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LETTER

Methane emissions from typical paddy fields in Liaohe Plain and Sanjiang Plain, Northeast China

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Abstract

Methane is one of the most important greenhouse gases and is emitted from waterlogged rice regions. Eddy covariance was used to obtain long-term (2017–2018), high-frequency data for the CH4 flux at typical rice paddy fields in Sanjiang Plain and Liaohe Plain. This study examines the CH4 emission process, emission characteristics at different time scales and the intensity of different emission sources. The relationships between CH4 emissions and temperature and photosynthetically active radiation were also analysed. CH4 emissions from Sanjiang Plain and Liaohe Plain in 2017 were 26.77 and 16.17 g CH4 m−2 year−1, respectively. The CH4 fluxes in 2017 and 2018 were similar; emissions were higher at the Sanjiang site in 2018. Peak CH4 emissions (daily average: 0.127 μmol CH4 m−2 s−1) corresponded to the tillering stage in Sanjiang Plain. CH4 emissions peaks corresponding to the jointing–heading and mature stages at Liaohe Plain. Monthly CH4 emissions were highest in July. The daily averaged flux was 0.065 μmol CH4 m−2 s−1 during the growing season. The field-transplanting stage exhibited the highest CH4 emissions. No CH4 was emitted during the greening–tillering stages. Emissions increased again during the jointing–heading stages to 0.102 μmol CH4 m−2 s−1, and then decreased gradually. A significant linear correlation was observed between PAR and the methane flux in Liaohe Plain. CH4 emissions in Sanjiang Plain were more sensitive to changes in temperature. The temperature coefficient value was highest from the sowing period to the green period. The CH4 flux at Sanjiang Plain depended on the stage of rice growth, the temperature, the degree of irrigation and drainage, and the soil pH. In November (during the non-growing season) at Liaohe Plain, a slight negative flux (a weak CH4 sink) was observed. CH4 emissions were also found to be inhibited in the growing season by increased soil alkalinity.

1. Introduction

The World Meteorological Organization (WMO) Statement on the Status of the Global Climate in 2015 [1] confirmed that greenhouse gases emitted by humans have caused a long-term rise in global temperature. Methane (CH4) is an important long-lived greenhouse gas, like carbon dioxide (CO2) and nitrous oxide (N2O), and is a lead contributor to thermal and chemical changes to the atmosphere [2]. According to the latest analysis of the WDCGG [3] observations from 120 worldwide greenhouse gas monitoring sites, the year-averaged methane concentration was 1853 ± 2 ppb in 2016. This concentration has increased by 6.8 ppb/year over the last decade [3]. Between 2014 and 2015, the rate of increase rate reached 10 ppb per year [4, 5]. Since the 1950s, various techniques have been used to monitor and record changes in CO2 concentrations, but CH4 is less well understood. Measured data for CH4 emission sources in wetland are insufficient to accurately locate CH4 sources, and this leads to uncertainty in the estimations made by global ecosystem models and carbon databases that are created following inversion simulations of remote sensing data [6].

The average mole fraction of CH4 in the atmosphere is only 1/217 of that of CO2. However, the global warming potential (GWP) of CH4 is about 21 times greater than that of CO2 [7]. Methane contributes about 19%
of the total GWP, second only to CO₂, which contributes about 63% [8]. Therefore, CH₄ is a major contributor to global climate change and it is important for research to study the role of CH₄ emissions from different ecosystems.

The total amount of CH₄ emitted by rice (Oryza sativa L.) accounts for about 12%–26% of global CH₄ emissions (IPCC, 2007) [8–10]. Under submerged conditions, methanobacteria reduce and decompose organic matter into CH₄ which is transported to the air by bubbles and diffusion through plants and soil [11]. China contains about 18.85% of the world’s rice-growing area [12]. Without proper mitigation measures, CH₄ emissions from paddy fields are likely to increase.

The eddy covariance technique (EC) that has been developed in recent years can sustain long-term monitoring to obtain more accurate flux estimations, and can decrease interference in the monitoring environment. EC also enjoys a high analysis frequency, making the obtained data more representative. Because of the interannual variability of meteorological factors and the complexity of environmental variables, more long-term EC data are needed to fully understand the relationships governing CH₄ exchanges in different environments.

