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# Temporal and spatial variability of methane emissions in a northern temperate marsh



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# HIGHLIGHTS

• We investigated the variability of methane emissions from a cool temperate marsh.

- Surface soil temperature exerted dominant control on the seasonal variability.
- The spatial variability was mainly controlled by the changes of water table level.
- The upscaled chamber based model overestimated methane emission by 28%.

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# ABSTRACT

Although methane (CH<sub>4</sub>) fluxes from northern wetlands in Asia have been described in previous research at different temporal and spatial scales, integrated studies at the ecosystem scale were scarce. In this study, CH<sub>4</sub> fluxes were measured using eddy covariance (EC) technique and the chamber method in a cool temperate marsh in northeast China during the growing season (May-September) of 2011. CH<sub>4</sub> emissions were highly variable, both temporally and spatially during the measurement period. According to the EC observation data, CH<sub>4</sub> fluxes showed a significant diurnal cycle during the mid-growing season with nighttime average flux about 67% of the average daytime values. Daily cumulative CH4 fluxes varied from 54 to 250 mg CH<sub>4</sub> m<sup>-2</sup> d<sup>-1</sup> with an average flux of 136.2 mg CH<sub>4</sub> m<sup>-2</sup> d<sup>-1</sup>. The observations of chamber method showed that CH<sub>4</sub> emissions differed markedly among the three main plant communities. Average flux at the Carex lasiocarpa site was about 4 times and 13.5 times of that at the Glyceria spiculosa site and Deveuxia angustifolia site, respectively. The spatial variability of CH4 flux was mainly controlled by the varying water table level as well as the spatial distribution of different vascular plants, while the seasonal dynamic of CH<sub>4</sub> emission could be best explained by the change of surface soil temperature and air pressure. A comparison was made between EC measurements and the upscaled chamber based model. The results from the model overestimated CH<sub>4</sub> emission by 28% compared to the EC data. Considering the large variability of methane emission, it is necessary to conduct continuous observations on CH<sub>4</sub> emission from northern wetlands at different temporal and spatial scales to comprehend the variability and also to predict responses to climate change.

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# 1. Introduction

Northern wetlands are the primary natural source of methane (CH<sub>4</sub>) into the atmosphere and they contribute between 6 and 40 Tg CH<sub>4</sub> yr<sup>-1</sup> with a wide variation in rates (Worthy et al., 2000; Houghton et al., 2001; Zhuang et al., 2006; Roulet et al., 2007). Because CH<sub>4</sub> gas emitted is 25 times more effective in absorbing heat in the atmosphere than CO<sub>2</sub> on a 100-year time scale and

contributes to over 20% of global warming (IPCC, 2007), even a modest change in methane sources can change the sign of the greenhouse gas budgets of northern wetlands. A wetland can be a carbon sink and greenhouse gas source at the same time (Whiting and Chanton, 2001; Friborg et al., 2003; Rinne et al., 2007). Because of the high temperature sensitivity of the biogeochemical processes of northern wetlands,  $CH_4$  emission from these ecosystems should be given sustained attention considering the spatial pattern and magnitude of current and anticipated changes in climate (Schlesinger, 1997; IPCC, 2007).

For natural wetlands,  $CH_4$  is produced by microbes in anaerobic sediments and transported to the atmosphere by both physical





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(diffusion and ebullition) and biological (plant-mediated) processes (Lai, 2009). Measuring CH<sub>4</sub> flux which is produced solely by microbes is much more complicated than measuring CO<sub>2</sub> flux which is mainly routed through plants. Microbes produce hot and cold sources across a landscape that may vary by two to three orders of magnitude within a few meters (Baldocchi, 2003, 2012). Therefore both temporal and spatial variability in CH<sub>4</sub> emission should be concerned when investigating ecosystem scale CH<sub>4</sub> dynamics within a wetland site.

