RESEARCH ARTICLE

Spatial and temporal variations of nitrous oxide flux between coastal marsh and the atmosphere in the Yellow River estuary of China

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

Abstract To investigate the spatial and seasonal variations of nitrous oxide (N_2O) fluxes and understand the key controlling factors, we explored N_2O fluxes and environmental variables in high marsh (HM), middle marsh (MM), low marsh (LM), and mudflat (MF) in the Yellow River estuary throughout a year. Fluxes of N_2O differed significantly between sampling periods as well as between sampling positions. During all times of day and the seasons measured, N_2O fluxes ranged from −0.0051 to 0.0805 mg N_2O m^{-2} h^{-1}, and high N_2O emissions occurred during spring (0.0278 mg N_2O m^{-2} h^{-1}) and winter (0.0139 mg N_2O m^{-2} h^{-1}) while low fluxes were observed during summer (0.0065 mg N_2O m^{-2} h^{-1}) and autumn (0.0060 mg N_2O m^{-2} h^{-1}). The annual average N_2O flux from the intertidal zone was 0.0117 mg N_2O m^{-2} h^{-1}, and the cumulative N_2O emission throughout a year was 113.03 mg N_2O m^{-2}, indicating that coastal marsh acted as N_2O source. Over all seasons, N_2O fluxes from the four marshes were significantly different ($p<0.05$), in the order of HM (0.0256±0.0040 mg N_2O m^{-2} h^{-1})>MF (0.0107±0.0027 mg N_2O m^{-2} h^{-1})>LM (0.0073±0.0020 mg N_2O m^{-2} h^{-1})>MM (0.0026±0.0011 mg N_2O m^{-2} h^{-1}). Temporal variations of N_2O emissions were related to the vegetations (*Suaeda salsa*, *Phragmites australis*, and *Tamarix chinensis*) and the limited C and mineral N in soils during summer and autumn and the frequent freeze/thaw cycles in soils during spring and winter, while spatial variations were mainly affected by tidal fluctuation and plant composition at spatial scale. This study indicated the importance of seasonal N_2O contributions (particularly during non-growing season) to the estimation of local N_2O inventory, and highlighted both the large spatial variation of N_2O fluxes across the coastal marsh (CV=158.31 %) and the potential effect of exogenous nitrogen loading to the Yellow River estuary on N_2O emission should be considered before the annual or local N_2O inventory was evaluated accurately.

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

Z. Sun · L. Wang · X. Mou · W. Sun
Shandong Provincial Key Laboratory of Coastal Zone Environmental Processes, YICCAS, Yantai, Shandong 264003, People's Republic of China

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

Nitrous oxide (N_2O) is an important greenhouse gas (GHG) that has 298 times the global warming potential of CO_2 over a 100-year time period and has been recognized to contribute global warming by 5 % (Mosier 1998). N_2O has an atmospheric lifetime of 114 years (Mosier 1998) and also contribute to the depletion of ozone in the stratosphere (Crutzen and Ehhalt 1977). The globally averaged atmospheric N_2O concentration increased from 270±7 ppb in pre-industrial times (before 1750) to 319±0.12 ppb in 2005, and is increasing approximately 0.26 % per year (IPCC 2007). In 2010, the globally averaged N_2O concentration reached 323.2 ppb, which exceeded the highest annual mean abundance so far (World Meteorological Organization 2011). Emission of N_2O from various natural ecosystems has significant influences on global climate change since they account for 44~54 % of the

total N_2O emissions (9.6~10.8 Tg N_2O year^{-1}) (IPCC 2007). Tropical soil and wetlands play an important role in the global nitrogen (N) biogeochemical cycles and are considered significant natural sources of N_2O, contributing approximately 22~27 % towards this inventory (Whalen 2005).

Coastal marsh is characterized by high temporal and spatial variation involved with topographic feature, environmental factors, and astronomic tidal fluctuation, and is very sensitive to global climate changes and human activities. The intertidal zone between terrestrial and coastal ecosystems may represent a high dynamic interface of intense material processing and transport, with potentially high GHG emission (Hirota et al. 2007). Considerable efforts have been made in the past two decades to quantify the N_2O fluxes in different coastal ecosystems, especially in estuarine salt marshes (Shingo et al. 2000; Magalhães et al. 2007), mangrove swamps (Muñoz-Hincapié et al. 2002; 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; Moseman-Valtierra et al. 2011). In China, the studies on N_2O emission from coastal marshes started quite late and the related research mainly focused on the coastal tundra marshes in Antarctica (Sun et al. 2002; Zhu et al. 2008) and the coastal marshes in the Yangtze River estuary (Yang et al. 2006; Wang et al. 2006, 2007a,b; Wang et al. 2010a) and the Min River estuary (Yang et al. 2012; Mou et al. 2012; Zhang et al. 2012, 2013), while information on the coastal marshes in northern estuaries (such as Liao River estuary and Yellow River estuary) was 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 (Xu et al. 2002; 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 the water or salinity gradient and the succession of vegetation from the land to the sea. In recent years, the N and organic matter (OM) loadings of the Yellow River estuary have significantly increased due to the effects of human activities, and approximately 4.22×10^4 tons of nutrients and 4.39×10^5 tons of OM were discharged into Bohai Sea every year (State Oceanic Administration of China 2013). 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, information on N_2O emission from the coastal marsh in the Yellow River estuary is very limited, and the potential effects of exogenous N loading on N_2O emission remains poorly discussed.

In this paper, we investigated N_2O fluxes and environmental variables in the coastal mash of the Yellow River estuary throughout a year (during 2010/2011). The aims of this study are: (1) to determine the spatial and temporal variations of N_2O fluxes and the annual average N_2O emission from the coastal marsh, and (2) to investigate the key factors influencing N_2O variations and assess the potential effects of exogenous N loading on N_2O emission.

Materials and methods

Study site

The study was carried out in the coastal marsh 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 annual average temperature is 12.1 °C, and the average temperatures in spring, summer, autumn, and winter are 10.7, 27.3, 13.1, and −5.2 °C, respectively. 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 to 15 cm. The annual evaporation is 1,962 mm, the annual precipitation is 551.6 mm, with about 70 % of precipitation occurring between June and August.

With an area of 964.8 km^2, 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). In the intertidal zone, natural geomorphology and depositing zones are distinct, and high marsh (HM), middle marsh (MM), low marsh (LM), and mudflat (MF) develop from the land to the sea. The soil in the intertidal zone is dominated by salt soil. *Suaeda salsa*, an annual C_3 plant, is the most prevalent halophytes in the coastal marshes of the Yellow River estuary (Tian et al. 2005). Due to the differences of water and salinity conditions, three *S. salsa* phenotypes are generally formed in HM, MM, and LM, respectively. The HM is predominated by *S. salsa* (>90%) and *Phragmites australis* (<10%), MM is predominated by *S. salsa* (>95%) and *Tamarix chinensis* (<5%), while LM is pure *S. salsa* community (100%) (Mou 2010). 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).