The climate in Northeast China has been warming since 1960 [13]. The average temperature is beneficial for growing rice [14]. The speed of rice ripening has accelerated and the growth period of rice has been prolonged [14]. Northeast China is the country’s main rice-production area where a single-cropping rice type is grown. Rice plantations cover 17.78% of China’s landmass, and the total output is 19.27% (2015). In the past 30 years, the area used for rice planting has increased from 1.193 to 5.306 million ha. Over the same period the proportion of grain crops in the northeast has increased from 8.9% to 23.6%. Rice plantations are expanding and gradually replacing areas previously used to grow soybean and corn. From 1988 to 2006, the area of paddy fields in Liaohe Plain increased by 977.1 km² [15]. Sanjiang Plain is one of the largest black soil zones in the world. However, since the 1990s, dry land has been converted into paddy fields at a large scale, with the area increasing very rapidly over the past 10 years. Changes to the area dedicated to paddy fields affects the accuracy of the national inventory of CH₄ emissions from paddy fields. The wetland extent could contribute 30%–40% on the estimated range for wetland emissions [16]. However, research that focuses on CH₄ emissions from the north-eastern region remains limited.

Through conditional monitoring at stationary EC sites this study aimed to determine the key period characteristics, emissions and exchange intensity of CH₄ emissions from rice wetlands in Northeast China following changes brought about by global climate change and human activities. Data on rice growth factors were considered to examine the environmental factors affecting CH₄ emissions. Meteorological data were also analysed to explore the main driving force of long-term CH₄ seasonal changes in rice ecosystems. In summary, this study aimed to obtain emission coefficients of two wetlands, study their CH₄ emissions, the factors influencing these emissions, and formulate reasonable measures to decrease CH₄ emissions from this region.

2. Materials and methods

2.1. Site description

Liaohe Plain is situated in the southern tip of Northeast China (figure 1). Liaohe Plain Rice Field Station is located in the centre of Liaohe Plain (40°41′–41°27′N, 121°30′–122°41′E) with an area of cultivated land of 144 million ha. The annual, July and January average temperatures are 8.6, 24.4 and −9.8 °C, respectively. The frost-free period lasts 171 days, and the area has an accumulated temperature of 3509 °C and an average annual precipitation of 631 mm [17]. Sanjiang Plain is situated in the north-eastern tip of Northeast China. Sanjiang Plain Rice Field Station is located in the centre of Sanjiang Plain. Here, the altitude is 30.0–40.0 m, the average temperatures in July and January are 21–22 and −21 to −18 °C, respectively, the accumulated temperature is 2300 °C–2500 °C, the frost-free period is 140 d, and the average precipitation is 500–650 mm [18].

The 2017 growth season on Liaohe Plain lasted from April 18th to September 27th, and that on Sanjiang Plain from April 15th to September 12th. The onset of the 2018 fertility period was close to that in 2017. However, the 2018 tillering periods for rice were delayed compared with those in 2017 (table 1). Irrigation at Liaohe Plain was in early May, later than that at Sanjiang Plain (late April). Drainage occurred between the end of the rice the milk maturity stages (August 23rd–27th). Harvest was in early October.

The soil in Liaohe Plain is characterised as a salinized paddy with an alkaline pH value. The tested rice variety was Yanfeng 47. The soil in Sanjiang Plain is characterised as black soil (Chernozem) and has a weakly acidic pH value (table 2). The typical redox value of the paddy field was in the range +300 to −200 mV [19]. The tested rice variety was Longjing 31. Sowing was carried out in the middle of April, but transplanting and reaching maturity in Liaohe Plain occurred later than in Sanjiang Plain.
2.2. EC measurements

The EC method calculates the flux of CH$_4$ and CO$_2$, LE, and $H$ by measuring the scalar of both vertical wind speed and turbulent fluctuation, and then calculates the covariance between them \[20\].

EC observation systems were installed at Liaohe Plain (40°55’53″N, 121°57’66″E) and Sanjiang Plain (47°9’7″N, 131°56’19″E) (figure 1). The Kormann & Meixner (KM) footprint model was used to estimate the contribution to a regional measurement \[21\]. The KM model allowed the estimation of net fluxes for non-uniform wind directions. The main parameters driving the KM model include the observation time, friction wind speed ($u^*$), Monin–Obukhov length and wind direction. Output parameters include the zero plane displacement.