The chamber method and the micrometeorological eddy covariance (EC) technique are the main two techniques for CH<sub>4</sub> measurements. Existing studies of CH<sub>4</sub> fluxes from wetlands were mostly based on the chamber method. Although the labor intensive chamber technique provides discontinuous measurements representative on the very small scale ( $\leq 1 \text{ m}^2$ ), it is still quite useful when conducting some process-based research. The applications of EC technique for CH<sub>4</sub> flux observation before 2000 were relatively few (Verma et al., 1992; Suyker et al., 1996; Hargreaves and Fowler, 1998; Kim et al., 1998). In recent years, the number of CH<sub>4</sub> measurements using eddy covariance technique is increasing (Rinne et al., 2007; Riutta et al., 2007; Hendriks et al., 2008, 2010; Jackowicz-Korczyński et al., 2010; Schrier-Uijl et al., 2010; Herbst et al., 2011; Parmentier et al., 2011). The EC technique can provide continuous and area-integrated flux at the ecosystem scale  $(10^2 -$ 10<sup>4</sup> m<sup>2</sup>). Compared with the chamber method, the use of EC technique for CH<sub>4</sub> emission from natural ecosystems is guite limited at present due to the high cost, the difficulty of maintenance in the harsh field environment and, in some cases, the infeasibility of power supply.

CH<sub>4</sub> fluxes from natural wetlands in Asia have been described in previous research at varying temporal and spatial scales (Ding et al., 2004; Ding and Cai, 2007; Song et al., 2009, 2011; Miao et al., 2012), however, integrated studies at the ecosystem scale has not yet been reported. In this research, based on micrometeorological EC technique and closed-chamber method, the growing season CH<sub>4</sub> emission was measured from a permanently inundated freshwater marsh in northeast China. The three aims of this study were (1) to elucidate the temporal and spatial variability of CH<sub>4</sub> flux at the ecosystem scale; (2) to identify the most relevant factors that influence CH<sub>4</sub> emission from this wetland type; and (3) to make a preliminary comparison between CH<sub>4</sub> emissions derived from EC technique and those from the upscaled chamber measurements.

# 2. Material and methods

#### 2.1. Site description

The study site is a permanently inundated and eutrophic marsh in the Sanjiang Plain, northeast China  $(47^{\circ}53' \text{ N}, 133^{\circ}30' \text{ E})$  at a latitude representative of the natural freshwater wetlands in this area (55 m a.s.l.). The Sanjiang Plain inhibits approximately 10,400 km<sup>2</sup> freshwater wetland area in China and is at present divided into many zones by cultivated lands (Zhao, 1999; Song et al., 2011).

Although the marsh is a dish-like depression, its slope grade is quite low (about 1:5000) and the topography is flat with *Carex lasiocarpa* as the dominant vegetation. Other plants in the marsh include *Carex pseudo-curaica, Glyceria spiculosa, Carex limosa, Deyeuxia angustifolia and Carex meyeriana.* The morphological appearance of the herbaceous vegetation in the marsh looks quite similar. The three main types of vegetation community dominated by *C. lasiocarpa, G. spiculosa* and *D. angustifolia*, respectively, successively show a concentric distribution pattern with the gradual decrease of water table level along the center to the edge of the marsh.

The climate is a temperate continental monsoon type with annual mean temperature 2.5 °C. The mean temperature in July and January is 22 and -21 °C, respectively. The mean annual precipitation is approximately 558 mm with approximately 80% occurring during the growing season from May to September. Precipitation is the main water source in freshwater marshes in normal years. Water and soil in marshes are completely frozen from late October to next April and begin to melt from late April till July.

#### 2.2. Eddy covariance measurements

An instrument mast was erected in the marsh at the beginning of May 2011. To measure wind speed and sonic temperature, a three-dimensional ultrasonic anemometer (CSAT-3 Campbell, Scientific, USA) was installed on the mast at a height of 2.5 m above the ground. At the same height, with a separation of 15 cm, an inlet was situated where air was drawn down toward the fast greenhouse gas analyzer (FGGA, Los Gatos Research, Mountain View, CA, USA). CH<sub>4</sub>, CO<sub>2</sub> and H<sub>2</sub>O concentrations were measured by FGGA based on off-axis integrated cavity ringdown spectroscopy (Baer et al., 2002). All measurements were taken at a nominal frequency of 10 Hz and the data were stored on a datalogger (CR3000, Campbell, scientific, USA).