Experimental design

Four sampling positrons were laid in HM, MM, LM, and MF in the intertidal zone of the Yellow River estuary. N_2O fluxes across the sediment–atmosphere interface were measured by using opaque, static, manual stainless steel chambers, and gas chromatography techniques. The chamber (50×50×50 cm) and its base (50×50×20 cm) were made from 0.4-mm thickness stainless steel. Inside the chamber, an electric fan was fixed to stir the air, a thermometer sensor was installed to

measure temperature, a trinal-venthole 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. Outside of the chamber was covered with 2 cm thickness white foam to reduce the impact of direct radiative heating during sampling, which generally caused very little change in temperature between the inside and the outside of the chamber (Teiter and Mander 2005; SØvik and KlØve 2007; Song et al. 2008; Jiang et al. 2010). All the connections were made "air tight" and sealed by silicon rubber. The stainless steel bases enclosed an area of 0.25 m^2 and were inserted into the ground to a depth of 20 cm below the soil on August 2010. During observations, the chamber was placed over the base filled with water in the groove to prevent leakage, and the plant was covered within the chamber.

Sampling campaigns were undertaken in September, October, November, and December in 2010, and April, May, June, and July in 2011 (the sampling in January, February, and March were canceled due to the frequent effects of storm tide and bad weather and that in August was canceled due to the damage of most chambers and instruments). Each measurement campaign consisted of 12 chambers set up at above-mentioned four positions (three chambers per site). On each sampling date, measurements were conducted at 0700, 0930, 1200, 1430, and 1700 hours (represented different times of day). About 60 ml gas sample inside the chamber was collected every 20 min over a 60-min period by using 100-ml syringe (total of four samples), and then stored in pre-evacuated gas sampling bags (100 ml). Since the tide in the Yellow River estuary is irregular semi-diurnal tide, the sampling campaigns in the LM and MF were sometimes affected by tidal inundation. The sampling campaigns in the LM and MF in May and the MF in June were not carried out due to the great influence of tide.

N$_2$O concentrations of gas samples were analyzed within 36 h using gas chromatography (Agilent 7890A) equipped with ECD. The N$_2$O portion was separated using a 1-m stainless steel column with an inner diameter 2-mm Porapak Q (80/100 mesh), and was measured using the ECD, which was set at 330 °C. The ECD also used high-pure nitrogen as a carrier gas, a flow rate of 35 ml min^{-1}. The column temperatures were maintained at 55 °C for all separations. Gas concentrations were quantified by comparing peak areas of samples against standards run every eight samples, ensuring each sample run maintained RSD below 6 %.

N$_2$O fluxes were calculated according to the following equation:

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

where dc/dt is the slope of the gas concentration curve variation along with time. M is the mole mass of each gas.

P is the atmospheric pressure in the sampling site. T is the absolute temperature during sampling. V_0, T_0, P_0 are the gas mole volume, air absolute temperature, and atmospheric pressure under standard conditions, respectively. H is the height of chamber above the ground/water surface.

The annual average N$_2$O flux was calculated by the data determined in the eight sampling months (covered four seasons). The N$_2$O emission/absorption (milligram of N$_2$O per meter squared) per sampling month was calculated by the average value (milligram of N$_2$O per meter squared per hour) of all sampling data and the time (hour) in each month. The N$_2$O emissions/absorptions in January and February were estimated by the average value in winter (December), and those in March and August were estimated by the average values determined in spring (April and May) and summer (June and July), respectively. The cumulative N$_2$O emission/absorption throughout a year was estimated by the values in 12 months.

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 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 2010 since the topsoil was frozen. Because the measurements in LM and MF in September 2010 were partly and slightly affected by tide (from 1200 to 1700 hours), the flooding depths were measured to discuss the effects of tidal inundation on N$_2$O emission. On each sampling date, two soil samples per layer (0–10, 10–20 cm) were taken in each site 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).

Statistical analysis

The Shapiro–Wilk test was applied to identify the normality of data before the related statistical analyses were conducted. The results were presented as means of the replications, with standard error. Statistical significance of differences at $p<0.05$ between samples was 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

Environmental variables in coastal marsh

Similar variations of air temperature and ground temperature in the four marshes were observed over all sampling period (Fig.1a). Air temperature did not show significant difference among the four marshes ($p > 0.05$) and the means were 19.69, 17.90, 15.74, and 15.00 °C, respectively. Ground temperatures generally decreased with increasing soil depth, but no significant differences were found within the four marshes ($p > 0.05$). Dissimilar variations of soil moisture and EC in the four marshes were observed (Fig.1b, c). With increasing depth, soil moisture increased while EC generally decreased. Soil moisture did not show significant differences among the four marshes ($p > 0.05$), while significant differences of EC were observed ($p < 0.05$). Seasonal dynamics of soil substrate in the four marshes were observed over all sampling period (Fig. 2). TC, TN, and NH_4^+–N in the surface and subsurface soil of MF were generally higher than those in other marshes (Fig.2a–c). Both TC in surface and subsurface soil had significant differences within the four marshes ($p < 0.05$), while only TN in subsurface soil showed significant difference ($p < 0.01$). Both NH_4^+–N and NO_3^-–N in soil were not significantly different among the four marshes ($p > 0.05$).

Spatial variations of N$_2$O fluxes

Variation of N$_2$O fluxes in spring

N$_2$O fluxes in spring averaged between −0.0102 and 0.0982 mg N$_2$O m^{-2} h^{-1} and differed significantly among the four marshes ($p < 0.01$) (Fig. 3). With the exception of MM, the other sites were found to release N$_2$O during all times of day sampled. In HM, N$_2$O fluxes in April and May were similar except for 1700 hours sampling, and significantly higher emission occurred in April compared to May ($p < 0.01$). N$_2$O fluxes from MM in April and May were opposite except for 1200 hours sampling, and the ranges were −0.0065~0.0131 and −0.0102~0.0081 mg N$_2$O m^{-2} h^{-1}, respectively. N$_2$O fluxes from LM in April had no significant variation before 1430 hours and a significant peak occurred in 1700 hours. In MF, N$_2$O fluxes in April ranged from 0.0285 to 0.0512 mg N$_2$O m^{-2} h^{-1} and the maximum occurred in 1200 hours. The mean N$_2$O fluxes from HM, MM, LM, and MF in spring were 0.0582, 0.0026, 0.0069, and 0.0384 mg N$_2$O m^{-2} h^{-1}, and the cumulative N$_2$O emissions were 128.02, 5.75, 15.19, and 84.74 mg N$_2$O m^{-2}, respectively, indicating that coastal marsh represented N$_2$O source.

Variation of N$_2$O fluxes in summer

N$_2$O fluxes in summer ranged from −0.0147 to 0.0262 mg N$_2$O m^{-2} h^{-1} and differed significantly among the four marshes in June ($p < 0.05$) (Fig. 3). With the exception of HM that were found to release N$_2$O during all times of day sampled, the other sites showed consumptions in some sampling times. N$_2$O fluxes from HM in June and July were 0.0085~0.0262 and 0.0117~0.0206 mg N$_2$O m^{-2} h^{-1}, respectively, and no significant difference was found between them ($p > 0.05$). In MM, N$_2$O fluxes in June and July were significantly different ($p < 0.05$) and the means were −0.0051 and 0.0052 mg N$_2$O m^{-2} h^{-1}, respectively. Although N$_2$O fluxes from LM in June and July were opposite except for 1430-hours sampling, they had no significant difference ($p > 0.05$). The MF was found to release N$_2$O before July 1430 hours sampling and significant consumption occurred in 1700-hours. The average N$_2$O fluxes from HM, MM, LM, and MF in summer were 0.0165, 0.00002, 0.0035, and 0.0055 mg N$_2$O m^{-2} h^{-1}, and the cumulative N$_2$O emissions were 36.54, 0.17, 7.92, and 12.14 mg N$_2$O m^{-2}, respectively, indicating that coastal marsh acted as weak N$_2$O source.