Rice was grown on 90% of the contribution area of the EC observation system. The EC system consisted of a precision 3-axis sonic anemometer, an open-path CO$_2$/H$_2$O analyser (Li-7700, Licor, Lincoln, NE, USA) and a data collector (Li-7550, Licor). The sampling frequency was 10 Hz and the installation height was 4 m. The output data included horizontal wind speed ($U_x$, $U_y$), vertical wind speed ($U_z$), CO$_2$ absolute density, water vapour absolute density, ultrasonic virtual temperature ($T_s$), atmospheric pressure and CSAT3 diagnostic value ($\text{diag\_csat}$). The system calculates the flux in real time (every 30 min), and saved in 30-min intervals (every 30 min); and data was measured at 10 Hz and then averaged over 30 min periods (10 Hz; figure 2).

The calibration of the LI-7700 open-path gas analyzer was checked at semiannual intervals, using a calibration cell provided by the manufacturer. The LI-7700 was calibrated span against a 5 ppm CH$_4$ standard gas and calibrated zero by ‘zero gas’.

The CH$_4$ flux ($F_C$) was calculated as:

$$ F_C = w'\rho'_C $$

where $w'$ is the instantaneous deviation between the vertical wind speed and the mean value, i.e., the disturbance value; $\rho'_C$ is the instantaneous disturbance value of the CH$_4$ density; and $w'\rho'_C$ is the covariance of the vertical wind speed and the CH$_4$ density (Baldocchi et al 1988) \[20\]. Raw data were manipulated using Eddypro 5.1.1 (Licor) software to carry out coordinate rotation, spiking removal, time-delay removal and compensation of density fluctuations (WPL) corrections.

Quality assurance and quality control (QA/QC) was performed to remove outliers. The calculation of spectral correction factors was performed in different ways for ‘small’ and ‘large’ fluxes using the software’s ‘minimum’ and ‘unstable’ values. During periods of precipitation, the observation error of the EC observation system was relatively large. Observational data collected during precipitation events were, therefore, excluded.

To accurately calculate the annual values of net ecosystem exchange (NEE) at the sites, gap-filling was required to account for missing data. A commonly used method for filling in small sections of missing data (1–3 points) is MS Excel’s interpolation FORECAST function. For longer datasets, the mean diurnal variation is an interpolation technique where the missing NEE value for a certain time period (30 min) is replaced with the averaged value of the adjacent days at exactly that time of day. Windows of 7 and 14 days were chosen for averaging for daytime and night time missing data, respectively. Missing half-hourly data points (owing to, e.g., instrument and power failure) during the winter season were filled using the mean diurnal variation approach.
Table 1. Observations of the rice growth period and irrigation/drainage start dates.

| Station       | Year  | Sowing | Emergence | Irrigating water | Trefoil | Transplanting | Turning Green | Tilling | Jointing | Booting | Flowering | Drainage | Milky maturity | Fruit ripening |
|---------------|-------|--------|-----------|------------------|---------|---------------|---------------|---------|----------|---------|------------|----------|----------------|----------------|
| Liaohe plain  | 2017  | Apr.18 | Apr.26    | May.01           | May.5   | May.27        | Jun.1         | Jun.16  | Jul.15    | Jul.27  | Aug.7      | Aug.20   | Aug.27        | Sep.27         |
|               | 2018  | Apr.15 | Apr.23    | May.13           | May.3   | May.24        | May.26        | Jun.22  | Jul.11    | Jul.29  | Aug.8      | Aug.18   | Aug.23        | Sep.25         |
| Sanjiang plain| 2017  | Apr.15 | Apr.24    | Apr.25           | May.4   | May.15        | May.22        | Jun.21  | Jul.04    | Jul.10  | Jul.23     | Aug.20   | Aug.24        | Sep.12         |
|               | 2018  | Apr.13 | Apr.23    | Apr.25           | May.2   | May.18        | May.22        | Jul.02  | Jul.12    | Jul.18  | Aug.23     | Aug.26   | Sep.14        |                |
and a 14-day window [22]. Half-hourly data and the CH₄ mole-fraction were thus calculated and time series standard deviation analysis of the CH₄ flux was performed using MS Excel 2013.