For the closed-path gas analyzer, a dry vacuum scroll pump (XDS35i, BOC Edwards, Crawley, UK) was adopted to draw the sampling air through a 7 m tube (inner diameter 6.4 mm, made of fluorinated ethylene propylene to minimize sorption or desorption) into the measuring cell at the operating pressure of approximately 19 kPa. The air was filtered through a filter with a pore size of 10  $\mu$ m to prevent dust and insects from entering the system and through two 2  $\mu$ m Swagelok filters (one internal and one external) before entering the measuring cell. Because the pump and the gas analyzer had a high power requirement, the EC system ran on AC power supply during the measurement period.

The average height of marsh plants changed from about 0.0 m to about 0.5 m. The terrain around the instrument mast was flat and uniform with a fetch of at least 280 m in the prevailing southeast wind direction and at least 200 m in all other directions.

Processing of high-frequency EC data was performed with EddyPro 4.1 (www.licor.com/eddypro). Raw data were filtered for spikes and linear detrending was used. Double coordinate rotations were performed to align the mean vertical velocity measurements normal to the mean wind streamlines before scalar flux calculations. Using the maximum cross-correlation method (relative to the vertical velocity or temperature), time lags were determined for each half-hourly period. Half-hourly fluxes of  $CH_4$ ,  $CO_2$  and  $H_2O$  were calculated as the mean covariance of vertical wind velocity and scalar fluctuations. The WPL correction for density fluctuations arising from variations in water vapor was applied according to Ibrom et al. (2007b) by using the uncorrected covariances of water vapor mass density with the vertical wind velocity when correcting the dilution effect. The low-pass filtering effects were assessed and corrected using the method of Ibrom et al. (2007a) based on in site determination of water vapor attenuation and on a model for the corresponding spectral correction factor. Quality control criteria according to Mauder and Foken (2004) were used to reject bad data. Additionally, data were excluded when the pump stopped working due to maintenance or high temperature in summer and when the transmission of sound on the sonic anemometer was blocked by heavy rain. CH<sub>4</sub> flux was calculated by adding the rate of CH<sub>4</sub> storage change (S) to the turbulent flux. S was estimated from the changes in the average CH<sub>4</sub> concentrations at the sensor height over the 30-min intervals assuming that the CH<sub>4</sub> concentrations were representative for the entire air column below the sensors. The data collected during weak turbulence were removed from further analyses by filtering all half hour fluxes with friction velocity ( $u^*$ ) below 0.1 m s<sup>-1</sup>. The threshold was determined by analyzing the relationship between  $u^*$  and methane flux during the growing season (Long et al., 2010). By these procedures, 40% of the data were removed. Data gaps less than 2 h were filled by linear interpolation. Longer gaps were filled with average data derived from compiling the 30 min ensemble average trend of five days prior to and following the gap. Two long data gaps (June 15–18 and August 5–8) occurred during the measurement period because of the maintenance of pump.

#### 2.3. Chamber measurements

During the growing season of 2011, independent methane flux measurements by static chamber method were performed. Along the center to the edge of the marsh, three sites were set up for chamber measurements within 200 m distance southeast of EC mast. The main plant community at each site was *C. lasiocarpa*, *G. spiculosa* and *D. angustifolia*, respectively. There were three replicates in each site and boardwalks were constructed around the sample plots to minimize disturbance.

CH<sub>4</sub> emission measurements were made at biweekly intervals and in each measurement day, gas was sampled from 6:00 to 18:00 at 2 h interval using stainless steel made chambers (50 cm  $\times$  50 cm  $\times$  50 cm). During each observation, the chambers were placed into the collars with water to prevent leakage, and the vegetation was included within the chambers. Inside each chamber, a small fan and a thermometer sensor were installed. Gas sampling lasted half an hour and four gas samples were took in 10-min intervals.