Variation of N$_2$O fluxes in autumn

N$_2$O fluxes in autumn averaged between −0.0092 and 0.0312 mg N$_2$O m^{-2} h^{-1} and differed significantly among the four marshes in October ($p < 0.01$) (Fig. 3). In HM, with the exception of September 0700 hours sampling, the other times were found to release N$_2$O. Significantly higher emission occurred in October compared to September and November ($p < 0.01$). N$_2$O fluxes from MM in autumn showed both emission and consumption, but no significant difference was found among sampling periods ($p > 0.05$). N$_2$O fluxes from LM in September and October were opposite except for 0700 and 0930 hours sampling. N$_2$O fluxes from MF in September and November were similar, but they had significant difference ($p < 0.01$). The LM in November and MF in October were found to release N$_2$O over all sampling times, with the maximums occurred in 1200 and 0700 hours, respectively. The mean N$_2$O fluxes from HM, MM, LM, and MF in autumn were 0.0118, 0.0031, 0.0038, and 0.0053 mg N$_2$O m^{-2} h^{-1}, and the cumulative N$_2$O emissions were 26.01, 6.90, 8.16, and 11.70 mg N$_2$O m^{-2}, respectively, indicating that coastal marsh represented weak N$_2$O source.

Variation of N$_2$O fluxes in winter

N$_2$O fluxes in winter ranged from −0.0114 to 0.0442 mg N$_2$O m^{-2} h^{-1} and differed significantly among the four marshes ($p < 0.01$) (Fig. 3). The average N$_2$O fluxes from HM, MM, LM, and MF were 0.0195, 0.0059, 0.0257, and
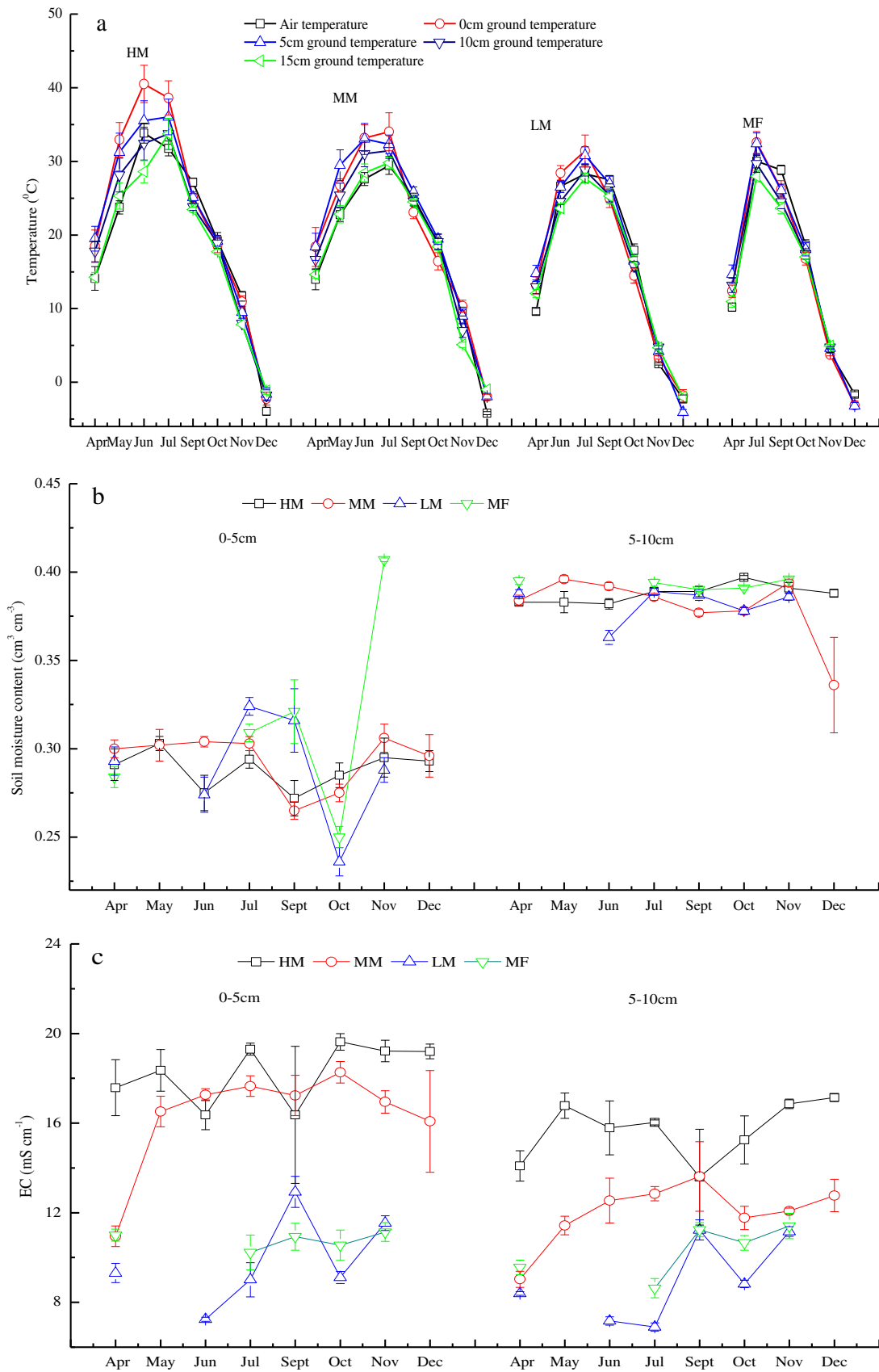
Fig. 1 Variations of environmental temperatures (**a**), soil moisture content (**b**), and electrical conductivity (*EC*) (**c**) in high marsh (*HM*), middle marsh (*MM*), low marsh (*LM*), and mudflat (*MF*)
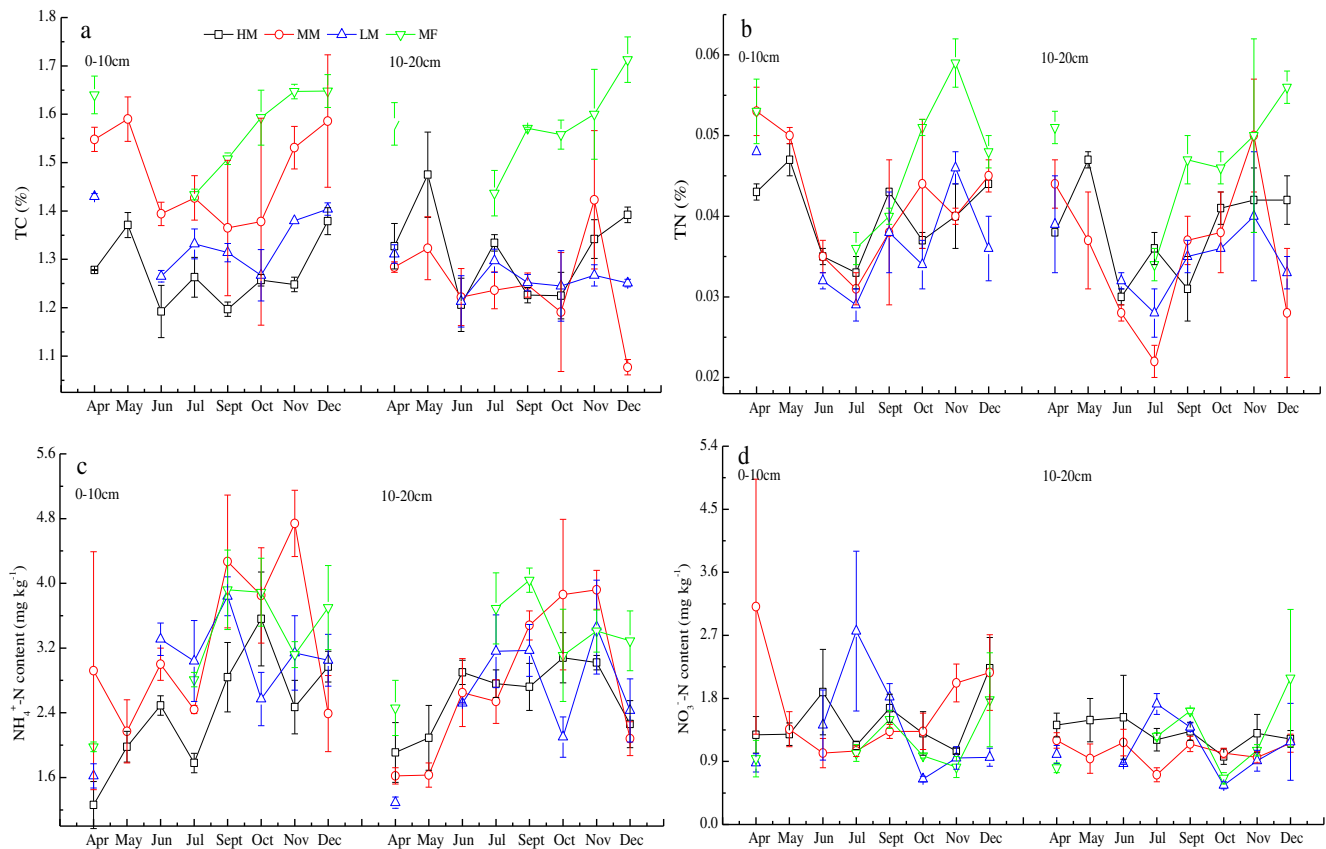
Fig. 2 Variations of TC (**a**), TN (**b**), NH_4^+–N (**c**), and NO_3^-–N (**d**) contents in high marsh (*HM*), middle marsh (*MM*), low marsh (*LM*), and mudflat (*MF*) soils