2.3. Meteorological measurements
An automatic weather observation system (A753 WS-X, Adcon, Vienna, Austria) was installed at 2.5 m. The soil temperature sensor (SM1) was installed at −5 cm. Photosynthetically active radiation (PAR) was observed by a PAR sensor (PAR1, Adcon). The main meteorological factors were monitored throughout the rice’s growth period to establish relationships between meteorological factors and CH₄ emissions.

3. Results

3.1. CH₄ flux near the surface of paddy fields
In 2017, the CH₄-release cycle ran from May 2nd (three-leaf stage) to September 12th (late-heading stage) for rice fields at the Sanjiang site. The CH₄ flux peaked at 0.52 g CH₄ m⁻² s⁻¹ on June 26th (rice tillering period; figure 2(A)). The rest of the time, CH₄ emissions were essentially zero. The annual CH₄ emission flux from paddy fields in Sanjiang Plain was 26.77 g CH₄ m⁻² year⁻¹, of which 79.57% was emitted in June and July.

The CH₄-release period at the Liaohe site was from April 18th (sowing date) to October 27th (the end of the growing season). Annual CH₄ emissions from paddy fields in Liaohe Plain were 16.17 g CH₄ m⁻² year⁻¹ (figure 2(A)), of which the highest CH₄ emissions occurred in July (34.78% of annual total). There was weak CH₄ absorption in winter. CH₄ emissions from Liaohe Plain were lower than those from Sanjiang Plain. Three peaks in the CH₄ flux were observed. These occurred during the early–transplanting stage, the jointing and booting stage and the mature stage. These peaks were in accordance with the results of Qingyu’s 2013 study [23].

The maximum CH₄ flux appeared at the jointing stage, reaching 0.33 μmol CH₄ m⁻² s⁻¹. CH₄ emissions were low

Table 2. Soil pH values and redox potentials in August.

| Station          | 0–20 cm  | 20–50 cm | mV    |
|------------------|----------|----------|-------|
| Liaohe plain     | 8.0–8.2  | 8.3–9.0  | 44.8–68.2 |
| Sanjiang plain   | 5.9–6.5  | 6.0–7.0  | 12.1–16.1 |

Figure 2. Methane fluxes to/from rice fields in 2017(A) and 2018 (B). Positive values indicate emissions while negative values indicate absorption.
during the tillering stage. The lowest CH$_4$ emissions were observed during the regeneration stage; this was different to the lowest-emitting stage in Sanjiang Plain.

The net fluxes of CH$_4$ in 2018 were similar to those in 2017. In 2018, the CH$_4$ emission fluxes from paddy fields were 32.76 and 20.51 g CH$_4$ m$^{-2}$ year$^{-1}$ in Sanjiang Plain and Liaohe Plain, respectively.

To determine the diurnal variation of the CH$_4$ fluxes during the different stages of the growing season, the data were divided into five stages, corresponding with observed changes in CH$_4$ emission data (figure 2). Stage A represented the time until soil thawing occurred (1–76 days) (figure 3(A)). Stage B covered soil thawing to seeding (77–104 days) (figure 3(B)). Stage C started after sowing and continued to the green period (105–141 days) (figure 3(C)). Stage D ran from reviving to the onset of booting (141–190 days) (figure 3(D)). Stage E covered the period from booting to the onset of milk ripening (191–234 days) (figure 3(E)). Stage F started at milky ripening and ran until the end of growing season (235–273 days) (figure 3(F)). Stage G (274–365 days) represented the period from the end of the growing season until the end of the year (figure 3(G)). Data for each stage were grouped according to the time interval. The stage’s half-hourly average was then calculated. The peak CH$_4$ fluxes (mean ± standard deviation) for each stage of the fallow period in Liaohe Plain were 0.12 ± 0.13, 0.17 ± 0.12 and 0.04 ± 0.07 μmol CH$_4$ m$^{-2}$ s$^{-1}$ (figure 3(G)) for stages A, B and G, respectively. During the growing season, the peak values were 0.15 ± 0.14, 0.19 ± 0.07 and 0.15 ± 0.24 μmol CH$_4$ m$^{-2}$ s$^{-1}$ for stages C, D and E, respectively. Different trends of diurnal (within day) variation of the CH$_4$ emissions flux from paddy fields were observed for different periods in 2017. Diurnal variations were obvious for Liaohe Plain for stages A–E (from the first day of measurements to the onset of milk ripening). Diurnal variations were very small during the fallow period (stages A, B and D). During this period, the CH$_4$ flux began increasing from 09:00, peaked at 13:00, and then fell back to a minimum around 15:00. Weak absorption of CH$_4$ during the night could offset daytime emissions in Liaohe Plain. In the growing season, the CH$_4$ flux began to increase at 08:00, peaked at 12:00–14:00, and fell to a minimum value by 17:00–18:00. There was little or no CH$_4$ emissions at night. The average CH$_4$ flux for the growing season was 0.065 μmol CH$_4$ m$^{-2}$ s$^{-1}$.