The gas samples were stored in syringes less than 12 h before being measured in the laboratory of the Sanjiang Station. Gas chromatography (Agilent 4890D, Agilent Co., Santa Clara, CA, USA) was used to measure the CH<sub>4</sub> concentrations; then the gradient of CH<sub>4</sub> concentration during sampling was used to calculate the CH<sub>4</sub> flux. Sample sets were rejected unless they yielded a linear regression of  $R^2$  greater than 0.85. Average CH<sub>4</sub> flux and standard deviation were calculated from the three replicates for each observation in each site.

#### 2.4. Auxiliary measurements

Supporting meteorological data of net radiation ( $R_n$ ), photosynthetic active radiation and barometric pressure (P) at a height of 2 m, air temperature, relative humidity and wind speed and direction at 1, 2 and 3 m, soil temperature at 5, 10, 15, 20, 30, 40 and 70 cm depths below the surface and precipitation were obtained from a long-term automatic weather station about 200 m away from the EC system in the marsh. Continuous and automatic recordings of water table level (WTL) by pressure transducers (Odyssey, Dataflow Systems Pty Ltd., Christchurch, New Zealand) and soil temperature at 5, 10, 15, 20 and 30 cm depths by multichannel soil thermometer (YM-04, Yimeng Inc., China) were made at the three chamber sites during the growing season. The sampling frequency was 2 h for WTL and 0.5 h for soil temperature.

#### 3. Results

#### 3.1. Environmental conditions

Large variation in monthly average air temperature was observed at the study site with the highest average temperature occurring in July and the lowest in January (Fig. 1a). Except July, the months in 2011 had average temperature close to the long-term mean (based on measurements during 1991–2011). Average temperature of 23.9 °C in July 2011 was higher than the long-term mean ( $\pm$ SD) of 21.7  $\pm$  0.9 °C in this month (Fig. 1a). Annual precipitation of 508 mm in 2011 was slightly lower than the long term average of 565  $\pm$  101 mm, which was mainly caused by the low precipitation in July. Monthly precipitation in July fell greater than one standard deviation below the long term average (Fig. 1b).

Due to the slightly depressional landform, the marsh was permanently inundated and the spatial variation of water table level was distinct (Fig. 1c). From the center to the edge of the marsh, the average WTL declined from 29 to 14 cm during the measurement period. Since precipitation is the main water source for the marsh, WTL generally increased with rainfall and decreased with evapotranspiration. For example, the relatively warm and dry conditions in July resulted in a gradual decrease of WTL until an 18 mm rainfall occurred in early August (Fig. 1c).

The vegetation in the marsh began to leaf-out in May and reached the maximum leaf area index around 2.4 in late July. The plants showed visible signs of senescence in late August. Senescence accelerated in September and there was almost no green leaves remaining by late September.

# 3.2. Diurnal variation of CH<sub>4</sub> emission

Based on the stage of plant growth, we divided the EC data set into three separate time periods: (period 1) early growing season (DOY 124–171): (period 2) mid-growing season (DOY 172–232) and (period 3) late growing season (DOY 233–273). Fig. 2a shows the diurnal cycle of CH<sub>4</sub> flux averaged for each period. During the early growing season, the rate of CH<sub>4</sub> emission was relatively low and steady throughout the day with an average rate of 3.8 mg  $CH_4$  m<sup>2</sup> h<sup>-1</sup>.  $CH_4$  emission rate increased significantly during the mid-growing season and there was a distinct diurnal pattern with higher fluxes observed during the daytime than nighttime. The clear increase of CH<sub>4</sub> emission coincided with the start of sunrise and the opposite occurred at the start of sunset (Fig. 2a, b). CH<sub>4</sub> emission rate kept relatively steady throughout the daytime and generally, there was no remarkable emission peak appearing at some regular time. The average emission rate was 7.9 mg  $CH_4$  m<sup>2</sup> h<sup>-1</sup> and the ratio of nighttime to daytime  $CH_4$ flux averaged 0.67 during the mid-growing season. The magnitude of CH<sub>4</sub> flux during the late growing season was similar to that of the early stage. CH<sub>4</sub> emissions were lower during the nighttime than daytime with an average ratio of 0.86 (Fig. 2a). Using bivariate correlation analysis, we found no significant influence of environmental factors (such as soil or air temperature, radiation and friction velocity) on the diurnal cycle of CH<sub>4</sub> emission for the three periods.