0.0047 mg N_2O m^{-2} h^{-1}, and the cumulative N_2O emissions were 42.12, 12.79, 55.47, and 10.11 mg N_2O m^{-2}, respectively, indicating that coastal marsh acted as N_2O source. Significantly higher emissions were observed in HM and LM compared to MM and MF during all times of day sampled ($p<0.05$).

Temporal variations of N_2O fluxes

Significant temporal variations of N_2O fluxes throughout a year were observed in HM, LM, and MF ($p<0.01$) (Fig. 4). During

all times of day and the seasons measured, N_2O fluxes from the intertidal zone averaged between −0.0051 and 0.0805 mg N_2O m^{-2} h^{-1}, and the annual average N_2O flux was 0.0117 mg N_2O m^{-2} h^{-1}. The maximum and minimum were observed in April (in HM) and June (in MM), respectively. High N_2O emissions generally occurred during spring (0.0278 mg N_2O m^{-2} h^{-1}) and winter (0.0139 mg N_2O m^{-2} h^{-1}) while low fluxes were observed during summer (0.0065 mg N_2O m^{-2} h^{-1}) and autumn (0.0060 mg N_2O m^{-2} h^{-1}) (Fig. 4). Over all seasons, N_2O fluxes from the four marshes were significantly different ($p<0.05$), in the order of HM (0.0256±0.0040 mg N_2O

Fig. 3 Spatial variations of N_2O fluxes (milligram of N_2O per meter squared per hour) from high marsh (*HM*), middle marsh (*MM*), low marsh (*LM*), and mudflat (*MF*) in spring (April and May), summer (June and July), autumn (September, October, and November), and winter (December)
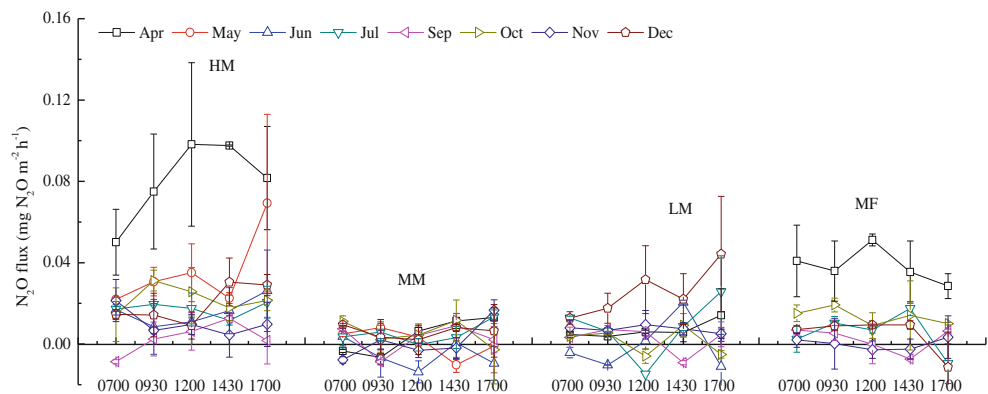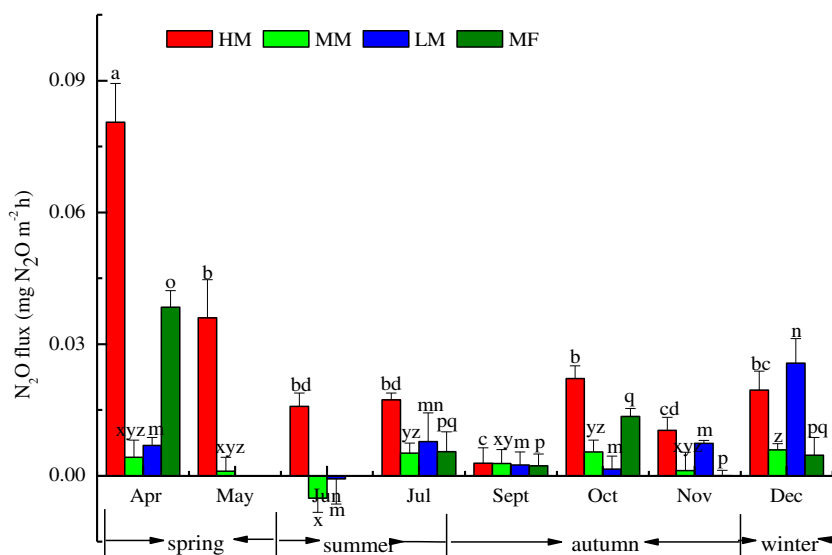
Fig. 4 Temporal variations of N$_2$O fluxes (milligram of N$_2$O per meter squared per hour) from high marsh (*HM*), middle marsh (*MM*), low marsh (*LM*), and mudflat (*MF*). Bars with *different letters* (*a, b, c, d* for *HM*; *x, y* for *MM*; *m, n* for *LM*; *o, p, q* for *MF*) are significantly different at the level of *p*<0.05; bars with *same letters* are not significantly different at the level of *p*<0.05

m^{-2} h^{-1})>MF (0.0107±0.0027 mg N$_2$O m^{-2} h^{-1})>LM (0.0073±0.0020 mg N$_2$O m^{-2} h^{-1})>MM (0.0026±0.0011 mg N$_2$O m^{-2} h^{-1}). The cumulative N$_2$O emission from the intertidal zone throughout a year was 113.03 mg N$_2$O m^{-2}, indicating that coastal marsh acted as N$_2$O source. Comparatively, the N$_2$O emissions measured during spring and winter contributed 49.65 and 26.65 % of the total emission, respectively, which were higher than the contributions measured during summer (12.03 %) and autumn (11.67 %).