There were obvious diurnal variations during the rice’s growing period in Sanjiang Plain (figures 3(C)–(E)), but no diurnal variations were observed during the fallow period. The peak CH$_4$ flux was 0.07 ± 0.09, 0.43 ± 0.13 and 0.18 ± 0.11 μmol CH$_4$ m$^{-2}$ s$^{-1}$ (figure 3(E)) in stages C, D and E, respectively. The CH$_4$ flux began to increase at 08:00, peaked at 12:00–13:00, and decreased to a minimum value around 15:00–18:00. Night time emissions of CH$_4$ from the tillering to jointing stages were above 0.2 μmol CH$_4$ m$^{-2}$ s$^{-1}$, and were
above 0.1 $\mu$mol CH$_4$ m$^{-2}$ s$^{-1}$ from the booting to the onset of milk ripening stages. The average CH$_4$ flux for the growing period was 0.127 $\mu$mol CH$_4$ m$^{-2}$ s$^{-1}$ in Sanjiang Plain. As shown in figure 4, methane emissions were higher in the afternoon (when the maximum temperature was observed) than in the morning.

### 3.2. Driving factors

In 2017, the average air temperature at Sanjiang Plain Station was 3.43 °C, and the accumulated $\geq 10$ °C temperature (figure 4(A)) was 2782 °C. The average air temperature at Liaohe Plain Station was 10.16 °C, and the accumulated $\geq 10$ °C temperature was 3924 °C. These air temperatures were well suited to rice growth, with those in Sanjiang Plain more favourable than those in Liaohe Plain. The maximum air temperature of the two stations was observed from late June to July, corresponding to the peak in CH$_4$ emissions in Sanjiang Plain. The optimum air temperature for CH$_4$ production in the Northeast Plain was 25 °C – 30 °C. The cumulative precipitation at Sanjiang Plain Station was 587.9 mm (figure 4(B)), which was higher than that at Liaohe Plain Station (521.7 mm). The frequency of precipitation was also higher at Sanjiang Plain Station than that at Liaohe Plain Station.

The PAR was 296.86 and 400.86 W m$^{-2}$ year$^{-1}$ at Sanjiang Plain Station and at Liaohe Plain Station (figure 4(C)), respectively. The PAR peak was from late May to early June, which corresponds to the first peak in CH$_4$ emissions in Liaohe Plain but does not correspond to the first CH$_4$ emissions peak in Sanjiang Plain. There was a significant linear correlation between PAR and methane flux (figure 5). The average wind speed at Liaohe Plain Station was 12.23 m h$^{-1}$, which was higher than that at Sanjiang Plain Station (11.64 m h$^{-1}$) (figure 4(D)). In the tillering and jointing stage, the average wind speed was lower than that in the other stages; CH$_4$ emitted from rice during this stage tended to accumulate nearer to the ground.

The annual average temperature of the soil surface at Sanjiang Plain Station was 6.06 °C. The ground was frozen for 138 days, from January 1st to March 17th and from November 2nd to December 31st. The temperature was 4.53 °C lower than that at Liaohe Plain (10.59 °C) where the soil was frozen for less time (114 days); from January 1st to March 16th and from November 23rd to December 31st. Cold winter temperatures inhibit microbial activity across the large landmasses and very low CH$_4$ fluxes were observed from the paddy fields when the soil was frozen. CH$_4$ was absorbed at night.