#### 3.3. Seasonal variation of CH<sub>4</sub> emission

Daily cumulative EC CH<sub>4</sub> flux varied from 54 to 250 mg CH<sub>4</sub> m<sup>-2</sup> d<sup>-1</sup> with an average of 136.2 mg CH<sub>4</sub> m<sup>-2</sup> d<sup>-1</sup> during the growing season from May to September (Fig. 3). CH<sub>4</sub> emission rate kept relatively smooth during May and began to increase significantly since early June until the peaks appeared in early July. For the period from DOY 191 to DOY 222 when daily average air temperature remained higher than 20 °C, CH<sub>4</sub> emission rates fluctuated around 212 mg CH<sub>4</sub> m<sup>-2</sup> d<sup>-1</sup>. Gradual decrease began since mid August with small fluctuations until the end of the measurement period (Fig. 3). The total emission was 20.4 g CH<sub>4</sub> m<sup>-2</sup> during the growing season.



**Fig. 1.** Seasonal patterns of (a) monthly mean air temperature ( $T_a$ ), (b) monthly cumulative precipitation and (c) daily cumulative precipitation and daily average water table level (WTL) at the three chamber sites. Mean represents the average of 1991–2011 (based on measurements from the long-term weather station in the marsh) and error bars indicate standard deviations.

On a daily time scale, a simple regression (Person correlation, 2tailed test for significance) showed that CH<sub>4</sub> flux correlated significantly with environmental variables of temperature and air pressure (P < 0.01). No significant relationship was found between CH<sub>4</sub> flux and other variables such as water table level, wind speed and radiation (P > 0.1). Of the temperature measured at different heights or depths, soil temperature at 5 cm depth  $(T_{s5})$  explained the highest proportion of variation in CH<sub>4</sub> flux (Fig. 4a). We then performed a multiple regression analysis with log transformed CH<sub>4</sub> emission as the explained variable and  $T_{s5}$  and P as the explanatory variables. Linear ordinary least square regression was used to find the best fit and estimate parameters. Results showed that  $T_{s5}$  was the dominant factor influencing the seasonal variation of CH<sub>4</sub> emission ( $R^2 = 0.77$ , P < 0.01). When the variable of air pressure was added, the model could give a 4% increase in its predictive ability ( $\Delta R^2 = 0.04$ ,  $\Delta F = 23$ , P < 0.01). Overall, 81% of the seasonal variation of CH<sub>4</sub> flux (log transformed) could be explained by soil temperature (5 cm depth) and air pressure ( $CH_4$  flux = exp (0.09)  $T_{\rm s5}$ -0.13P + 16.26),  $R^2$  = 0.81, P < 0.01).

# 3.4. Spatial variation of CH<sub>4</sub> emission

There were significant differences among the CH<sub>4</sub> fluxes measured by chamber method at the three sites (Mann–Whitney *U* test, P < 0.01). Highest CH<sub>4</sub> emission was observed at the *C. lasiocarpa* site near the center of the marsh with an average water table level of 29 cm, while lowest CH<sub>4</sub> emission was detected at the *D. angustifolia* site near the edge of the marsh with an average WTL of 14 cm, and intermediate CH<sub>4</sub> emission at the *G. spiculosa* site with an average WTL of 20 cm (Figs. 5and 1c). The growing season average CH<sub>4</sub> flux at the *C. lasiocarpa* site was 14.9 mg CH<sub>4</sub> m<sup>2</sup> h<sup>-1</sup>), and 13.5 times of that at the *D. angustifolia* site (1.1 mg CH<sub>4</sub> m<sup>2</sup> h<sup>-1</sup>). The large differences in flux magnitude between the chamber sites suggested considerable spatial variation of CH<sub>4</sub> emission within the marsh.