Relationships between environmental variables and N$_2$O fluxes

Most correlations between N$_2$O fluxes and temperatures in different marshes were not significant (*p*>0.05; Table 1). Although both positive and negative impacts of soil moisture (or EC) on N$_2$O emissions were observed within the four marshes, only the correlation between soil moisture (5–10 cm) and N$_2$O fluxes in LM was significant (*p*<0.05; Table 2). Lacks of correlations between N$_2$O fluxes and substrate variables were observed (*p*>0.05) except the correlations occurred in subsurface soil of HM and MF (*p*<0.05; Table 3). The environmental variables determined in the four coastal marshes were all excluded in the stepwise liner regression. NH$_4^+$–N content (X_1) and TC content (X_2) were the dominant factors that controlled the N$_2$O emissions (Y) in HM (Y=0.129−0.040X_1, R^2=0.539, *p*=0.038) and LM (Y=−0.109+0.090X_2, R^2=0.798, *p*=0.016), respectively, while in MM and MF, the environmental variables determined during sampling periods were all excluded, indicating that N$_2$O fluxes were controlled by multiple site-specific factors.

Discussion

Temporal variations of N$_2$O fluxes

Many studies have demonstrated that the seasonal patterns of N$_2$O fluxes were generally governed by seasonal variability in temperatures since it influenced water availability, production of substrate precursors, and microbial activity (Whalen 2005; Zhu et al. 2008). However, we have drawn a different result. Although significant temporal variations in N$_2$O fluxes from the coastal marshes in the Yellow River estuary were observed, the influence of seasonal variability in temperatures on N$_2$O emissions seemed not significant (Figs. 1a and 4). Moreover, most correlations between N$_2$O fluxes and temperatures in different marshes were not significant (*p*>0.05) (Table 1). These indicated that the influences of temperatures on N$_2$O emissions might be covered by other biotic/abiotic parameters (such as vegetation, soil moisture, and substrate) in most sampling periods. Because the environmental variables determined in coastal marsh were all excluded in the stepwise liner regression, we considered that N$_2$O emissions in different seasons might be controlled by the interactions of multiple controlling factors.

In this paper, we observed that N$_2$O emissions in spring, summer, autumn, and winter were 0.0278, 0.0065, 0.0060, and 0.0139 mg N$_2$O m^{-2} h^{-1}, respectively (Fig. 4), and low values occurred during summer and autumn. The result was similar with Allen et al. (2007) but was different with Wang et al. (2007a) and Søvik and Kløve (2007). There were two possible reasons. Firstly, the presence of vegetations (*P. australis*, *S. salsa*, and *T. chinensis*) across the coastal marsh might have significant effects on the low N$_2$O emissions during the growing season. Many studies have demonstrated that, under flooding or anaerobic conditions, the three mash

Table 1 Pearson correlation analysis between N_2O fluxes and temperatures

Month	Air temperature				0-cm ground temperature				5-cm ground temperature				10-cm ground temperature				15-cm ground temperature			
	HM	MM	LM	MF	HM	MM	LM	MF	HM	MM	LM	MF	HM	MM	LM	MF	HM	MM	LM	MF
April	0.598	−0.062	0.035	0.529	0.783	0.345	−0.288	0.131	0.936*	0.743	0.292	−0.101	0.800	0.885*	0.548	−0.232	0.719	0.927*	0.587	−0.318
May	0.118	−0.179	–	–	0.329	0.251	–	–	0.462	−0.632	–	–	0.469	−0.762	–	–	0.573	−0.783	–	–
June	0.285	−0.562	0.396	–	−0.100	−0.503	0.480	–	0.336	−0.492	0.530	–	0.461	−0.441	0.333	–	0.619	−0.400	0.456	–
July	−0.554	−0.626	−0.848	0.218	−0.453	−0.534	−0.826	0.354	−0.287	−0.545	−0.915*	0.116	−0.214	−0.297	−0.506	−0.066	−0.214	0.090	−0.184	−0.228
September	0.948*	0.062	0.040	−0.014	0.785*	0.016	0.387	0.363	0.878*	0.094	−0.209	−0.449	0.723	0.238	−0.573	−0.629	0.616	0.372	−0.705	−0.813
October	0.314	0.391	0.385	−0.176	0.514	0.350	0.185	−0.079	0.530	−0.153	0.255	−0.295	0.474	−0.238	0.002	−0.351	0.559	−0.448	−0.076	−0.192
November	−0.103	0.181	−0.435	−0.682	0.048	−0.180	0.367	−0.364	0.204	0.385	−0.047	−0.776	0.496	0.605	−0.173	−0.791	0.583	0.731	−0.173	−0.061
December	0.343	0.165	0.290	0.444	−0.108	−0.209	0.488	0.344	0.413	−0.554	0.721	−0.468	0.733	–	–	–	0.808	–	–	–

Pair sample size, $n=8$ for air temperature and ground temperatures (0, 5, 10, and 15 cm) in HM; $n=8$ for air temperature and 0-, 5-, and 15-cm ground temperatures, and $n=7$ for 10-cm ground temperature in MM; $n=7$ for air temperature and ground temperatures (0, 5, 10, and 15 cm) in LM; $n=6$ for air temperature and ground temperatures (0, 5, 10, and 15 cm) in LM; $n=5$ for air temperature and 0- and 5-cm ground temperatures, and $n=7$ for 10- and 15-cm ground temperatures in MF

HM high marsh, MM middle marsh, LM low marsh, MF mudflat

*$p=0.05$ correlations are significant

Table 2 Pearson correlation analysis between N_2O fluxes and soil moisture or electrical conductivity (EC)

Sites	Soil moisture		Electrical conductivity (EC)	
	0–5 cm	5–10 cm	0–5 cm	5–10 cm
HM	0.296	−0.419	−0.053	−0.321
MM	−0.382	−0.532	−0.192	−0.049
LM	0.577	0.839*	0.329	0.135
MF	−0.605	0.165	0.153	−0.407

Pair sample size, $n=8$ for soil moisture and EC in 0–5 and 5–10 cm depths in HM and MM; $n=6$ for soil moisture and EC in 0–5 and 5–10 cm depths in LM; $n=5$ for soil moisture and EC in 0–5 and 5–10 cm depths in MF