The CH$_4$ emission flux was low or zero when the soil temperature was lower than 0 °C. The night-time temperature sensitivity in Sanjiang Plain was obviously greater than that during the daytime. The night-time temperature sensitivity followed an exponential relationship (figure 6). The temperature coefficient Q10 was calculated using the exponential equation. At night in stages C, D and E, the values of Q10 were 0.24, 0.17 and 0.09, respectively. Q10 was highest between the sowing and the green periods of the growing season (0 °C–20 °C). The CH$_4$ emission fluxes in Liaohe Plain were more sensitive to changes in temperature than those in

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**Figure 4.** Meteorological parameters through 2017. (A) Temperature, (B) precipitation, (C) PAR, (D) wind speed, (E) surface soil temperature and (F) soil heat flux.
Sanjiang Plain (figure 7). At night during these stages, the value of Q10 was 0.10. The emissions fluxes of CH$_4$ in stages C, D and E were generally below 0.2 μmol · CH$_4$ m$^{-2}$ s$^{-1}$ during the daytime and below 0.1 μmol CH$_4$ m$^{-2}$ s$^{-1}$ at night.

4. Discussion
Sanjiang Plain was affected by temperature changes. The most temperature-sensitive period occurred between sowing and the green period. The CH$_4$ flux in Sanjiang Plain during the tillering period had a distinct peak; this was consistent with observed temperature changes. Gas transport via the vascular bundle during rice’s rapid growth period and vigorous respiration during the tillering period contributed to CH$_4$ emissions. CH$_4$ emissions in Liaohe Plain were affected by the soil pH during in the growing season.
The rice in the plains of Northeast China was grown in the suitable temperature range (table 2), and the soil was reductive during the flooding period. The flooding periods of the two areas were similar but that in Sanjiang Plain occurred slightly earlier, which helped to accelerate the increase of soil temperature. The growth period in Liaohe Plain was one month longer than that of Sanjiang Plain. In the growing season, a thin water layer of 3–7 cm was maintained; any excess water following precipitation events was drained away. Water was drained from both fields once the rice was mature, and there were almost no CH$_4$ emissions after the fields had been drained. CH$_4$ emissions from the Sanjiang site were larger than those from the Liaohe site. Peak CH$_4$ emission fluxes in the afternoons were consistent with data reported for other rice fields [24–26]. During the afternoon, the increase in temperature and solar radiation enhance plant respiration and transpiration. This can promote the transfer of CH$_4$ through the plant to the atmosphere. These conditions can also accelerate the diffusion rate of CH$_4$ through the water layer and the rate at which CH$_4$ gas in the soil bubbles to the water’s surface. The higher afternoon temperatures that favour CH$_4$ emission through soil and plant pathways are the main reasons for the observed peak in CH$_4$ emissions at this time of day. These temperature-driven changes are the most
common cause of diurnal variation, as has been observed in other parts of the world. For example, Werle and Kormann (2001) observed maximum CH4 fluxes during rice’s initial development stages. Their results for the first half of the vegetative growth period were similar to those observed for Sanjiang Plain. The absorption of CH4 is related to microbial physical reactions and weather conditions. Specifically, in paddy fields, the absorption of CH4 is related to the activity of methylotrophs, a group of oxidizing bacteria. CH4 production is related to the growth period and peaked in Sanjiang Plain during rice’s tillering stage. This peak in CH4 emissions occurred during the rice’s rapid-growth stage owing to gas-exchange processes taking place in the rice’s vascular bundles. After the paddy fields had been flooded, sufficiently anaerobic conditions in the soil permitted a peak in the methanation of organic matter and rice stalks. Meanwhile, changes to the soil’s pH value during the growth period also affected the ability of rice to aerate the soil. Sim Jiangying et al (2006) found that a low pH value could induce the formation of aeration tissue within rice roots and increase the activity of ATPase. The low level of CH4 emissions during the tillering stage in Liaohe Plain was not consistent with results from Sanjiang Plain, indicating the presence of factors that restricted CH4 formation in this period. Methane production requires acidic conditions to functionalise its precursor materials, such as H2/CO2, acetic acid, formic acid, methanol and methylamine. In particular, the coenzyme F420 is an important contributor to the formation of CH4. F420 is a flavin mononucleotide analogue that is a low-molecular-weight (630) and fluorescent compound, which is held by methanogens. F420 is liable to be photolysed and inactivated under neutral or alkaline conditions. Thus, the activity of soil-borne CH4-oxidizing bacteria is inhibited by alkaline conditions. The pH value of the rice fields in Liaohe Plain was 8.0–9.0 (table 2). The alkalinity of the soil thus likely had an inhibitory effect on the precursor substances and on the production of CH4 by F420. This inhibiting effect was more apparent below a soil depth of 20 cm.