A bivariate correlation analysis using daily averages at the three sites showed a significant relationship between CH<sub>4</sub> flux and water table level ( $R^2 = 0.54$ , P < 0.01, n = 27), with linearly increasing CH<sub>4</sub>



**Fig. 2.** The average daily pattern of (a)  $CH_4$  emission, (b) net radiation ( $R_n$ ) and (c) air temperature during different periods of the 2011 growing season. The different growing periods were early growing season (DOY 124–171), mid-growing season (DOY 172–232) and late growing season (DOY 233–273). Flux data were binned by time of the day and then averaged for all days during the time period.

emission with rising WTL. The relation between average  $T_{s5}$  and the average CH<sub>4</sub> fluxes at the three sites was not significant (P > 0.01, n = 27). Since  $T_{s5}$  generally decreased with the increase of WTL, the effects of  $T_{s5}$  on the spatial variation of CH<sub>4</sub> emission might be obscured by the effects of WTL.

#### 3.5. Comparison of EC measurement and chamber-based model

Similar to the response of EC measured  $CH_4$  flux to temperature, hourly  $CH_4$  fluxes measured by chambers at each site also showed best relationship with  $T_{s5}$  compared with soil temperature at other depths. Using continuous  $T_{s5}$ , hourly  $CH_4$  flux for the whole growing season were estimated and daily emissions were then calculated for each site. To scale up the chamber data to the ecosystem level, daily  $CH_4$  fluxes were weighed according to the



**Fig. 3.** Seasonal variation of daily cumulative CH<sub>4</sub> emission during the growing season of 2011.



**Fig. 4.** The relationship between daily cumulative  $CH_4$  flux and 5 cm depth soil temperature  $T_{s5}$  (a) and data series of daily cumulative  $CH_4$  flux and daily averaged air pressure (P) (b).

coverage fractions of the three main plant communities (Table 1). The results of the upscaled regression model generally followed the seasonal trend of EC measured CH<sub>4</sub> flux although the former had a relatively smooth temporal variation (Fig. 6a,b). Average daily emission by the upscaled regression model was 174.6 mg CH<sub>4</sub> m<sup>2</sup> d<sup>-1</sup>, 28% higher than that of the EC measurements during the growing season.

#### 4. Discussion

# 4.1. Diurnal and seasonal variability of CH<sub>4</sub> emission

Diurnal variation of CH<sub>4</sub> emission showed different patterns at different growth periods (Fig. 2a). Clear diurnal variation of CH<sub>4</sub> emission during the mid-growing season (period 2) was observed in this research, as has been observed in many other studies (Morrissey et al., 1993; Thomas et al., 1996; Kim et al., 1998; Van der Nat et al., 1998; Garnet et al., 2005; Long et al., 2010). During the mid and late growing season, there were synchronous changes of CH<sub>4</sub> emission and radiation at sunrise and sunset hours while their



**Fig. 5.** Seasonal variations of  $CH_4$  emissions at the three chamber sites dominated by different marsh plants during the growing season. The error bars represent standard deviations.

Table 1
Results from the regression analyses of CH <sub>4</sub> fluxes with $T_{s5}$ at each chamber site. The exponential equation was CH <sub>4</sub> flux = $a \exp(bT_{s5})$

Dependent variable	Independent variable	Plant community	Coverage fraction	п	а	b	Р	$R^2$
CH <sub>4</sub> flux (mg CH <sub>4</sub> m <sup><math>-2</math></sup> h <sup><math>-1</math></sup> )	<i>T</i> <sub>s5</sub> (°C)	Carex lasiocarpa Glyceria spiculosa Deyeuxia angustifolia	0.46 0.32 0.22	63 63 63	5.18 0.70 0.21	0.07 0.11 0.08	<0.01 <0.01 <0.01	0.50 0.67 0.52

 $T_{s5}$ , soil temperature at 5 cm depth.

daytime fluctuations were quite different (Fig. 2a,b), indicating that radiation might impose a short term and indirect influence on  $CH_4$  transport by affecting stomatal conductance.