HM high marsh, MM middle marsh, LM low marsh, MF mudflat

*$p=0.05$ correlations are significant

plants could transport oxygen from aboveground parts to roots by aerenchyma (Han et al. 2005; Ling et al. 2008; Kong et al. 2008; Ge and Zhang 2011), which generally formed oxidizing microenvironment around rhizosphere soil (Kong et al. 2008). Moreover, the roots of the three plants could excrete some small molecular compounds (glucide, organic acid, and amion acid), which caused the microorganism amount and microbial activity in rhizosphere soil to be much higher than those in non-rhizosphere soil (Ling et al. 2008; Cheng 2009; Wang et al. 2010b; Ge and Zhang 2011). These indicated that although the coastal marshes were frequently influenced by tidal inundation, the nitrification–denitrification rate still could be accelerated by the three plants since the soil microbes in rhizosphere were generally supplied with available C and N and proper amount of oxygen. Thus, N_2O, to a great extent, was reduced to N_2 by denitrification regardless of whether N_2O was produced by nitrification or denitrification or both, which resulted in the great decrease of N_2O emissions. Secondly, as mentioned before, N_2O production was through nitrification and denitrification, in which microorganism used the C and mineral N (NH_4^+–N and NO_3^-–N) as substrates (Conrad 1996). The low N_2O emissions in summer might also be due to the limited C and mineral N in the soils caused by low mineralization rates of organic C and N. As shown in Fig. 2, compared to other seasons, TC and TN in different coastal marsh soils (0–10, 10–20 cm) were lower in summer and NH_4^+–N were lower during spring and summer, indicating that the shortage of C and mineral N during summer was unfavorable for N_2O production. Chapuis-lardy et al. (2007) also pointed out that N_2O production seemed to be suppressed by low mineral N and high moisture contents in soil from analysis a large number of literatures. The weak N_2O emissions in summer were probably because that the mineral N was almost used up by plants. This speculation could be supported by the evidence that the biggest biomass

Table 3 Pearson correlation analysis between N_2O 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
HM	0.292	0.250	0.317	0.232	−0.611	−0.734[*]	−0.211	0.219
MM	0.198	−0.310	0.230	−0.020	−0.079	−0.059	0.369	−0.194
LM	0.673	0.194	0.078	−0.120	−0.056	−0.028	−0.125	0.281
MF	0.298	−0.293	0.251	0.019	−0.700	−0.851[*]	−0.316	−0.515

Pair sample size, $n=8$ for TC, TN, NH_4–N, and NO_3^-–N in 0–10 and 10–20 depths in HM and MM; $n=7$ for TC, TN, NH_4–N, and NO_3^-–N in 0–10 and 10–20 depths in LM; $n=6$ for TC, TN, NH_4–N, and NO_3^-–N in 0–10 and 10–20 depths in MF

HM high marsh, *MM* middle marsh, *LM* low marsh, *MF* mudflat

*$p=0.05$ correlations are significant

accumulation rate (Mou et al. 2010) coincided with the lowest level of C and mineral N in the soils (Fig. 2) at this period. This study also showed that high N_2O emissions occurred during spring and winter (Fig. 4). Many studies have demonstrated 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). Because the coastal marsh of the Yellow River estuary located in the mid-latitude region (37 °35′N~38 °12′N) and the freeze/thaw cycles frequently occurred in topsoil in majority days during spring and winter (frozen depth, 0~15 cm), high N_2O emissions might be attributed to the frequent freeze/thaw cycles. For one thing, frequent freeze/thaw cycles destroyed the size and stability of soil aggregate (van Bochove et al. 2000), released abundant bioavailable C and N (Ludwig et al. 2006), and altered the course and intensity of soil N transformation (Jarvis et al. 1996), which enhanced the denitrification and N_2O emission (Priemé and Christensen 2001). For another, since the frozen water film on the soil matrix represented a diffusion barrier which reduced oxygen supply to the microorganisms and partly prevented the release of the N_2O (Teepe et al. 2001), high emissions occurred due to the quick release of N_2O trapped by ice layer and/or denitrification during frequent freeze/thaw cycles (Goodroad and Keeney 1984; Teepe et al. 2001). Similar results were drawn by Zhang et al. (2005) and Jiang et al. (2010) who found significant N_2O emissions from freshwater marsh in the Sanjiang Plain and Alpine meadow in the Qinghai–Tibetin Plateau during the freeze/thaw cycle as the temperature increased.

In this study, the N_2O emission per sampling month was calculated by the average value and the time in each month and the emissions in absent months were estimated by the average values in the corresponding seasons. Similar method was adopted in some studies to estimate the N_2O emissions in absent months. Hao et al. (2007) studied the effects of freshwater marshes (*Carex lasiocapa* marsh and *Deyeuxia*

angustifolia marsh) reclamation on N_2O fluxes and estimated the emission in absent month (February) by the average value in winter (December and January). Sun et al. (2009) investigated the N_2O fluxes from *Calamagrostis angustifolia* marsh in the Sanjiang Plain and estimated the emissions in absent months (February and October) by the average values in winter (December and January) and autumn (September and November), respectively. Since the measurements in this study covered four seasons [spring (April and May), summer (June and July), autumn (September, October, and November), and winter (December)], we considered that based on the average values in the corresponding seasons, the extrapolation of N_2O emissions in absent months, to a great extent, was valid as the local N_2O inventory throughout a year was assessed roughly. Although only the roughly accumulated N_2O emission was given in our study, considering it was estimated for the first time, it still would provide valuable information on understanding the N_2O inventory in the coastal marsh of the Yellow River estuary. Overall, across all the seasons sampled, the coastal marsh was a net source of N_2O (113.03 mg N_2O m^{-2} year^{-1}) and the N_2O emissions measured during non-growing season (spring and winter) comprised the principal part to the total emission (76.30 %), indicated that the importance of seasonal N_2O contributions (particularly during non-growing season) to the estimation of local N_2O inventory should be paid more attentions. In the following studies, in order to assess the regional budget of N_2O emissions precisely, measurements should be conducted at all months and the sampling frequency should be increased.
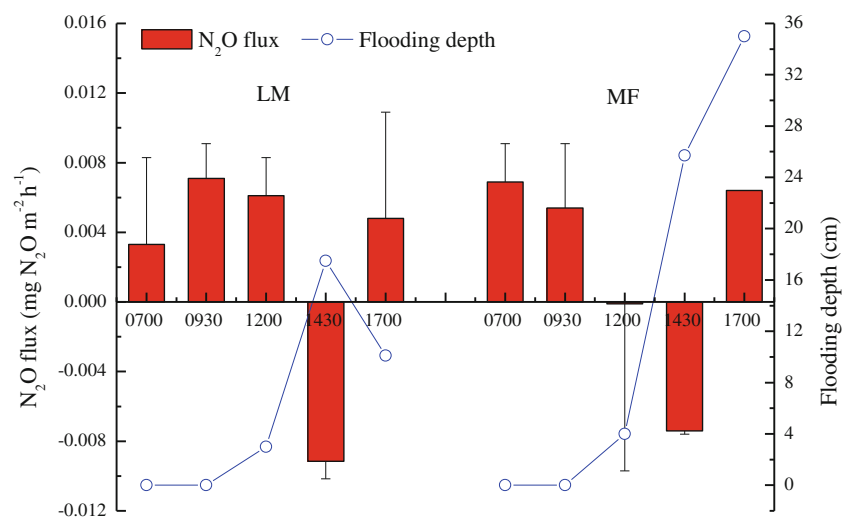
Spatial variations of N_2O fluxes