High levels of CH4 emissions were observed from Sanjiang Plain. The CH4 flux term must be considered in addition to emissions of CO2 to accurately describe the region’s annual net carbon balance. During November (the non-growing season at Liaohe Plain), absorbance of CH4 yielded a weak CH4 sink at this time. The beginning and end of net CH4 emission cycles at Sanjiang Plain and Liaohe Plain occurred in tandem with the irrigation and drainage cycles. CH4 production was also inhibited by lower temperatures. As the climate warms, the growing season of crops will shift, and methanation of organic matter deeper in the soil will be accelerated. Therefore, it is necessary to conduct longer-term measurements of CH4 fluxes in the rice regions of Northeast China. The effects of spring melting and ageing, winter-ice dissolution and rice growth on CH4 emissions should be more closely studied to better quantify the overall exchange of carbon in wetlands.

Figure 7. Soil temperature sensitivity of methane emissions in Liaohe Plain.
5. Conclusion

The EC method was used to measure the CH$_4$ flux in representative inland (Sanjiang Plain) and coastal, saline–alkaline (Liaohe Plain) paddy fields in Northeast China. The relationships developed from this analysis were used to explore how emissions varied across the rice-growing cycle. Data from two years of continuous measurements showed that CH$_4$ emission fluxes from Sanjiang Plain were greater than those from Liaohe Plain. The fluxes depended on the stage of the rice-growing process, the temperature, and the irrigation and drainage processes in place. For the same climatic conditions, CH$_4$ emissions from paddy fields in northern China vary depending on the rice’s growth period and the soil’s pH.

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Author Contributions

Qingyu Jia contributed to the conception and design, data analysis and interpretation, draft and revision of the manuscript. Wenying Yu collected the data. Li Zhou contributed to the draft of the manuscript. Rihong Wen, Yanbing Xie and Nina Chen contributed to the observation and analysis data.

Conflicts of Interest

The authors declare no conflict of interest.

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References

[1] WMO Statement on the status of the global climate in 2015, WMO, 2016, 1167
[2] Christen A et al 2013 Integral emission factors for methane determined using urban flux measurements and local-scale inverse models EGU General Assembly 15 6143
[3] WMO 2018 WDCGG data summary WDCGG No. 42. Volume IV—Greenhouse gases and other atmospheric gases Japan Meteorological Agency https://gaw.kishou.go.jp/static/publications/summary/sum42/sum42.pdf
[4] Ed Dlugokencky, NOAA/ESRL Annual Increase in Globally-Averaged Atmospheric Methane www.esrl.noaa.gov/gmd/ccgg/trends_ch4
[5] Dalsøren S B et al 2016 Atmospheric methane evolution the last 40 years Atmos. Chem. Phys. 16 3099–126
[6] Baldocchi D D 2003 Assessing the eddy covariance technique for evaluating carbon dioxide exchange rates of ecosystems: past, present and future Glob. Change Biol. 9 479–92
[7] Ehhalt D et al 2001 Atmospheric Chemistry and Greenhouse Gases Climate Change 2007: The Physical Science Basis. Contribution of Working Group I to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change ed W Robert et al (Cambridge, United Kingdom/New York, NY, USA: Cambridge University Press) pp 243–80
[8] Forster P et al 2007 Changes in atmospheric constituents and in radiative forcing Climate Change 2007: The Physical Science Basis. Contribution of Working Group I to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change ed S Solomon (Cambridge, United Kingdom/New York, NY, USA: Cambridge University Press) pp 131–243
[9] Hua X, Zucong C and Yagi K 2008 Methane production potentials of rice paddy soils and its affecting factors Acta Pedologica Sinica 45 89–104
[10] Ramesh R et al 1997 Anthropogenic forcing on methane eflux from polluted wetlands (adyar river) of Madras city, India Ambio 26 369–74 (https://www.jstor.org/stable/4314620)
[11] Zu-Chong C, Xu H and Ma J 2009 Methane and Nitrous Oxide Emissions from Rice-based Ecosystems (Hefei: University of Science and Technology of China Press)
[12] Xia J and Amp Y V 2014 Study on China’s Food Security and the ‘Three Rural Issues’ J. Shanxi Agri. Sc. 8 771–85
[13] Ruo-Tong L 2017 Impacts of climate change on crop production Chinese Agricultural Digest: Agriculture Engineering 29 25–8
[14] Xulan Z 2000 Influence of climate change on agriculture in Northeast China in recent 50 years J. Northeast Agric. Univ. 41 144–9
[15] Yu-He J and Guang-Sheng Z 2010 Transformation of vegetation structure in China’s Liaohe Delta during 1988–2006 Chin. J. Plan. Ecol. 33 359–67
[16] Chang-Chun S 2004 Advance in the studies on methane emission from wetlands *Ecology and Environment* **13** 69–73

