluxes from coastal marsh were determined by soil moisture and salinity. As mentioned previously, although the formation of CH_4 sink in coastal marsh at temporal scale was negative influenced by soil moisture and salinity, both positive and negative effects of them on CH_4 fluxes were observed in different positions (Table 4), which induced the formation of different CH_4 emission patterns at spatial scale. Similar result was drawn by Hirota et al. (2007) who found that, in coastal ecosystems subjected to the fluctuation of soil moisture by astronomic tide, there would be both positive and negative impacts on CH_4 emissions. Site-level control of CH_4 emission was also related to the effects of nutrient status (Ding and Cai, 2002). In this study, both positive and negative correlations between CH_4 emission and nutrient status were observed

(Table 4). Especially, significantly higher relationships between CH_4 emission and NH_4–N in HSM ($p < 0.05$) and between CH_4 emission and NO_3–N in MSM ($p < 0.05$) occurred (Table 4). Hence, local nutrient differences due to topography, aspect, slope, hydrology and vegetation influenced the spatial differences of CH_4 emissions at spatial level.

In order to better understand the differences of trace gas fluxes from the coastal marsh in the Yellow River estuary and up-scale trace gas emissions to annual inventories, our study highlights the need to strengthen the frequency of sampling *in situ* at long-term and different spatial scales, which identifies the magnitude of N_2O and CH_4 variations at seasonal, diurnal and spatial scales, before values are up-scaled to annual inventories; and to further identify key drivers of trace gas emissions in coastal marsh to reveal the causalities of spatial and temporal variations.

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