During all times of day and the seasons measured, we found that the physical and chemical parameters of soil differed in their magnitude among the four marshes, and significant differences in TC, TN, and EC in soil were observed ($p<0.05$). Such differences within the four marshes would be due to the site-specific conditions (such as topography, slope, hydrology, and species composition) which influence the magnitudes and

variations of N_2O at spatial scale (Allen et al. 2007; Hirota et al. 2007). Over all sampling seasons, we observed that N_2O fluxes from the four marshes were significantly different ($p < 0.05$), in the order of HM>MF>LM>MM (Fig. 4). Also, a large spatial variation of N_2O fluxes in the coastal marsh of the Yellow River estuary was observed. The coefficient of variations (CVs) of N_2O fluxes in the four marshes were 98.47, 278.29, 164.56, and 138.12 %, respectively, while the value across the coastal marsh was 158.31 %, indicating that to assess the regional budget of N_2O emissions precisely, measurements should be designed at fine scales and the number of spatial replicates should be increased. Previous studies have indicated that temperatures had great influences on N_2O emissions at spatial scale (Alongi et al. 2005; Gregorich et al. 2006), but this study has drawn a different result. During all the seasons measured, air temperature and ground temperatures did not show significant difference among the four marshes ($p > 0.05$), most correlations between N_2O fluxes and temperatures in different marshes were not significant ($p > 0.05$) and only few significant positive or negative correlations were found in HM or MM (Table 1). This indicated that the function of thermal condition might be covered by the interactions of other biotic or abiotic factors (such as moisture, salinity, sediment substrate, and vegetation) though it was considered an important factor affecting N_2O emission. Although EC showed significant differences within the four marshes ($p < 0.05$), the correlations between EC and N_2O emission were not significant ($p > 0.05$) (Table 2). By comparison, soil moisture did not show significant differences among the four marshes ($p > 0.05$), but significant positive correlation between soil moisture (5–10 cm) and N_2O emission was found in LM ($p < 0.05$) (Table 2). Generally, both positive and negative impacts of soil moisture (or EC) on N_2O emissions were observed in coastal marshes (Table 2), which were different with mostly previous studies that N_2O emissions had negative correlation with moisture (Regina et al. 1996; Wang et al. 2005) or EC (Law

et al. 1991; Dalal et al. 2003; Chen et al. 2010). One possible explanation for the positive correlations between EC and N_2O emissions was that the salinity in LM and MF was much lower than that in HM and MM (Fig.1c), which might not completely inhibit the activities of nitrifiers and denitrifiers in soil (Lv et al. 2008). The positive correlations between soil moisture and N_2O emissions might be partly dependent on the fluctuation of soil moisture (or water level) by astronomic tide. As shown in Fig. 5, dissimilar variations of N_2O emissions and flooding depths in LM and MF were observed. Both LM and MF were found to release N_2O at 0700 and 0930 hours sampling (before flood), indicating that the proper soil moisture might contribute to a favorable aerobic–anerobic status for N_2O production. As the flood began at 1200 hours sampling, the N_2O flux in LM decreased and the value in MF became negative, indicating that the soil moisture in LM and MF might be greatly changed due to the different flooding depths, which produced different impacts on N_2O emission. When the flooding was deeper at 1430 hours sampling, both LM and MF showed great consumptions, indicating that the tidal inundation produced an unfavorable anaerobic status for N_2O production and the limited N_2O emission might be severely prevented by flooding seawater. Similar result was drawn by Zhang et al. (2013) who found that tidal inundation significantly decreased the N_2O emission in the coastal marsh of the Min River estuary. Although the flooding depth in LM decreased and that in MF increased greatly at 1700 hours sampling, both LM and MF were found to release N_2O greatly, which was mainly dependent on the N_2O transportation from surface seawater to the two marshes by tidal fluctuation (Senga et al. 2001; Hirota et al. 2007). In addition, the decrease of flooding depth in LM might cause the dissolved N_2O in seawater to be released, which partly contributed to the significant N_2O emission. Similar result was drawn by Hirota et al. (2007) who found that in coastal ecosystems subjected to such short-term fluctuation of water level (or soil moisture) by astronomic tide, the

Fig. 5 Variations of N_2O flux and flooding depth in low marsh (*LM*) and mudflat (*MF*) in September 2010

spatial variations in N_2O flux was controlled by fluctuation of water lever ($r=0.58$, $p<0.05$).

Site-level control of N_2O emission was also attributed to the effects of vegetation and nutrient status. In this study, the plant distributed continuously across the coastal marsh and the plant compositions in the four mashes were different. The coverage and maximum biomass of S. salsa–P. australis community (HM) were 1.19- and 1.90-folds and 1.60- and 3.57-folds of S. salsa–T. chinensis community(MM) and S. salsa community (LM), respectively (Mou 2010; Dong et al. 2010). Also, the presence of P. australis, T. chinensis, and S. salsa had great impacts on N_2O emission as mentioned previously. These indicated that the vegetations across the coastal marsh might play an important role in controlling the N_2O emissions at spatial scale. Although both TC and TN in soils showed significant differences among the four marshes ($p<0.05$), lacks of correlations between N_2O fluxes and TC (or TN) were observed ($p>0.05$) (Table 3). By comparison, although both NH_4^+–N and NO_3^-–N had no significant differences within the four marshes ($p>0.05$), significant correlations between N_2O fluxes and NH_4^+–N could be observed in subsurface soil of HM and MF ($p<0.05$) (Table 3). C and N were very important substrates for N_2O production (participated in nitrification and denitrification processes) (Tauchnitz et al. 2008) and they generally influenced N_2O emissions by C/N regulations and interactions with other abiotic variables (Blackwell et al. 2010). In this study, negative correlations between N_2O emissions and nutrient variables were generally observed in the four marshes (Table 3), which was different with mostly previous studies (Aelion et al. 1997; Muñoz-Hincapié et al. 2002; Chen et al. 2010). One possible reason was related to the interaction of vegetation and microorganism (nitrifiers and denitrifiers) during N_2O production (Li et al. 2002). N_2O production might be greatly inhibited as the available N was significantly competed by both vegetations and microorganisms in S. salsa–P. australis community, S. salsa–T. chinensis community, and S. salsa community, which partly contributed to the difference of N_2O emissions at spatial scale. Since there was no vegetation in MF, the negative correlations between N_2O emissions and nutrient variables were possibly correlated with the high soil moisture (Fig.1b). Under high moisture condition, the NO_3^-–N in topsoil could be easily transferred to the deep soil layer, which decreased the chance to participate in nitrification and denitrification processes. As shown in Fig.2d, the NO_3^-–N in topsoil of MF was very low, which could partly explain the negative influences on N_2O production and emission.