[17] Guangsheng Z, Li Z, Enkai G and Fangwen Z 2006 Brief introduction of Panjin wetland ecosystem research station *J. Meteorol. Environ.* **22** 1–6

[18] Wen-Fu L et al 2012 Temperature variation of jiamusi in sanjiang plain *J. Anhui Agri. Sci.* **40** 437–40

[19] Xiao-Bo Qin et al 2013 The effect of soil oxygen availability on greenhouse gases emission in a double rice field *Acta Ecol Sin* (https://doi.org/10.5846/stxb201304110682)

[20] Baldocchi D D, Hincks B B and Meyers T P 1988 Measuring biosphere-atmosphere exchanges of biologically related gases with micrometeorological methods *Ecology* **69** 1331–40

[21] Kormann R and Meixner F X 2001 An analytical footprint model for non-neutral stratification *Boundary-Layer Meteor* **99** 207–24

[22] Sagerfors J, Lindroth A, Grelle A, Klemedtsson L, Weslien P and Nilsson M 2008 Annual CO₂ exchange between a nutrient-poor, minerotrophic, boreal mire and the atmosphere *J. Geophys. Res.* **113** G01001

[23] Qingyu J et al 2015 The characteristics of CH₄ concentration and flux of the near surface in Liaoh Delta rice region *Ecology and Environment* **24** 804–10

[24] Tseng K H, Tsai J L, Alagesan A, Tsuang B J, Yao M H and Kuo P H 2010 Determination of methane and carbon dioxide fluxes during the rice maturity period in Taiwan by combining profile and eddy covariance measurements *Agric. For. Meteorol.* **150** 852–9

[25] Miyata A, Leuning R, Denmead O T, Kim J and Harazono Y 2000 Carbon dioxide and methane fluxes from an intermittently flooded paddy field *Agric. For. Meteorol.* **102** 287–303

[26] Neue H U, Wassmann R, Kludze H K, Bujun W and Lantin R S 1997 Factors and processes controlling methane emissions from rice fields *Nutr. Cycl. Agroecosyst.* **49** 111–7

[27] Werle P and Kormann R 2001 Fast chemical sensor for eddy correlation measurements of methane emissions from rice paddy fields *Appl. Opt.* **40** 846–58

[28] Steinkamp R, Butterbach-Bahl K and Papen H 2001 Methane oxidation by soil of an N limited and N fertilized spruce forest in the Black Forest, Germany *Soil Biol. Biochem.* **33** 145–53

[29] Jiang-ying S, Xiao-li W and Ke F 2006 Effects of medium pH and N forms on aerenchyma formation in roots of different rice genotypes *Plant. Nutr. Soil. Sc.* **12** 193–200

[30] Yan F et al 1998 Adaptation of active proton-pumping and plasmalemma ATPase activity of corn roots to low root medium pH *Plant Physiol.* **117** 311–9

[31] Hua S, Maoping H and Mingcheng H 2015 Impact of global climatic warming on agricultural production in china *Chinese Journal of Agricultural Resources and Regional Planning* **36** 51–7