For wetlands dominated by vascular plants, plant transport of CH<sub>4</sub> was found to be a very import pathway. CH<sub>4</sub> transport by C. lasiocarpa had been proved to account for over 80% of the total emission from the permanently inundated marsh (Ding et al., 2004). There are two major mechanisms involved in the plantmediated transport of CH<sub>4</sub> from wetland to the atmosphere, namely molecular diffusion and bulk flow (Joabsson et al., 1999). Previous research found that aerenchymatous sedges like Carex spp. released  $CH_4$  by diffusive transport which was influenced by stomatal conductance (Morrissey et al., 1993; Thomas et al., 1996; Van der Nat et al., 1998; Kutzbach et al., 2004). The research by Morrissey et al. (1993) found that CH<sub>4</sub> emission rate corresponded well to stomatal conductance in a Carex-dominated tundra and the importance of stomatal pathway relative to cuticular pathway was expected to vary through the growing season. In this research, we considered plant-mediated diffusion as the major contributing mechanism for the strong diurnal cycle of CH<sub>4</sub> emission during the mid-growing season. This could account for the relatively low CH<sub>4</sub> flux before (after) sunrise (sunset) when the minimum stomatal conductance weakened ventilation and consequently transport of CH<sub>4</sub> through plants. Besides plant transport, diffusion through the soil and water from deeper soil layers could also be an important pathway for CH<sub>4</sub> emission, which could partly explain the relatively stable and low CH<sub>4</sub> emission rates during the early and late growing season when the living vascular plants available for CH<sub>4</sub> transport



Fig. 6. Comparison of daily cumulative  $CH_4$  flux by EC measurements and those simulated by the upscaled chamber based model.

and organic material supply were less than those of the midgrowing season.

Daily CH<sub>4</sub> emission increased exponentially with the increase of  $T_{s5}$  which could explain about 77% of the seasonal variation of CH<sub>4</sub> fluxes (Fig. 4a). Since methanogenesis occurs across a range of soil depths which have different soil temperatures and diurnal lags, the close relationship between CH<sub>4</sub> emission and  $T_{s5}$  in this research could be ascribed to that soil temperature around 5 cm depth represented the average temperature condition conducive to methanogenesis.

In this research, air pressure was found to significantly improve the exponential model based on soil temperature. The drop of air pressure generally correlated with the increase of CH<sub>4</sub> emission (Fig. 4b), which suggested that ebullition could also be an important pathway for CH<sub>4</sub> emission from the marsh. This could be further proved by our visual observations of ebullition from the marsh at times. Ebullition has been showed to release a large amount of CH<sub>4</sub> and contribute to a significant portion (50%–64%) of total CH<sub>4</sub> flux measured in northern peatlands (Glaser et al., 2004; Tokida et al., 2007). However, the sporadic occurrence and distribution of ebullition in the field make it difficult to be independently observed and quantified. CH<sub>4</sub> emission by ebullition could often be ignored when flux was measured by chamber method because of its discontinuous samplings.

Water table level is generally considered to be a physical parameter of major importance for CH<sub>4</sub> emission from wetlands (Kettunen et al., 1999; Frenzel and Karofeld, 2000; Updegraff et al., 2001; Treat et al., 2007). In this study, the bivariate correlation analysis showed that WTL was not significant in predicting the seasonal variation of CH<sub>4</sub> fluxes. This is probably due to the fact that the marsh soil and vegetation roots layer are permanently inundated during the growing season (Fig. 1c) and thus the anaerobic environment for methanogenesis and transport is kept relatively stable. Similar results could also be found in the researches of Jackowicz-Korczyński et al. (2010), Hargreaves et al. (2001), Christensen et al. (2003), and Sachs et al. (2008).

# 4.2. Spatial variability of CH<sub>4</sub> emission

According to the chamber measurements, growing season average of CH<sub>4</sub> flux near the center of the marsh was 13.5 times of that near the edge of the marsh (Fig. 5), which indicated that even in a permanently inundated marsh with homogenous vegetation, CH<sub>4</sub> emission still showed large spatial variability. Although we didn't find significant influence of WTL on the seasonal variation of CH<sub>4</sub> emission, WTL could be one of the major controlling factors for the spatial variation of CH<sub>4</sub> emission with higher CH<sub>4</sub> flux corresponding to higher WTL. Besides WTL, vegetation type could be another important controlling factor for the spatial variability of CH<sub>4</sub> release from wetlands (Schimel, 1995; Petrescu et al., 2008). Ding et al. (2004) found that CH<sub>4</sub> emissions from the C. lasiocarpa community dropped by 81% while those from the D. angustifolia community dropped by 21% after the plants were clipped below the water surface of the inundated marsh. In this research, different CH<sub>4</sub> transport capacities among different plants, especially between Carex spp. and Deyeuxia spp. could explain the observed spatial variability of CH<sub>4</sub> emission to a large extent.