Comparisons with other measurements and potential of N loading on N_2O emissions

Previous studies that have examined the release of N_2O from different coastal marshes and mangrove swamps reported about the fluxes in the range of −0.1298 to 0.1953 mg N_2O m^{-2} h^{-1} (Alongi et al. 2005; Xie et al. 2011) (Table 4). The magnitudes of N_2O fluxes determined in this study (−0.0051~0.0805 mg N_2O m^{-2} h^{-1}) were in the range, which were higher than those from coastal marshes in the Yangtze River estuary (−0.0096~0.0079 mg N_2O m^{-2} h^{-1}) and the Min River estuary (0.0037~0.0157 mg N_2O m^{-2} h^{-1}), and mangrove swamps in the Moreton Bay (−0.002~0.014 mg N_2O m^{-2} h^{-1}), but matched emissions recorded at coastal tundra marshes in the eastern Antarctica (−0.0206~0.0856 mg N_2O m^{-2} h^{-1}), coastal marshes in the coastal lagoon of Lake Nakaumi (−0.01~0.06 mg N_2O m^{-2} h^{-1}) and mangrove swamps in the Brisbane River (−0.004~0.065 mg N_2O m^{-2} h^{-1}) and Magueyes Island (0.0022~0.0616 mg N_2O m^{-2} h^{-1}) (Table 4).

In this paper, we found that the coastal marsh acted as a N_2O source (cumulative N_2O emission throughout a year was 113.03 mg N_2O m^{-2}) in the present N loading of the Yellow River estuary. Numerous studies have demonstrated that exogenous N generally had great stimulatory effects on the production and emission of N_2O (Lee et al. 1997; Muñoz-Hincapié et al. 2002; Liikanen et al. 2003; Song et al. 2006; Zhang et al. 2007; Stadmark and Leonardson 2007; Li et al. 2009, 2010; Zhang et al. 2012, 2013), but the promoted magnitude of N_2O flux to N enrichment varied due to the N addition level (Song et al. 2006; Zhang et al. 2007; Li et al. 2009, 2010; Zhang et al. 2012; Mou et al. 2012) and N forms (NH_4^+–N and NO_3^-–N) (Smith et al. 1983; Lindau and DeLaune 1991; Cartaxana and Lloyd 1999; Muñoz-Hincapié et al. 2002; Wan et al. 2009). Wang (2011) studied the responses of N enrichment (NH_4^+–N) on the N_2O production of coastal marsh soil in the Yellow River estuary, and found that the additions of NH_4^+–N had great stimulation on N_2O production, with approximately 1.93~3.71-folds of N_2O production being observed with increasing NH_4^+–N addition. Denitrification was the most important process for N_2O production and its contribution to total N_2O production would also be elevated with increasing NH_4^+–N addition (Wang 2011). The increase in N_2O emission under N addition was probably caused by the enhancement of both nitrifiers and denitrifiers activities (Wrage et al. 2004). At present, the exogenous N loading (NH_4^+–N is dominated) of the Yellow River estuary is increasing due to human activities (State Oceanic Administration of China 2013). 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 N_2O emission. As shown in Table 4, the mean N_2O flux from Futian mangrove (0.7572 mg N_2O m^{-2} h^{-1}) and the maximum N_2O emission from Mai Po mangrove (0.4176 mg N_2O m^{-2} h^{-1}) in the Deep Bay region of South China recorded by Chen et al. (2010) were 9.41 and 5.19 times greater than the maximum N_2O emission reported by our study, and the reason

Table 4 Literature data of N$_2$O emissions from different marshes

Marsh types	Location	Vegetations		N$_2$O fluxes[a] (mg m^{-2} h^{-1})	Observation period	References
Coastal marsh	Yellow River estuary, China	*Phragmites australis, Suaeda salsa, Tamarix chinensis*		0.0117 (−0.0051~0.0805)[b]	September 2010~July 2011	This study
	Yangtze River estuary, China	*Scirpus mariqueter*		0.0699	July~August 2004	Wang et al. (2007)
		Mudflat		−0.0276		
	Min River estuary, China	*P. australi, S. mariqueter*		(−0.0096~0.0079)[b]	April~November 2005	Wang et al. (2010a)
		Cyperus malaccensis		(0.0195~0.0514)[b]	September~October 2011	Zhang et al. (2012)
		Spartina alterniflora		0.0108 (−0.0230~0.0466)[b]	September~October 2011	Zhang et al. (2013)
				0.0092 (0.0037~0.0157)[b]	March 2012	
	Dagu estuary (Jiaozhou Gulf), China	*P. australis*		(−0.1289~0.1511)[b]	September 2009~Feburary 2010	Xie et al. (2011)
	Eastern Antarctica (coastal tundra marsh)	*Wolong*	Wet tundra	0.0069 (−0.0206~0.0856)[b]	December 2005~February 2006	Zhu et al. (2008)
			Mesic tundra	0.0018 (−0.0053~0.0508)[b]		
			Dry tundra	0.0198 (0.0064~0.0271)[b]		
		Tuanjie	Ponds	0.0041 (−0.0059~0.0126)[b]		
			Shallow fens	0.0105 (−0.0036~0.0121)[b]		
			Dry tundra	0.0248 (0.0128~0.0398)[b]		
	Lake Nakaumi (coastal lagoon), Japan	*Carex rugulosa, P. australis, Solidago altissima*		0.02 (−0.01~0.06)[b]	August 2003	Hirota et al. (2007)
		Sand shore		0.03 (0.02~0.05)[b]		
	Fildes Peninsula, Maritime Antarctica	*Adenocystis utricularis*		0.0018 (0.0008~0.0025)[b]	January~March 2000	Sun et al. (2002)
Mangrove swamp	Jiulongjiang estuary, China	*Kandelia candel*		(0.0029~0.1953)[b]	July and September 2002	Alongi et al. (2005)
	Brisbane River, Queensland, Australia	*Avocennia marina, Aegiceras corniculatum*		(−0.004~0.065)[b]	April 2004; February and July 2005	Allen et al. (2007)
	Deep Bay region, South China	Futian	*Kandelia obovata, Acanthus ilicifolius, Bruguiera gymnorrhiza*	0.7572	July and August 2000	Chen et al. (2010)
		Mai Po		(0.1509~0.4176)[b]		
	Magueyes Island, Puerto Rico	*Rhizophora mangle*		0.022 (0.0022~0.0616)[b]	2000	Bauza et al. (2002)
	Moreton Bay, Queensland, Australia	*A. marina*		(−0.002~0.014)[b]	July and August 1998; September and October 1999	Kreuzwieser et al. (2003)

[a] Means in different observation periods

[b] Values in bracket are the range of N$_2$O fluxes

was probably dependent on the higher nutrient loadings of the Deep Bay region compared to the Yellow River estuary. According to the Ocean Environmental Quality Communique of China in 2012, approximately 4.22×10^4 tons of nutrients and 4.39×10^5 tons of OM were discharged into Bohai Sea by Yellow River, while approximately 6.42×10^5- tons of nutrients and 4.65×10^5 tons of OM were imported into Deep Bay region by Pearl River (State Oceanic Administration of China 2013). Because the Futian and Mai Po mangroves were located in inner Deep Bay receiving discharges from the Pearl River Delta and nearby polluted rivers in Shenzhen and Hong Kong (Ong Che 1999), significantly higher fluxes of N_2O were mainly related to the high nutrient inputs from the polluted rivers that flooded into Deep Bay, such as Pearl River (Chen et al. 2010).

Based on the above analysis, we concluded that the N_2O emission in the future will be enhanced with increasing N loading to the Yellow River estuary (especially NH_4^+–N is the major pollutant) and denitrification will play a very important role in contributing the total N_2O emission. With increasing N loading, the magnitude of N_2O emission in the Yellow River estuary should be paid more attention as the annual or local N_2O inventory was assessed accurately.

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