# 4.3. Comparison of EC and regression model

CH<sub>4</sub> emissions were compared between EC measurements and the regression model based on chamber data and  $T_{s5}$ , taking into account the relative coverage of the three main plant communities in the marsh. Although the results from the model generally showed good agreement with the EC data (Fig. 6a,b), the former overestimated daily CH<sub>4</sub> emission with approximately 28%. The overestimation could be ascribed to the following: (1) The regression model was not sensitive enough to simulate changes of ecosystem behavior and there will be no predicted response to factor other than  $T_{s5}$  in the current upscaling. However, as we have found when analyzing factors controlling the seasonal variation of CH<sub>4</sub> flux measured by EC technique, air pressure could impose a negative effect on CH<sub>4</sub> release (Fig. 4b). Since the influence of air pressure can not be considered in the chamber measurement, the absence of parameter such as air pressure could partly lead to the overestimation. Besides, the circadian rhythms of stomatal activities might have effect on CH<sub>4</sub> diffusion, as had been indicated by the diurnal cycle of CH<sub>4</sub> emission during the mid and late growing season (Fig. 2a). Because chamber measurements were only conducted during daytime when CH<sub>4</sub> fluxes were relatively high, the relatively low nighttime CH<sub>4</sub> release could not be simulated and thus resulted in an overestimation of the daily emission. (2) Coverage fractions of different plant communities were dynamic during the growing season, while a fixed ratio was used in the model. This might lead to uncertainties in the upscaling process since CH<sub>4</sub> emissions from different plant communities varied greatly, and a small change in the coverage fraction could significantly influence the up-scaled results.

In this research, we didn't use the footprint models to evaluate  $CH_4$  fluxes in different source areas. The application of footprint models depends on the meteorological conditions. Splitting up the model in small footprint area could result in limited data available for each area and the footprint calculations have uncertainties of their own. Furthermore, the concentric distribution of the vegetation types in the marsh made it difficult to estimate  $CH_4$  flux from some plant community using the footprint models. To investigate the spatial variability of  $CH_4$  emission, the chamber method was the better way while when investigating the temporal variability of  $CH_4$  emission at the ecosystem scale, the eddy covariance technique was best.

# 5. Conclusions

The eddy covariance technique and the chamber method were used in this study to measure CH<sub>4</sub> flux at a cool temperate marsh site in northeast China. CH<sub>4</sub> emission showed to be highly variable, both temporally and spatially. Distinct diurnal pattern of CH<sub>4</sub> emission was found during the mid-growing season and highest daily emission appeared in early July. CH<sub>4</sub> emission differed markedly among the three main plant communities. For the seasonal dynamic of CH<sub>4</sub> emission, surface soil temperature exerted the dominant control and air pressure also showed some influence, while spatial variability of CH<sub>4</sub> emission was mainly controlled by the change of water table level as well as the spatial distribution of different marsh plants. Comparison of CH<sub>4</sub> fluxes between EC measurements and the upscaled chamber-based model generally showed good agreement although the latter overestimated average daily emission by 28%. To scale up the chamber measurement more accurately, a continuous and detailed knowledge of the spatial pattern of environmental and biotic variables was essential.

Both the temporal and spatial dynamic of  $CH_4$  emission in this research indicated that long-term and continuous  $CH_4$  flux measurements on various scales are needed to address seasonal, interannual and spatial variations of  $CH_4$  emission, to adequately quantify the emission amount and to identify the environmental and biotic controlling mechanism for  $CH_4$  emission from wetlands in the mid-high latitudes.

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